Studies in Environmental Science 44

HIGHWAY POLLUTION

Edited by
Ronald S. Hamilton
Roy M. Harrison

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FOREWORD

Long before "green" issues became a subject of widespread public concern, both of us were heavily involved in research relating to pollutants in the highway environment. This led in 1983 to the First International Symposium on Highway Pollution which attracted some 60 participants from 10 countries. Following on the success of this meeting, we jointly organised the Second and Third Symposia, held in London and Munich respectively, each of which grew in size relative to its predecessor. The success of these meetings has led us to the view that the subject of Highway Pollution is a coherent interdisciplinary sub–division of environmental science which engages the interest of a substantial research community.

The proceedings of the three conferences have been published respectively as volumes 33, 59, and 93 of The Science of the Total Environment. These provide individual snapshots of developing research projects, but do not present a coherent overview. We therefore perceived a need for a review volume to present the current state of knowledge and to put recent research results into context. We consequently invited some of the leading international authorities in the field to contribute. Through their input, we have, we believe, addressed the major issues of the causes, effects and control of pollution of the highway environment.

We are grateful to Mike Revitt and Bryan Ellis for advice and helpful comments. May George and Carl Kilroy took on the task of translating all the inputs into a common wordprocessing language and without their assistance we could not have produced this book in its present format.

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CONTENTS

Chapter Page

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Chapter 1

THE INFLUENCE OF HIGHWAY–RELATED POLLUTANTS ON ENVIRONMENTAL QUALITY

D.J. Ball, R.S. Hamilton and R.M. Harrison

Contents **Page**

1 EMISSIONS AND LEVELS

1.1 A Historical Perspective

The importance of highway–related pollutants has increased dramatically in the last forty years. Pollution of the environment is not new but it is only in recent years that transportation has made a major contribution. Since the Industrial Revolution, there has been an increase in the quantity of waste products generated by industry and society and current estimates are that a city of one million inhabitants in an industrialised country consumes daily 600,000 tons of water, 10,000 tons of fuel and 2,000 tons of food, leading to a daily production of 500,000 tons of sewage, 2,000 tons of refuse and 1,000 tons of air pollutants [1]. These emissions led to a deterioration of the environment, particularly the urban environment, for generations, but it was not until clear, unambiguous evidence linking exposure to pollution with adverse health effects that steps were taken to impose control procedures. The most infamous incident was the 1952 London smog, when levels of about 1 mg smoke m^{-3} and 2 mg SO_2 m^{-3} existed for four continuous days in England's capital city and were linked with up to 4,000 premature deaths. Control procedures involving technology and legislation have contributed to the reduction of emissions, particularly industrial and domestic emissions, in industrialised countries over the last forty years. Figure 1.1 shows the fall in smoke levels in the UK atmosphere since that period.

At the same time as industrial and domestic emissions have reduced, motor vehicle use has increased rapidly with the result that transportation is now a major source of pollution, particularly in the urban environment. Figure 1.2 shows the rate at which vehicle use is increasing. In 1950, there were less than 50 million cars worldwide and 85% of those cars were in the USA. Between 1950 and 1990, the USA car population increased by a factor of 3 and the worldwide car population increased by a factor of 8. Major population increases, particularly in Asia and South America, will probably be accompanied by increased urbanisation as social factors force the majority of the increased population into cities and away from rural areas. The overall effect of these trends is likely to be an increase in the number of cities experiencing major traffic–related pollution problems.

Air pollution is the most recent of traffic–related problems to be recognised. The adverse health effects of Pb, particularly on young children, only began to be recognised in the 1970s.

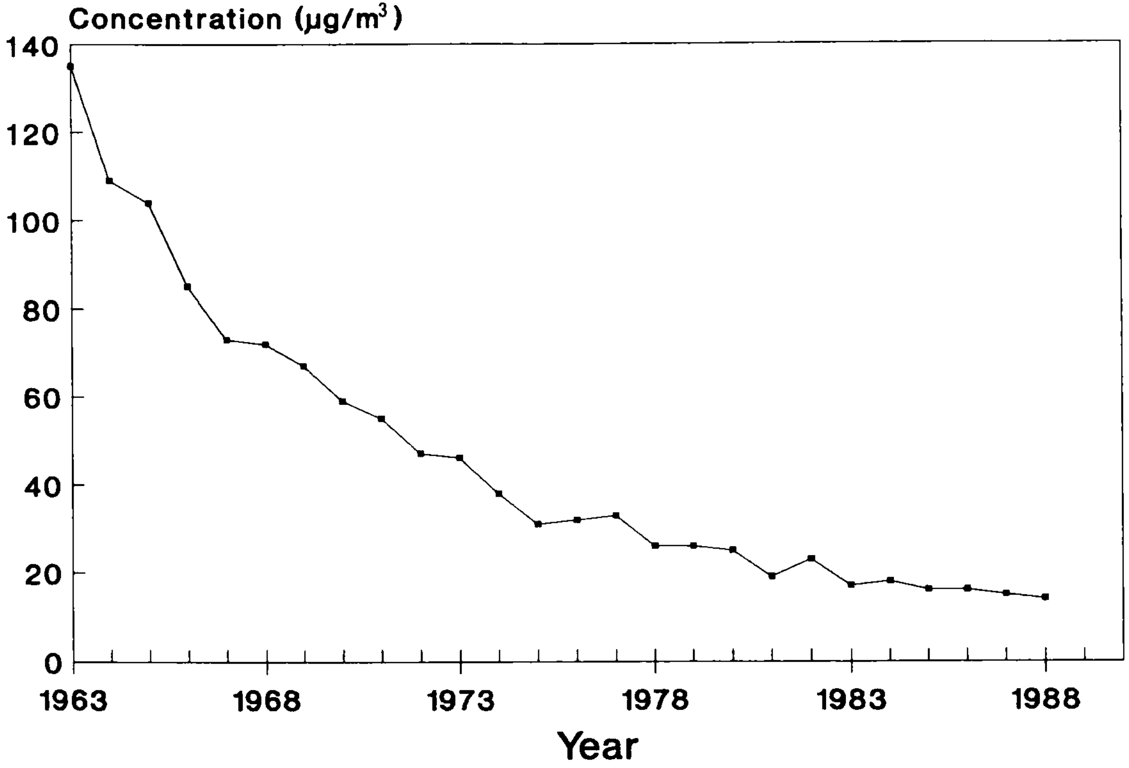
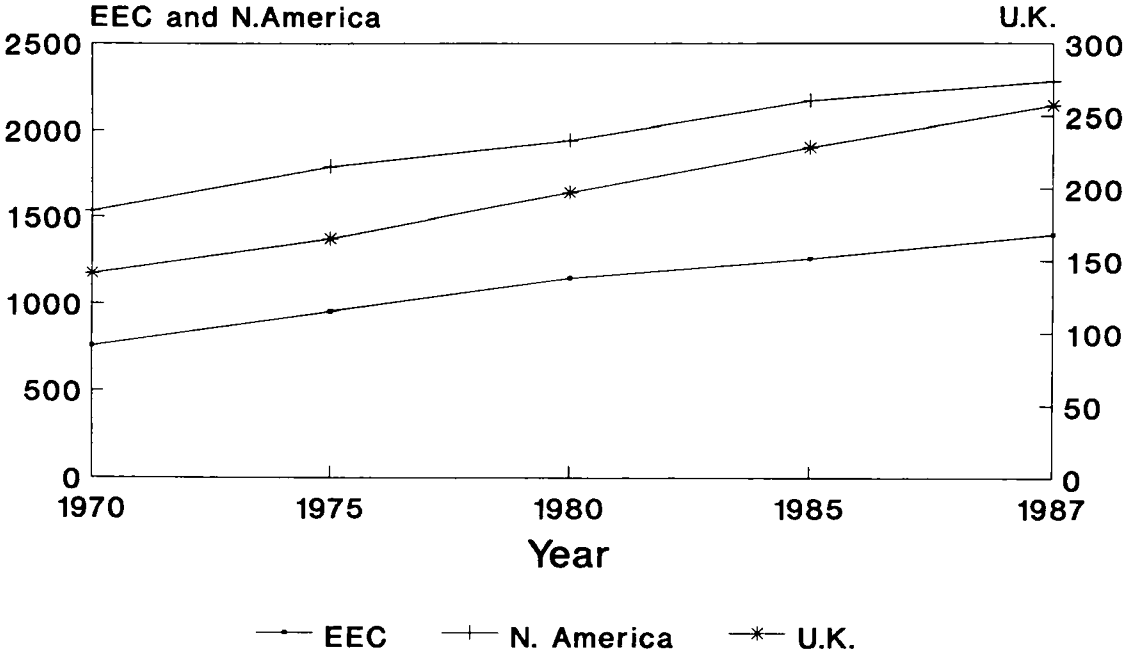


FIGURE 1.1 *U.K. AVERAGE URBAN SMOKE CONCENTRATIONS*



Units: billion vehicle-km

FIGURE 1.2 *ROAD TRAFFIC VOLUMES (passenger cars)*

More recently, vehicle emissions have been implicated in a range of health effects (see Chapter 7) and also in acid deposition, photochemical smog, global climatic change and the soiling of surfaces. The deposition of these vehicle–emitted pollutants has implications for ecosystems; this is reviewed in Chapter 8. Similarly, highway runoff has only recently been recognised as an important constituent of urban, non–point source pollution. Some studies have suggested that even though highways may only occupy 5–8% of the urban catchment area, highway drainage can contribute as much as 50% of the total suspended solids, 16% of the total hydrocarbons and 75% of the total metal inputs to a receiving stream [2]. These loadings are a result of both the pollutant input to the highway surface and modifications to local hydrology imposed by the design and construction of the highway (Chapters 4 and 5). Finally, noise from road traffic has been identified as a nuisance to a large proportion of the population (Chapter 9).

From this brief review, it is clear that highway sources are involved in pollution of the atmosphere, surface dust, soil and water (including rainfall and runoff). Material and acoustic forms of pollution are involved, both may exert adverse effects on the living and non–living environment and these effects are expected to increase in the years ahead. The aim of this book is to examine and quantify these pollutant sources, their transport through the environment, the monitoring of their levels, and their effects. Finally, the potential for technological and legal control is considered.

1.2 Sources, Transport and Levels

The levels of pollutants at any point in the environment are determined by:

i) source emission rates,
ii) dispersion characteristics,
iii) removal rates.

Emissions to the atmosphere near highways are mainly from the vehicle exhaust. This source is responsible for virtually all of the carbon monoxide, nitrogen oxides and lead compounds emitted. Most of the dark smoke and about 65% of the hydrocarbons are also from this source. The remainder of the hydrocarbons are emitted from crankcase blowby (about 20%), and evaporation from the fuel tank and carburettor (about 15%). Hydrocarbon data is generally presented as total hydrocarbon, specific or speciated hydrocarbons, or volatile organic compounds (VOCs). VOCs are non–methane hydrocarbons and other organic compounds including oxygenated and halogenated organics. Dark smoke is associated with diesel exhaust emissions, though resuspension by passing vehicles also makes a contribution to airborne particulate matter. The wear of automotive components and the corrosion of bodywork also contributes to atmospheric particulate loadings, particularly heavy metal inputs. The abrasion of tyres and brake linings gives rise to emissions of Zn, Cd, Cu, Ni, Cr and Fe. These emission rates can be quantified, though in some situations and for some pollutants there is substantial uncertainty in the values. This is discussed later in this section.

Deposition of these pollutants near the highway results in contamination of surface dust and soil. In addition, leakages of brake fluids, anti–freeze compounds, transmission box lubricating oils, engine oil and greases result in a direct pollutant input to the highway surface. Also, abrasion particles from the highway surface are produced by the passage of vehicles and this increases the dust loading on the surface. Resuspension processes result in a redistribution of dust on and near the highway; these processes are considered in detail in Chapter 4. Additional pollutant removal processes include washoff following rainfall and street cleaning practices.

A balance in the chain of events of emission → dispersion → deposition → removal, produces an environmental level of pollutant. However, each of these processes varies in space and time and as a result, pollutant levels also exhibit these variations. Table 1.1 records "typical" pollutant levels in the urban or highway environment, but the variations must be borne in mind before applying these values. A coefficient of variance (standard deviation ÷ mean) of 0.3 is common and 90% of recorded values might be expected to lie within a factor of 2 of the "typical" value. For trace constituents, greater variation can be expected. Pollutant levels in dust samples frequently exhibit variations by a factor of ten.

Modelling the transport processes which relate emissions, dispersion, deposition and removal are considered in later chapters of this book. A prerequisite for a good model is accurate emission rate data. For individual sources, particularly when dealing with a stationary source, this information can be obtained by monitoring. The methodology for vehicle emission monitoring is reviewed in Chapter 2. For large area and line sources, emission inventories are employed. These inventories rely on the use of emission factors. The emission factor for an activity is defined as the mass of pollution released to the *environment per unit (kg for solids, m^3 or litres for liquids) of fuel consumed.* For vehicle–generated pollution, the rate of pollutant generation is also a function of fuel consumption rate and this is combined with the emission factor to give the emission rate in

TABLE 1.1 Pollutant Levels in the Highway Environment

(Typical values are quoted but it must be remembered that levels exhibit strong spatial and temporal variations)

POLLUTANT	TYPICAL LEVEL	REFERENCE
Particulate Matter		
total airborne suspended particulates	100 μg m^{-3}	3
atmospheric dark smoke	30 μg m^{-3}	4, 5
total suspended solids in runoff	100 g m^{-3}	2
surface dust	50 g m^{-2}	6
airborne particulate elemental carbon	3 μg m^{-3}	4
Gaseous Atmospheric Pollutants		
CO	2 mg m^{-3} (1.7 ppm)	3
SO_2	60μg m^{-3} (22 ppb)	3
NO_2	50μg m^{-3} (25 ppb)	3,5
NO_x	150μg m^{-3} (100 ppb)	3,5
total hydrocarbons	1500μg m^{-3}	7
VOC	500 μg m^{-3}	7
Heavy Metals		
Pb in air	0.3 μg m^{-3}	8
Pb in dust	350μg g^{-1}, 20 mg m^{-2}	8
Pb in stormwater	100mg m^{-3}	8
Zn in air	0.1 μg m^{-3}	8
Zn in dust	350μg g^{-1}, 20 mg m^{-2}	8
Zn in stormwater	100mg m^{-3}	8
Cd in air	0.0010 μg m^{-3}	8
Cd in dust	1 μg g^{-1}, 0.1 mg m^{-2}	8
Cd in stormwater	2 mg m^{-3}	8
Cu in air	0.01 μg m^{-3}	8
Cu in dust	60 μg g^{-1}, 3 mg m^{-2}	8
Cu in stormwater	50mg m^{-3}	8
Polyaromatic Hydrocarbons		
benzo (a) pyrene in air	2 ng m^{-3}	9,10
benzo (a) pyrene in dust	0.5 μg g^{-1}	11
benzo (a) pyrene in runoff	0.1mg m^{-3}	12
fluoranthene in air	10 ng m^{-3}	10
fluoranthene in dust	1.1 μg g^{-11}	11
fluoranthene in runoff	0.3 mg m^{-3}	12

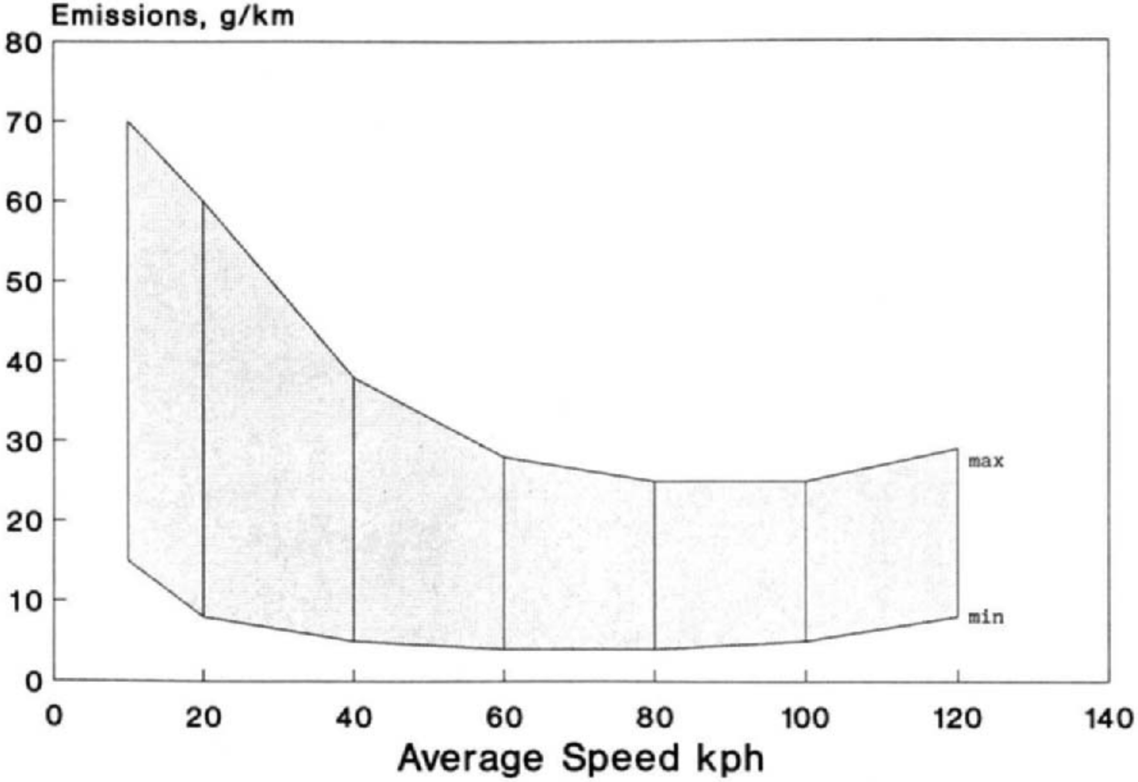


FIGURE 1.3 *VARIATION OF CO EMISSIONS WITH SPEED*

the units of mass per unit distance driven, g km^{-1}. Emission factors for pollutants in exhaust and evaporative emissions can be determined accurately by employing the laboratory and on–the–road techniques described in Chapter 2, but the factors show substantial dependence on engine size and driving mode. Figure 1.3 shows the variation observed for CO. Some dispersion models require an input of the vehicle type and range of vehicle speeds on the highway under consideration and calculate the emissions generated within each increment [13]. Most use a single value to cover all vehicle types and speeds, e.g. 10 g km^{-1} for CO. Pollutant generation by wear and abrasion cannot be accurately measured and has been inferred from mass loss estimates. As a result, there are major uncertainties in the emission factors for these pollutants. Values of emission rates for highway sources are recorded in Table 1.2.

TABLE 1.2 Emission Rates for Pollutants from Highway Sources (without control technology fitted)

	Emission Rate (g km^{-1} vehicle^{-1})		
POLLUTANT	GASOLINE	DIESEL	Reference
CO	10	1	5
total hydrocarbons	1	0.3	5
NO_x	3	6	5
SO_2	0.03	0.2	5
smoke	0.1	0.4	4,5
dark smoke	0.04	1.2	4,5
Pb (assuming 0.15g Pb/ℓ petrol)	0.01	0	14
PEC	0.001	0.13	4
benzo(a)pyrene	7×10^{-7}	2×10^{-6}	15
fluoranthene	2×10^{-5}	4×10^{-5}	15
Zn	0.003 (total)		8
Cd	1×10^{-6} (total)		8,15
Cu	4.5×10^{-5} (total)		8,15

The difference between "total suspended particulate matter", "smoke" and "dark smoke" or "black smoke" should be clarified. The following terminology is normally employed:

total suspended particulate matter (TSP): all airborne particulate matter measured gravimetrically;

smoke: the mass of $< 15\ \mu m$ particulate matter, emitted during a combustion process;

dark or black smoke: the level of particulate matter estimated by the smoke shade technique and therefore a function of both the mass and optical properties of smoke.

PM_{10} : particulate matter $\leq 10\ \mu m$ diameter, the basis of the primary standard in the United States since 1983.

1.3 Primary Air Pollutants

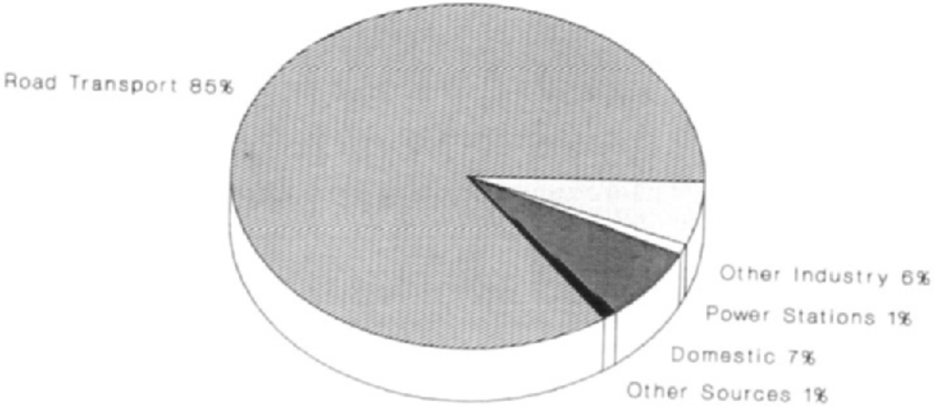
The pollutants emitted directly from motor vehicles are termed primary. Most of the primary vehicle–emitted pollutants are transformed in the atmosphere to other secondary pollutants, and these are considered in the next section of this chapter. The main primary pollutants are carbon monoxide, carbon dioxide, total hydrocarbons/VOCs, nitrogen oxides, dark smoke, lead and particulate elemental carbon (PEC). While vehicles are a major source of these pollutants, they are not the only source and the application of the emission inventory approach permits the quantification of the contributions by each type of source. Figure 1.4 shows the source contributions for some primary pollutants in the UK, based on emission data [16]. Data on total emissions are presented in Table 1.3.

TABLE 1.3 Total Anthropogenic Emissions of Some Primary Pollutants (tonnes)

	Global (ref. 16,17)	OECD Europe (ref. 19)	UK (ref.16,18)
CO	9×10^8	not reported	5.5×10^6
VOC	2.4×10^8	9.2×10^6	1.8×10^6
NO_x (NO_2 equivalent)	1.2×10^8	1.2×10^7	2.5×10^6

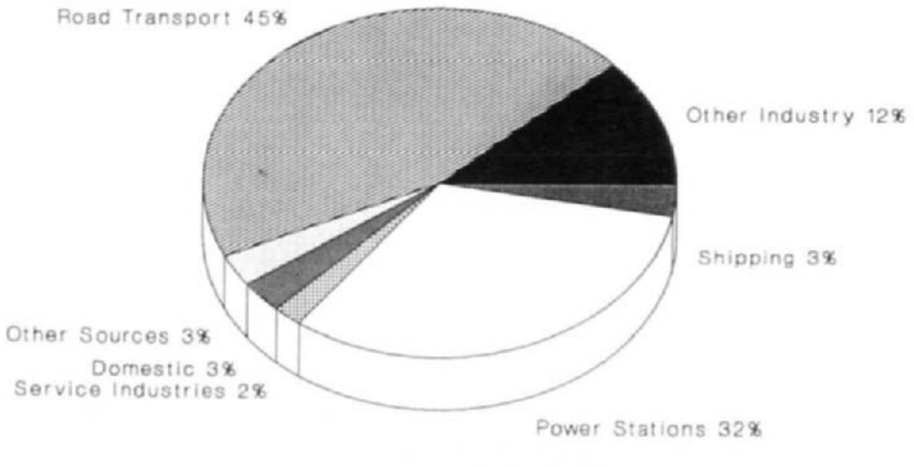
Transportation is responsible for approximately 85% of CO emissions, 40% of nitrogen oxides emissions and 25% of VOC emissions in the UK. In the USA, [20], the equivalent figures are 66%, 43% and 48%. Carbon monoxide and VOCs are the pollutants present at highest concentrations and it is for this reason that transportation is generally considered responsible for about 50% of current air pollution.

The growth in consumption of leaded gasoline and its recent rapid fall has recently been documented by Nriagu [14]. The trend is shown in Figure 1.5. The antiknock properties of organolead compounds were first observed during laboratory tests in 1921 and tetraethyllead additives were introduced into petrol for public sale two years later. An additional benefit of the lead additive was its ability to act as a valve lubricant. The development of a tetramethyllead additive extended the perceived benefits of this approach



Total = 5.51 million tonnes CO

FIGURE 1.4(a) *EMISSIONS OF PRIMARY POLLUTANTS, CO, U.K. 1988*



Total = 2.48 million tonnes NO_X

FIGURE 1.4(b) *EMISSIONS OF PRIMARY POLLUTANTS, NO_x, U.K. 1988*

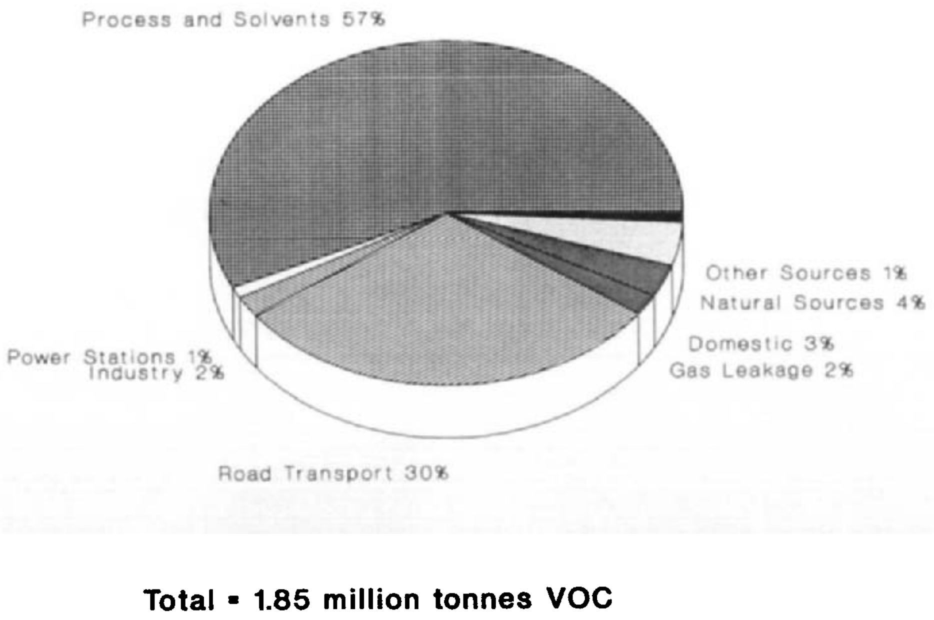
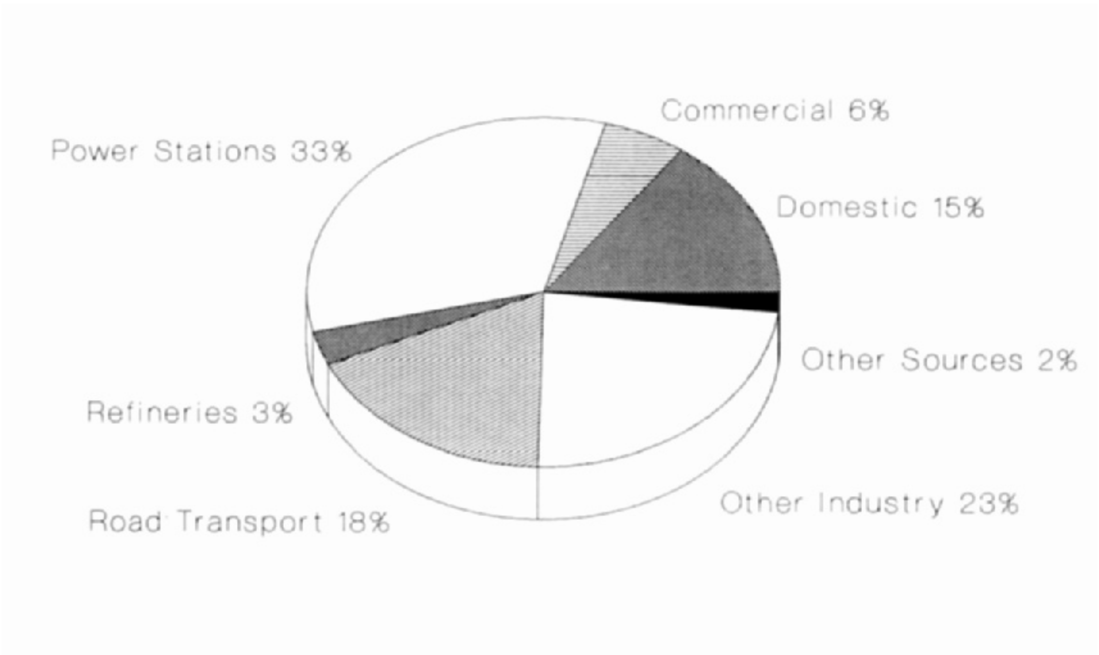
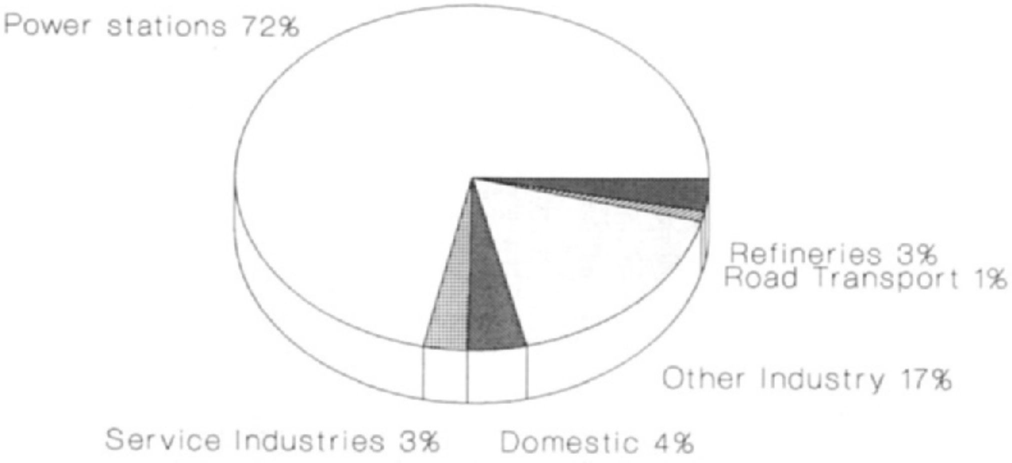


FIGURE 1.4(c) *EMISSIONS OF PRIMARY POLLUTANTS, VOCs, U.K. 1988*



TOTAL = 158 MILLION TONNES CO_2

FIGURE 1.4(d) *EMISSIONS OF PRIMARY POLLUTANTS, CO_2, U.K. 1988*



Total = 3.58 million tonnes SO_2

FIGURE 1.4(e) *EMISSIONS OF PRIMARY POLLUTANTS, SO_2, U.K. 1988* to improving engine performance. However, scientific evidence in the late 1960s eventually led to widespread concern about the effects to children of automobile Pb exposure. This concern led to legislation reducing the level of Pb in gasoline. Also, the introduction of catalytic converters to reduce emissions of other primary pollutants requires that the petrol is free of Pb which would otherwise contaminate the precious metals in the converter.

The overall effect of these developments was a reduction in the quantity of Pb emitted from vehicle exhausts and a rapid, approximately proportional decrease in the level of airborne Pb. Levels in most major cities have fallen from about $2\mu g$ m^{-3} to about $0.3\mu g$ m^{-3}. A comparable reduction is anticipated to occur with Pb levels in dust, soil, vegetation and runoff but will take a longer time to materialise because of the relatively slow removal processes.

The first section of this chapter mentioned the changes in atmospheric smoke levels which have occurred since 1952 and the London smog. At that time, emissions were due mainly to the combustion of coal in domestic premises. These were subsequently controlled by technology and legislation although changing fuel use patterns were also influential. Changes in emission patterns can be clarified by employing the emission factor and emission inventory approach. Emission factors were recorded in Table 1.2. For smoke, the emission factor for diesel engines is about four times that for gasoline engines. On a mass–for–mass basis, diesel smoke is about 7.5 times darker than gasoline smoke with the result that the dark smoke emission factor for diesel engines is thirty times that for gasoline engines [4]. An emission inventory for London in 1980, using these factors, is shown in Table 1.4 and the results show that diesel emissions were

TABLE 1.4 **The quantities of smoke and dark smoke emitted in London (1980) according to fuel type (based reference 21)**

Fuel Type	**Amount consumed (tonnes)**	**Approx. soiling factor relative to coal**	**Emission Factor % by mass**		**Smoke emitted (tonnes)**	**Dark smoke emitted (tonnes)**
			smoke	**dark smoke**		
Coal (domestic)	5 x 10^4	1.0	3.5	3.5	1750	1750
Coal (industrial)	2.5 x 10^5	1.0	0.25	0.25	625	625
Solid smokeless	3.5 x 10^5	1.0	0.56	0.56	1960	1960
Gas oil	1 x 10^6	1.55	0.025	0.039	250	387
Fuel oil	6 x 10^5	0.43	0.10	0.043	600	258
Motor spirit	2.2 x 10^6	0.43	0.15	0.065	3300	1419
Diesel fuel	5.2 x 10^5	3.0	0.60	1.8	3120	9360

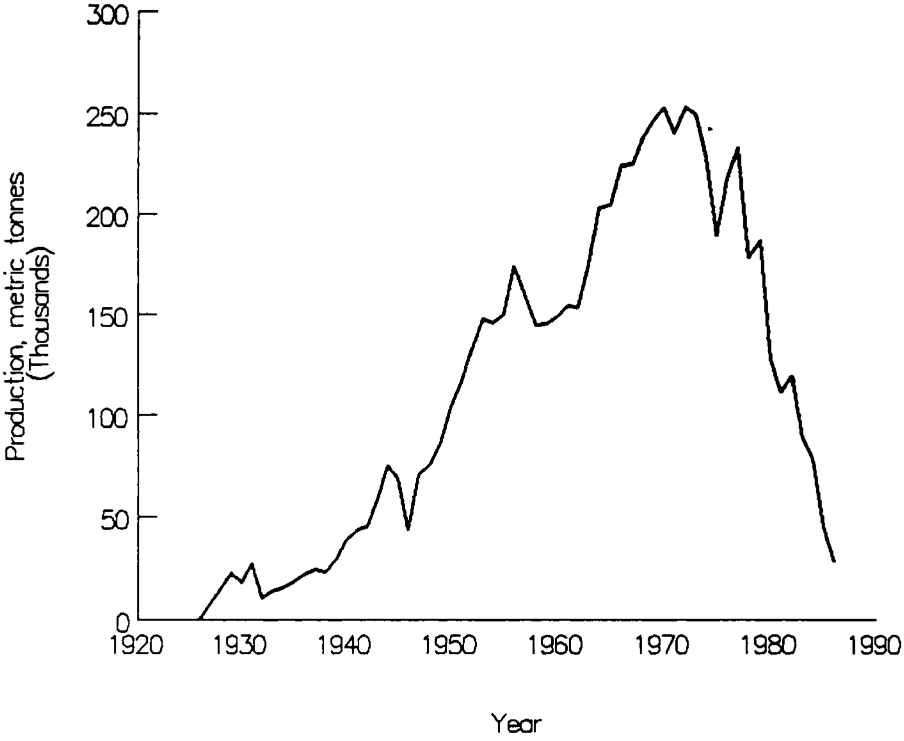


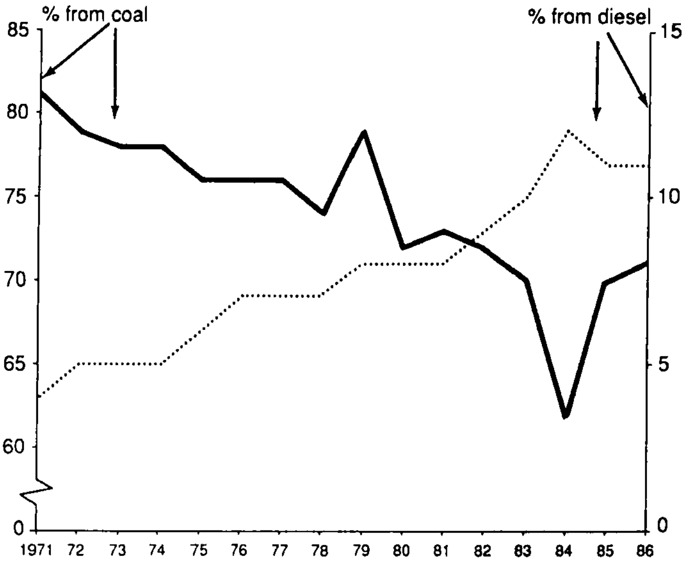
FIGURE 1.5 *HISTORIC TRENDS IN THE CONSUMPTION OF LEADED PETROL U.S.A.* (*ref. 14*)

responsible for about 60% of the dark smoke in the city in that year. The same approach has been applied to UK emissions [4, 22] and the results are presented in Figure 1.6. On the basis of these results, diesel emissions are responsible for 12% of smoke and 30% of dark smoke emissions in the United Kingdom.

The reason for the darkness of diesel smoke is the presence of particulate elemental carbon (PEC) in the emissions. The sources and transport of PEC and its potential importance as an environmental pollutant have recently been reviewed [4, 23]. Its emission factors for gasoline and diesel sources were recorded in Table 1.2 and an emission inventory using these factors and values for other combustion processes showed that diesel emissions were responsible for between 85% and 95% PEC emissions for all countries in Western Europe. Values for London, 1980, are presented in Table 1.5. PEC is generally considered responsible for more than 90% of light absorption and about 30% of visibility reduction in the atmosphere [4].

In recent years, the contribution of transportation sources to greenhouse gas emissions has received considerable attention because of concern over climatic changes [24, 25]. The relative importance of the various pollutant greenhouse gases is shown in Figure 1.7. The contribution of traffic sources to carbon dioxide has been quantified using the emission inventory approach and results for the UK [24] were presented in Figure 1.4. These results indicate that vehicles are responsible for 14% of the carbon dioxide emissions. The equivalent figure for the USA is 24% [25], and on the global scale, a value of 15% has been estimated. Contributions from vehicle sources to other greenhouse gases are less well known. The contribution to methane emissions is less than 1%. To date, little progress has been made in assessing the influence of vehicle emissions on nitrous oxide and tropospheric ozone concentrations because of lack of knowledge of nitrous oxide emission factors and the complexity of the chemical reactions involving hydrocarbons, nitrogen oxides and carbon monoxide in ozone formation. On the basis of the best available information, it appears that transportation sources are responsible for between 8% and 10% of greenhouse gas levels.

(a) Smoke



(b) Dark smoke

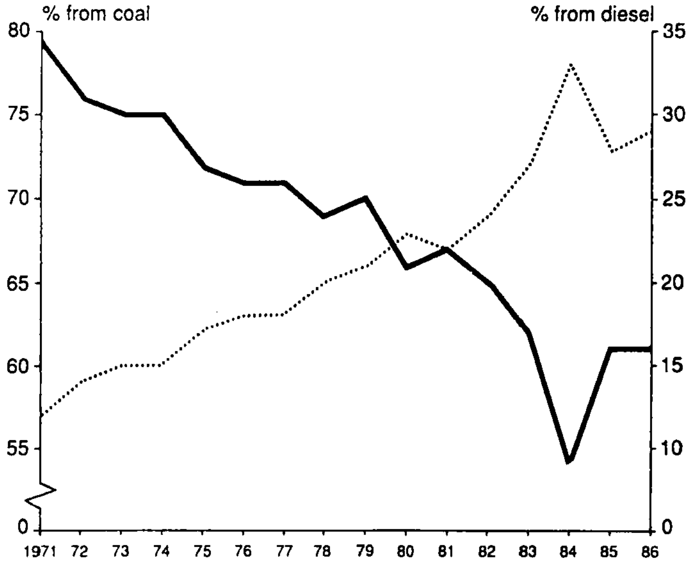


FIGURE 1.6 *SOURCES OF SMOKE AND DARK SMOKE IN THE U.K.*

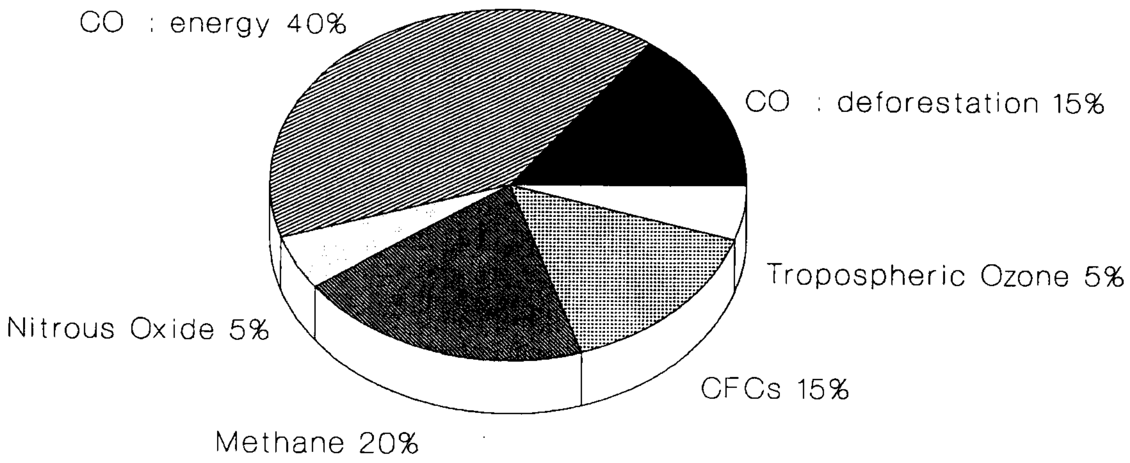


FIGURE 1.7 *CAUSES OF GLOBAL WARMING*

TABLE 1.5 PEC Emissions, London 1980

FUEL TYPE	Amount of fuel consumed (10^3 tonnes)	PEC emission factor (% by mass)	Amount of PEC factor (tonnes)	% of PEC Total by fuel type
Coal: domestic	50	0.0001	0.05	0.004%
Coal: industrial	250	0.0001	0.25	0.02%
Solid smokeless	350	0.0001	0.35	0.03%
Gas oil	1000	0.01	0.1	0.009%
Fuel oil	600	0.002	12	1%
Petrol	2200	0.002	44	4%
Diesel	520	0.2	1040	94.8%

2 SECONDARY POLLUTANTS

2.1 Secondary Pollutant Formation

The problem of secondary pollutants first came to light in Los Angeles in the 1950s. A "smog" bearing some similarity, but showing many differences from the London wintertime smogs was observed with growing frequency. The most obvious symptom was visibility degradation, hence the term smog (literally smoke + fog) was applied. Table 1.6 compares the two types of smog and exemplifies the differences between the two phenomena. The Los Angeles smog, now more generally known as a photochemical smog, is now known to be prevalent in areas with high sunlight intensities and high levels of hydrocarbon and oxides of nitrogen pollutants, and is thus commonly associated with regions such as the Los Angeles Basin and Mexico City. Its presence has also been established in higher latitude regions such as western Europe, much of which may be affected simultaneously during a single episode [26].

TABLE 1.6 Characteristics of London* and Los Angeles smogs

London – type	Los Angeles – type
Low temperatures (0–5°)	High temperatures (25–35°)
Fog formation	Clear sky
Surface–based (radiation) temperature inversion	Overhead (subsidence) inversion
Chemically reducing (due to SO_2)	Chemically oxidising (due to ozone etc.)
Causes respiratory irritation (due to smoke + SO_2)	Causes respiratory and eye irritation

**Readers should note that the London–type smog no longer occurs in London.*

There are both chemical and meteorological requirements for formation of appreciable photochemical pollution. The meteorological factors are:

a) a restriction on vertical mixing, normally by a subsidence inversion associated with an anticyclone. This permits good daytime mixing close to the ground, but with a stable layer at 500–1000 metres restricting exchange with air at higher levels;

b) high intensity sunshine, again typically associated with an anticyclone.

When the above meteorological factors are combined with high emissions of precursor pollutants, most notably oxides of nitrogen and hydrocarbons, photochemical air pollution develops. The most commonly used indicator of smog formation is the concentration of ozone. Mid–latitude northern hemisphere ozone concentrations at ground–level in the absence of appreciable regional or local photochemistry are typically around 10–50 ppb (1ppb = 10^{-9} by volume).

Concentrations in excess of 80 ppb are normally taken as indicative of photochemical air pollution, although other causes such as enhanced stratospheric intrusions are sometimes responsible. In 1988, ozone concentrations in southern California exceeded the US ambient air quality standard of 120 ppb on 172 days. In comparison, UK levels above 120 ppb are comparatively rare; over the period 1977–1983 ground–level ozone at one site exceeded 80 ppb on 4.2% of days (i.e. about 15 days per year) with 3.9% attributed to photochemical pollution and 0.3% to enhanced stratospheric intrusions. The comparable figures for days exceeding 100 ppb were 1.5% total (i.e. about 5 days per year) of which 1.4% were due to photochemical pollution [27].

Secondary pollutant formation processes have two essential starting points : photolysis of ozone and of nitrogen dioxide – the former is responsible for formation of the hydroxyl radical and in polluted atmospheres the latter gives rise to a build–up of ozone and other secondary pollutants. The hydroxyl radical (OH) is a species central to much atmospheric chemistry, which, due to its high reactivity, is responsible for the breakdown of many pollutant species and consequently for formation of many secondary pollutants. Ozone photolysis by sunlight ($h\nu$) can lead to the formation of excited state oxygen atoms, termed $O(^1D)$ which then react with water vapour to form hydroxyl:

$$O_3 + h\nu \rightarrow O_2 + O\,(^1D) \quad \lambda < 315\ \text{nm} \qquad (1)$$

$$O\,(^1D) + H_2O \rightarrow 2OH \qquad (2)$$

Photolysis of nitrogen dioxide NO_2 by sunlight leads to formation of ground–state oxygen atoms, termed $O(^3P)$, which react with oxygen molecules, O_2, in the presence of a third body reactant (termed M) to form ozone:

$$NO_2 + h\nu \rightarrow NO + O(^3P) \qquad (3)$$

$$O(^3P) + O_2 + M \rightarrow O_3 + M \qquad (4)$$

The other product of the NO_2 photolysis is nitrogen oxide (also known as nitric oxide), NO, which reacts rapidly with ozone to reform NO_2:

$$O_3 + NO \rightarrow NO_2 + O_2 \qquad (5)$$

The overall effect is to produce a small steady–state ozone concentration which can be predicted by equating the rate of reaction (3) which removes NO_2 with the rate of reaction (5) which reforms it.

$$\text{Rate of } NO_2 \text{ loss} = \text{Rate of } NO_2 \text{ production}$$

$$k_3\,[NO_2] = k_5\,[NO]\,[O_3]$$

$$[O_3] = \frac{k_3}{k_5}\,\frac{[NO_2]}{[NO]}$$

Square brackets indicate concentrations and k_3 and k_5 are the rate constants of reactions 3 and 5 respectively. Whilst k_5 may be considered as a constant over small ranges of temperature, k_3 is directly related to sunlight intensity and thus has a maximum on a clear day at midday, falling to zero at night.

As indicated above, elevated concentrations of ozone are formed in photochemical air pollution. This requires not only a high value of k_3 (consistent with sunny anticyclonic conditions) but also a high ratio of NO_2:NO). The simple $NO/NO_2/O_3/O_2$ system will not produce the latter. This is where hydrocarbons and carbon monoxide play an important role. Whilst CO is much less important than hydrocarbons in this regard, its chemistry is less complex and it will be used initially to explain the processes occurring. The hydroxyl radical (OH) is capable of oxidising CO to CO_2. The hydrogen atom (H) also formed can react with oxygen molecules to form the hydroperoxy radical, HO_2.

$$CO + OH \rightarrow CO_2 + H \quad (6)$$
$$H + O_2 \rightarrow HO_2 \quad (7)$$

This radical can react with nitrogen oxide.

$$HO_2 + NO \rightarrow NO_2 + OH \quad (8)$$

In doing so, it not only regenerates the OH radical, but also converts NO to NO_2 without directly consuming ozone (that consumed by ozone photolysis to produce OH is insignificant as OH is regenerated in these essentially catalytic cycles. Once a high NO_2:NO ratio exists, a high ozone concentration is predicted in the presence of strong sunlight (high k_3).

More important than CO in enhancing NO_2:NO ratios are hydrocarbons. The most abundant atmospheric hydrocarbon, methane (CH_4) is rather unreactive. It does not play a major role in the polluted urban atmosphere, but contributes to the formation of an ozone background at northern hemisphere sites. An example of hydrocarbon chemistry can be provided by butane, C_4H_{10}; the initial reaction is with OH:

$$CH_3(CH_2)_2CH_3 + OH \rightarrow CH_3(CH_2)_2CH_2 + H_2O \quad (9)$$
$$CH_3(CH_2)_2CH_2 + O_2 \rightarrow CH_3(CH_2)_2CH_2O_2 \quad (10)$$
$$CH_3(CH_2)_2CH_2O_2 + NO \rightarrow CH_3(CH_2)_2CH_2O + NO_2 \quad (11)$$
$$CH_3(CH_2)_2CH_2O + O_2 \rightarrow CH_3(CH_2)_2CHO + HO_2 \quad (12)$$

Thus reaction (10) has produced an alkylperoxy radical, which in reaction (11) converts NO to NO_2. Reaction (12) forms butanal, a relatively stable product, and a hydroperoxy radical (HO_2) which by reaction (8) will further enhance the NO_2:NO ratio, and consequently the ozone concentration.

Figure 1.8 shows typical diurnal profiles of primary and secondary pollutants in a polluted atmosphere such as Los Angeles. At night–time, ozone levels fall to zero as k_3 is zero and no NO_2 photolysis occurs. As the sun rises, so hydroxyl radicals are formed from processes such as photolysis of nitrous acid, HNO_2, formed overnight from reactions of oxides of nitrogen:

$$HNO_2 + h\nu \rightarrow NO + OH \quad (13)$$

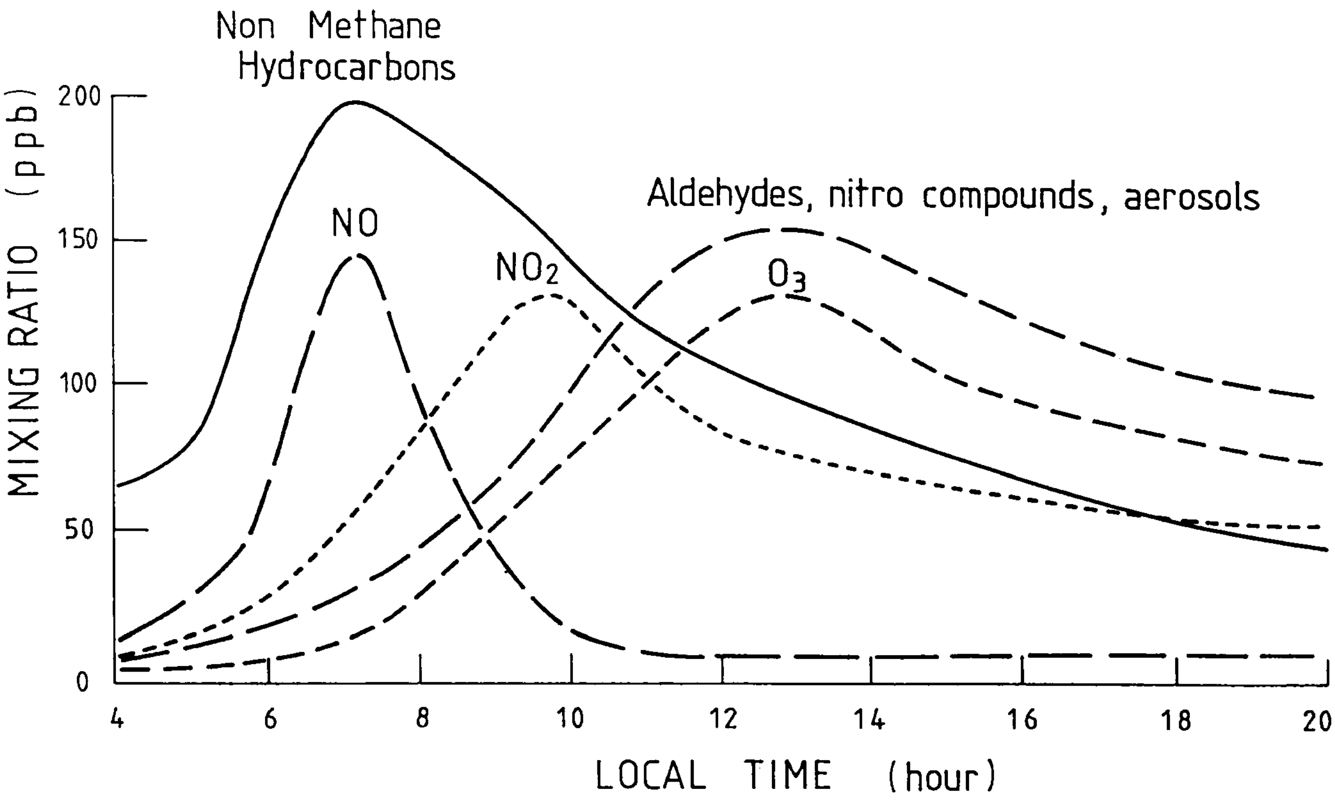


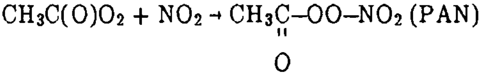
FIGURE 1.8 *Typical diurnal cycles of primary and secondary pollutants in an urban photochemical smog.*

This is followed by production of NO_2 (by reactions such as 6,7,8) and a progressive increase in the NO_2:NO ratio which in turn leads to an increase in ozone. In southern California, this increase in ozone typically occurs around midday whilst in Europe peaks at around 1500 hours are more common, perhaps reflecting in part the need for a longer time interval to maximise the NO_2/NO ratio.

Another product of the photochemical reactions included in Figure 1.8 is peroxyacetyl nitrate, known as PAN. This compound is highly characteristic of atmospheric photochemistry, having no known primary emission sources. It has several routes of formation, most notably oxidation of ethanal (acetaldehyde) by the hydroxyl radical:

$$CH_3CHO + OH \rightarrow CH_3CO \qquad (14)$$

$$CH_3CO + O_2 \rightarrow CH_3C(O)O_2 \qquad (15)$$



$$CH_3C(O)O_2 + NO_2 \rightarrow CH_3\underset{\underset{O}{\|}}{C}{-}OO{-}NO_2\ (PAN) \qquad (16)$$

Table 1.7 shows concentrations of ozone and PAN typically observed in highly and moderately polluted atmospheres. Other pollutants are also included. Hydrocarbons tend to show a peak in the early morning (Figure 1.8) corresponding to heavy vehicle traffic, diminishing somewhat during the day as photochemical processes leading to breakdown and removal by advection (i.e. movement by the wind) more than compensate for fresh emissions.

TABLE 1.7 Typical concentrations of primary and secondary pollutants in polluted air

Pollutant	Concentration (ppb) (1 hour averages)	
	Highly polluted	Moderately polluted
NO	50–750	10–50
NO_2	50–250	10–50
CO	1,000–25,000	500–5,000
O_3	500	80–200
PAN	5–35	0.5–5
Hydrocarbons (excluding methane)	500–1,200	50–300
Sulphate aerosol ($\mu g\ m^{-3}$)	10–50	5–15
Nitrate aerosol ($\mu g\ m^{-3}$)	10–50	5–15

The photochemical "smog" derives its name from the visibility–reducing aerosols produced within it. These are mainly ammonium sulphate and nitrate produced from primary sulphur dioxide, nitrogen oxides and ammonia. The main origin of ammonia is release from animal excrement; in the Los Angeles basin, cattle feed lots are a major ammonia source. Mechanisms of oxidation of SO_2 and NO_2 are described in more detail in the next section, but in simple terms the chemistry is as follows:

$$SO_2 \rightarrow H_2SO_4 \tag{17}$$

$$H_2SO_4 + 2NH_3 \rightarrow (NH_4)_2SO_4\,(\text{aerosol}) \tag{18}$$

$$HNO_3 + NH_3 \rightarrow NH_4NO_3\,(\text{aerosol}) \tag{19}$$

The ammonium sulphate and nitrate salts are present as an aerosol of fine particles which scatter light extremely effectively. The approximate relationship between aerosol mass and visibility is shown in Figure 1.9. These particle–forming reactions occur widely in the atmosphere, but are greatly enhanced in a photochemical smog by the high concentrations of precursor pollutants and the very active photochemistry contributing to rapid oxidation of SO_2 and NO_2.

One facet of recent research into photochemical air pollution in both Europe and North America is the observation of ozone concentrations in rural areas which are as high, or frequently higher than in the urban source regions [26]. This phenomenon is partially explained by the presence of a strong primarily vehicular source of NO in urban areas which tends to limit NO_2:NO ratios and thus suppress ozone somewhat. Rural areas are without this NO source and thus under anticyclonic conditions which prevent extensive vertical dispersal of pollutants, ozone concentrations may exceed those in upwind urban areas. In the longer range, dilution and deposition processes will compete with ozone formation, and ozone levels will diminish.

Ozone concentrations in summer months at rural sites typically show an apparently similar diurnal variation to urban sites (Figure 1.10, cf Figure 1.8). This has a different cause, however. Rather than arising from the chemistry of the $O_3/O_2/NO/NO_2$ system, it has its origin in meteorological processes. The absence of a strong NO source in rural areas allows high O_3 levels to persist overnight. However, dry deposition removal of ozone at the ground surface under nocturnal conditions of little boundary–layer mixing causes a depletion of measured concentrations close to ground–level, and hence an apparent diurnal cycle. However, if concentrations are measured at, say, 50 metres above the surface, little nocturnal decay of ozone is seen and no strong diurnal fluctuation occurs [28,29].

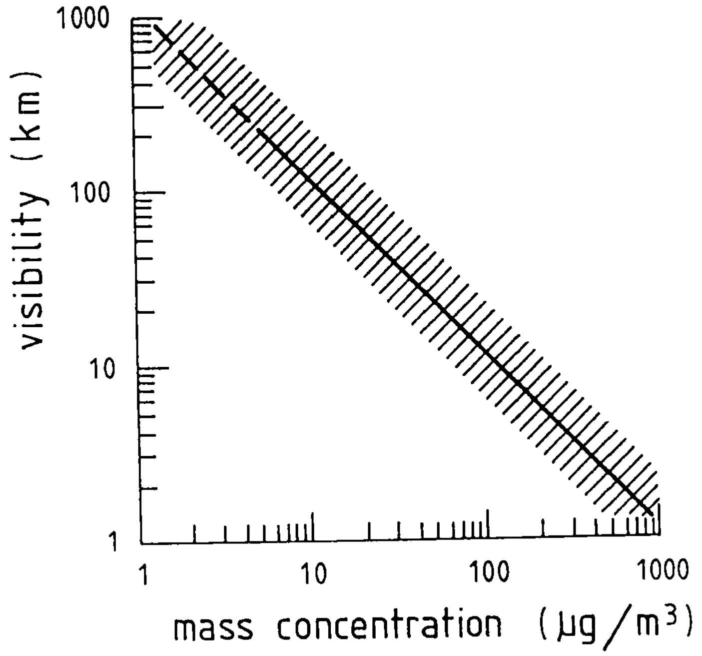


FIGURE 1.9 RELATIONSHIP BETWEEN AEROSOL MASS LOADING AND VISIBILITY

The relationship is approximate only, but most data fit the shaded area

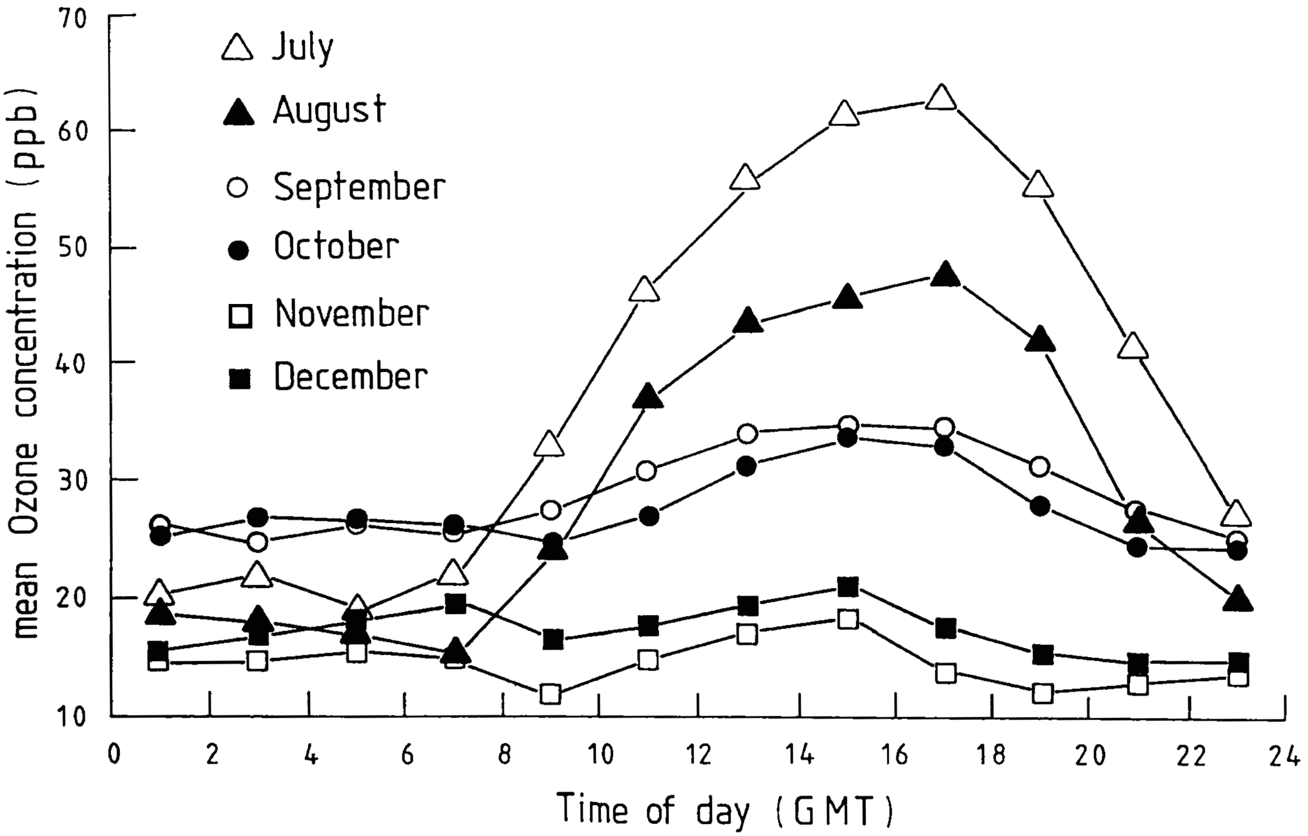


FIGURE 1.10 *Diurnal variations in ozone measured at a rural site, Stodday near Lancaster, U.K. averaged for the months indicated (from ref. 1)*

2.2 Control of Secondary Pollutants

For primary pollutant control, the "linear rollback" concept is applicable. This concept states that for an x% reduction in emissions of a pollutant ground–level concentrations will also diminish by x%. Thus, for example, for carbon monoxide which is primarily vehicle–generated, a 50% reduction in vehicle emissions should give a corresponding 50% fall in atmospheric concentrations. There is evidence in the form of monitoring data that this approach works well for emissions of lead, where in the UK the maximum permitted lead content of gasoline was reduced from 0.40g l^{-1} to 0.15g l^{-1} (actual gasoline lead fell from *ca* 0.38g l^{-1} to 0.14g l^{-1}) at the end of 1985. Atmospheric concentrations of lead fell by almost the same proportion, any differences being explicable by industrial emissions of lead which were not subject to additional control, and lead in air arising from resuspension of street dusts which take some time to adjust to a change in the air lead level [14, 16].

The linear rollback approach cannot be applied to secondary pollutants by extrapolation from reductions in primary pollutant emissions, since secondary pollutant concentrations do not have a simple linear relationship with precursor emissions. Indeed, under some circumstances, a reduction in precursor emissions can lead to an increase in secondary pollutant concentrations.

The influence of changing precursor emissions upon secondary pollutant concentrations can be predicted from computer modelling of atmospheric photochemical reaction processes incorporating some description of atmospheric transport and dynamics. These models vary greatly in their complexity and spatial coverage. Generally, at least 200 chemical reactions are included and whilst some models include only a few representative hydrocarbons whose concentrations are adjusted to take account of other compounds which are not included, other models include explicit chemical reactions to many hydrocarbons. The treatment of pollutant transport and atmospheric dynamics also varies. At one extreme are simple moving box models in which a hypothetical box of air moves along a trajectory, receiving emissions and losing pollutants by deposition processes; at the other extreme are large three–dimensional Eulerian grid models which cover substantial geographic areas in terms of smaller grid squares into which pollutants are emitted and between which air is advected. The vertical mixing of pollutants may also be included in all types of model by use of horizontal layers of air between which pollutant transfer occurs at rates which vary according to atmospheric mixing processes and time of day.

The results of a box model are exemplified by Figure 1.11. This model [30] seeks to describe the changes in composition of initially rural air transported across London. Before entering London, the model is run at typical rural pollutant emission rates to generate a

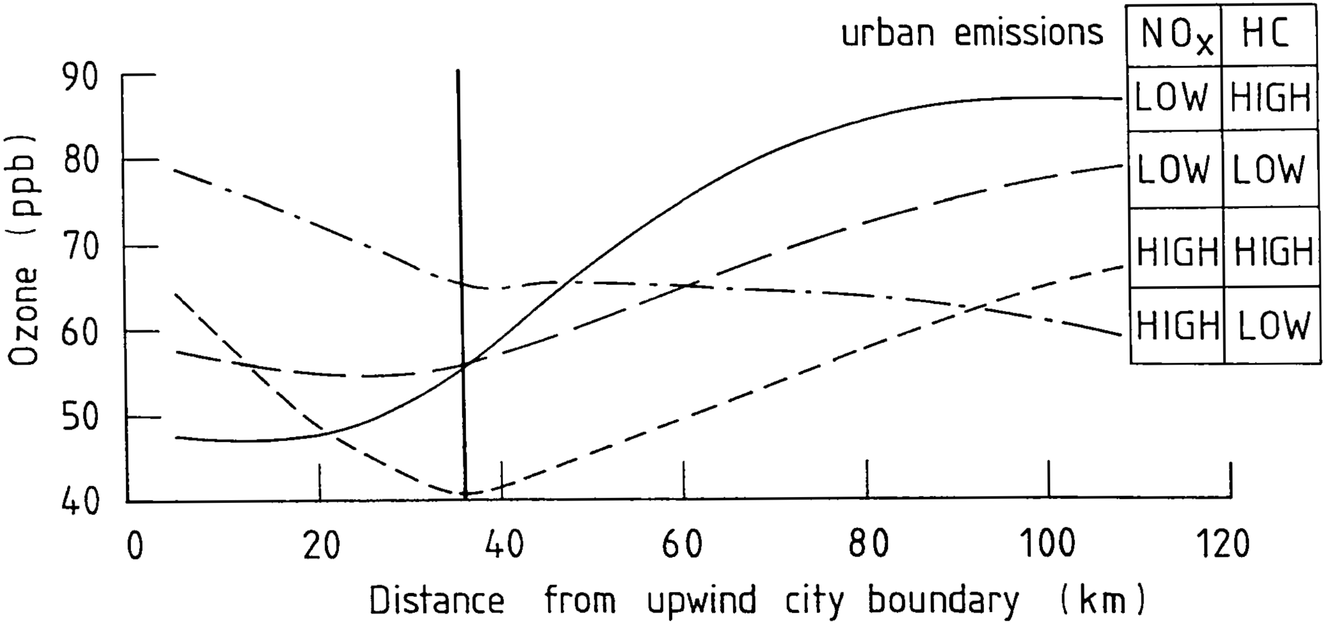


FIGURE 1.11 *Model predictions of ozone formation in an initially rural air mass which traverses London. The mean horizontal wind speed in the mixed layer is* $4 m\ s^{-1}$

background of ozone. It then enters London, encountering enhanced pollutant inputs, subsequently (36 km in Figure 1.11) and again encountering rural levels of emission. The model may be run for different source strength scenarios, shown in Figure 1.11 as "low" and "high". In all cases, as the air enters London it suffers a loss in ozone due to reaction of O_3 with NO; high NO_x emissions (mainly NO) give the most rapid loss. Subsequently, in all but one scenario, ozone concentrations recover and increase. The points which come from this exercise include the following:

i) the predicted degree of control of ozone is dependent upon the downwind distance at which it is measured;

ii) the wind speed, which affects both travel times and the extent of dilution of pollutants, is a crucial factor;

iii) measured 100 km downwind, with this model, maximum emission reductions of both NO_x and hydrocarbons led to higher ozone concentrations than maintaining high emissions of both pollutants;

iv) on the time–distance scale of this experiment, high NO_x emissions are associated with lower ozone concentrations. This is probably not the case at appreciably larger downwind distances.

A convenient form of presentation of the predictions of a computer model is exemplified by Figure 1.12. This shows ozone concentrations calculated as an average for 15 receptor points as a function of NO_x and hydrocarbon emissions on a base (1.00) of the 1980 European emission level. In general, reductions in hydrocarbon emissions are seen to be highly beneficial. This is, however, often not the case with NO_x. Take, for example, a hydrocarbon reduction of 50% (HC emission = 0.50) and progress on a horizontal line from initial NO_x = 1.00. From a starting level of around 70 ppb, ozone concentrations increase to 80 ppb at a NO_x emission level of ~ 0.60, then returning to 70 ppb at $NO_x \simeq 0.30$ and decreasing to 60 ppb at $NO_x \simeq 0.20$.

A consequence of the strong non–linearities of the precursor–ozone system is that predictions of ozone abatement are extremely complex and as yet not entirely reliable. Nonetheless, abatement of precursor hydrocarbon and NO_x emissions has begun in many countries. Although this may be justifiable in terms of reducing environmental effects of the primary pollutants and some of their atmospheric reaction products, the overall effect upon production of secondary pollutants is uncertain. Whilst our ability to model ozone production is reasonably good, at least in as far as may be judged by comparison of the outputs of different models with a common input scenario, model performance for other secondary pollutants such as PAN and hydrogen peroxide is more uncertain.

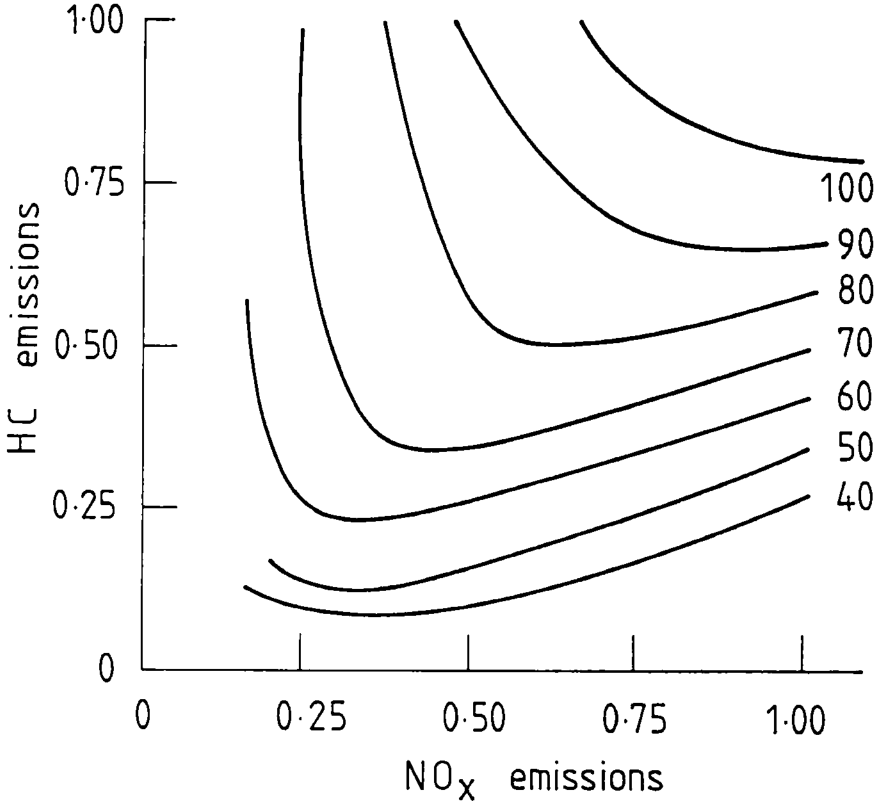


FIGURE 1.12 *Predicted ozone concentrations (isopleths, mean of 15 receptor points) in relation to reductions in European NO_x and hydrocarbon emissions (1980 level = 1.00) (from ref* 31)

2.3 Secondary pollutants and acid rain

As mentioned above, sulphate and nitrate compounds are formed by atmospheric photochemistry and are largely responsible for the visibility reduction associated with photochemical smog. These compounds are formed also under other atmospheric conditions and by other mechanisms and contribute to the existence of "acid rain". Unfortunately, the term "acid rain" covers a number of distinct, but connected phenomena and is thus not clear–cut. Literally, acid rain is rainwater of high acidity (low pH). However, the term has also become associated with other phenomena such as forest dieback which may, or may not, be connected with atmospheric acidity.

The main strong acids in polluted atmospheres are the following:

H_2SO_4	sulphuric acid
HNO_3	nitric acid
HCl	hydrochloric acid

In Western Europe and eastern North America, sulphuric acid is generally thought to be of greatest importance, whilst on the American west coast, nitric acid normally predominates.

The main precursor of sulphuric acid is sulphur dioxide. In the United Kingdom, motor vehicles account for only about 1% of SO_2 emissions [16] and thus contribute very little directly to atmospheric acid. It may be argued, however, that vehicular emissions of oxides of nitrogen and hydrocarbons contribute appreciably to the oxidising capacity of the atmosphere by leading to production of compounds such as ozone and hydrogen peroxide and thus to hydroxyl radicals. In doing so, motor vehicles may make an indirect contribution to atmospheric acidity.

In dry air (i.e. air without liquid water in the form of clouds or raindrops) the main mechanism of sulphur dioxide oxidation is by reaction with the hydroxyl radical [32–34].

$$SO_2 + OH \rightarrow HOSO_2 \tag{20}$$

$$HOSO_2 + O_2 \rightarrow HO_2 + SO_3 \tag{21}$$

$$SO_3 + H_2O \rightarrow H_2SO_4 \tag{22}$$

The sulphuric acid so formed exists as small aerosol particles (generally $< 1\ \mu m$ diameter) which are neutralized by atmospheric ammonia, if available

$$H_2SO_4 + 2NH_3 \rightarrow (NH_4)_2SO_4 \tag{18}$$

This is by far the most rapid route of SO_2 oxidation in dry air, accounting for conversion of SO_2 at about 1.6% h^{-1} for a typical peak daytime concentration of the OH radical (5×10^6 cm^{-3}) [32].

If water droplets are available, rather different mechanisms can operate within the aqueous phase [30–32].

$$SO_2\,(gas) + H_2O\,(liq) \rightleftharpoons SO_2.H_2O \qquad (23)$$

$$SO_2.H_2O \rightleftharpoons HSO_3^- + H^+ \qquad (24)$$

$$HSO_3^- \rightleftharpoons SO_3^{2-} + H^+ \qquad (25)$$

$$HSO_3^- + H_2O_2 \rightarrow HSO_4^- + H_2O \qquad (26)$$

$$HSO_3^- + O_3 \rightarrow HSO_4^- + O_2 \qquad (27)$$

Thus both hydrogen peroxide H_2O_2 and ozone can oxidise disulphite (HSO_3^-) to disulphate (HSO_4^-) thus converting sulphurous acid (H_2SO_3) to sulphuric acid, since HSO_4^- is a dissociation product of H_2SO_4:

$$H_2SO_4 \rightleftharpoons HSO_4^- + H^+ \rightleftharpoons SO_4^{2-} + H^+ \qquad (28)$$

These chemical processes have a potential to convert SO_2 to H_2SO_4 very rapidly but in the atmosphere the overall rate of the process is limited by the rate of mass transfer of the reactants SO_2,H_2O_2 and O_3 into the water droplets and thus typically conversion proceeds at around 1–2% h^{-1} as a result of this mechanism. Concentrations of both oxidising species, O_3 and H_2O_2 are influenced by vehicular pollutants through atmospheric photochemistry. Hydrogen peroxide is formed mainly from the hydroperoxy radical [30]:

$$HO_2 + HO_2 \rightarrow H_2O_2 + O_2 \qquad (29)$$

Formation of nitric acid is more directly connected with highway pollution as the source is nitrogen dioxide; in the UK 40% of oxides of nitrogen emissions arise from motor vehicles as shown in Figure 1.4. During daytime, nitric acid is formed from oxidation of NO_2 by the hydroxyl radical

$$NO_2 + OH \rightarrow HNO_3 \qquad (30)$$

In a moderately polluted atmosphere, conversion of NO_2 can proceed at ~ 20% h^{-1} in bright sunlight. During the hours of darkness, the concentration of OH radicals falls to effectively zero, and a different route for HNO_3 formation is important. This involves initial reaction of NO_2 with ozone, the resultant NO_3 radical reacting reversibly with NO_2 to form N_2O_5 which produces nitric acid by reaction with water [32–34]:

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \qquad (31)$$

$$NO_3 + NO_2 \rightleftharpoons N_2O_5 \qquad (32)$$

$$N_2O_5 + H_2O \rightarrow 2HNO_3 \qquad (33)$$

There are considerable uncertainties in the rate of this process, but it appears that the reaction may account for appreciable overnight production of HNO_3.

Nitric acid is neutralised by atmospheric ammonia in a reversible equilibrium process:

$$NH_3 + HNO_3 \rightleftharpoons NH_4NO_3 \qquad (19)$$

The position of equilibrium depends upon the ambient temperature and relative humidity [30–32] and thus these variables, as well as the atmospheric concentration of ammonia, control the level of free nitric acid in the air. The product ammonium nitrate aerosol, together with ammonium sulphate (reaction 18), is largely responsible for atmospheric visibility reduction.

Motor vehicles are a very minor source of hydrochloric acid emissions. These arise from decomposition of 1,2–dichloroethane added to gasoline as a scavenger in alkyllead additives. In the UK, at present levels of lead in gasoline this source may account for about 0.16 kt a^{-1}, or $< 0.1\%$ of total HCl emissions [35]. As lead levels in gasoline reduce, this source should also diminish approximately *pro rata.*

The pH of water in equilibrium with atmospheric carbon dioxide is around 5.6, which is weakly acidic. The presence of strong acids such as H_2SO_4, HNO_3 and HCl can suppress the pH appreciably and rainwater pHs have occasionally been observed to fall below a value of pH2. Over the United Kingdom, annual average rainwater pH varies from pH $>$ 4.6 on the west coast to pH $<$ 4.3 to the east [36]. The actual annual deposition of acidity, however, (i.e. how much acid falls per unit area per year) is greatest in the upland areas of northern England and western Scotland where the depth of rainfall is greatest, as the deposition is the product of hydrogen ion concentration and rainfall volume [37].

Whilst acidity is deposited to the earth's surface in rainwater, it can also be transferred there by the process of dry deposition. This is the transfer of airborne particles or gases to surfaces of plants, soil, water, etc. without the intervention of rain, sleet, hail or snow. The rate of the process is quantified through the deposition velocity, v_g:

$$v_g\,(\mathrm{cm\ s^{-1}}) = \frac{\text{flux to the surface } (\mu g\ \mathrm{cm^{-2}s^{-1}})}{\text{atmospheric concentration } (\mu g\ \mathrm{cm^{-3}})}$$

For small aerosol particles, such as atmospheric sulphuric acid, v_g is small, around 0.1 – 0.2 cm s^{-1}, and deposition is inefficient. The particles do not contribute appreciably to deposition of acidity, but potentially have a rather long (~ 7 days) atmospheric lifetime. During this time, they will probably be neutralised by ammonia. Both nitric acid and hydrochloric acid exist in the atmosphere in the form of vapours and have a high deposition velocity of ~ 3 cm s^{-1} [34]. The implication is that they are removed extremely efficiently by dry deposition processes and can contribute appreciably to the deposition of acidity to the terrestrial environment.

Environmental damage attributed to "acid rain" may be variously due to acidity *per se* in air or rain, or to primary gaseous pollutants such as NO_2 and hydrocarbons, or secondary pollutants such as ozone and PAN. Whilst some phenomena have a single cause, many are linked with a number of causative agents acting in combination. In many instances, vehicle–related primary or secondary pollutants are thought to be an important component.

3 THE SOILING OF BUILDINGS

3.1 The Extent of the Problem

In October 1987, Britain's National Society for Clean Air (NSCA) presented evidence to the (UK) House of Lords Select Committee on the European Communities on the subject of *particulate emissions from diesel–engined cars* [38]. The policy of the Society on this issue, which was formulated by its Technical Committee in February 1987, emphasises the need for new and more stringent smoke emission standards for all categories of diesel vehicles, from cars to trucks and buses, as a matter of priority. In the view of the Society, "the main argument for curbing diesel smoke relates to the *costs of soiling and the lowering of the quality of life in cities*". Although the link between diesel particulate emissions and the soiling of the built environment has not been rigorously proven, there is considerable circumstantial evidence in support of this proposition. Particulate emission standards from diesel vehicles exist in the USA (Chapter 10, Table 10.3). European standards have been proposed for introduction in the early 1990s.

Despite the declining use of coal as a source of heating and power in European cities and the decrease in smoke levels shown in Figure 1.1, buildings and materials continue to soil at a discernible rate. Visual evidence of this is easily found. For example, Figure 1.13 shows heavy soiling of the Queen Elizabeth Hall in central London. The streaking is caused by the redistribution, by rain runoff, of the deposited soot. External walls of concrete have a marked tendency to become unevenly, and hence more obviously, soiled due to the high water absorption of concrete which inhibits rain washing of the entire surface. Figure 1.14, of a partially–shielded external stairway at the National Theatre in central London, shows that comparatively new buildings are still subject to soiling, as does Figure 1.15 of the Shell Centre in London, which was last cleaned in the mid–1970s. The macroscopic variations in soiling clearly visible in Figure 1.15 can, on closer inspection, be seen to be attributable to differences in roughness of the surface stone.

FIGURE 1.13 *SOILING OF THE FACADE OF THE QUEEN ELIZABETH HALL IN LONDON, 1988.*

FIGURE 1.14 *SOILING OF THE PARTIALLY SHIELDED EXTERNAL STAIRWAY OF THE NATIONAL THEATRE, LONDON*

FIGURE 1.15 *THE FACADE OF THE SHELL CENTRE IN LONDON, LAST CLEANED IN THE MID 1970s AND NOW SHOWING RENEWED SOILING*

This phenomenon is, of course, not restricted to either London or the UK. Very similar effects have been documented in, for example, other parts of Britain [39] as well as in Sweden [40] and France [41].

Subjective observations on the rate of soiling of building facades in these countries indicate that serious disfigurement can occur within a timescale of about 10 years, though this is dependent on individual circumstances, such as the type and detailing of the surface material, as well as the levels of air pollution.

3.2 Origins of Soiling

The soiling of buildings is caused by the deposition and adherence of particles from the atmosphere, which may originate from industrial emissions, fuel combustion by stationary sources, vehicular emissions, tyre wear, resuspension of dust, and other sources. There is some controversy over the size range of particles most responsible for soiling. Haynie and Spence, in the United States, report that coarse particles ($> 10\mu$m) dominate soiling of smooth surfaces such as glass, but comment that for rough surfaces sub–micron particles may be more significant because of their higher deposition velocity [42]. Certainly, there is strong evidence that most of the *dark–coloured* material in the urban atmosphere is of micron or sub–micron size. Caswell, in a series of measurements in central London, used a particle size fractionating device to demonstrate that, on a deposited mass per unit area basis, particles of 1.4 μm diameter or less contributed far more to the blackening of filter media than larger particles (Table 1.8).

TABLE 1.8 **Aerosol particle size data for central London, collected by cascade centripeter (ref. 43). A comparison of the surface loadings on each stage with the corresponding darkness index of the deposit, shows that the finer particles are the more powerful darkening agents.**

Particle size band (μm)	**Mass collected (μg)**	**Surface loading ($\mu g/cm^2$)**	**Darkness Index**
> 12.5	272	120	23
3.8 to 12.5	740	326	51
1.4 to 3.8	590	260	86
< 1.4	310	146	86

Ultimately, it is to be expected that both large and small particles are responsible for different aspects of the soiling process, and that their relative importance as soiling agents depends even upon the definition of 'soiling'. It has been suggested that the US EPA definition of soiling, namely, "that which is undone by cleaning" might be modified to "what is not undone by meteorological factors but by manual processes", in recognition of the fact that larger particles are less adherent and are relatively easily removed by rain and wind.

Despite uncertainty over emission factors, Section 1.1.3 showed there is strong evidence that road vehicles are the largest source of the fine, dark material in the atmosphere. Early indications of this were provided by air quality data gathered in central London in the mid–1970s at a non–street side location [44]. These data revealed a strong correlation between the airborne concentration of black smoke, as measured by the standard optical reflectance technique, and airborne lead concentrations. It was inferred that road vehicles, and especially diesels, were the main source of dark smoke, perhaps contributing as much as 80% to dark smoke concentrations. This finding is supported by the emission estimates for London shown in Table 1.4. As discussed earlier in this chapter, the likely importance of diesel vehicle emissions as a soiling agent can be traced to their high content of particulate elemental carbon (PEC). Further confirmation of the role of elemental carbon as a soiling agent in urban atmospheres is provided by the close correlation noted between black smoke concentrations and EC concentrations [45].

$$\text{EC } (\mu\text{g m}^{-3}) = 0.13 \text{ black smoke } (\mu\text{g m}^{-3}) - 0.1$$

This is consistent withthe values quoted in Table 1.1.

3.3 Models for Soiling

Two procedures for modelling the soiling process have been proposed [44,45]. Both assume that soiling is due to deposition of particulate matter from the atmosphere. The effects of dust resuspended from the road surface and impacting directly on the surface experiencing soiling have not been considered, nor have optical changes associated with damage to the material caused by polluting gases and acid deposition. The approaches to modelling the soiling process involve:

i) Surface Area Coverage

This model assumes that loss of reflectance is directly proportional to area covered.

If the exposed surface has area A_o, of which area A is covered, then the mass deposition rate to the uncovered surface is given by

$$\frac{dm}{dt} = (A_o - A)\, C\, v_g$$

therefore, $$\frac{dA}{dt} = \frac{3}{4r\rho}\frac{dm}{dt} = k\,(A_o - A)$$

therefore, $$A = A_o\,(1 - e^{-Kt})$$

therefore, $$R = R_o e^{-Kt} \quad \text{where } R_o \text{ is the initial reflectance} \qquad \text{(I)}$$

with $$K = \frac{3}{4}\frac{C}{r}\frac{v_g}{\rho} \qquad \text{(II)}$$

This model can be extended to account for the reflectance R_p of the settled particulates so

$$R = R_p + (R_o - R_p)\, e^{-Kt}$$

ii) **Thickness of Deposited Film**

This model assumes that the loss in reflectance is proportional to the thickness (z) of the deposit, so that

$R = R_o e^{2z\mu}$, where μ is the linear absorption coefficient.

The thickness of the deposit increases with time according to

$$C\, v_g\, t = z\,\rho$$

so $$R = R_o e^{-Kt}$$

where $$K = \frac{2\, C\, \mu\, v_g}{\rho} = 2\, C\, \alpha\, v_g \qquad \text{(III)}$$

with $\alpha = \frac{\mu}{\rho}$ = mass absorption coefficient

In both models,

C = airborne concentration
v_g = deposition velocity
ρ = density of particulate matter
r = radius of particulate matter

Haynie [46] employed the surface area coverage approach, with C referring to TSP concentration. He considered the deposition process appropriate to each size range and the soiling constant K was evaluated by summing across the particle size spectrum. The result was dependent on the size distribution, meteorological conditions, surface roughness and surface orientation. For example, for glass at 3 m s^{-1} windspeed, $K = 0.3$ yr^{-1} for a vertical surface and 0.96 yr^{-1} for a horizontal surface when $C = 100$ μg TSP m^{-3}. Higher values were predicted for painted surfaces.

Lanting [47] considered both processes but assumed that PEC was responsible for all soiling. Appropriate values are:

$v_g = 2 \times 10^{-4}$ m s^{-1} $\quad$ $R = 0.025$ μm

$\rho = 2 \times 10^{3}$ kg m^{-3} $\quad$ $\alpha = 1 \times 10^{4}$ m^2 kg^{-1}

so $K = 0.095$ C (μg EC m^{-3}) yr^{-1} according to equation II

and $K = 0.13$ C (μg EC m^{-3}) yr^{-1} according to equation III

Using the values reported in Table 1.1, 100 μg TSP m^{-3} corresponds to 3 μg EC m^{-3}. The predicted soiling constants are:

0.28 yr^{-1} from equation II and 0.4 yr^{-1} from equation II

The Lanting predictions are in reasonable agreement with the Haynie predictions, bearing in mind that both models of necessity simplify the complex process of particulate deposition to, and retention on, rough surfaces and also the optical properties of the deposit.

Experimental programmes [4] reported measurements of soiling constants in a variety of environments and the results observed in a road tunnel were found to lie between values predicted by the two models.

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Chapter 2

HIGHWAY POLLUTANT MONITORING

R.S. Hamilton, R.M. Harrison, I.S. McCrea and N.I. Ward

Contents Page

1 GENERAL CONSIDERATIONS

1.1 Monitoring Objectives

Chapter 1 considered the emissions of pollutants from sources on highways, the transport of these pollutants away from source and their effects on the living and non–living environment. Because effects are exposure–dependent, legislation is framed in terms of limit and/or guideline values for pollutant levels. The formulation of legal standards is considered in Chapter 12 and quality standards are reported in Appendix 1. The responsibility of government to meet legal obligations requires the establishment of monitoring programmes which recognise, and take account of, the need to obtain representative samples and perform accurate analyses on these samples. Carefully designed monitoring programmes are also required by research scientists investigating the transport, properties and effects of specific pollutants and by engineers investigating improved environmental control technology, for example, in vehicle emission control and road drainage design. the aim of this chapter is to review the approaches and techniques used for monitoring pollutant levels in vehicle emissions and near highways. A chapter of this size cannot provide full practical details, many of which are given in environmental handbooks [1,2,3]; rather, the aim is to identify the relevant approaches, describe the principles and limitations and refer the reader to sources of up–to–date practical information.

Before establishing a monitoring programme, the following factors have to be considered:

1. What are the objectives of the programme?
2. Which environmental sectors are to be monitored?
   Measurements may be required at the vehicle exhaust, in air (indoors or out–of–doors), in water, soil, dust or vegetation.
3. What pollutants are to be measured?
   And to what level of sensitivity and accuracy?
4. What resources, financial and manpower, are available?

These factors are often interrelated, as shown diagrammatically in Figure 2.1. In some situations, as, for example, with many consultancy contracts or with a programme to see if a site meets existing environmental standards, the answers to the above questions are known in detail with the result that the monitoring programme is clearly defined and has little flexibility. In most cases, particularly with research programmes, this is not the situation and the open–ended nature of the investigation means that the monitoring programme will be modified as the research progresses.

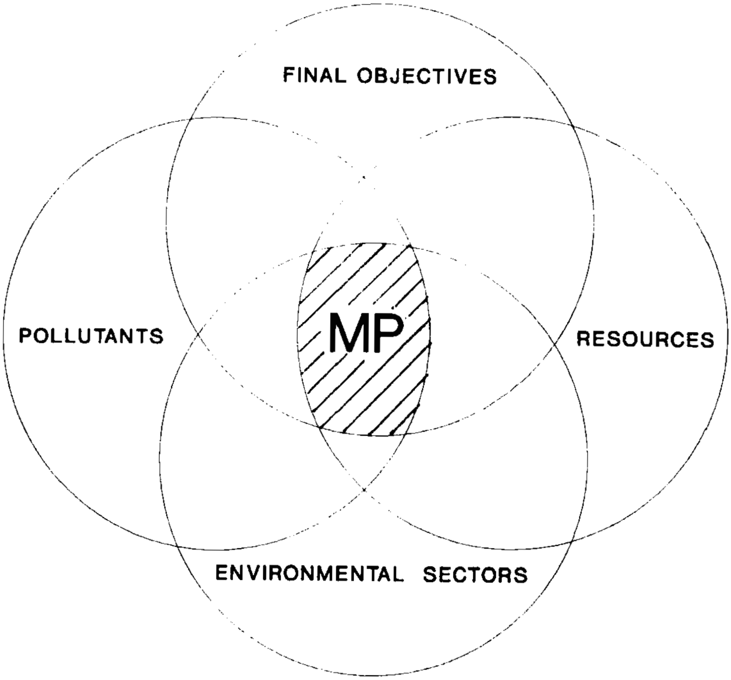


FIGURE 2.1 *FACTORS TO BE CONSIDERED IN THE DESIGN OF A MONITORING PROGRAMME (MP)*

1.2 Sampling Statistics

The frequency of sampling, averaging time and the total duration and spatial extent of the monitoring programme need to be carefully considered. Pollutant concentrations tend to be log–normally distributed and this distribution is preserved for all averaging times [4,5]. A consequence of the log–normal distribution is that an increase in averaging time will lead to a decrease in maximum concentration. An analysis of air quality data collected from seventeen monitoring stations in California showed that the pollutant maximum concentration was related to the averaging time by the equation:

$$C_{(t)} = C_{(1)} \cdot t^{b} \quad \text{(ref 6)}$$

where $C_{(t)}$ was the maximum concentration with averaging time
t (hours),
$C_{(1)}$ was the maximum hourly average,
b was a constant characterising the relation.

Values of b were site specific and pollutant specific,
for example,

$-0.158 < b < -0.068$ for CO
$-0.270 < b < -0.167$ for NO_x

The consequence of these results is that if a set of pollutant data stretching over several years is time–averaged, for CO the maximum yearly average is approximately one–third the maximum hourly average and the difference for NO_x is even greater.

For averaging times greater than five days, differences in mean or maximum concentrations are small so this effect is important mainly for pollutants monitored by fast–response instrumentation.

Prior knowledge of the extent of temporal and spatial variations in pollutant levels is helpful in deciding the total duration and spatial extent of the monitoring programme. This may be determined from a pilot study or from published information. For example, a study of Pb levels in the London atmosphere [7] showed considerable variations in weekly samples, collected over ten consecutive weeks, with the highest level approximately four times the lowest level and a coefficient of variation (standard deviation ÷ mean) of 0.3. Dust samples showed a similar coefficient of variation and runoff samples exhibited a similar effect [8]. A rule–of–thumb is that the duration of the study should be at least ten times as long as the averaging time so an average weekly level can be reliably estimated from the result of ten weekly samples. If a cyclic variation is involved, for example a seasonal trend, the sampling period should be at least ten times shorter than the period of the cycle so ten weekly samples would be sufficient to give a seasonal average weekly value. Annual averages could be determined from monthly samples and annual trends require data collected over a ten–year period.

Although long sampling and averaging times may be involved, it is often more convenient, particularly with automatic analysers for gaseous and some aquatic pollutants, to have shorter sampling times, to store the data using loggers and to process the data at a later stage with microprocessors. For the reasons discussed earlier, the operator of a monitoring programme may have no choice over sampling and averaging times, particularly for studies established to test compliance with legal standards. The European Commission air quality limit value for Pb is 2 μg m^{-3}, measured as an annual average of weekly values while the NO_2 limit value is 200 μg m^{-3}, measured as the 98th percentile of hourly averages. Therefore a Pb monitoring programme requires 7–day samples while the NO_2 programme requires measurements to be taken at a maximum of 1 hourly intervals.

Spatial variations must also be considered and here again legislation generally indicates appropriate sampling locations. As the pollutants disperse away from the highway, their levels in air decrease. These processes are discussed in Chapter 3. Deposition of pollutants results in dust, soil and vegetation contamination so these pollutant levels also tend to decrease with increasing distance away from the highway. A survey of pollutant–in–dust loadings (expressed in g m^{-2}) and concentrations (expressed in μg g^{-1}) showed that the loading exhibited greater variation than the concentrations [9]. Representative pollutant–in–dust concentrations can be achieved by sampling over a relatively large area, for example, 5 m^2 [7].

2 SAMPLING AND ANALYSIS

2.1 Vehicle Emissions

Pollutant emissions from vehicles are monitored for four main reasons:

- to fulfil manufacturers' legal obligations to meet emission standards,
- to provide information on emission rates of non–regulated pollutants,
- to investigate the physical, chemical and biological characteristics of the emissions,
- to test the effects of alternative engine and control technologies.

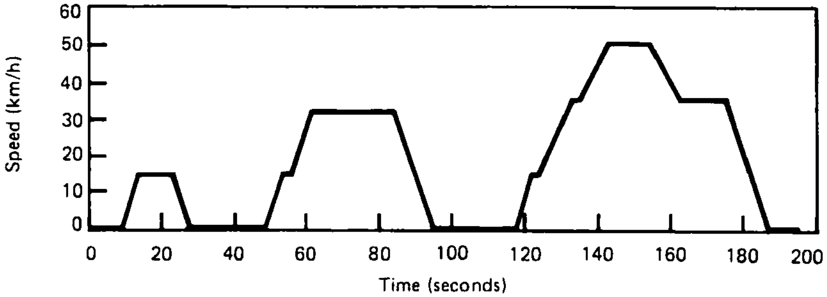
Manufacturers' legal obligations include type approval tests for new vehicles and engines before their mass production and conformity of production tests for vehicles on sale to the public. Gasoline engined vehicle emission standards from 1970 to the present day are summarised in Table 2.1. Within Europe, recent reviews have considered the regulatory approach of the EEC [10] and the rate of international harmonisation [11]. Emission standards for diesel vehicles tend to be introduced later than for gasoline vehicles, though their importance is now recognised, as discussed in Chapter 3.1.

Table 2.1 Exhaust Emission Standards, Gasoline Engined Vehicles

Year	Pollutant	EEC Standard (test cycle corresponds to 4.05 km)	USA Standard	Japan Standard
1970	CO	100 – 200 g/test	21 g/km	2.5%
	HC	8 – 12.8 g/test	3.4 g/km	–
	NO_x	–	3.1 g/km	–
1980	CO	65 – 143 g/test	4.4 g/km	2.7 g/km
	HC	6 – 9.6 g/test	0.25 g/km	0.39 g/km
	NO_x	8.5 – 13.6 g/test	1.2 g/km	0.48 g/km
1990	CO	19 – 45 g/test	2.1 g/km	2.7 g/km
	HC	–	0.25 g/km	0.39 g/km
	NO_x	3.5 – 6.0 g/test	0.62 g/km	0.48 g/km
	HC + NO_x	5 – 15 g/test	–	–
To be introduced 1st January 1993	CO	2.7 g/km		
	HC + NO_x	0.97 g/km		

Exhaust emission tests are performed with the vehicle on a chassis dynamometer equipped with rollers to simulate road driving. Because of the dependence of emission rate on driving conditions, the vehicle on the dynamometer is taken through a specific driving cycle. Since 1970, countries within the European Economic Community (EEC) have used the ECE 15 driving cycle which is a low speed cycle designed to simulate urban conditions but in 1990, an extra–urban driving cycle was added to include the effects of high–speed driving (Figure 2.2). Tests for the regulated emissions–hydrocarbons, CO and NO_x – are

a) ECE 15 Cycle



b) Extra Urban Driving Cycle (EUDC)

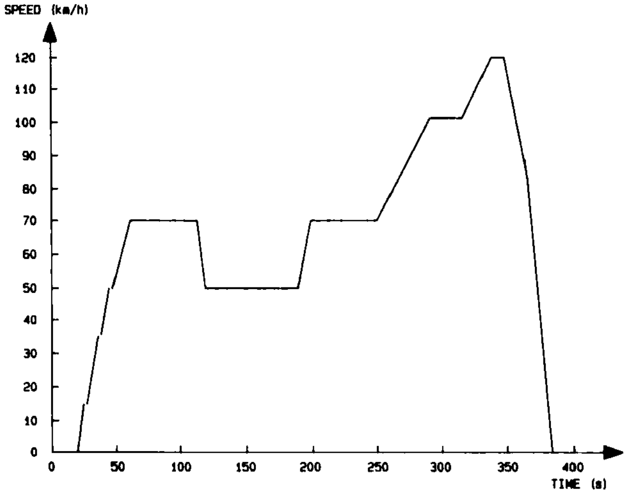


FIGURE 2.2 *EUROPEAN DRIVING CYCLES*

carried out by diluting the exhaust gas with background air under constant volume conditions and small samples are passed to a set of analysers. The exhaust gas collection system is shown in Figure 2.3. Emission rates are expressed as "grams per test" or "grams per km". The ECE 15 test cycle corresponded to a driven distance of 4.05 km. Chapter 10 examines the emissions of some current models and considers the improvements which may be possible with improved engineering control systems.

High pollution levels are often associated with specific driving conditions such as high speed or congested traffic, and reliable information on emissions in these situations cannot be gained from dynamometer tests using standard driving cycles. It is better to make measurements with the vehicle travelling on the road and this is achieved by drawing off a fraction of the exhaust gas, which is either collected in a Tedlar bag for subsequent laboratory analysis [12] or monitored by on–board sensors [13].

In addition to exhaust emissions, legal standards are also applied to hydrocarbon evaporative emissions. These emissions are important because of their role in secondary pollutant air quality, as reviewed in Chapter 1 [see also ref. 14]. The measurement of evaporative emissions is carried out with the vehicle in a special room equipped for sealed house evaporative determination (hence the acronym, SHED test). The procedure, which is outlined in Figure 2.4, is in two stages. First, and following preconditioning of the vehicle but before the exhaust emission test is carried out, the fuel in the car is heated from 16°C to 30°C over one hour. Then, after the exhaust emission test has been completed and while the engine is still hot, the vehicle is returned to the enclosure for one hour. Hydrocarbon concentrations are measured throughout both stages of the test and from these values the total hydrocarbon emission can be calculated. The current evaporative emission standard is 2 g per test under EEC and US Federal legislation.

Full details of the procedures for vehicle emission determinations are given in official technical publications [15, 16]. Methods of gaseous pollutant analysis are considered in Section 2.3.

2.2 Ambient Air

Air sampling systems tend to have three components:

a) **An intake and transfer component.** This allows for a representative sample of air to be passed to the collector(s) and/or analyser(s), and normally consists of a pump and a section of tube. The size of the pump depends on the flow rate required, which in turn is determined by the sampling time, the characteristics of the analytical instrumentation and

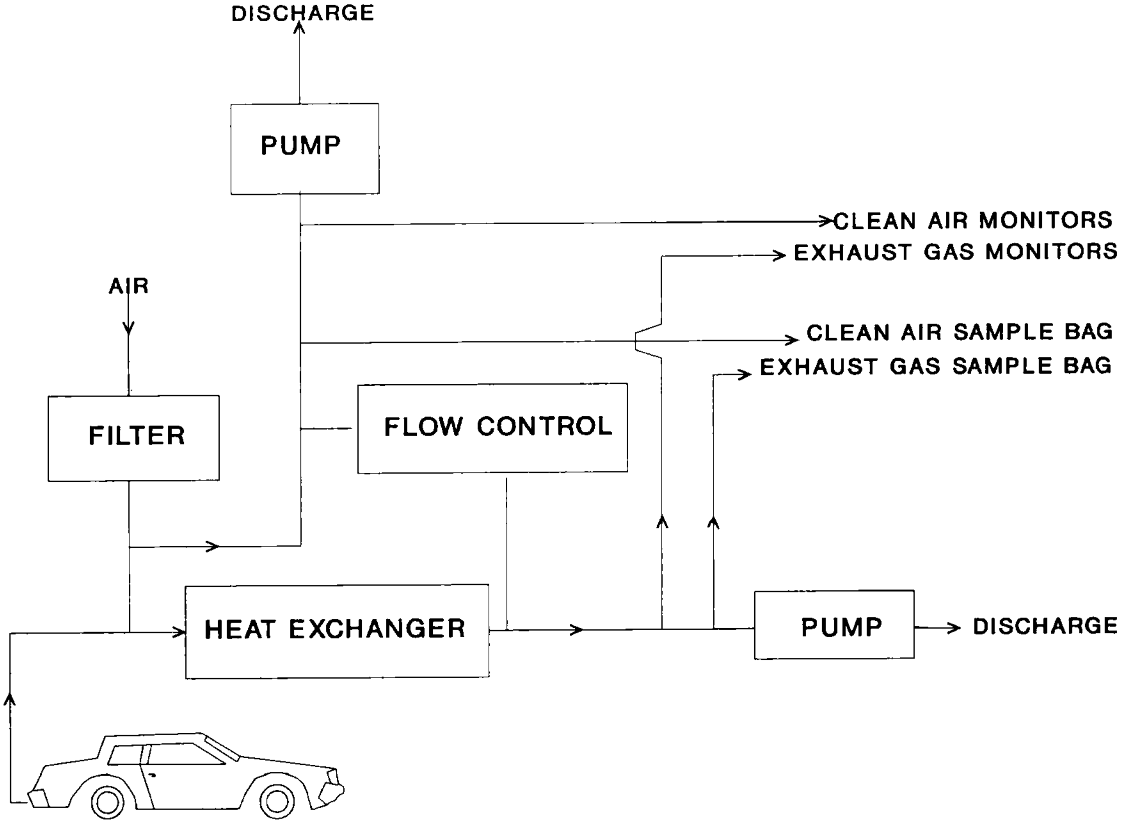
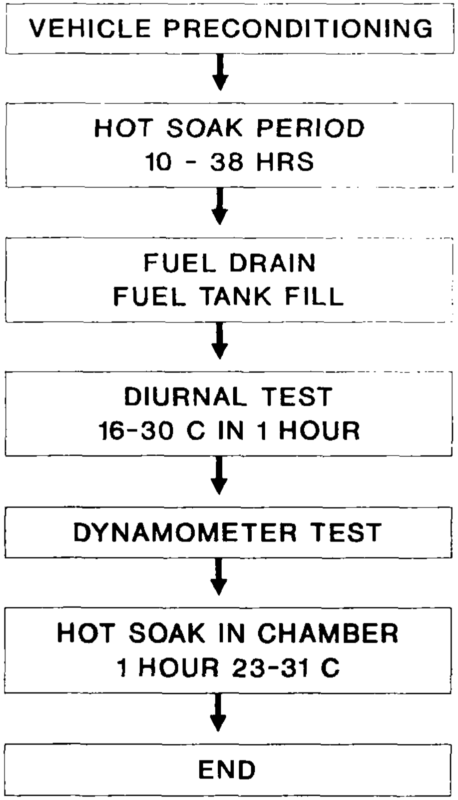


FIGURE 2.3 *EXHAUST EMISSION DETERMINATION*



Including overnight soak at 16° C

5 minutes max.

Ambient temperature
20 – 30° C.

40% ± 2% of tank capacity
Fuel temperature 10 – 14° C

1 hour max.

ECE ± EUDC driving cycles

Test result (grams) = dirunal result (gms) + hot soak result (gms)

FIGURE 2.4 *EVAPORATIVE EMISSION DETERMINATION* the level of the pollutant in the atmosphere. In practice, flow rates vary from 2 m^3 per day as used in the UK National Survey of Air Pollution [17] for collecting daily samples, to 2 m^3 per minute as used in high–volume samplers which can collect sufficient sample in a matter of hours. The intake has to be sheltered from rainfall, for example, high volume samplers have a fitted "roof" open at the base to allow air flow, while low–volume samplers have an inverted filter funnel at the open end of the sampling line. Problems associated with inappropriate design of the intake and transfer component include:

- erroneous sampling due to faulty inlet design,
- adsorption, condensation and adhesion on the transfer lines,
- chemical reactions with the sampling materials.

Further details and preventative measures are described in the specialist literature [1].

The siting of the intake needs to be considered carefully. With a ground level source, concentration will normally decrease with height though topography, including street canyons, can lead to complex concentration patterns. It is recommended that the intake is at least 2 m away from a surface. Factors such as access to electricity and possible vandalism must be taken into consideration.

b) **A flow measuring component.** This is essential if the pollutant level in g m^{-3} is required but not for aerosol related concentrations in μg g^{-1}. The measurement is carried out with either a ratemeter or a volume meter. The latter is more regularly used and the conventional dry gas meter is the commonest form. The only advantage of ratemeters is their relatively small size. However, they are more expensive and require more frequent calibration than volume meters. Examples of ratemeters include venturi meters, orifice meters, flow nozzle meters, rotameters, pitot tubes and hot wire anemometers.

c) **A sample collector or sensor component.** This is the component which connects the sampler with the analyser. "Collection" implies a separate stage in the overall process, with the pollutant collected on a surface or in a medium for subsequent analysis, either within the instrument or subsequently in a laboratory. "Sensor" implies that the pollutant is directed into a zone where its presence gives rise to a change in a physical property, usually optical or electrical, and this change is a direct measure of the pollutant parameter, e.g. concentration or particle size. Calibration is required.

The most frequently used particulate collection systems are based on either filtration or impaction. The most common type of filters are:

- cellulose, e.g. Whatman No.1 and No.41, used in many routine air pollution studies including the UK Smoke Shade method;
- glass fibre, preferred for use with high–volume samplers because of their low flow resistance;
- cellulose acetate membrane, e.g. Millipore, which has a low level of impurity and is preferred for trace element analysis;
- Nuclepore, produced by neutron bombardment of polycarbonate sheet giving clear, uniform pores and high surface collection efficiency and so ideally suited to subsequent microscopy examination.
- teflon membrane, due to very low impurity blank and lack of chemical reactivity.

Impactors allow particulate matter to be size–selectively sampled and so yield information on the size distribution of airborne particulate matter. Systems range from 2–stage dichotomous samplers to 10–stage devices. Low and high volume systems are available.

Gas collection systems employ the principles of adsorption, absorption, condensation and grab–sampling. Though fast–response sensor systems are available for many gaseous air pollutants, collection and subsequent analysis is generally cheaper and examples of the use of this procedure include the determination of SO_2 by absorption in H_2O_2, NO_2 determination by absorption in triethanolamine diffusion tubes and the determination of organics by adsorption on to porous polymers such as Tenax. Grab sampling involves the collection and isolation of a sample in an impermeable container. The use of a Tedlar bag for this purpose was referred to in Section 2.1. Examples of the use of sensing techniques include:

- fluorescence, particularly for SO_2 monitoring;
- adsorption spectroscopy, involving infrared and ultraviolet radiation with either dispersive or non–dispersive analysis, for CO, O_3 and total hydrocarbon monitoring;
- chemiluminescence, for O_3 and NO_x monitoring.

A major advantage of sensor–based systems is that they offer the potential for real–time display. Figure 2.5 shows the telemetry system operated by the Warren spring Laboratory for continuous monitoring of NO_x, O_3 and SO_2 at seventeen sites across the British Isles.

Automatic gas analysers and techniques for particle analysis are reviewed later in this chapter. Traditional laboratory methods for the analysis of collected samples are well documented in the literature [1].

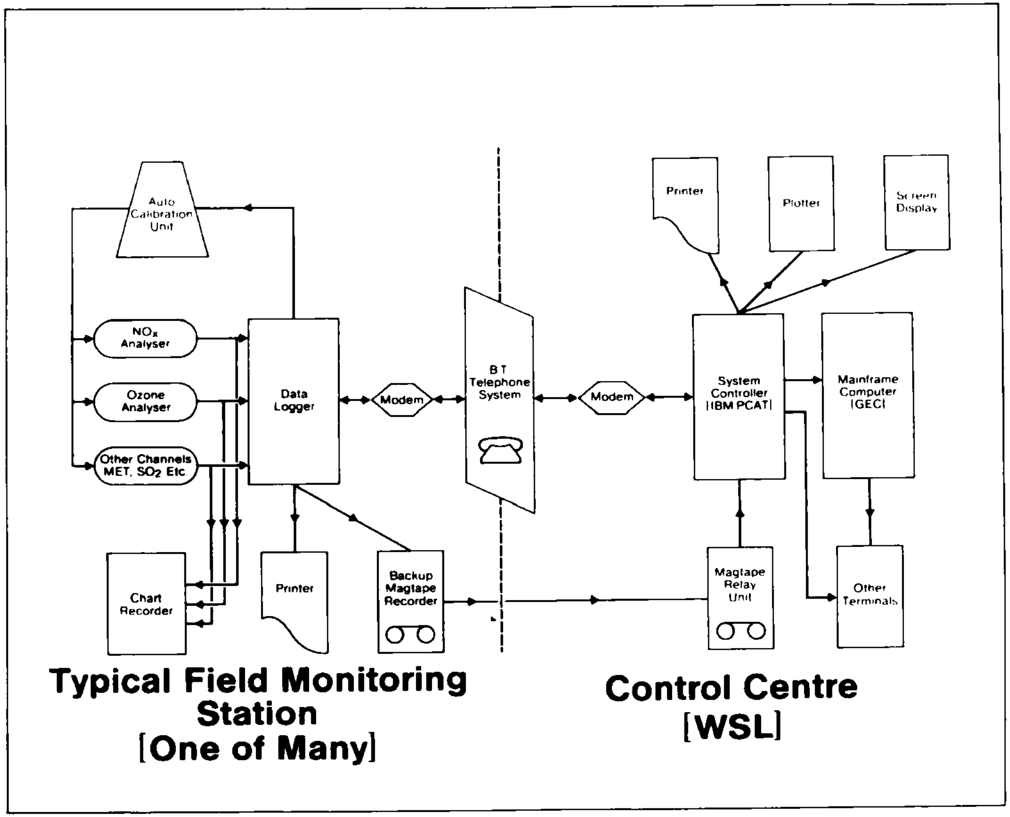


FIGURE 2.5 *THE U.K. AIR QUALTIY MONITORING TELEMETRY SYSTEM*

2.3 Deposition

Pollutants are deposited from the atmosphere to solid and aqueous surfaces by wet deposition (which includes rainout and washout) and by dry deposition. The monitoring of deposition rates may involve the measurement of wet–only, dry–only or total (bulk) deposition. Total deposition is the easiest and cheapest parameter to measure, and most monitoring programmes will measure total and either wet–only or dry–only deposition, with the other parameter calculated by subtraction.

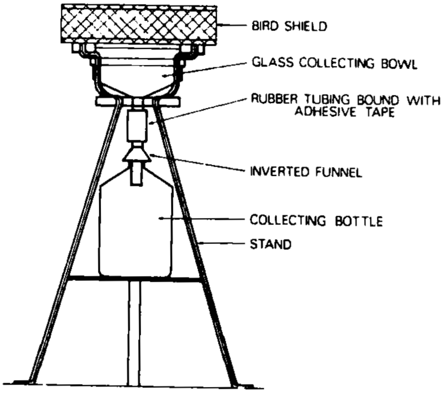
The different designs for total deposition gauges, often referred to as deposit gauges, are shown in Figure 2.6. Recent tests have shown that an inverted Frisbee is an effective collector [18]. Wet–only collectors have an inlet covered with a shield which opens only when rainfall begins. Plastic, pyrex and polythene containers have been found satisfactory for storing the deposited matter. A simple dry deposition collector consists of a Whatman 541 filter paper held horizontally 10–15 cm below a plastic rain cover. Chemical analysis on total deposition can be performed on an aliquot from the bulk sample or, following filtration, on the dissolved and particulate–associated fractions separately.

2.4 Dust and Soil Monitoring

The heterogeneous nature of dust and soil increases the problem of obtaining a representative sample. Higher pollutant concentrations tend to be associated with smaller particle sizes but even within a well–defined size range, environmental particulates are assemblages with widely varying chemical composition. The established methods of reducing this variability are by analysing a large sample mass and/or by grinding the sample prior to analysis. There is some evidence that grinding increases the measured pollutant level, presumably because of the greater surface area exposed and the consequent greater ease of pollutant extraction. Soil and dust samples are normally dried and sieved prior to analysis. There is little conformity in the use of sieve sizes; the literature contains examples of different sieve sizes in use, with 250 μm, 500 μm and 1 mm being the most common cut–off sizes.

Dust samples are collected either by dustpan and brush or by vacuum techniques. The former method is preferable if the dust is damp or present in large quantities, but care has to be taken to avoid particle loss by resuspension. Soils are collected by surface scrapings or by using a corer if depth profiles are required. A plastic or chromium–plated steel tube with 2–3 cm internal diameter makes a satisfactory corer.

a) British Standard BS 1747 gauge



b) directional gauge

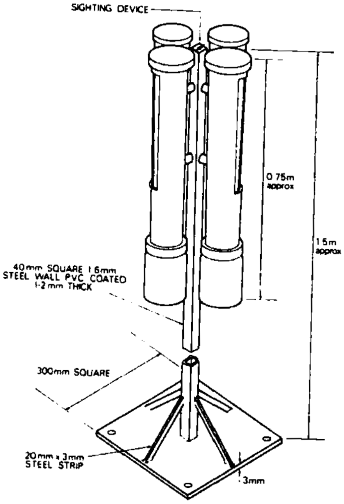


FIGURE 2.6 *TOTAL DEPOSITION GAUGES*

2.5 Road Drainage Sampling and Analysis

Highway or urban runoff pollution studies may involve sampling the surface runoff, within the gullypot (or catchpit) or in the stormwater sewer. The below ground systems may involve sampling deposited particulate matter and/or runoff/supernatant liquor containing dissolved or particulate– associated pollutants. Grab sampling is normally sufficient for obtaining samples from a gullypot or deposited matter from a sewer.

As with ambient air sampling, sewer sampling involves an intake and transfer component, a flow measuring component and a sample collection or sensor component. Examples of properties normally measured on site include turbidity, pH, electrical conductivity, temperature, flow velocity and dissolved oxygen. Collection systems operate in either the discrete or composite mode, with the former more common. A typical discrete sampler contains between 24 and 48 sample bottles each with about 600 ml capacity, a control gear, timer and a sample distribution mechanism. Water samples are transferred to the collecting bottle by a connecting tube and either a vacuum pump or a peristaltic pump. An external float switch, capable of making or breaking a circuit in the sampler, is fixed to a support above the base of the runoff sewer. The height above the base is chosen to represent the required flow above base level. Once the flow reaches the float, the circuit is completed and sampling commences. Samples are directed to the collection bottles and each aliquot is collected either at fixed time intervals or on a flow proportional basis [19].

The parameters most frequently recorded in urban runoff quality studies are:

pH
conductivity
total suspended solids
total dissolved solids
total volatile solids
volatile dissolved solids
dissolved oxygen
biochemical oxygen demand (BOD)
chemical oxygen demand (COD)
total carbon/total organic carbon
sulphate
nitrate
ammonia
orthophosphate

chloride
phenol
total hydrocarbons/speciated hydrocarbons
heavy metals, especially Pb, Zn, Cu, Cd and Hg.

Analytical techniques are described in Reference 2. For some pollutants, particularly for hydrocarbon and heavy metal analysis, the dissolved and particulate–associated forms are separated by filtration prior to analysis, with 0.45 μm filters normally used. "Dissolved" and "particulate" pollutants are operationally defined as those which are passed through, or were trapped on, the filter, respectively.

Data processing and storage facilities for water quality monitoring are similar to those for air quality monitoring, and involves:

i) the recording of data from automatic analysers on magnetic tape or floppy disc. It is essential that accurate time indicators are provided so that variations in recorded parameters can be related.

ii) the transmission of the recorded data to a central store, normally a computer. For real–time monitoring, this involves a telephone link, as shown in Figure 2.5 for air quality systems. Water quality systems are considered in Chapter II.

iii) the incorporation of data from laboratory analyses into the central store;

iv) display, from the central store, on screen or paper.

2.6 Particle Analysis

The main parameters of importance with particulate pollution are mass, size and chemical composition. Analysis can be performed either on bulk samples or by summing the properties of a large number of single particles. The latter approach often means microscopy, though light scattering and electrical zone techniques for particle size analysis also involve single particle parameter summation.

Optical and electron microscopy have been used extensively in particle analysis [20]. Optical microscopy is suitable for particles greater than 1 μm in diameter and so is an appropriate technique for dust, soil, suspended solids in runoff and coarse airborne particulates. It is unsuitable for fine airborne particulates and these samples are best studied by scanning electron microscopy. The collection and analysis of the X–rays emitted during electron bombardment means that the elemental composition of each particulate can be semi–quantitatively determined. Recent developments in signal and image processing, including low atomic number detection, have extended the scope of this method of analysis and reduced the time required for the analysis of a large number of particulates [21]. The application of automated scanning electron microscopy for receptor modelling is considered in Chapter 6.

The size of airborne particulate matter is determined either by size–selective sampling or by light–scattering techniques [1, 22]. Suspended solids in urban runoff have been sized using a Coulter Counter operating on the electrical zone method, where the passage of a particulate between two electrodes connected to a constant current supply gives rise to a voltage pulse [22, 23]. The relatively new Coulter LS series operates on laser diffraction and can size across the range 0.1 to 800 μm with 100 size channels.

The importance of particulate elemental carbon (PEC) was discussed in Chapter 1 and it was shown that, in most countries, over 80% of airborne PEC came from diesel emissions.

The most common techniques for atmospheric PEC analysis are:

i) **thermal methods**, involving the sequential combustion of the particulate matter and the analysis of the products produced,

ii) **optical methods**, particularly the integrating plate technique, which rely on the well–documented evidence that PEC is the dominant light absorbing species in the atmospheric aerosol, and

iii) **photoacoustic spectroscopy**, which measures light absorption directly by passing ambient air into a photoacoustic cell. This is the only technique which offers real–time monitoring capability [24].

Other examples of the chemical analysis of environmental particulate matter are considered in Section 4 of this Chapter.

3 AUTOMATIC ANALYSERS FOR GASEOUS POLLUTANTS

Table 2.2 summarises the commonly employed methods for the measurement of gaseous air pollutants and gives information on the required minimum sample collection period, instrument response time and detection limit. The following sections consider the techniques appropriate for each pollutant.

3.1 Carbon Monoxide

The main analytical method used in the determination of carbon monoxide has traditionally been non–dispersive infrared analysis (NDIR). This method, which is capable of operating on a real time basis, is based on the absorption of infrared radiation by CO. Infrared radiation is split into two parallel beams, one passing through the sample compartment and one through a reference cell. The two beams finally pass into the absorption chamber which consists of two matched cells, both containing CO separated by a diaphragm. The CO in the detector cell only absorbs radiation corresponding to its characteristic frequency.

The two detectors are balanced electronically with non–absorbing gas in the reference cell and no CO in the sample cell. The introduction of any CO gas into the sample cell will absorb radiation which increases temperature and pressure in the detector cell of the absorption chamber opposite the sample cell and displaces the diaphragm. This displacement can be monitored electronically.

With this analytical system a calibration curve must be constructed using standard CO samples, in order to find the steady linear relationship between the diaphragm displacement and the CO concentration.

The NDIR technique can suffer from a number of interferences according to the nature of the sample. For example water vapour can produce an interference. This can be minimized by initially passing the air sample through a drying agent such as silica gel. Interferences can be further reduced by maintaining a constant humidity in both the sample and the calibration gases through refrigeration and finally with the use of narrow band optical filters. Minimum detectable limits for this technique are nominally 0.5 ppm. An improvement to the technique is gas–correlation IR.

Gas chromatographic techniques in combination with flame ionization detectors have been used for the evaluation of CO. With the sequential use of both oxidation and reduction catalysts, an air sample can be freed of all of its components except for methane and unmodified CO. The methane (CH_4) and CO can then be separated using chromatography. Carbon monoxide is catalytically reduced to CH_4 and measured in a flame ionization detector [25]. Minimum detectable limits for this technique are nominally of the order of 10 ppb and sample collection is on a cyclic basis over periods of a few minutes.

Table 2.2
A summary of some of the most commonly employed methods for the measurement of gaseous air pollutants

Pollutant	**Measurement technique**	**Sample collection period**	**Response time (continuous techniques)**	**Minimum detectable concentrations**
Total Hydrocarbons	Flame		0.5 sec	10ppb C
Specific hydrocarbons	Glc	b		< 1 ppb
Carbon monoxide	Ndir		5 sec	0.5 ppm
	Catalytic methanation/fid	c		10 ppb
	Electrochemical cell		25 sec	1 ppm
Sulphur dioxide	Absorption in hydrogen peroxide/titration	4 hr		2 ppb
	Absorption in tetrachloromercurate/spectrophotometry	15 min		10 ppb
	Flame photometric analyser		25 sec	0.5 ppb
	Fluorescent analyser		2 min	0.5 ppb
Oxides of nitrogen	Diffusion tube		1 week	2 ppb
	Chemiluminescent reaction with ozone		1 sec	0.5 ppb
Ozone	Chemiluminescent reaction with ethene		3 sec	1 ppb
	UV absorption		30 sec	3 ppb
Peroxyacetyl nitrate	Glc/electron capture detection	c		1 ppb
	Glc/conversion to NO_2/chemiluminescence	c		30 ppt

a) Time taken for a 90% response to an instantaneous concentration change
b) Grap samples of air collected in an inert container and concentrated prior to analysis.
c) Instantaneous concentrations measured on a cyclic basis by flushing the contents of a sample loop into the instrument.

Electrochemical techniques with a response time of up to 25 seconds and minimum detectable concentrations of 1.0 ppm can also be used for CO. These techniques are generally less accurate in comparison with NDIR and catalytic methanation–FID techniques. The main advantages of the electrochemical technique are the compact size of the instruments and the marked price advantage over its competitors.

A recently introduced method for the evaluation of CO has been developed at the UK National Physical Laboratory. This system operates on spectroscopic principles using tunable lasers as the radiation source. Every species in the atmosphere has a characteristic absorption spectrum. By alternately tuning a laser to and away from a characteristic line in this spectrum and measuring the resulting absorption, the pollutant concentration can be calculated. A range of pollutants can be assessed with this technique including CO, HCl, NO, NO_2, HNO_3, NH_3 and H_2S as well as some chlorofluorocarbons (CFCs) and light hydrocarbons. Sensitivities exceeding one part in 10^8 by volume, dependent on the particular gas under study, have been achieved using this technique [26,27].

3.2 Oxides of Nitrogen

The direct measurement of NO is difficult due to a lack of distinct properties associated with it. Thus early analytical techniques were based on the conversion of NO to NO_2, which is more easily measured. A number of techniques have been used including non–dispersive and dispersive infrared (IR), ultraviolet (UV), electrochemical sensors and chemiluminescence.

The non–dispersive infrared adsorption method (NDIR) is based upon the infrared absorption spectrophotometry principle, discussed in the preceding section. NDIR for NO_2 is subject to interferences from water vapour, CO, CO_2 and some hydrocarbons. A number of these can be removed by scrubbing but this can often inadvertently remove both NO and NO_2. NDIR instruments typically operate over a range of between 1 to 10^6 ppm, with response times in the region of 1 to 10 seconds.

The non–dispersive ultraviolet absorption method (NDUV) is used for the measurement of NO_2 only. Typical measurements are based on the absorption of radiation at 436 nm. NDUV instruments are more sensitive than NDIR instruments and do not suffer from the same interferences. They are affected by the presence in the test sample of SO_2, particulate matter and by the loss of nitrogen dioxide due to deposition on the walls of the sample cell. These problems can be eliminated by electronic filtering, removal of fine particles and the heating of the sample cell [3].

The currently preferred analytical technique is based on chemiluminescent reaction between NO and O_3 response times for this type of instrument are fast and typically between 1 and 2 seconds. The disadvantages are its high cost and the need for the production and disposal of the ozone gas reagent [28].

Following the oxidation of NO to NO_2, the excess ozone may react with NO_2 to form nitric acid. Water can enter the instrument through the air intake and thus HNO_3 can be formed which is deposited on the coolest part of the analyser, the optical filter. Recently it has been found that during continuous operation, typically over periods of approximately 6 months, the NO_x chemiluminescence analyser undergoes a sensitivity loss of about 50%. This has been found to be caused by a white deposit (NH_4NO_3 and unidentified organic material) forming on the glass filter which divides the reactor and photomultiplier tube. Cleaning this surface, however, restores the original sensitivity of the analyser [28].

Grosjean *et al* [29] have observed that NO_x chemiluminescence analysers respond to a number of organic pollutants and mixtures. Two interferences were observed. A number of nitrogenous pollutants including alkyl nitrates and nitrites may be reduced to NO by some catalytic converters and are thus recorded as NO_x. Secondly, many organosulphur compounds are seen as NO, due to the fact that they undergo a chemiluminescent reaction with ozone. Thus the NO_x data derived using a chemiluminescent NO_x analyser should be treated with more caution than originally considered necessary.

Electrochemical cell instruments use a sealed module with a semi–permeable membrane through which an air sample is passed. NO_x can diffuse on the surface of a sensing electrode. The electrochemical oxidation of NO and NO_2 into nitrate produces electrons that cause a current to flow between the sensing electrode and a counter electrode. The current produced is thus proportional to the NO_x concentration. The results derived from this method are positively affected by SO_2 and therefore this needs to be scrubbed from the sample prior to examination. These instruments tend not to perform as well as infrared monitors but have the advantage of being compact, simple to maintain and operate and they also display short response times, typically in the region of 5 seconds. Electrochemical cell analysers can also be used for the measurement of sulphur dioxide and carbon monoxide [1, 3].

The Palmes Tube has been widely used for the low level determination of NO_2 [30]. It consists of a hollow acrylic tube, open at one end, with coated stainless steel screens at the closed end. The nitrogen dioxide is trapped as nitrate ions in triethanolamine which coats the screens. The Palmes Tube is normally used as a passive sampler, the tubes traditionally exposed for weekly periods, in both indoor and outdoor locations [31]. Diffusion tubes are a cheap and reliable method. They require no power and need only limited attention at the start and end of each sampling period. Minimum detection dose limits of about 300 ppb–hr have been reported using colour developing reagents and a spectrophotometer. This corresponds to a detection concentration limit of 2 ppb with a tube exposed for 7 days. Ion chromatography combined with the use of concentrator columns, to allow the total sample to be analysed, can achieve minimum detection dose limits in the order of 30 ppb–hr.

3.3 Sulphur Dioxide

The measurement of sulphur dioxide has involved the use of a number of operating principles, several of which are discussed in this section. The conductometric method relies on the scrubbing of SO_2 through a sulphuric acid/hydrogen peroxide reagent. This produces an increase in the ionization of the solution which can be detected as a change in electrical conductivity. Conductometric SO_2 analysers are not recognised as a standard or 'equivalent' method by the US EPA. The widespread use of these analysers can be credited to their high sensitivity, fast response, minimal maintenance and simple operation. The main disadvantage is that they have poor selectivity for SO_2 but can be used in situations where SO_2 is assumed to be the principal pollutant. Concentrations in the parts per billion range can be monitored with this technique [32].

Continuous conductivity analysers have been developed in which the reacting agent is replenished after each sample determination. Interference from ammonia reduces the conductivity and thus removal of ammonia from the sample stream prior to analysis should be considered in environments with high ammonia concentrations [33].

Two relatively new techniques for the detection of SO_2 are flame photometry and more recently pulsed fluorescence. When sulphur compounds are burnt in a hydrogen flame the sulphur is raised to an excited diatomic state. Upon return to its normal state the sulphur emits energy in the UV region between 300 – 430 nm. This can be detected using a light sensitive photomultiplier which produces an analogue readout proportional to the SO_2 concentration. To analyse SO_2 selectively, a narrow band filter (394 nm) is placed directly in front of the photomultiplier tube.

The most recent continuous technique for SO_2 is based on the principle of pulsed fluorescence. With this technique the air sample is passed through a permeation drier and then into a short fixed path length cell which is subjected to a source of pulsed UV light within the range of 190 – 203 nm. The measurement is based on the fluorescence resulting from the excitation and de–excitation of SO_2 molecules between 240 – 420 nm. The fluorescence is detected by a photomultiplier tube and is proportional to the concentration of SO_2 in the air sample. Table 2.3 summarises the relative advantages and disadvantages of flame photometric and pulsed fluorescence methods.

TABLE 2.3

Advantages and Disadvantages of Flame Photometric and Pulsed Fluorescence Techniques for SO_2 determinations [ref. 34].

Type	**Advantages**	**Disadvantages**
Hydrogen flame photometry	US EPA approved High sensitivity Not flow dependent. Detects other sulphur compounds when coupled with GC.	Dependent on sample pressure Critical hydrogen flow dependent. Uses compressed hydrogen. Pneumatic compressed air operated solenoid. Bulky due to cylinder requirements.
Pulsed fluorescence	US EPA approved. No consumables Portable Fast start up Not flow dependent. Low zero drift.	Subject to O_2 quenching Subject to partial pressure effects in sample cell.

3.4 Total Hydrocarbons

Ambient measurements of gaseous hydrocarbons are normally divided into methane and nonmethane components. Methane (CH_4) constitutes the major component of THC and often comprises over fifty per cent of the total background concentrations. Thus any analytical technique used to monitor THCs does not need to be particularly sensitive. Nonmethane hydrocarbon (NMHC) levels within the atmosphere are considered more of a pollution problem than methane levels, since methane reacts comparatively slowly within the atmosphere. Hydrocarbons can either be monitored on a continuous real time basis or samples can be bulk collected and analysed at a latter date.

An increasingly popular method for the collection and analysis of organic pollutants involves passing polluted air through an adsorbing material (Table 2.4). The pollutant is then desorbed from the adsorbing material using heat or a suitable solvent such as carbon disulphide (CS_2).

TABLE 2.4

Adsorbing Materials used in Hydrocarbon Collection

Type	Description
Porapak P	Porous polymer of styrene and divinyl benzene – limitations in the temperature it can withstand before it bleeds onto the chromatographic column.
Carbon molecular sieve	Carbosieve thermally cracked vinylidene chloride. – temperatures of > 400°C are required to desorb some organic volatiles. Could cause pyrolysis in some compounds.
Tenax	2,6–diphenyl–p–phenyleneoxide polymer – Difficulties in removing some of the higher molecular weight compounds.
Polsorbimide–1 & Carbonchrome K5	Thermally graphited carbon black modified with polymethylsiloxane. – Good up to n–dodecane but problems with desorbing HC of higher molecular weight

Tenax is commonly regarded as the most suitable material, due to its excellent performance in adsorbing and desorbing pollutants, and also its ability not to lose materials during storage. Desorption can be achieved by using a thermal desorber connected in line to a cryogenic trap and gas chromatography.

The organics are liberated from the Tenax by the thermal desorber and concentrated by the cryogenic device prior to being sent to the chromatographic column as a single spike.

Charcoal tubes, developed by the National Institute of Occupational Safety and Health have been extensively used for the collection of hydrocarbons such as benzene. The NIOSH tubes have two sections, one to collect the sample and the other section to monitor breakthrough. At the end of a sampling period the ends are filed off the tube and the HCs are liberated from the charcoal with CS_2. Air samples can also be collected using sampling pumps, with the resulting samples stored under pressure prior to analysis within stainless steel canisters (volumes of up to 6 litres). Prior to use the canisters must be repeatedly flushed with zero grade air [35]. Sampling with porous polymer adsorbents and stainless steel canisters have been shown to be complementary methods [36]. Polymers tend to be preferable for hydrocarbons greater than C_8 while the stainless steel canisters were best suited to low molecular weight compounds.

Two distinct methods have been widely used to measure THCs utilising non–dispersive infrared and flame ionization techniques. Flame ionization detectors (FIDs) consist of a hydrogen–air flame burning between two electrodes. Any substances introduced into this flame produces charged ions, which are subsequently collected on the electrodes and converted to an electrical signal. The magnitude of this signal is proportional to the concentration of the material, measured in terms of carbon counts, being monitored.

Non–dispersive IR analysers use the principle that HC molecules absorb IR radiation at particular frequencies. Accurate measurements of total hydrocarbons are only possible when the sample is mainly composed of aliphatic alkanes of C_2 upwards. Thus it should be noted that measurements made using FIDs are not strictly comparable with those using NDIR analysers [37].

Flame ionization detectors are generally more sensitive than NDIR, as NDIR detectors suffer from interferences from high concentrations of CO_2, CO and water vapour. A number of problems have been noted with flame ionization detectors particularly with possible correlation discrepancies between different commercial instruments due to differences in operating conditions. The relative importance of a number of conditions such as instrument sample pressure, bypass flow rate, diameter and length of sample lines and operating temperatures have been identified. [38].

Nonmethane hydrocarbon data can be obtained from the collection of air samples which are subsequently analysed using chromatography. This would identify and quantify a number of individual HCs which could be summed to produce a value for both NMHC and total hydrocarbons [39]. This method can provide more reliable and accurate determinations in comparison with continuous methane – total hydrocarbon analysers. However this technique of hydrocarbon separation followed by summation is very time–consuming and thus expensive. In the majority of methane – nonmethane hydrocarbon FID analysers, the methane level is determined following the catalytic removal of the NMHC component. A separate measurement is made of the total hydrocarbon level and the NMHC level is determined by subtraction.

The majority of continuous methane – nonmethane instruments do not provide reliable data for concentrations of less than 1 ppm. They present a number of problems particularly with the subtractive nature of the results and with the possible non–uniform response per carbon for different compounds due to O_3 interferences [40]. A simple method for the accurate determination of low levels of total NMHC has recently been developed. This involves collection of air samples in tedlar bags and cryogenic concentration of the nonmethane species followed by thermal desorption of the sample into a flame ionization detector [39]. The vapour pressure of methane is higher than that of any other HC. Thus within ambient air methane can be separated from other HCs by condensing the heavier ones on a suitably cold surface. The method described by Cox *et al* [39] requires manual operation and thus cannot be used to detect the rapid concentration fluctuations found within ambient air. An oxygen or argon trap can be used to collect the NMHC component selectively; this is then passed to the FID without chromatographic separation [41].

3.5 Individual Hydrocarbons and PAH.

Chromatographic analysis is commonly used to analyse individual hydrocarbon compounds. With standard gas–solid chromatography the sample is injected into a column containing a packing material. This packing material (alumina, charcoal, silica gel, plastic granules (porapak) or molecular sieves) has different affinities for each particular component in the sample, and separates each component upon heating at characteristic temperatures. The column can be subjected to heating in controlled steps with temperature programming typically increasing the temperatures from a two minute holding phase up to 300°C for ten minutes, with a ramping rate of 10°C per minute.

As the temperature increases different hydrocarbons are released to the detector. Thus the detector can monitor the amount of hydrocarbons released at a particular time and temperature. This is plotted on a chromatograph, as a series of peaks. The area under each peak allows a quantitative determination of each HC present. The identification of these HCs can be established by referring to standard chromatographs given that individual HCs are released at characteristic temperatures.

Practically any mixture can be separated using this technique. The separation achieved depends on the column, the temperature, the detector and flow rate. For a quantitative determination these controlling variables have to be kept constant for both the test sample and standard calibration sample. With this method two very similar HCs can be represented by an individual peak on the chromatograph. To separate these substances further, the column length can be increased. This increases the column efficiency but also increases analysis time. Many hundreds of compounds can typically be determined in a single sample using GC, and thus it represents a very powerful analytical technique.

The choice of detector to use in combination with chromatographic analysis is dependent primarily on the specific compounds under study. Certain detectors are more sensitive to particular compounds.

Flame ionization detectors are in general the most commonly used instrument. Another detector, using the principle of electron capture has been increasingly adopted for the study of PAHs. This detector has a high affinity for compounds with free electrons such as benzo(a)pyrene and benzo(k)fluoranthene. It displays a better response than FID, even though it varies from compound to compound but is limited by a narrower linear range of response. Direct photoionization detectors linked to gas chromatography (PID/GC) display lower detectable limits of 0.3 ppb for C_6H_6 and 1.0 ppb for C_8–alkylbenzenes. This technique is however inconsistent at low concentrations of higher alkylbenzenes.

With paper, thin layer or high pressure liquid chromatography the most commonly used detectors are ultraviolet and fluorescence spectro-photometers. All PAHs, for example, absorb UV radiation with sharp characteristic absorption peaks. However, fluorescence spectrophotometry is often 100 times more sensitive than UV and most PAHs are fluorescent. This technique has a number of problems with interferences from trace levels of unknown compounds and variations in the fluorescent spectra can occur with different solvents. This technique thus requires skilled operation and is time consuming and less precise than UV spectrophotometry.

Mass spectrometry linked to gas chromatography is an extremely powerful and sensitive method for the determination of organic compounds [42]. High performance liquid chromatography is another technique which has found applications for hydrocarbon analysis [1], particularly in situations where gas chromatography may not be appropriate. Compounds which come into this category include thermally unstable polyaromatic hydrocarbons and higher molecular weight species with low volatility.

3.6 Secondary Pollutants

Chemiluminescent analysers have also been adapted for the measurement of ozone concentrations. Ozone analysers can be based on the chemiluminescent reaction (585 nm) between O_3 and Rhodamine B, absorbed on silica gel. The resultant emission produces a measure of the mass of ozone flowing over the dye, per unit time. The detection limit of this method is between 1 and 10 ppb.

Gas chemiluminescent reactions are also utilized such as the reaction between ethene and ozone. The chemiluminescent reaction is outlined in the following reaction sequence.

$$C_2H_4 + O_3 \rightarrow \text{OZONIDE} \rightarrow CH_2O^*$$
$$CH_2O^* \rightarrow CH_2O + h\nu$$

Emissions are centred on 435 nm and hence no interference from nitrogen dioxide is encountered. This method is sensitive to ozone levels as low as 1 ppb.

Ozone can also be monitored using its ability to absorb UV light at 254 nm. The amount of absorbance is directly proportional to the concentration of ozone in the sample which is determined using a precalculated calibration curve. For monitoring situations where a dusty atmosphere is likely, for example near roads, prefiltering of the air sample should be undertaken as suspended particulates cause an increase in absorbance, particularly in the region below 300 nm. This method can be used for ozone monitoring at levels as low as 3 ppb. The background concentrations of O_3 lie in the region of 10 to 50 ppb and thus both the UV absorption and the chemiluminescence methods are sufficiently sensitive for ambient measurements.

Interferences can occur for UV absorption techniques from other pollutants such as mercury and hydrocarbons which both absorb UV light. However, this problem can be eliminated by taking two measurements, one of the absorbance of the natural air sample and one after the O_3 has been changed to O_2 using a catalyst. The differences in absorbance represents the ozone content of the sample.

Peroxyacyl nitrates, a group of secondary nitrogen—based pollutants, are commonly collected in the field through cryotrapping. The resulting samples, stored at liquid nitrogen temperatures, are analysed using gas chromatography linked with electron capture detectors (GC–ECD) or infrared techniques [43] or more recently, by liquid phase chemiluminescence after conversion to NO_2.

4 ELEMENTAL ANALYSIS

The most important aspect of improved analytical methodology is the great advance in specificity which has been achieved in recent years. Many of the techniques in use twenty or more years ago suffered from major interferences and have been shown subsequently to yield unreliable results for environmental monitoring. Advances in specificity, precision, and detection capability, coupled with major developments in sampling and preparative methods have enhanced the importance of investigating the associations between environmental contamination and potential health risk. In review, analytical methodology has dramatically changed from basic wet chemical, colorimetric and polarographic methods to sophisticated instruments, such as atomic absorption spectrometry (utilising both flame, F–AAS and electrothermal, E–AAS modes); atomic emission (AES) and fluorescence (AFS); anodic stripping voltammetry (ASV); X–ray fluorescence (XRF); mass spectrometry techniques, such as ion probe and spark source (MS); neutron and proton activation analysis (NAA, PAA) utilising both instrumental, pre–chemical separation, and radiochemical methods; inductively coupled plasma–source atomic emission (ICP–AES) and mass spectrometry (ICP–MS).

Overall, the ideal analytical technique for undertaking environmental elemental analysis must offer; (a) qualitative, semi–quantitative and quantitative analysis, (b) high degree of specificity, (c) very low detection limits, (d) simple sample preparation, (e) simple interference–free spectra, (f) high throughput and low cost per determination, and (g) unattended batch–mode operation. Whilst there are a large number of methods of analysis available for determining chemical pollutants in biological and environmental matrices most fail to satisfy the above criteria. In fact as the following review will show whilst some methods are impressive in terms of good detection limits or interference–free precision this is not always synonymous with high levels of accuracy – especially for values determined in real environmental matrices and under routine experimental conditions.

4.1 Wet Chemical and Sample Preparation Methods

Titrations and gravimetry have long proved to be too insensitive for most environmental analytes of interest, and require lengthy analysis times. In terms of general wet chemical sample preparation, many basic methods require the sample to be dissolved before analysis. Extractive leaching by chelation can be achieved using such typical solvents as esters or ketones, i.e. MEK or methyl–isobutyl–ketone (MIBK) in conjunction with typical chelating agents such as EDTA or ammonium pyrolidine dithiocarbamate (APDC). This method has been widely used for the extraction of many heavy metals from acidic solution [44] but is associated with poor precision or reproducibility levels. Wet ashing with strong oxidising agents, such as aqua–regia, sulphuric acid–perchloric acid, or a mixture of acids including HF, perchloric, phosphoric and sulphuric acids, is normally useful for analysing most environmental samples. For rapid and low temperature digestion, peroxide is often used. In contrast dry ashing has the problem of inorganic species (Hg, Cd, Cu, As, Pb, and

Ag) being lost during preparation; especially at temperatures reaching between 500 and 600°C [45]. Also the resulting residue is normally difficult to dissolve in acid only. The use of all pre–analysis chemical procedures introduces the prospects of possible analyte loss, contamination, and 'blank correction' problems.

4.2 Polarography and Voltammetry

Both methods have been widely applied to the measurement of various metals and metalloids (Cd, Pb, Hg, Cu, Zn, Ni, Co, Se, etc.) with very good low detection limits [46] provided sample preparation is meticulously undertaken so as to eliminate possible masking effects. Simultaneous determination of trace metals is possible usually by various versions of stripping (SV) and differential pulse stripping voltammetry (DPSV) utilising various working electrodes. DPSV competes favourably with other basic trace element methods with definite cost advantages. Voltammetry has a low vulnerability to systematic errors, although contamination problems can occur for some metals as a result of pre–digestion procedures.

4.3 Atomic Absorption Spectrometry

F–AAS and E–AAS have been widely used for numerous applications in the assessment of highway pollution [47]. Many factors influence both sensitivity and detection capability of AAS including nebulisation efficiency [48–50], flame composition for F–AAS [51], optical efficiency – especially in terms of the use of hollow cathode or electrodeless discharge lamps [52], and for E–AAS, the overall programming of the drying, pyrolysis, and atomisation stages [53]. Improved detection does occur by the use of background correctors which reduce both light scattering and molecular absorption effects [48,53]. In order to improve both selectivity and sensitivity for AAS, various pre–treatment separation procedures have been used to separate the element from the sample matrix, thereby reducing possible interference effects and possibly pre–concentrating the analyte [54–56]. Pre–concentration methods used for AAS include ion–exchange [57], electrodeposition [58–63], co–precipitation [64], and evaporation methods. Whilst E–AAS is considered to be a more sensitive method there can be numerous interference effects rendering poor precision or reproducibility. These include pyrolysis losses, anion/cation interferences, condensation effects, carbide and nitrate formation [52]. Various techniques have been used to compensate for these interferences, including standard addition, internal standard methods, and the use of protective and reducing agents.

Hydride generation [65, 66] provides lower detection limits for some elements (Sb, As, Bi, Se, Te, Sn, ...); in fact 50 to 200 times lower than those of direct F–AAS and 5 to 10 times lower than E–AAS methods. Elemental Hg is also routinely determined using the cold–vapour AAS method which for the resonance line of 236.7 nm can provide detection limits of 0.002 ng/mL Hg [67,68].

In summary F–AAS is basically limited to single element analysis, giving μg/g detection limits in solids, but involves numerous problems for environmental analysis, for example, carbide matrix interferences and signal suppression by Na^+,Cl^-,K^+. This is of major importance in the analysis of highway samples which may contain high salt levels due to winter weather salting programmes. Although E–AAS in conjunction with Zeeman background correction can now provide ng/g detection limits, all AAS methods (including hydride generation and cold–vapour) lack speed and convenience for multi–element determinations, and for analysing large sample numbers. Limited dynamic range and poor reproducibility at low levels require multiple dilution steps to cover the wide analyte range of many elements in water, soil, and vegetative material.

4.4 Atomic Fluorescence Spectrometry

AFS has been used for environmental analysis far less than AAS or AES mainly as there are only about 30 elements for which intense sources are available [69]. The most important interferences in AFS are those due to nebulisation, dissociation, and quenching[70]. Modifications in flame compositions or mixtures, and the use of releasing agents [71] can lead to a reduction in these interferences. Non–flame sources for AFS have been used, such as ICP [72] with improvements for some elements (Mg, Ag, Pb, Bi, Th, Ga, and Zn) in terms of absolute detection capability.

4.5 X–Ray Fluorescence Spectrometry

XRF has overall good precision and is therefore widely used for environmental fingerprinting and elemental ratio studies. However it is restricted by matrix problems and is not suitable for ultratrace element investigations due to a somewhat limited detection power. XRF also needs careful standardisation and very frequently the use of preconcentration procedures, such as ion exchange materials as surface accumulators [73].

4.6 Atomic Emission Spectrometry

Flame–AES is widely used like AAS and suffers from similar interferences, such as, background effects, spectral and vapourisation interferences, and ionisation effects. Most of these can be minimised by using background correctors or alternative emission lines (with the relative change in detection limits), careful choice of flames, nebulisers and chemical additives (especially La or Sr), and the use of ionisation suppressants (such as K).

The development of inductively coupled plasma atomic emission spectrometry (ICP–AES) has dramatically enhanced the potential of undertaking multi–elemental environmental studies. The advent of specialised types of nebulisers [74], modifications in the position of the ICP source and the gas flow systems have enhanced the performance of ICP–AES. Alternatives to nebulisation have been explored including thermal and electrothermal vapourisation devices [75–59]. These devices enable samples of 1–200 μL to be used and yield approximately an order of magnitude improvement in detection power compared with nebulisation.

Laser ablation [80] has also been used in conjunction with ICP–AES though poor precision has meant that its use has been limited. Other alternative sample–introduction techniques used for ICP–AES include gas or liquid chromatography, hydride generation [81]; the latter leads to improved sensitivity for P, Pb, and Se [82].

A major problem with ICP–AES is the overlapping of emission line interferences, particularly for heavy ions. The wide choice of usable lines means that the spectral interference can be minimised by the use of a different line though of course this will affect the detection limit obtained for that element. Ionisation effects such as when an increase in electron density in the source (due to the sample matrix) causes an enhancement in the emission of neutral atoms, can be important. During analysis they can be eliminated by the use of sample and standard matrix matching, though for signal optimisation it is arguably preferable to extract the analytes into a more favourable matrix. Various reports also suggest that the use of high concentrations of organic and inorganic acids in the sample matrix may lead to enhancement or reduction of the signal as a result of modification in solution uptake rate [83] or due to nebulisation effects.

4.7 Mass Spectrometry

MS is a very powerful tool for trace and ultratrace analysis; especially spark source mass spectrometry (SSMS) which has a comprehensive element coverage with a determination range to below 10 pg (ref. 84). Major disadvantages are the low daily sample throughout numbers and the relative poor levels of accuracy with the exception of the time consuming and cumbersome method of isotope dilution mass spectrometry (IDMS) [85].

4.8 Neutron Activation Analysis
(instrumental, pre–chemical separation and radiochemical)

NAA provides good precision and accuracy at the ultra–trace (ng/g) level, especially for radiochemical methods [86] but is severely restricted for small sample sizes, or liquid samples. The latter is due to fears of radioactive leakage after activation within the irradiation sites of a thermal reactor. Most radiochemical methods are also very time–consuming with complex separation schemes applied directly to radioactive solutions.

Multi–element analysis based upon gamma–ray spectrometry is of limited usefulness for undertaking large sample number studies, as many elements are measurable only after long irradiation and analysis times; for example, ^{60}Co ($t^1/_2$ = 5.27y), ^{51}Cr ($t^1/_2$ = 27.7d), ^{59}Fe ($t^1/_2$ = 45.1d), and ^{65}Zn ($t^1/_2$ = 244d). The measurement of many important environmental elements using short–lived nuclide irradiation conditions (^{28}Al, ^{49}Ca, ^{66}Ca, ^{20}F, ^{128}I, ^{27}Mg, ^{56}Mn, ^{101}Mo, ^{86m}Rb, ^{77m}Se, and ^{52}V) is normally subject to reduced precision as a result of increased gamma–ray background activity induced by the presence of numerous major matrix interferences, such as those from ^{32}P ($t^1/_2$ = 14.3d), ^{24}Na ($t^1/_2$ = 14.96h), and ^{38}Cl ($t^1/_2$ = 37.18m) (refs. 87,88). Similar precision problems arise from the high elemental content of Al and Sc whose respective nuclides ^{28}Al ($t^1/_2$ = 2.3m) and ^{46}Sc ($t^1/_2$ = 83.8d) induce pulse pile–up counting errors, especially for the analysis of soil and sediment samples. Acceptable precision and accuracy for NAA is achieved by cyclic activation (for very short–lived nuclides) or by the use of Compton suppression for low gamma–ray energy nuclides [89]. Various complicated pre–irradiation chemical separations and/or pre–concentration steps can be used in conjunction with NAA methods but this introduces the prospects of possible analyte loss, contamination and blank–reagent correction problems. A basic disadvantage of routine thermal NAA for environmental and in particular highway pollution studies is the inability to detect certain important elements especially Pb, Si, Li, B, and P. This is due in part to poor nuclear cross–sections, non–activation by neutrons, or as in the case of P, non–production of a gamma–ray for analysis. Overall NAA is extensively used for multi–element studies, especially for solid samples where complicated and imprecise digestion procedures are not necessary. NAA also has the ultimate sensitivity for most elements, but as outlined above there are many areas where reservations must be considered before using this method. Also it is a very expensive technique requiring sophisticated equipment such as a nuclear research reactor with associated gamma–ray spectrometry counting systems.

4.9 Inductively Coupled Plasma–Source Mass Spectrometry

In recent years ICP–MS has developed as a new multi–element method which offers exceptional ultra–trace element sensitivity which is vital for measuring the concentrations of many biologically and environmentally significant elements (Li, B, Al, Co, Cr, Mn, V, Pb, and U). A number of applications of ICP–MS to environmental analysis have been published: soil and vegetation [90, 91] marine sediments and waters [92–95], and geochemical samples [96]. Specific studies on the multi–element contamination of the surroundings of major British motorway environments have been reported based on the work undertaken using the VG Elemental ICP–MS Plasmaquad [97].

There are many features which make ICP–MS an attractive technique for environmental trace–element studies. ICP–MS provides:

a) good multi–element capability for determining a large number of elemental concentrations over the range of μg/g to sub–ng/g, especially in the presence of major environmental matrix components, such as Na, Cl, S, P, and Si.

b) low detection limits in aqueous solution typically of the order 0.01 to 0.1 μg/L which are desirable for evaluating water quality in fresh, groundwater and seawater samples [95].

c) the potential for undertaking small sample analysis, from, for example, plant extracts, seeds, or air particulate filters, where the lower detection limits of ICP–MS enables smaller sizes to be analysed than can be achieved by other methods. This avoids the necessity for preconcentration steps which introduce the possibility of contamination or analyte loss.

d) the direct analysis of many toxic metals (Cd, Hg, Pb), heavy metals (As, Sb, Be, Cr, Cu, Ni, Se, W, Ag, Tl, Th, Zn, U, etc.), and many other elements not readily determined by other analytical techniques (Li, B, Be, Si, and rare–earth metals).

e) extremely simple spectra–enabling routine qualitative, semi-quantitative and quantitative analysis.

f) the ability to determine isotopic ratios or undertake isotope dilution studies on multiple or single elements at levels below 0.1 μg/L with precisions of 0.2 to 1% relative standard deviations [98].

g) the opportunity to investigate a wide range of water–related matrices as a result of the expanding range of sample introduction techniques, including conventional pneumatic, ultrasonic, and high–solids nebulisers; and electrothermal volatilization or laser ablation capability for undertaking the direct analysis of solids.

h) routine sample throughput can be achieved with 2–3 minutes required for analysis and subsequent washout to minimize 'memory' effects between successive sample analyses. A typical workday enables 100–150 samples to be measured in the multi–element mode. This provides an excellent basis for undertaking environmental studies where large sample numbers are normally involved.

i) the dynamic range of ICP–MS extends from below 0.1 to 10^6 ng/mL with matrix tolerance in solutions of 0.1% solids content.

Overall ICP–MS appears to be the 'ideal' trace element technique. However various analytical problems can be encountered during the analysis of biological samples. It is very important to consider the potential risk of interferences below 80 m/z, especially in relation to the type of acid which may be used for sample dissolution prior to analysis. Overall, the use of H_2SO_4 and HCl as a solvent is undesirable. Sulphur has a low ionisation energy (10.36eV) and yields various oxide and hydroxides of sulphur which occur at masses of 48, 49, 50, 51 and 52 m/z; coincident with the various isotopes of Ti, V, Ca, and Cr. Hydrochloric acid (as oxides and hydroxides of chlorine) produces a series of interference peaks, the most important being at mass 51 m/z ($^{35}Cl^{16}O$) and 53 m/z ($^{37}Cl^{16}O$) which coincide with mono–isotopic vanadium and a small isotope of chromium (9.55%). Due to the high ionisation energy of nitrogen, 1% nitric acid causes few undesirable molecular peaks and is therefore commonly used. Another problem is direct sample analysis which can result in ion rate suppression due to the presence of high dissolved solids content. Direct analysis of samples containing high levels of protein or fat can result in solids deposition on the extraction aperture, skimmer or the plasma torch injector tube; which will ultimately shut–down the system. For some matrices which contain various analytes at high concentrations the existence of memory effects,mainly due to volatilisation of excess analyte previously deposited upon the walls of the spray chamber or associated nebuliser transfer lines, will result in decreased precision and accuracy. The overall detection capability of ICP–MS therefore allows an easy solution to this problem through the simple dilution of such problematic samples. Although ICP–MS has only been used for a limited number of pollution applications it appears to have a very bright future, especially with the development of laser ablation and electrothermal volatilisation techniques [99,100].

4.10 Summary

Many of the reviewed analytical methods provide adequate levels of specificity and precision for undertaking both mono– and multi–element analyses. However, a number still suffer from various matrix interference problems, or lack of speed and convenience for undertaking routine environmental analysis. The multi–element techniques provide the greatest advantages, especially ICP–AES, NAA, and ICP–MS. Table 2.5 lists the detection limits for various analytical techniques (F–AAS, E–AAS, NAA, ICP–AES, and ICP–MS) which are routinely used to analyse a difficult environmental matrix; namely, natural water. Comparison is made with reported elemental values for river water (ref. 101). Whilst these values illustrate the impressive detection limits of ICP–AES and especially ICP–MS, in general less attention should be paid to improving (or quoting) detection limits, but instead should be given to improving accuracy for most analytical techniques.

5 QUALITY CONTROL

In chemical trace analysis quality control of analytical activities may be based upon

1) the use of stable and homogeneous reference samples (reference materials, standard materials, SRMs),
2) inter–laboratory elemental comparisons (with analysis using varying analytical methods, analysts, and instruments), and
3) methodology comparisons, such as the use of different sample preparation methods.

In evaluating the accuracy of an analysis, biased data may result from poor methodology; improper instrument calibration; variable experimental techniques; and a lack of a reliable control. The traditional method used for assessing systematic errors in analytical procedures is the determination of recovery of known amounts of analytes added to the matrix under investigation. However, this procedure for many techniques does not allow for the fact that some analytes, when added as solutions, behave differently from the same analytes present naturally in the sample matrix (especially for those techniques which require excitation of the analyte before quantification). A review of the literature reveals that most published papers using the various analytical techniques already outlined for assessing the elemental status of particular polluted environments lack any form of quality control. Such assessments are extremely important since the results of these analytical measurements may be the basis upon which economic, administrative, medical or legal decisions are taken.

5.1 Reference materials (RMs)

Reference materials are homogeneous stable materials with certified chemical or physical properties for use in calibrating instruments, validating laboratory data, developing methods of known accuracy, and referring data from different laboratories to a common base. In practice, RMs are substances for which one or more chemical, physical, biological or technical property have been measured reliably and are therefore considered to be known. Each material has a certificate which includes the nature and amount of various components, methods used, and a statement of reliability. For example, certification of

TABLE 2.5 **Detection limits for analytical techniques routinely used to analyse natural water**

Element	Concentration in river water [108]	Detection limit (μg/L)				
		F–AAS‡	E–AAS*	NAA‡‡	ICP–AES**	ICP–MS‡‡‡
Al	50	10	6	30	0.8	0.01
As	1.7	100	20	1	2	0.01
Ba	60	8	10	10	0.01	0.01
B	18	700	600	–	0.1	0.01
Br	20	–	100	20	–	0.01
Cd	0.02	2	0.02	5	0.2	0.01
Ca	13300	.5	0.06	500	0.01	10
Cr	1	5	1	0.5	0.8	0.01
Co	0.2	10	1	0.5	3	0.001
Cu	1.5	2	1	3	0.6	0.01
Fe	40	4	0.6	10	0.09	0.1
Mg	3100	0.3	0.01	50	0.003	0.01
Mn	8.2	5	0.1	0.1	0.02	0.01
Hg	–	(.2)***	20	1	10	0.01
Mo	0.5	30	8	0.2	0.1	0.01
Ni	0.5	8	2	0.6	0.1	0.01
K	1500	2	0.2	20	200	10
Se	–	(1)***	20	1	30	0.1
Sr	60	24	5	50	0.03	0.01
Sn	–	30	10	30	3	0.01
V	1	40	20	0.5	0.06	0.01
Zn	30	2	0.02	5	0.01	0.01

‡ Flame atomic absorption spectrometry [102]
* Electrothermal atomic absorption spectrometry [102–104]
‡‡ Neutron activation analysis [105,106]
** Inductively coupled plasma atomic spectrometry [102]
‡‡‡ Inductively coupled plasma–source mass spectrometry [107]
*** Special method

some reference materials at the US National Institute of Standards and Technology (NIST), formerly known as National Bureau of Standards (NBS) is done in one of three ways [108].

1) measurement by a method of known accuracy, by at least two different laboratories,
2) measurement by two or more independent and reliable methods, which give errors that are small compared to that required for certification, and
3) measurement with the co–operation of a group of qualified laboratories using a number of methods.

Following preparation all RMs are assessed for their homogeneity by determining numerous key elements (P, Fe, Mn, Zn, Rb, Cu, Cr, As, U) utilising samples of 500 mg or less taken at various locations of the available bulk material. It is important to note that most RM samples should be air–dried (at 85°C for up to 7 days) as they may contain a moderate content of hygroscopic moisture (weight loss ~ 3% on drying at 105°C). The analytical methods used for determining the levels of uncertainty of the elemental composition (or 'certification') are AAS, IDMS, IDSSMS, NAA, ICP–AES and now ICP–MS.

There are numerous agencies who are responsible for providing such materials; among them the major ones are NIST, the International Atomic Energy Agency, Vienna (IAEA), National Research Council of Canada, Ottawa (NRCC), National Institute for Environmental Studies, Japan (NIES), and Community Bureau of Reference, Commission Bureau of Reference, Commission of the European Communities, Brussels (BCR). Table 2.6 lists the various biological and environmental reference materials, and quotes the certified (C) or non–certified (NC) value for the most commonly measured environmental toxic metal, namely Pb. Ward [109] as part of the quality control assurance of a multi–element study which assessed the degree of environmental contamination along British motorways measured 23 elements (μg/g, dry weight) in NIST : SRM 1575 Pine Needles by ICP–MS. Figure 2.7 shows the mean concentrations of measured values as a function of certified values. Overall this study showed excellent agreement obtained between analytical and NIST values, providing a degree of confidence in the overall accuracy of the multi–element levels reported in this study. Similar results have been reported by Ward for Bowen's Kale [97], NIST:SRM 1577 Bovine Liver [110], IAEA–soil 5 [111], NIST:SRM 1571 Orchard Leaves [90, 91], and NIST:SRM 1643b Trace Elements in Water [112].

5.2 <u>Interanalytical Comparison Studies</u>

Such comparisons may be either inter–laboratory or inter–analytical techniques in which samples of homogeneous nature are analysed in order to establish a level of accuracy which

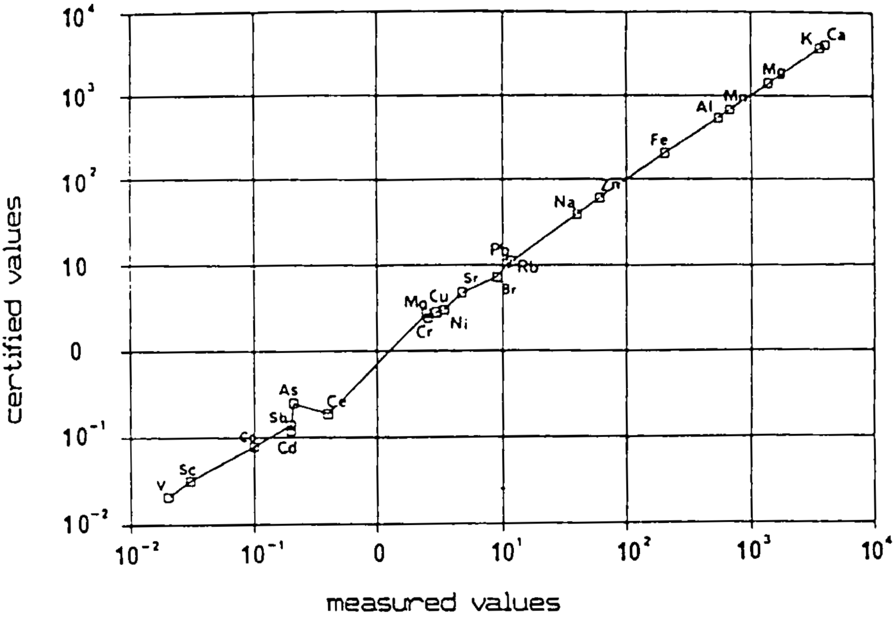


FIGURE 2.7 ***QUALITY CONTROL ASSESSMENT***
Between ICP–MS and Certified Elemental Values
for NIST : SRM 1575 Pine Needles Reference Material

provides an estimate of analytical bias or variability in experimental techniques. Some inter–laboratory comparative studies have reflected wide variances in the results, as shown by a reported study [113] of inorganic constituent analyses using various analytical techniques for milk powder. The ranges of mean values obtained by participating laboratories for the same concentrations of typical elements were: As (5 to 544 ng/g, Cd (1 to 1660 ng/g), Co (0.005 to 51 ng/g), Cr (0.02 to 52 μg/g), Se (25 to 313 ng/g), and V (0.002 to 20 μg/g). Analysis of these particular elements is generally considered to yield values with high uncertainties using most analytical techniques. Uncertainties of the elemental concentrations in reference materials are also based upon inter–analytical method comparisons. Gladney et al [114] reported the lead values determined by various analytical methods, namely atomic absorption (AAS), anodic stripping voltammetry (ASV), inductively coupled plasma emission spectrometry (ICP–AES), neutron activation analysis (NAA), photon activation analysis (PAA), X–ray fluorescence spectrometry (XRF), polarography (POL), isotope dilution mass spectrometry (IDMS), and spark source mass spectrometry (SSMS) for a series of NBS (NIST) reference materials (Table 2.7). More recent results for other reference materials have been reported for lead (Table 2.8) and other trace elements using inductively coupled plasma–source mass spectrometry in both the conventional solution nebulisation (ICP–MS) and laser ablation (LA–ICP–MS) modes; comparison is made with electrothermal (EAAS) and flame (F–AAS) atomic absorption spectrometry values [91, 115].

5.3 Methodology Comparisons

As part of the analytical process, the pre–analysis steps need to be critically evaluated, especially various dissolution or extraction procedures. The necessity of clean laboratories for sample preparation steps is of vital importance in order to minimize contamination during all steps of pre–analysis. As part of a regularly monitored laboratory control programme all analytical procedures should be routinely checked. As an example, Loring [119] compared the various dissolution and extraction procedures involving HF + (HNO_3 : HCl)–aqua regia or HF + ($HClO_4$) for determining the trace metal levels (Al, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Tl and Zn) of marine sediments. Metal concentrations were measured using flame and graphite furnace AAS, and ICP–AES. This study showed not only that HF+ aqua regia dissolution was the most effective procedure for removing the total metal concentrations from estuarine and coastal marine sediments, but also provided a regularly high standard of precise and accurate measurements. Similar inter–laboratory comparisons of sequential extraction procedures for the analysis of heavy metal in soils and sediments have been reported [120].

TABLE 2.6
(1 of 3)

Certified (C) or non-certified (NC) lead concentrations in international biological and environmental reference materials

Reference material		Lead content‡‡		Certified (C)/ Non-cert, (NC)
Biological				
Body fluids				
Animal blood	IAEA-A-13	0.18 ± 0.0	*	NC
Bovine blood	BCR-CRM-194	126 ± 4	‡	C
	BCR-CRM-195	416 ± 9	‡	C
	BCR-CRM-196	772 ± 11	‡	C
Blood serum	NIST-SRM-909	20.2 ± 0.5	‡	C
Milk powder	IAEA-A-11	0.27	*	NC
	NIST-SRM-1549	0.019 ± 0.003	*	C
Skim milk powder	BCR-CRM-063	0.1045 ± 0.0030	*	C
Urine	NIST-SRM-2670	0.01	*	NC
Animal and human tissues				
Animal bone	IAEA-H-5	3.1 ± 0.5	*	C
Animal muscle	IAEA-H-4	NVA		
Bovine muscle	BCR-CRM-184	239 ± 11	‡	C
Bovine liver	NIST-SRM-1577a	0.135 ± 0.015	‡	C
	BCR-CRM-185	501 ± 27	‡	C
Horse kidney	IAEA-H-8	NVA		
Pig kidney	BCR-CRM-186	306 ± 11	‡	C
Human hair	NIES-CRM-5	6.0	*	NC
Marine animals				
Albacore tuna	NIST-SRM-50	0.46	*	NC
Copepod	IAEA-MA-A-1	2.1 ± 0.3	*	C
Fish flesh	IAEA-MA-A-2	0.58 ± 0.06	*	C
Lobster	NRCC-Tort-1	10.4 ± 1.9	*	C
Mussel	NIES-CRM-6	0.91 ± 0.04	*	C
Mussel tissue	IAEA-MA-M-2	1.92	*	NC
Oyster tissue	NIST-SRM-1566	0.48 ± 0.04	*	C
Aquatic plants				
Aquatic plant	BCR-CRM-060	64.4 ± 3.5	*	C
	BCR-CRM-061	63.8 ± 3.2	*	C
Chlorella	NIES-CRM-3	0.60	*	NC
Sea lettuce	BCR-CRM-279	13.48 ± 0.36	*	C
Terrestrial plants				
Bowen's kale	–	2.49 ± 0.57	*	C
Citrus leaves	NIST-SRM-1572	13.3 ± 2.4	*	C
Cotton cellulose	IAEA-V-9	0.25 ± 0.06	*	NC
Hay powder	IAEA-V-10	1.6 ± 0.5	*	C
Olive leaves	BCR-CRM-062	25.0 ± 0.06	*	C
Pepper bush	NIES-CRM-1	5.5 ± 0.8	*	C
Pine needles	NIST-SRM-1575	10.8 ± 0.5	*	C
Tomato leaves	NIST-SRM-1573	6.3 ± 0.3	*	C

Table 2.6 (2 of 3)

Others				
Brewers yeast	NIST-SRM-1569	350	‡	NC
Wheat flour	NIST-SRM-1567	18	‡	NC
Rye flour	NIST-SRM-1568	32	‡	NC
Mixed human diet	IAEA-H-9	0.15 ± 0.07	*	C
Environmental				
Ash and aerosols				
Air filter	IAEA-Air-3/1	0.105 ± 0.008	*	C
City Waste Ash	BCR-CRM-176	10.87 ± 0.17	**	C
Cool fly ash	IRANT-ECH	70.7 ± 11.3	*	NC
	IRANT-ENO	41.7 ± 9.6	*	NC
	IRANT-EOP	40.7 ± 8.1	*	NC
	NIST-SRM-1633a	72.4 ± 0.4	*	C
	NIST-SRM-1633	70 ± 4	*	C
Fly ash	BCR-CRM-038	262 ± 11	*	C
Urban particulate	NIST-SRM-1648	262 ± 80	*	C
Fuels				
Coal	BCR-CRM-040	24.2 ± 1.7	*	C
Coal (sub-bituminous)	NIST-SRM-1635	1.9 ± 0.2	*	C
Coal	NIST-SRM-1630	4	*	NC
	NIST-SRM-1632a	12.4 ± 0.6	*	NC
	NIST-SRM-1632	30 ± 9	*	C
(Bituminous)	NIST-SRM-16326	3.67	*	C
Coking coal	BCR-CRM-181	2.59 ± 0.16	*	C
Fuel oil	NIST-SRM-1643a	2.80 ± 0.08	*	C
Gas coal	BCR-CRM-180	17.5 ± 0.5	*	C
Steam coal	BCR-CRM-182	15.3 ± 1.2	*	C
Sediments				
Estuarine	NIST-SRM-1646	28.2 ± 1.8	*	C
Lake	IAEA-Sl-1	37.7 ± 7.4	*	C
Marine	IAEA-SD-N-$^1/_2$	120 ± 12	*	C
Marine	NRCC-BCSS-1	22.7 ± 3.4	*	C
River	NIST-SRM-1645	714 ± 28	*	C

Table 2.6 (3 of 3)

Soils and sludges				
Sewage sludge	BCR–CRM–143	1333 ± 39	*	C
	BCR–CRM–144	495 ± 19	*	C
	BCR–CRM–145	349 ± 15	*	C
	BCR–CRM–146	1270 ± 28	*	C
Soil	BCR–CRM–141	29.4 ± 2.6	*	C
	BCR–CRM/142	37.8 ± 1.9	*	C
	IAEA–Soil–5	129 ± 26	*	C
	IAEA–Soil–7	60 ± 8	*	C
Waters				
Fresh	IAEA–W–4 (w–5)	0.025	*	C
	NIST–SRM–1643	20 ± 1	‡	C
	NIST–SRM–1643a	27 ± 1	‡	C
	NIST–SRM–1643b	23.7 ± 0.7	‡	C
Seawater	NRCC–CASS–1	0.2251 ± 0.028	*	C
	NRCC–NASS–1	0.039 ± 0.006	*	C

‡‡	concentration units,
‡	µg/L, ng/g, ng/mL
*	µg/g, µg/mL
**	mg/g

Agencies:	NIST	National Institute of Standards and Technology, Gaithesburg, MD, USA
	IAEA	International Atomic Energy Agency, Vienna, Austria.
	BCR	Community Bureau of Reference, Commission of the European Communities, Brussels
	NRCC	National Research Council of Canada, Ottawa
	IRANT	Institute of Radioecology and Applied Nuclear Techniques, Czechoslovakia
	NIES	National Institute for Environmental Studies, Japan.

References [114, 116–118]

NVA: no values available

TABLE 2.7 Lead concentrations in NBS (NIST) materials determined by various analytical methods.‡

Reference material NBS:SRM	Certificate	Lead concentration : analytical method			NAA	PAA	XRF	POL	IDMS	SSMS
		AA	ASV	ICP-AES						
1570 Spinach	1.2 ± 0.3 μg/g	1.20 ± 0.13	1.11 ± 0.10	0.87 ± 0.12	–	–	–	–	–	–
1571 Orchard Leaves	45 ± 3 μg/g	45 ± 3	46 ± 1	44 ± 2	43 ± 2	46 ± 4	46 ± 4	44 ± 1	–	–
1573 Tomato Leaves	6.3 ± 0.3 μg/g	5.5 ± 1.1	5.9 ± 2.1	5.9 ± 2.1	–	–	–	–	–	–
1575 Pine Leaves	10.8 ± 0.5μg/g	10.8 ± 0.4	10.7	10.1 ± 0.8	–	–	–	–	–	–
1577 Bovine Liver	340 ± 80 ng/g	350 ± 60	382 ± 8	360 ± 50	–	–	–	–	331	–
1632 T.e in Coal	30 ± 9 μg/g	28 ± 3	–	21 ± 6	–	30 ± 2	25 ± 8	–	28.7	30 ± 3
1633 Coal Fly Ash	70 ± 4 μg/g	74 ± 7	–	74.9	–	70 ± 2	67 ± 3	67	78	59 ±17

‡ reference (120)

TABLE 2.8 **Comparison of lead concentrations (μg/g) in various reference materials determined by ICP–MS and AAS methods.**

Reference material		Certified values	Lead concentration (mean ± std dev.)			
			ICP–MS‡	LA–ICP–MS‡	E–AAS*	F–AAS*
NIST–SRM–1571	Orchard leaves	45 ± 3	44.7 ± 4.5	47.6 ± 6.7	46 ± 8	48 ± 9
1572	Citrus leaves	13.3 ± 2.4	12.6 ± 1.7	12.2 ± 1.8	13.1 ± 3.6	13 ± 4
1573	Tomato leaves	6.3 ± 0.3	4.7 ± 1.5	4.8 ± 1.8	6.8 ± 2.1	7 ± 2
1575	Pine needles	10.8 ± 0.5	10.3 ± 0.8	11.7 ± 0.9	11.6 ± 11.1	12 ± 2
1577	Bovine liver	0.34 ± 0.08	0.25 ± 0.08	0.29 ± 0.18	0.36 ± 0.11	0.4 ± 0.1
BCR–CRM–062	Olive leaves	25 ± 1.5	24.6 ± 1.8	23 ± 1	24 ± 3	24 ± 6
NIES–1	Pepperbush	5.5 ± 0.8	5.3 ± 0.6	4.7 ± 0.7	5.6 ± 1.2	6 ± 2
NIES–7	Tea leaves	0.80 ± 0.03	0.83 ± 0.04	1.0 ± 0.2	1.0 ± 0.02	1.2 ± 0.3
IAEA–V–10	Hay powder	1.6 ± 0.5	1.24 ± 0.14	1.4 ± 0.2	1.6 ± 0.9	2.0 ± 0.7
IAEA–Soil 5		129 ± 26	131.2 ± 8.1	133 ± 12	131 ± 13	135 ± 25
IAEA–SL1	Sediment	37.7 ± 7.4	36.8 ± 42	38.4 ± 11.2	38 ± 8	39 ± 12

‡ Inductively coupled plasma–source mass spectrometry, conventional solution nebulisation and laser ablation (LA)

* Atomic absorption spectrometry, electrothermal (E–AAS), flame (F–AAS)
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Chapter 3

ATMOSPHERIC DISPERSION AND DEPOSITION MODELLING

B.E.A. Fisher

Contents **Page**

1. INTRODUCTION

This review illustrates in a simple way the main kinds of models which have been used to tackle air pollution problems associated with traffic pollution. The application of the similarity, diffusion approximation and Gaussian methods are described. All the models are shown to have limitations. Regional pollution questions are also addressed. A common approach is used based on the application of results and ideas from boundary layer meteorology and applied mathematics. The intention is to reveal the thinking behind dispersion calculations and not to produce a rulebook. The methods discussed apply to the whole range of traffic–related pollutants provided certain conditions are satisfied.

This chapter is concerned with methods for calculating atmospheric dispersion and deposition of pollutants released from motor vehicles. A practical view is taken of how a dispersion calculation should be undertaken. It is assumed that source strengths and meteorological conditions are specified and what is required is an assessment of the consequences of a release. Dispersion or mixing in the atmosphere is controlled by turbulence and the average flow is itself subject to considerable variability in both space and time. This means that the spread of a pollutant cloud is subject to considerable natural variability in apparently identical atmospheric conditions. Although there has been extensive study of the lowest few hundred metres of the atmosphere (the planetary boundary layer) only qualitative descriptions exist. There is moreover no rigorous theory of turbulence especially for the non–uniform conditions expected in the atmosphere. For these reasons no fully validated theory exists and any calculation is expected to be subject to considerable uncertainty. In principle all studies should be accompanied by an observational programme to test predictions. In practice this is rarely possible. By a sensible choice of what is calculated and a careful interpretation of results, modelling can be a useful tool. At the very least it should clarify thinking about the important processes influencing the transport of material in the atmosphere.

Since atmospheric dispersion models play an important role in air quality management there have been a number of attempts to review knowledge of the planetary boundary layer and to see how it may be applied to dispersion calculations (Nieuwstadt and van Dop, 1982; Weil, 1985). These reviews recognised the dilemma of matching practical models with the latest scientific results in the field. These problems are outlined in this chapter so that the reader gets an impression of the basic understanding underpinning practical atmospheric dispersion calculations. Conditions and available information will vary considerably from application to application, so that a broader understanding rather than a "recipe book" approach is essential.

For applications to highway pollution, atmospheric dispersion theory need only consider near ground-level sources. Significant plume rise, associated with say chimneys of large industrial sources, need not be considered. Very often the emission can be treated as though originating from one, or a number, of uniform line sources. The calculation is therefore essentially a two-dimensional one. The presence and movement of vehicles is likely to disturb the airflow in the immediate vicinity of the vehicles and the presence of nearby buildings and of local topography, such as road embankments and cuttings, will distort airflow on a wider scale. It is not the intention to include specifically urban pollution models or larger scale topographic influences. Only the dominant aspects of the life cycle of vehicle related pollutants, namely dispersion in the near-field (out to a few kilometres) and removal in the far-field (from a few tens to a few thousand kilometres) will be included. Local influences on the airflow are too specific to be included within general modelling treatments. The source strength associated with vehicles travelling along a road is effectively a uniform line source. Some stirring of the pollution at the point of emission is expected. This can be taken into account by displacing the line source to a virtual position, some way upwind of its actual position, to ensure realistic dilution initially. In the next section the fundamentals of modelling a ground-level line source are considered.

2. ATMOSPHERIC DISPERSION THEORY

2.1 Fundamentals of Atmospheric Dispersion

To orientate thinking we start by considering a clearly two-dimensional case when the wind is perpendicular to a road represented by a uniform ground-level source (see Fig.3.1). All dispersion calculations are based on the principle of conserving mass. In this idealised case this is best expressed in terms of a mass balance. Dispersion is determined solely by the increasing depth of the polluted layer with distance from the road. It is assumed that there is no variation in time of the source strength or of the meteorological conditions. Then a mass balance for the horizontal flux of pollution per unit length (of road) perpendicular to the road is

$$\frac{d}{dx}\left[\int_{o}^{d} u\,C\,dz\right] = -R \qquad (1)$$

where u(z) is the windspeed perpendicular to the road in the direction x and C(x,z) is the concentration with the z-axis taken vertically upwards. d is taken to be a distance above

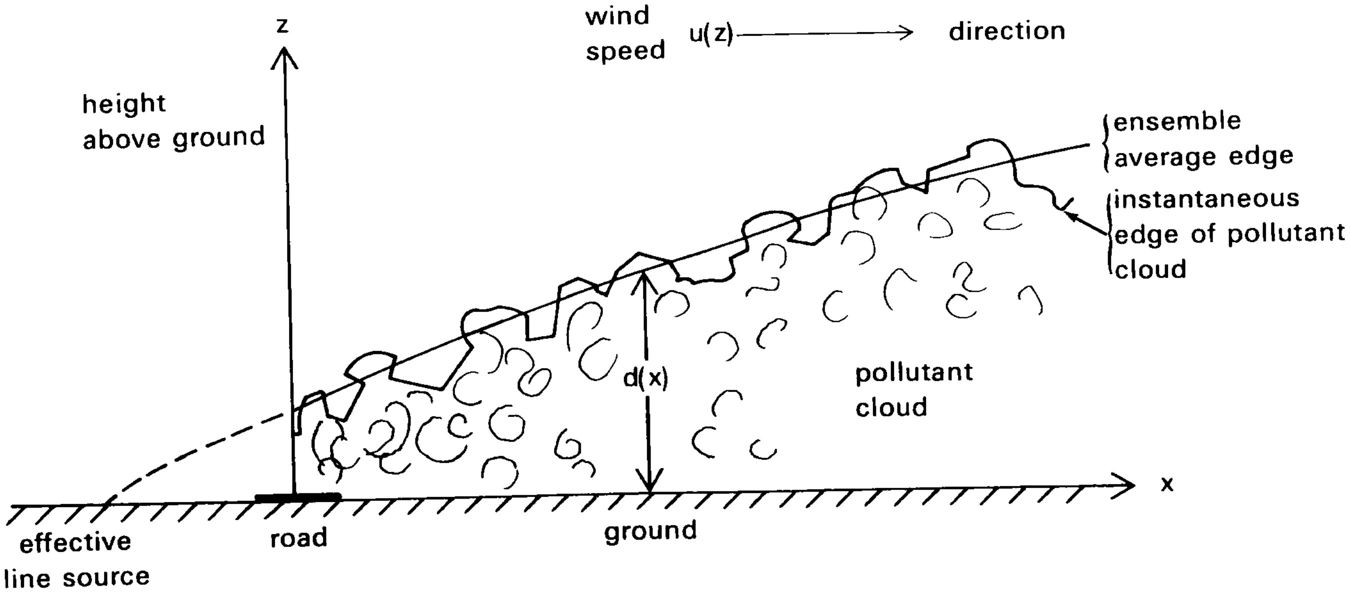

FIGURE 3.1 *SCHEMATIC REPRESENTATION OF SPREAD OF POLLUTION DOWNWIND OF A LINE SOURCE*

the cloud at which concentrations are zero. R includes all removal processes. The time–average concentration is calculated, using an averaging period of 30 minutes to 1 hour, since this removes the variability associated with turbulent eddies on a scale smaller than the plume depth.

The edge of the pollutant cloud, if made visible, would have a sharp but very ragged edge. An ideal measurement probe near the edge of the cloud will intermittently measure zero and positive instantaneous concentrations. If the time–average concentration is measured the mean concentration at the edge of the cloud will decay smoothly with height. The essential problem in describing dispersion of highway related pollution is to obtain an accurate estimate of the edge of the cloud, $z = d$.

In many situations mixing along the direction of the road will smooth out variations caused by non–uniform traffic flows. The concentration of pollutant in situations where there is more than one road is obtained by adding contributions from each road, or line of traffic. It will be seen that even in the very idealised two–dimensional dispersion situation there is no exact model. There are three well–known approaches to the problem and these are outlined in the next sections.

The term R in equation (1) includes all the removal processes, which in simple terms can be separated into dry deposition at the ground and wet deposition associated with precipitation. Generally close to the source, atmospheric dispersion is the main process by which concentrations are reduced and the removal term can be neglected. Equation (1) then becomes

$$\int_0^d u\, C\, dz = Q \qquad (2)$$

where Q is the source strength per unit length of road. If it is possible to identify reasonably accurately the depth of the cloud, d, then the magnitude of the ground level concentration can be specified.

2.2 Similarity Theory

In neutral stability conditions when thermal effects can be neglected, it is well established that the wind profile near the ground follows a logarithmic profile similar to those in boundary layers of engineering flows. The part of the atmospheric boundary layer where the log profile applies is the surface layer of depth of order 100 m. The wind direction does not vary with height in these lowest layers, but the windspeed above the surface roughness elements follows the well-known log law:

$$u(z) = \frac{u_*}{k} \ln (z/z_o) \tag{3}$$

z is defined to be the vertical distance from the so-called displacement height of the roughness elements, k is von Karman's constant (= 0.4), z_o is the roughness length describing the aerodynamic nature of the surface, which depends on the geometry of the roughness elements at the surface (trees, grass, buildings etc.) and u_* is the friction velocity, which is a characteristic turbulent velocity near the surface. The roughness length z_o is a fictitious height at which the windspeed appears to tend to zero.

The basis of similarity arguments used to derive the ground-level concentration is that the concentration is proportional to the product of a velocity scale and a length scale. The appropriate length scale is the mean depth of the pollutant cloud $\bar{z}(x)$ at a distance x from the source and the appropriate velocity scale is $\bar{u}$, the mean speed with which the cloud is advected. The ground-level concentration is given by

$$C(x,0) = \frac{a\ Q}{\bar{z}\ \bar{u}} \tag{4}$$

where a is a constant of order one (~ 0.7). $\bar{u}$ is equal to the windspeed at the effective height of plume transport, $c\bar{z}$, since this height is proportional to the cloud depth. c is another constant of order one (~ 0.6). Thus an expression for $\bar{u}$ can be written down

$$\bar{u} = \frac{u_*}{k} \ln (c\bar{z}/z_o) \tag{5}$$

which can then be substituted into equation (4).

Formula (4) is not of practical use since it does not explicitly give the variation of concentration with distance. It is however valuable as it shows the expected functional form of the concentration and how it is likely to depend on other important variables. This argument leads naturally into various types of box model which have been very useful in the description of urban pollution. Further progress can be made in the spirit of the similarity approximation by assuming that the vertical growth of the cloud, $\frac{d\bar{z}}{dt}$, must be proportional to u_*,namely

$$\frac{d\bar{z}}{dt} = bu_* \tag{6}$$

from which the relationship

$$\frac{d\bar{z}}{dt} = \frac{1}{bk} \ln (c\bar{z}/z_o) \tag{7}$$

may be derived, where b is a further constant of order one (~ 0.4). Integration of equation (7) leads to an implicit formula for $\bar{z}$ as a function of distance x, which when substituted into equation (4) gives a formula for the ground–level concentration.

The similarity solution contains a number of constants whose value have to be fixed by experiments. In addition it does not tell one anything about the shape of the vertical concentration distribution (Horst, 1979). It is therefore not surprising that other methods have been favoured which give explicit formulae for the concentration, even if they are based on somewhat weaker physical arguments.

2.3 Diffusion Approximation

For sources near the ground the length scale of eddies producing mixing increases in proportion to the vertical growth of the cloud. Hence turbulent mixing is caused by eddies smaller than the depth of the cloud and the time–averaged behaviour will appear like a diffusion process, described by an effective diffusion coefficient. Equation (1) for the conservation of mass can be rewritten in the absence of removal terms as a diffusion equation

$$u \frac{\partial C}{\partial x} = \frac{\partial}{\partial z} \left[K \frac{\partial C}{\partial z} \right] \tag{8}$$

where K is the eddy diffusivity in conventional notation. Assuming perfect reflection at the ground z = 0 and no restriction on vertical dispersion, a solution for the concentration profile C(x,z) can be derived by approximating the windspeed and eddy diffusivity by power laws

$$u(z) = u_1 \left[\frac{z}{z_1}\right]^m \tag{9a}$$

$$K(z) = K_1 \left[\frac{z}{z_1}\right]^n \tag{9b}$$

where u_1, K_1, m and n are constants. The solution was originally derived by Roberts (1923). It is instructive to write it in the similarity form (Van Ulden, 1978) as

$$C(x,z) = \frac{A\ Q}{\bar{z}\ \bar{u}} \exp\left[-(Bz/\bar{z})^s\right] \tag{10}$$

where $s = 2 + m - n$ is a shape factor. A and B depend only on s. The mean depth of the cloud $\bar{z}$ is proportional to the standard deviation of the vertical concentration distribution σ_z. The constant of proportionality is another factor dependent on s alone (~ 1.3).

In principle the parameters s, $\bar{u}$ and $\bar{z}$ should be related to the basic variables used to describe the atmospheric boundary layer. For practical applications they need to be related to easily measurable meteorological variables, such as u_{10} the windspeed at a height 10 m above ground (or more exactly 10 m above the displacement height) and geometrical variables such as distance downwind. These two requirements are not necessarily compatible. To satisfy the first requirement a basic understanding of the atmospheric boundary layer is needed.

2.4 Specifying Parameters in the Diffusion Equation

The fundamental advance in boundary layer meteorology has been recognition of the need to scale measurements in the appropriate way. Data on the atmospheric boundary layer, though very scattered and variable, can be made to look consistent when presented in terms of non–dimensional plots, provided the correct length, velocity and temperature scales are chosen. For example z_0, the roughness length, is an important lengthscale near the ground, while G the freestream windspeed at the top of the boundary layer is an important external velocity scale determining the nature of the atmospheric boundary layer.

The surface heat flux Q_o (defined as positive upwards) is an important variable describing the stability of the lowest layers of atmosphere. Under horizontally uniform conditions, which of course are rarely satisfied exactly, there are well established schemes for parameterising the wind profile and the eddy diffusivities for heat and momentum in the surface layer of the atmosphere. In terms of the roughness length, z_o, and the friction velocity, u_*, the Monin–Obukhov length scale L can be defined

$$L = -\frac{\rho\, c_p\, T\, u_*^3}{kgQ_o} \tag{11}$$

where ρ is the air density, c_p is the specific heat of air, T is the surface temperature and g is the acceleration of gravity. All the terms in L are effectively constant apart from u_* and Q_o. L represents the height at which the rate of generation of turbulence by mechanical forces near the ground, u_*^3/z, equals the non–dimensionalised surface heat flux Q_o. In neutral conditions $Q_o = 0$ and $L = \infty$. For stable conditions $Q_o < 0$ and L is positive. It is argued that the only way that L can appear in non–dimensional relations describing properties of the surface layer is through the non–dimensional height z/L. Similarity theory has no methods for deciding the shape of the wind and diffusivity profiles. These have to be provided by measurements.

The commonly used similarity forms of the wind and diffusivity profiles are not simple laws directly related to parameters m and n in equation (7). The diffusion equation has to be solved numerically if the usual similarity forms of the wind and diffusivity profiles are applied (Nieuwstadt and van Ulden, 1978).

2.5 Example of Application of Diffusion Approximation

It is instructive to obtain approximate analytical solutions of the diffusion equation in terms of the basic variables (van Ulden, 1978). This is particularly easy in neutral conditions when the eddy diffusivity takes the simple form

$$K(z) = k\, u_*\, z \tag{12}$$

One still has to choose a power law to approximate the logarithmic wind profile. The wind profile can be made exact at 10 m by setting u_l in equation (9a) equal to the windspeed at 10 m ($u_l = u_{10}$) and z_l equal to 10 m. Alternatively the wind profile can be made to be exact at the mean height of the pollutant cloud $\bar{z}$ at a distance x from the source by setting $z_l = \bar{z}$ and $u_l = \bar{z}$ and $u_l = \frac{u_*}{k} \ln (\bar{z}/z_0)$. If $m = 1/\ln (\bar{z}/z_0)$ the power law wind profile becomes (Hunt and Weber, 1979)

$$u(z) = u_l \exp (m \ln (z/z_l)) \tag{13}$$

$$= u_l (1 + m \ln (z/z_l) + O(m^2))$$

$$= \frac{u_*}{k} \ln \left[\frac{z}{z_0}\right] + O(m) \tag{14}$$

so a power law is a good approximation provided $m << 1$. Either way of choosing m shows that the solution is dependent on the roughness length z_0, though as it always appears with a logarithmic dependence, the sensitivity is not great.

Once m is chosen, the diffusion equation (8) may be solved to give a concentration profile of the form

$$C(x,z) = \frac{Q}{ku_*x \; (1 + m)} \exp - \frac{z \; u(z)}{ku_*x \; (1 + m)^2} \tag{15}$$

This solution has the same dimensional dependence as the similarity solution (equation (4)), but it emphasises that the ground level concentration is inversely proportional to distance x given the approximations made. It shows that concentrations are very sensitive to the initial dilution at the kerbside which determines the minimum distance at which x is defined. The shape factor $s(= 1 + m)$ is approximately one, since the exponent m in the wind profile power law is generally much less than one. Thus the concentration profile is a negative exponential which decreases approximately linearly with height.

2.6 Gaussian Approximation

It is attractive to the meteorologist to base dispersion calculations on the basic variables u_*, Q_0, z_0 etc. which control the boundary layer structure. This is one of the recommendations of a Workshop on updating applied diffusion models (Weil, 1985). Use of schemes based on boundary layer variables describing stable boundary layers have been successfully tested against results from field programmes. Despite this, most practical dispersion calculations in Europe and North America are still based on the Gaussian approximation (Hanna, Briggs, Deardorff, Egan, Gifford and Pasquill, 1977; Middleton, Butler and Colwill, 1979)

$$C = \sqrt{\frac{2}{\pi}} \frac{Q}{u_{10} \sigma_z} \exp - \frac{z^2}{2\sigma_z^2} \qquad (16)$$

which amongst other possible limitations does not have the correct dependence (theoretically or experimentally) on height in the concentration profile. Its attraction is its ease of use, since σ_z, the standard deviation of the vertical distribution, is a specified function of distance. By contrast in the USSR the K–diffusion model is commonly adopted (Berlyand, 1983).

In functional form the Gaussian approximation is like the similarity and K–diffusion models. The ground–level concentration is inversely proportional to the product of a windspeed and a length scale (σ_z). *It is clear that since the windspeed is a strong function* of height above ground, the x dependence of σ_z depends on what height the windspeed is defined. In principle therefore the windspeed should always be referred to the same height as that chosen to derive the original forms of σ_z from experimental data.

Great diversity in Gaussian models arises from the variety of forms of σ_z available. These used to be expressed as curves (Hanna et al., 1977) but now are commonly given as algebraic formulae dependent on distance, stability and possibly surface roughness height (Pasquill and Smith, 1983). Generally 6 or 7 discrete stability categories are used, ranking from unstable, through near neutral, to very stable atmospheric conditions. These originally gained acceptance since they were related to easily observable properties of the boundary layer, such as the surface windspeed, cloud cover and insolation. Implicitly the choice of dispersion category should depend on the internal parameters of the atmospheric boundary layer, such as u_* and L, but these are hard to measure and are not usually available. It has been suggested that in stable boundary layers $L = 1.1 \times 10^3 u_*^2$ is an adequate approximation, in which case only windspeed measurements are required (Venkatram, 1980).

Smith (see Clarke, 1979; Pasquill and Smith, 1983) has derived a nomogram which allows the surface heat flux to be derived as a function of time of year, time of day and cloud cover, from which atmospheric stability on a continuous scale may be derived. However much of the variation in prediction is associated with choosing the correct dispersion category appropriate to the prevailing conditions.

2.7 Example of Application of Gaussian Model

As an example of the Gaussian approach one may consider the σ_z curve used by Calder (1973) to fit dispersion measurements in near–neutral conditions

$$\sigma_z(x) = 0.08\,(x + 27)^{0.865} \tag{17}$$

where distances are expressed in metres and the non–zero intercept $\sigma_z(0) = 1.4$ m at the roadway $x = 0$, ensures some initial dilution produced by vehicle wakes. The form of the x dependence in equation (17) is consistent with the diffusion model formulation, considering the accuracy of the assumptions on which the formulations are based.

So far all discussion has referred to a two–dimensional problem in which the wind is perpendicular to the road. When the wind is not perpendicular to the road the problem can still be turned into a two–dimensional problem by defining the horizontal axis in the direction of the wind. A point at a perpendicular distance x from the road is now at an effective distance $x_\theta = x/\cos\theta$ from the source, where θ is the angle between the wind and the direction perpendicular to the road. The effective line source strength is $Q_\theta = Q/\cos\theta$. Replacing x and Q in equations (16) and (17) now gives the concentration at a distance x from the roadway in an oblique wind.

It may be seen from these equations that the ground level concentration is virtually independent of wind direction because the factor $(\cos\theta)^{-1}$ occurs in the numerator and the factor $(\cos\theta)^{-1}$ occurs in the denominator. This is a very useful result as it implies that for most wind directions the ground–level concentration in near neutral conditions, at a fixed distance, is either close to a non–zero constant (when the wind is blowing emissions towards the receptor) or zero (when the wind is blowing emissions away from the receptor). The same insensitivity to wind direction applies in near neutral conditions to the similarity and K–diffusion models. The argument applies to the Gaussian model in other stability conditions, though very stable conditions are unlikely to occur in the vicinity of moving vehicles. It thus can be seen that the study of dispersion from line sources, such as roads, is insensitive to two of the main factors which influence dispersion from point sources, namely wind direction and dispersion in the crosswind direction.

2.8 Scaling Arguments in Convective Conditions

Particular attention has been paid in recent years to convective daytime conditions characteristic of unstable dispersion categories. A major feature of the convective boundary layer is the presence of strong thermal updraughts and weaker downdraughts, which can promote rapid exchange of material throughout the depth of the boundary layer. This is not a situation which is likely to be well modelled as a diffusion process. The depth of the convective layer, h, is an important length scale from which a convective velocity w_* may be defined (Weil, 1985) using

$$w_* = \left[\frac{g\ Q_o\ h}{\rho\ c_p\ T}\right]^{1/3} \tag{18}$$

h and w_* have been found to be useful in ordering turbulence data over a broad range of scales. At mid–day, h is typically 1 to 2 km and w_* is 2 m s^{-1}. The diffusion approximation is only valid in the very lowest layers of the atmosphere, $z < -L$, where wind shear is still important. Above this layer it can no longer be applied. The Gaussian model because it is not dependent on underlying physical assumptions, is robust enough to handle this situation. The following formula has been suggested (Weil, 1985; Hanna, 1982)

$$\sigma_z = 0.6\, w_* x/u \tag{19}$$

(This may be compared with the form of equation (6).)

2.9 Limits on Vertical Diffusionn

Equations (15) or (16) apply provided σ_z is significantly less than h. At longer distances when σ_z becomes large it is no longer appropriate to neglect the influence of the top of the atmospheric boundary layer (see Fig. 3.2). In dispersion calculations it is assumed that material is not free to continue dispersing vertically indefinitely, but is reflected down from stable layers at the top of the boundary layer. Equation (16) can be generalised to include

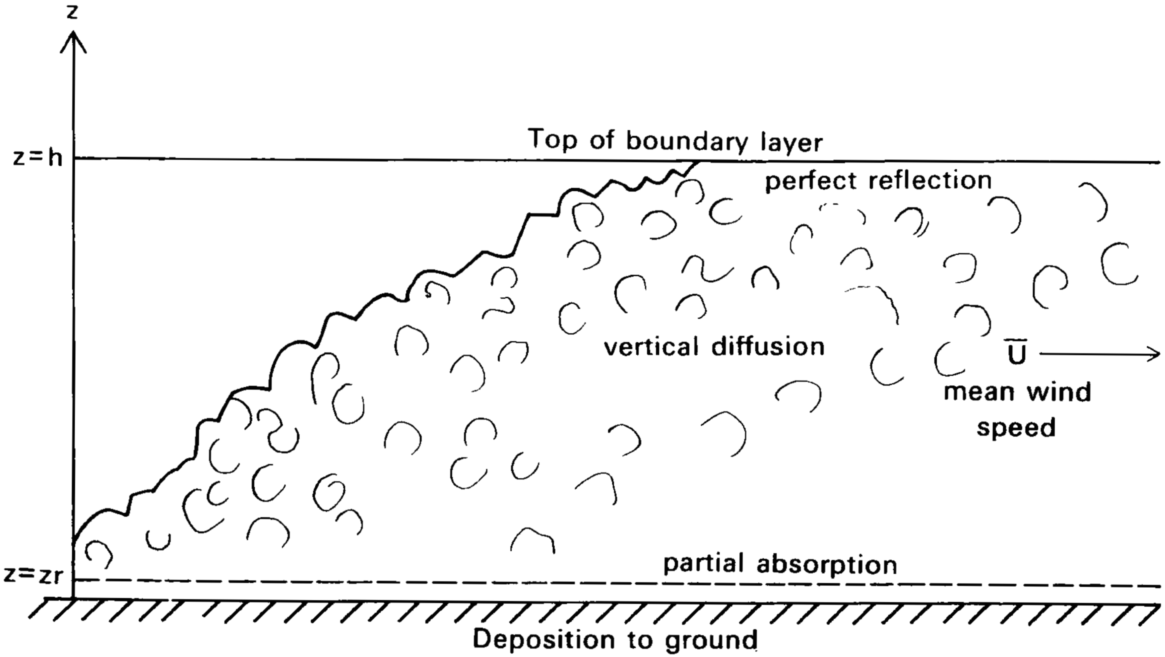

FIGURE 3.2 *SCHEMATIC REPRESENTATION OF POLLUTION ON A REGIONAL SCALE*

a series of terms representing multiple reflections at the top and bottom of the boundary layer. Usually it is only necessary to include the lowest order terms to get a formula which has acceptable accuracy.

At some stage, usually a few tens of kilometres downwind of the source, it is realistic to neglect multiple reflections and instead to assume that once $\sigma_z \sim 0.8\ h$ the vertical concentration distribution becomes effectively uniformly distributed throughout the boundary layer. Equation (16) needs modification and the concentration in the boundary layer tends towards

$$C = \frac{Q}{\bar{u}\ h} \tag{20}$$

$\bar{u}$ is the mean windspeed in the boundary layer, typically the windspeed at a characteristic height of several hundred metres. The change from using a windspeed, u_{10}, at ten metres to $\bar{u}$ may introduce a discontinuity in concentration gradient which is not real. It reflects uncertainty about concentration levels in an intermediate zone between two limits $\sigma_z << h$ and $\sigma_z \sim h$, in which the concentration is well defined. There is little observational data on σ_z over the intermediate distance range and it is not necessary to treat σ_z accurately over this range in practical applications.

In use, equation (20) would represent the contribution of a very wide line source to a monitoring site many tens of kilometres downwind. At these sorts of distances the individual effect of a particular road is not of interest. One needs to consider the combined effect of numerous sources. Discontinuities in concentration gradients will be smoothed out.

Often a formula like equation (20) will be used to estimate background levels, on which will be superimposed the much larger contribution of local sources. In these situations it may be necessary to generalise equation (20) in order to take into account the source distribution of distant regional sources.

2.10 Factors Determining Boundary Layer Depth

It is worth pointing out the contrast between the boundary layer depth in convective and stable boundary layers, viewed as possible scaling parameters. In the convective layer the depth of layer is dependent on surface heating since dawn and the extent to which this has eroded the nocturnal boundary layer. It is therefore not dependent on the current surface heat flux and must be an externally specified parameter. In stable boundary layers the depth of the boundary layer is determined by an equilibrium between windshear generated turbulence and its suppression because of stratification. In conditions which are not changing, h is therefore expected to be an internal parameter and dimensional arguments would suggest (Zilitinkevich, 1972) the relationship

$$h = \alpha(u_*L/f)^{1/2} \tag{21}$$

where α is a constant and f the Coriolis parameter (= 10^{-4} s^{-1} at mid–latitudes). It is the only externally defined timescale in the wind equations describing the balance between horizontal momentum and pressure gradient forces at the top of the boundary layer. In neutral conditions u_*/f is the appropriate height scale. Since f is a constant over the horizontal distances of a few tens of kilometres we are considering, the depth of the boundary layer is proportional to u_* and thus the wind speed.

2.11 Limitations of Current Methods

It is important to recognise the limitations of current methods and their inability to deal with certain special situations. In convective conditions for example, when $u < w_*$, diffusion in the alongwind direction cannot be neglected (Weil, 1985). Similarly in very stable conditions at low windspeeds ($u \sim 1$ m s^{-1}), there may not be a well–defined direction along which the wind is moving material away from a source. Increasing meandering of cloud parcels is to be expected. In boundary layers capped by stratus cloud, radiative cooling from cloud tops may initiate convection in the air below. The surface heat flux is then no longer the relevant parameter describing the boundary layer.

Apart from these difficulties even in ideal conditions one does not expect an exact prediction. The inherent variability of the atmosphere has led to the recognition that the prediction is only an estimate of the ensemble average of concentrations that could arise. The ensemble average observation is obtained by forming the mean of, say, hourly observations, which are characterised by the same meteorological conditions (windspeed, stability, etc.) and emissions. The standard deviation of observations within the same meteorological conditions is a measure of the natural variability, which can only be found from measurements and not from the model. Since experience shows that the natural variability is often a factor of two or more, this sets a limit on how well models can be expected to perform. Questions such as what is the highest hourly average concentration expected during a year, are not ones that can be answered using dispersion models alone.

3. APPLICATIONS OF DISPERSION THEORY

3.1 How Dispersion Methods Should be Applied

From estimates of the concentration within specified meteorological categories, the annual average concentration can be calculated by taking a weighted average of the concentration in each windspeed/stability category. One needs to know the frequency with which the various stability categories (A to F) occur. In most industrialised areas in coastal regions at temperate latitudes, the near–neutral categories with moderate winds occur most frequently. A site specific climatology of dispersion categories is unlikely to be available. However the diurnal variation of traffic flow (Chamberlain, Heard, Little and Wiffen, 1977) and the influence of vehicle wakes (Chock, 1978) will tend to increase the frequency of near–neutral categories in the vicinity of roads. Since highly unstable and very stable conditions tend to lead to higher concentrations than near–neutral conditions, because of the lower $u\sigma_z$ factor in the denominator of equation (16), this will lead to a certain pessimism in any calculation, which is often a desirable feature. Concentrations are also insensitive to wind direction apart from differences in the frequency with which winds blow towards one side or other of the road.

The above line of thinking leads to the conclusion that the annual average concentration is a rather robust measure of air pollution and not very dependent on specific atmospheric conditions. The uncertainty caused by atmospheric fluctuations over shorter time–scales is damped out.

Therefore despite the criticisms of models voiced in earlier sections, calculations based on them are likely to provide useful guidance on the levels of air pollution to be expected in the vicinity of roads. Experience based on observational data enables a measure of the concentration fluctuations around an ensemble average to be estimated.

3.2 Urban Pollution Levels

Consideration of air pollution at any rural monitoring site would take into account the background pollution levels produced by the transport of material from more distant sources in addition to those from a nearby roadway. The background pollution is the sum of contributions from other traffic sources as well as from sources not connected with vehicular emissions.

In an urban area the overall effect of multiple sources of traffic related pollutants, such as lead aerosol, NO_x or black smoke, contribute to ambient background levels. Inside the urban area a monitoring site is surrounded by sources in all directions. Taking Q to be the total source strength of the urban area and ℓ to be its characteristic width, the ambient ground–level background level is from equation (16)

$$C = \sqrt{\frac{2}{\pi}} \frac{Q}{u_{10}\ell^2} \int_0^{\ell/2} \frac{dx}{\sigma_z} \tag{22}$$

In near neutral conditions σ_z is approximately proportional to distance. Taking $\sigma_z \propto x^s$, where $s \simeq 1$ (equation (17)), this becomes

$$C = \sqrt{\frac{2}{\pi}} \frac{Q}{u_{10}\ell^2} \frac{\ell/2}{\sigma_z[\ell/2](1 - s)} \tag{23}$$

The integral is insensitive to the width of the urban area, ℓ, and the main factors determining ambient background concentrations are the emission density Q/ℓ^2 and the windspeed, u_{10}.

Applying this formula to lead concentrations in air over London, before the reduction of lead levels in petrol introduced at the end of 1985, one may make an estimate of airborne lead concentrations. Although equation (17) is not strictly applicable, since some account should be taken of the increased roughness length of urban areas (see Pasquill and Smith, 1983), a back of envelope calculation (using $Q = 1000$ tonnes y^{-1}, $u_{10} = 5$ m s^{-1} and $\ell = 40$ km) yields a concentration of 1 μg m^{-3}. This is not dissimilar from measured values (Jensen and Laxen, 1987).

The use of a power law expression for σ_z, such as that in equation (17), introduces no problem at the lower limit of the integration (where $x = 0$). Nearby sources contribute more in proportion than more distant sources and this is the reason why the exact width of the urban area is not important (Gifford and Hanna, 1973). The averaging of sources over the urban area has smoothed out the strong impact of very local traffic emissions (Bennett, 1987). This is apparent in measurements made at kerbsides within cities. In London, for example, smoke (Ball and Caswell, 1983), NO (Williams, Broughton, Bower, Drury, Lilley, Powell, Rogers and Stevenson, 1987), and lead concentrations (Jensen and Laxen, 1987) are a factor of about three higher than urban background values. The local contribution from diesel vehicles in London enhances concentrations by a factor of two for SO_2 measurements made along roads (Bennett, Rogers and Sutton, 1985).

The extra local contribution cannot be estimated using the methods applied above. There are street–canyon models (Sabottka and Leisen, 1981) in which the characteristic width and height of the street are important parameters. Upwind dispersion produced by vehicle induced disturbance of the airflow is another complicating factor (McCrae, Hamilton, Revitt and Harrop, 1988). These are examples where very localised factors play a role and it is not possible to apply a generic dispersion model.

3.3 Urban Pollution Episodes

In applying equation (23) to estimate the long–term average concentration, an average windspeed (strictly the harmonic mean) was used. It is clear that the highest short–term (episodic) concentrations would occur when the windspeed is low. The assumptions underlying equation (23) include the neglect of along–wind diffusion in relation to transport by the wind. Equation (23) is no longer applicable in very low winds.

Physically in these situations emissions supply pollution to an atmospheric "box" over the city. While weather conditions remain stagnant, concentrations in the city would rise. Some leakage at the edges of the city will occur, and deposition at the ground is another removal process. However, concentration would be likely to remain high until there is a change in the weather. These conditions are the worst from the point of view of air pollution and can be of concern from a public health point of view. The main factors determining their frequency are high source strength and stagnant weather conditions occurring in combination.

The depth of pollution over a city on such occasions is determined by the extent to which the stable lower atmosphere is mixed by surface heating. An elevated temperature inversion is usually present at a height of several hundred metres, above which turbulent mixing processes are damped out. Pollution emitted within the urban area below the inversion contributes to the buildup of high concentrations. Sources emitting above the inversion would not contribute. The presence of fog just below the inversion level would inhibit surface heating and thereby restrict the vertical extent of the mixed layer. This further exacerbates such pollution episodes. The presence of hills or mountains around the town or city would in addition suppress exchange at the edges.

The presence of hygroscopic particles amongst the smoke emissions can increase the incidence of fog formation. Pollutants would be absorbed by water droplets and new pollutants can be formed in solution. This may involve a whole range of further microphysical and chemical processes, the detailed treatment of which are outside the scope of this chapter.

3.4 Interpretation of Estimates Based on Dispersion Formulae

The review of urban pollution in the last section emphasised conditions which produced the highest atmospheric concentrations. From a modelling viewpoint one does not expect to be able to calculate the highest concentrations to great accuracy. Dispersion in rural areas depends on boundary layer structure described in terms of a broad classification of meteorological conditions. One expects departures from idealised boundary layer behaviour which may lead to higher than expected concentrations. For dispersion in urban areas there are many local features which complicate any prediction and can also lead to high concentrations. Modelling is often able to indicate which conditions would lead to the worst conditions. Predicted values are useful quantifications, provided the inherent uncertainties are recognised. For example, interpretation of measurements (Williams, 1987) should take into account the whole life cycle of the pollutant in the atmosphere and how and where, on average, it is removed, even if dilution processes in the atmosphere provide great reductions in concentrations before the influences of deposition processes are felt. It is therefore necessary to consider how removal leads to modifications of the basic structure of equation (8).

4. DRY DEPOSITION

Dry deposition on to land and sea surfaces is one of the most important processes by which pollution is removed from the atmosphere. The rate of removal is conventionally described in terms of a deposition velocity. The deposition velocity of an airborne pollutant $v_g(z_r)$ at a reference height, z_r, above ground is defined as the ratio of the downward flux to the concentration at height, z_r. Over a uniform surface type far enough from the edge of surface and from the source of pollution, the downward flux near the surface (within the lowest meter or two) is expected to depend only on concentration, the characteristics of the particular pollutant and the type of surface. Assuming uptake to be an irreversible process the flux is taken to be proportional to concentration; the constant of proportionality, which has dimensions of velocity, is the deposition velocity.

Over a homogeneous surface it is convenient to visualise the inverse of the deposition velocity as a sum of resistances, viz.

$$\frac{1}{v_g(z_r)} = r_a(z_r) + r_b + r_s \tag{24}$$

where r_a is the "aerodynamic resistance" to transfer of a pollutant from the reference height, z_r, to the vicinity of the roughness elements, r_b is the "sub–layer" resistance across the boundary layers close to the roughness elements and r_s is the surface resistance, independent of concentration and atmospheric conditions, but dependent on properties of the surface. Atmospheric conditions can influence the surface indirectly by causing wetting of vegetation, for example. However the deposition velocity concept is only useful provided results obtained under specified ideal conditions can be applied to surfaces under equivalent conditions. If this were not the case one would have to treat all situations using the measured net downward flux at the surface. This would complicate the problem excessively, because of the extreme difficulty in making routine measurements of the flux of pollutants at the earth's surface.

$r_a(z_r)$ provides a useful upper bound on the deposition velocity as it can be calculated exactly in terms of the eddy diffusivity, K(z), from

$$r_a(z_r) = \int_{z_o}^{z_r} \frac{dz}{K(z)} \tag{25}$$

For grass with a typical value of z_o of 1 cm, one sees immediately that the deposition velocity at z_r = 2 m, a standard reference height, is a small fraction of the friction velocity, u_*. Thus upward vertical dispersion by turbulence is a much more effective process for reducing concentration from near ground–level sources than deposition. It turns out that for most pollutants over most kinds of surfaces that r_b or r_s are somewhat greater than r_a, so that this conclusion is reinforced further. It is therefore quite reasonable when considering airborne concentrations in the neighbourhood of a source to neglect removal by dry deposition, which was the assumption in earlier sections. It also has the advantage that when considering real terrain with many changes of surface type, the difficulty of defining r_a rigorously is not a serious problem. At changes of surface type, the non–uniformity of flow conditions can produce changes in downward flux which are not associated with changes in aerodynamic resistance. Broadly speaking traffic pollutants can be classified into those that behave like reactive gases, those that are non–reactive and those which have properties of small particles. Knowledge of the deposition velocity of many airborne materials is very imprecise, even for closely studied gases such as sulphur dioxide. In the real world one encounters marked variability in surface conditions apart from the expected variation in aerodynamic resistance caused by changes in atmospheric conditions.

Amongst reactive gases one would classify sulphur dioxide, associated with diesel exhausts (Association of London Chief Environmental Health Officers, 1985), nitrogen dioxide NO_2 and HNO_3 gas, formed in the atmosphere from NO_2. Amongst non–reactive gases one would include emissions of NO, CO and hydrocarbon compounds. Lead emissions from motor vehicles behave as submicron particles (Chamberlain, 1975; Little and Wiffen, 1978) in a variety of chemical forms. The sources of sulphate and nitrate in particles are primary emissions of sulphur and nitrogen oxides. The secondary pollutants are formed in the atmosphere sometime after emission and are submicron in size (of diameter D_p less than $1\mu m$).

Primary lead particles have a size between 0.01 and 0.1 μm diameter, but may coagulate during atmospheric transport. Particles of lead and carbon chain aggregates of diesel smoke will disperse in the same way as gases (Ball and Hume, 1977; Ball and Caswell, 1983). However deposition of submicron size particles is limited by a high sub–layer resistance r_b, with $r_s = 0$, while gases are generally controlled by a high surface resistance r_s. For some gases r_s is so high ($>>r_a$) that deposition may be neglected as a removal process.

The reason why small particles are deposited slowly is as follows. The particles follow the motion of the gas in which they are travelling very closely, adjusting very rapidly to changes in velocity in a relaxation time, t_r, of order

$$t_r \sim \frac{\rho_p D_p^{\ 2}}{18\ \mu} \sim 10^{-5}\ s \qquad (26)$$

for submicron particles, where ρ_p is the density of the particles, D_p their diameter and μ the viscosity of air. To be deposited at a surface these particles must have enough inertia to carry them across the viscous sublayer adjacent to surface roughness elements. Given a turbulent velocity u_* the thickness of the sublayer is of order ν/u_*, where ν is the kinematic viscosity of air, so the condition for effective inertial deposition is $u_* t_r \sim \nu/u_*$ or $t_r \sim \nu/u_*^2$. However typically $\nu/u_*^2 \sim 10^{-4}$ s and therefore submicron particles are not easily carried across the viscous sublayer. Larger sized particles would be and are therefore expected to have a larger deposition velocity, while very small particles will diffuse across efficiently in the manner of a gas. Particles of diameter 0.2 to 1 μm have the lowest deposition velocity of order 0.1 cm s^{-1} (Chamberlain, 1967).

For reactive gases the surface resistance depends on the nature of the surface, though the condition of the surface may depend on past and present weather conditions. Thus for natural vegetation it may vary with stomatal opening. Information on r_s depends entirely on measurements.

The variation in surface type and condition makes it difficult to generalise, but the presence of the resistance, r_a, in series ensures that the widespread occurrence of a deposition velocity much greater than 1 cm s^{-1} is unlikely. For reactive gases the deposition velocity is somewhat lower than 1 cm s^{-1}. For non–reactive gases it can be taken to be zero, provided there is some other more effective atmospheric removal mechanism.

4.1 Influence of Dry Deposition on Concentration

The attraction of the deposition velocity concept is that it fits naturally into the theory of water vapour and heat exchange at the surface of atmospheric boundary layers and is a natural boundary layer condition

$$K\frac{\partial C}{\partial z} = v_g C \qquad \text{at } z = z_r \tag{27}$$

for solving the diffusion equation (8). With unlimited vertical dispersion the effect of deposition would be rather small and most material would escape to the free atmosphere. However it has already been pointed out that the atmosphere is generally stable and the top of the boundary layer depth is effectively a barrier above which material only penetrates intermittently. Though not always exactly satisfied the boundary condition

$$K \frac{\partial C}{\partial z} = 0 \quad \text{at } z = h \tag{28}$$

is a useful approximation when assessing regional scale pollution out to 1000 km, involving travel times of several days.

The solution of the diffusion equation subject to these boundary conditions may be viewed as an unnecessarily complicated way of assessing the importance of deposition, especially as atmospheric conditions in the boundary layer are unlikely to remain steady over a time scale of several days. If instead one simply assumes that the whole of a region's or a nation's emissions can be taken to be a very wide line source of effective source strength Q and the emission had time to mix uniformly with depth throughout the boundary layer, then the mass balance equation (1) becomes

$$\bar{u} \frac{dC}{dx} = -\frac{v_g}{h} C \tag{29}$$

where $\bar{u}$ is the characteristic transport velocity in the atmospheric boundary layer. Integration of this equation shows that concentrations behave as

$$C = \frac{Q}{\bar{u} h} \exp\left(-\frac{v_g x}{h \bar{u}}\right) \tag{30}$$

The decay distance $\bar{u}h/v_g$ is typically 1000 km indicating that reactive gases will be deposited over distances of this order.

4.2 Dry Deposition Near Sources

Clearly the assumptions in deriving equation (30) are weakest near the source when material has not mixed to any great depth. This is taken into account in trajectory models, which apply equation (30) along an air trajectory by assuming that the actual source strength Q has been depleted by a certain fraction, α, representing deposition while material is diffusing vertically. The effective source strength is

$$Q_{eff} = (1 - \alpha)Q \tag{31}$$

Trajectory models have gained wide acceptance as a way of treating long–range transport, but the local deposition factor α is a parameter fixed from outside the model.

In the EMEP model (Eliassen and Saltbones, 1983) α in equation (31) is taken to be 0.15. This is a value which may be appropriate for sulphur dioxide with a large fraction of high level sources. For ground level sources α should be greater.

Within limits the Gaussian model can be modified to include deposition. The method, known as the source depletion method, essentially includes the first order correction term to the solution in which deposition has been neglected. The method does not take into account the effect that deposition has on the concentration profile. The effective source strength for a ground level source is given by

$$Q_{eff} = Q \exp - \int_0^{x_0} \frac{v_g \, dx'}{u_{10}\sqrt{\pi/2}\ \sigma_z} \tag{32}$$

Thus
$$\alpha \simeq \int_0^{x_0} \frac{v_g \, dx'}{u_{10}\sqrt{\pi/2}\ \sigma_z} \tag{33}$$

x_0 is the distance beyond which the concentration can be assumed to be approximately uniformly mixed. $\sigma_z(x_0)$ is close to the depth of the boundary layer ($\sim h/\sqrt{\pi/2} = 0.8$ h). The source depletion model would be exact in the limit of rapid vertical mixing for then the concentration throughout the plume rapidly adjusts to removal by deposition. In a diffusion model with constant vertical diffusivity K, $\sigma_z = \sqrt{2Kx/u}$. Prahm and Berkowicz (1978) have shown that the source depletion method is a reasonable approximation when

$$\beta = v_g h/K << 1 \ . \tag{34}$$

The removal of material in the source depletion model is generally overestimated close to the source.

An alternative way of estimating α is to solve the diffusion equation subject to boundary conditions (27) and (28). It has been shown for a ground source assuming a constant eddy diffusivity K that the fraction deposited in the near field may be approximated by (Fisher, 1981)

$$\alpha = 1 - \frac{2(1 + 0.4\beta)}{1 + (1 + \beta)(1 + 0.4\beta)} \tag{35}$$

Rapid mixing corresponds to the limit $\beta = \frac{h v_g}{K} \rightarrow 0$ in which case the extra fraction deposited in the near field compared with the box model is zero. In the limit $\beta \rightarrow \infty$ corresponding to the limit of very slow vertical mixing with a high deposition velocity, the extra fraction deposited in the near fields tends to 1, i.e. no material reaches the far field. Typical conditions for which $h \sim 10^3$ m, $K \sim 10\ m^2\ s^{-1}$ and $v_g \sim 0.01\ cm\ s^{-1}$ correspond to $\beta = 1$ and the extra fraction deposited is 0.26. Hence according to this estimate 74% of the emitted material is involved in long–range transport. This situation applies to reactive gases. In contrast for small particles $\beta = 0.1$ and $\alpha = 0.97$ and most of the material is involved in long–range transport.

5. WET DEPOSITION

For reactive gases and particles associated with traffic emissions, wet deposition, that is incorporation in rain or snow, is an important pathway by which the material is removed from the atmosphere. For small particles with their low deposition velocity, it is more important than dry deposition. A proper treatment of wet deposition involves consideration of scales much larger and much smaller than those considered hitherto. On the larger scale precipitation is only formed when moist air parcels are lifted out of the boundary layer, so that large–scale atmospheric dynamics are involved. At the same time the incorporation of material in cloud droplets involves cloud microphysics (Fisher, 1982).

In this chapter we have considered situations when high pollution levels occur and how these may be treated within atmospheric dispersion theory. It is not appropriate to consider all aspects of the scavenging by precipitation of traffic related gases and particles.

Most traffic related pollutants are not readily removed in rain at their point of release, though they may be incorporated in rain at a later stage. The primary gases require atmospheric reactions before being efficiently absorbed in cloud droplets. The longer the travel time the greater the dilution and the greater opportunity for these transformations to take place. These conditions are more likely to occur as pollutants are lifted out of the boundary layer and drawn into mid–tropospheric clouds, where the initial growth of cloud droplets to precipitation size drops occurs. Thus in–cloud processes within precipitating rain are of dominating importance. This involves much broader scales than those discussed hitherto. In simple terms it indicates that the most important timescale involved in removal of gaseous pollutants in precipitation, provided rainfall is not occurring close to emission, is the time until the start of the first period of wet weather.

During the period of dry weather, dispersion within the boundary layer and dry deposition at the ground are the main dilution processes. Little is known about dispersion in disturbed weather conditions associated with the onset of precipitation though considerable mixing should be involved.

The removal of particles is thought to be primarily involved with the nucleation process. Partially soluble submicron particles have the potential to be activated in clouds at typical supersaturation. This means that they have the potential to grow into cloud size droplets and be subsequently scavenged by falling raindrops. Which particles are activated depends on the size and number distribution of soluble material. Coagulation of particles of different origin and dilution may reduce the number concentration. At some stage it may be argued that the potential for nucleation is likely to be high, though it is not certain that all particles are nucleated during the first period of rain the release encounters. Nevertheless a strong potential for removal in rain exists. The atmospheric lifetime of these materials is therefore a matter of days.

Some traffic–related emissions are not readily removed by dry or wet deposition, but are readily converted into other forms which are deposited. For example NO is converted into NO_2 and subsequently dry deposited or transformed into other oxides, which may be more easily removed from the atmosphere (though the chemical pathways are not well understood (Fisher, 1987)). Some hydrocarbons are rapidly broken down into CO_2 and water through a series of chemical reactions in the atmosphere. Some species, the primary example being CO (Khalil and Rasmussen, 1984), are not broken down rapidly. In long–range transport terms CO acts as a practically inert tracer.

6. CONCLUSIONS

This review has discussed, in a simple way, how far modelling can be used to address problems of atmospheric dispersion associated with highway pollution. It has concentrated on the physical aspects of dilution and removal processes. Although there are many uncertainties it is shown that these processes are often controlled by a few physical parameters. By focusing attention on the main length and velocity scales within a problem considerable progress can be made. It is shown that by no means all problems are amenable to attack in this way. Some require case by case study by physical modelling or full–scale experiments. However there are classes of problem involving near–field dispersion, urban pollution, regional transport and deposition, which can be tackled in this way. In these, useful approximations, limiting values and bounds on concentration can be obtained. The methods can be applied to a range of traffic–related pollutants, subject to appropriate conditions being satisfied.

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Chapter 4

HIGHWAY RUNOFF TRANSPORT MODELLING

M.J. Hall and R.S. Hamilton

Contents **Page**

1. INTRODUCTION

The infrastructure needs of modern society, coupled with growing concern about the environment in general, have dictated that the quantity and quality of urban runoff are increasingly studied at a variety of temporal and spatial scales. The importance of sampling and averaging time was discussed in Chapter 2. For larger catchment areas containing a substantial proportion of urban development, monthly or even annual data may be required for the long–term monitoring of water resources and their quality impacts. However, the smaller the catchment area, the greater the importance of variability over much shorter time scales and the larger the emphasis on problems of drainage design.

As the urbanisation of what was once a predominantly rural area proceeds, the greater the likelihood that the natural drainage system of the area will be altered or even replaced completely by stormwater drainage systems consisting largely of closed conduits. Unlike the former natural drainage system, the entry to stormwater sewerage occurs only at a limited number of points, the location of which is usually dictated by the road layout. This situation is fortuitous from the engineering design viewpoint in that the modelling of urban stormwater runoff can conveniently be separated into an above–ground and a below–ground phase. The transformation of the incident rainfall into a surface flow towards the nearest road gulley may therefore be considered separately from the routing of multiple input hydrographs through the pipe system. When the modelling is extended to water quality, the existence of these above–ground and below-ground phases allows the mechanisms of dirt accumulation and washoff to be considered separately from mixing processes in the gulley–pot and the entrainment and deposition of in–pipe sediments.

The road gulley, or stormwater inlet, being the transfer point between the surface and the underground phases of runoff, therefore assumes considerable importance. If gulleys are spaced too far apart, surface water may be held back and cause inconvenience to residents. However, if the inlets to the sewers are too close, downstream peak flows may be reinforced to the point where pipe diameters, and therefore construction costs, are increased. The hydraulic behaviour of different types and configurations of road gulley has therefore been examined in laboratory studies with the object of replacing rules–of–thumb on inlet spacing with a more rigorous design method. However, when pipe sizes are to be determined, the details of surface flow to individual gulleys become less relevant, and the majority of modern methods for the design and analysis of stormwater drainage systems represent the above–ground phase of runoff by a conceptual hydrological model. The difficulties of modelling the quantitative aspects of urban runoff almost fade into insignificance when compared with those of modelling water quality. What progress has been made has been confined to conservative determinands and to first–order models. Although water quantity issues are difficult to separate from those of water quality in the modern context of planning, design and maintenance of stormwater drainage systems, this Chapter begins in Section 2 with a brief review of the hydraulic design of road gulleys. Section 3 continues with a description of the conceptual approach to the modelling of inlet areas. Water quality considerations are then addressed in Sections 4 and 5, and the Chapter concludes in Section 6 by identifying current deficiences in methodology that will become increasingly critical as public attitudes to the urban environment become more demanding.

2. THE HYDRAULICS OF ROAD RUNOFF

2.1 The Importance of Good Drainage Design

According to Russam (1969a,b), the spacing of road gulleys in UK design practice has largely been based upon precedent, weighted by personal experience. The same author quoted typical design rules then in use, including a standard spacing of 45 m; a maximum impermeable area per gulley of 200 m^2; and a spacing (m) given by 3588 times the quotient of the square root of the longitudinal gradient in percent and the drained width (m). The empiricism of these rules belies the importance of good road drainage, not only in relation to the design of sewerage systems, as described in Section 1 above, but also to construction and materials engineering, the road user and traffic engineering. Implications of the latter three aspects were discussed by Hamlin and Hobbs (1970), and have been summarised in Figure 4.1.

Despite the widespread ramifications of poor gulley spacing indicated by Figure 4.1, the hydraulic testing of gulley gratings is of comparatively recent origin. Larson and Straub (1949) investigated the capacities and self–cleaning abilities of some typical American grate inlets with the objective of developing an improved model. Their laboratory–based study employed a tiltable gutter into which full–sized gratings could be installed. A significant conclusion drawn by those authors was the increase in the capacity of the inlet that could be obtained by permitting a proportion of the flow to bypass the inlet. They concluded that the grating should extend well into the flow path, and that its efficiency depended upon the effective length of individual openings in the direction of flow.

Development of the principles of behaviour of road gulleys and the practical application of those principles to design practice formed one of the major objectives of the Johns Hopkins University Storm Drainage Research Project. According to Schaake (1969), this work began when the project was initiated in 1949 and was completed in 1956. Attention was

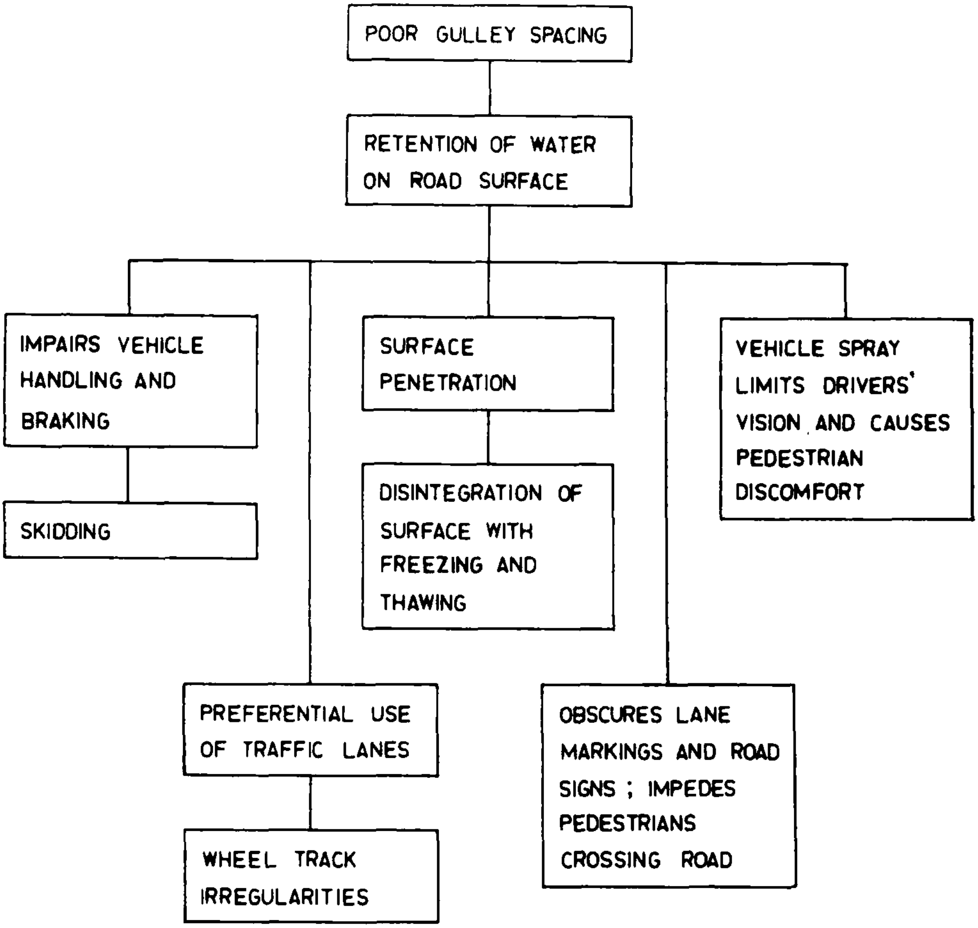


FIGURE 4.1 *THE IMPORTANCE OF GOOD DRAINAGE*

concentrated on one–half and one–third scale models, and dimensional analysis was applied to summarise the results for design purposes (see Li et al, 1951a,b; 1954a,b; Bock et al, 1956).

Cassidy (1966) also conducted a laboratory study of five basic types of inlet in order to determine the variables governing their efficiency. His results were expressed as a series of graphs of the grate efficiency, Q_i/Q, where Q_i is the flow intercepted by the grate and Q is the flow approaching the grate, against Froude number (based upon flow depth at the kerb) for a family of cross-slopes. Significantly, results were reportedly insensitive to the longitudinal slope, and grate length had only a minor effect. Gulleys with wide, flat bars were found to be inefficient, and those with curved vane–type bars were to be preferred.

In the UK, some work on the hydraulics of road gulleys was initiated at the University of Birmingham in 1961, and a similar programme of laboratory testing was undertaken at the Transport and Road Research Laboratory (TRRL) in the late 1960s. The object of the Birmingham experiments was to determine the values of three empirical constants contained in an analytical approach to describing the capacity of a road gulley previously proposed by Li (1954), a description of which is given below in Section 2.2. Tests were carried out with one type of heavy–duty gulley grating and both concrete and asphalt chipping surfaces (see Hamlin and Hobbs, 1970). In contrast, the TRRL work concentrated on determining the efficiency of the three principal types of gulley gratings to BS 497:1967 (British Standards Institution, 1967), where efficiency was again defined as the quotient of the flow entering and the flow approaching the gulley. The principal conclusions from the TRRL tests were summarised by Russam (1969a,b) as follows:

a. the hydraulic efficiency of properly–installed BS road gulleys is high; if the width of flow does not exceed 1.5 times the width of the grating and the gulley is directly in the path of the flow, efficiencies typically exceeded 95 per cent;
b. kerb inlets had a much lower efficiency, particularly for steep slopes, but their performance could be improved by angling the approach; and
c. further standardisation of road gulleys was possible.

The results obtained from these two studies in terms of drained area for the same type of grating differed by between 25 and 37 per cent. However, in order to appreciate the reasons for this discrepancy, the details of Li's theoretical approach must be elaborated.

2.2 Theoretical Framework

Figure 4.2 illustrates the general case of flow into a wide opening at two different discharges, Q and q, $Q > q$. If Figure 4.2 is assumed to represent a cross–section of the flow, the acceleration, a, is that due to gravity, g, and L' represents the length of opening that is just sufficient to capture the flow, Q. Retaining the general notation:

$$L' = u.t \text{ ; and } D = a.t^2/2 \qquad (1)$$

where D is the depth and u the mean velocity of the approaching flow, and t is a time variable. Eliminating t:

$$L' = u.(2.D/a)^{0{\cdot}5} \qquad (2)$$

so that, considering a smaller flow, q, with depth, d, (see Figure 4.2),

$$d/D = (L/L')^2 \qquad (3)$$

Figure 4.2 may also be considered to represent a plan view of the flow into a kerb inlet, a typical cross–section of which is shown in Figure 4.3. In this case, if y_k is the depth of flow at the kerb and Θ is the angle between the kerb and the road surface,

$$D = y_k.\tan\Theta; \text{ and } a = g.\cos\Theta \qquad (4)$$

Therefore, from equation (2),

$$u = L'.[(g.\cos\Theta)/(2.y_k.\tan\Theta)]^{0{\cdot}5} \qquad (5)$$

In addition,

$$Q = 0.5\, u.y_k{}^2.\tan\Theta = L'.y_k.[(g.y_k).(\sin\Theta/8)]^{0{\cdot}5} \qquad (6)$$

Since the above analysis neglects friction, equation (6) may be rewritten to include an empirical constant, K:

$$Q = K.L'.y_k.(g.y_k)^{0{\cdot}5} \qquad (7)$$

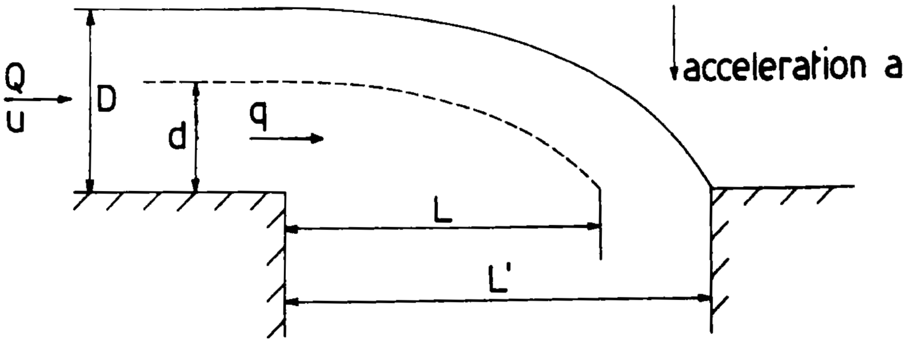


FIGURE 4.2 *FLOW INTO A WIDE OPENING*

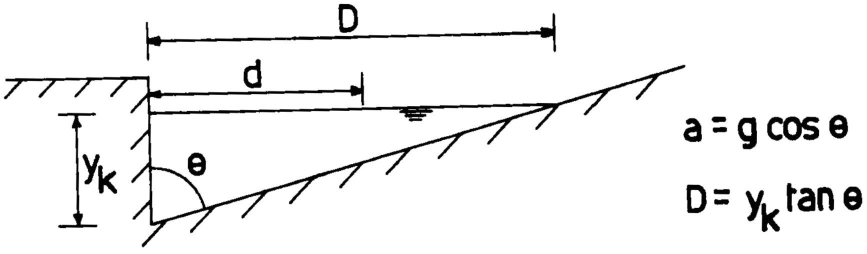


FIGURE 4.3 *CROSS–SECTION OF FLOW INTO A KERB INLET*

Using equation (3),

$$d/y_k.\tan \Theta = (L/L')^2 \tag{8}$$

and

$$q/Q = [y_k.d - (d^2.\cot \Theta)/2]/[(y_k{}^2.\tan \Theta)/2] \tag{9}$$

which, with the aid of equation (8), may be simplified to:

$$q/Q = 2.(L/L')^2 - (L/L')^4 \tag{10}$$

However, if L/L' equals or exceeds 0.6,

$$q/Q = L/L' \tag{11}$$

The above relationships may now be employed to describe the division of the flow, Q, approaching a gulley grating into the following components:

i) flow through the opening;
ii) flow between the kerb and the opening;
iii) flow past the opening or carry–past flow, Q_p; and
iv) flow across the opening or carry–over flow, Q_o.

Since the flow between the kerb and the opening is negligible and usually neglected, the gulley will capture a flow equal to $Q - (Q_p + Q_o)$. A plan and cross–section of the flow pattern is depicted in Figure 4.4.

An expression for the carry–past flow may be derived from equation (7):

$$Q = (u.s/2).y_k{}^2 = K.L'.y_k.(g.y_k)^{0.5} \tag{12}$$

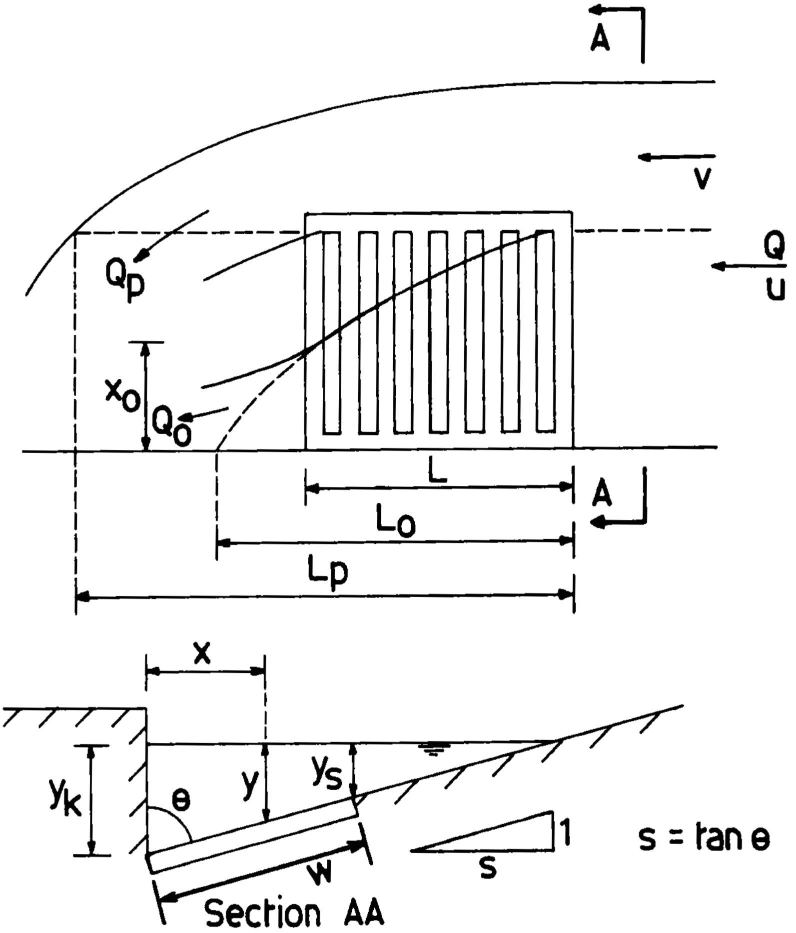


FIGURE 4.4 *FLOW INTO A GULLEY GRATING*

where $s = \tan \Theta$ (see Figure 4.4). From equation (12):

$$1/K = (2.L'/u.s).(g/y_k)^{0\cdot 5} \qquad (13)$$

The analogy of a kerb inlet is now applied to the flow on the road side of the grating. If v is the mean velocity of flow in this region, and the variable L_p is introduced as the length of a notional kerb inlet that will just capture the flow outside the line of the grating,

$$1/K = (2.L_p/v.s).(g/y_s)^{0\cdot 5} \qquad (14)$$

where y_s is the depth of flow at the up–slope edge of the grating (see Figure 4.4). Since u is a more convenient parameter than v, multiplying through equation (14) by v/u and rearranging gives

$$L_p = k_1.u.s.(y_s/g)^{0\cdot 5} \qquad (15)$$

Given w as the width of the gulley grating (see Figure 4.4), y_s may be computed from y_k sd $y_k - e \cos \Theta$, which may be approximated by $y_k - w/s$.

For a kerb inlet, equation (11) gives

$$q = (Q/L').L \qquad (16)$$

but, using an expression for Q/L' from equation (7), the flow NOT captured by a kerb inlet of length L is given by

$$Q - q = (Q/L').(L'-L) = K.(L'-L).y_k.(g.y_k)^{0\cdot 5} \qquad (17)$$

and, by analogy, the flow past the gulley

$$Q_p = k_2.(L_p - L).y_s.(g.y_s)^{0\cdot 5} \qquad (18)$$

A complementary expression for the carry–over flow may be derived by defining a new variable, L_y, as the gulley length required to capture a flow depth of y. By analogy with equation (3),

$$L_y = L_o.(y/y_k)^{0\cdot 5} \qquad (19)$$

but, since from Figure 4.4 $y = y_k - x/s$, this equation becomes

$$L_y = L_o.[1 - (x/s.y_k)]^{0{\cdot}5} \tag{20}$$

If $L < L_y$, the gulley will capture a depth $y' < y$, and

$$y - y' = y.[1 - (y'/y)] = y.[1 - (L/L_y)^2] = y_k.[1 - (L/L_o)^2] - x/s \tag{21}$$

The width of flow within which carry–over occurs may then be obtained by setting $y - y' = 0$ to give

$$x_o = s.y_k.[1 - (L/L_o)^2] \tag{22}$$

Since Q_o is the integral between limits of 0 and x_o of $(y - y').u.dx$,

$$Q_o = u.y_k.[1 - (L/L_o)^2].x_o - u.x_o^2/2.s \tag{23}$$

Substituting for x_o and noting that $Q = (u.s/2).y_k^2$,

$$Q_o = Q.[1 - (L/L_o)^2]^2 \tag{24}$$

where by analogy with equation (7),

$$L_o = Q/[k_3.y_k.(g.y_k)^{0{\cdot}5}] \tag{25}$$

The mean velocity of the approach flow, u, may be estimated from a uniform flow formula such as the Manning equation. Since $s >> 1$, the wetted perimeter, $y_k + y_k.\sec\Theta$ may be approximated by $y_k.s$, giving a hydraulic mean depth of $y_k/2$. Hence, if f is the gutter slope,

$$u = (c/n).(y_k/2)^{2/3}.(1/f)^{0{\cdot}5} \tag{26}$$

where c is a constant depending upon the units and n is the roughness coefficient. Therefore,

$$Q = (c/n).[s/(f)^{0{\cdot}5}].y_k^{8/3} \tag{27}$$

If the area drained by the gulley is A and the average rainfall intensity is i, then $Q = A.i$ and the following procedure may be employed to estimate the flow captured by the gulley:

1. given A, i, c, n, s and f, compute y_k from equation (27);
2. given A, i, y_k, k_3 and g, compute L_o from equation (25);
3. given A, i, L and L_o, compute Q_o from equation (24);
4. given u, s, g, k_1 and y_s (from y_k, w and s), compute L_p from equation (15);
5. given L, L_p, y_s, k_2 and g, compute Q_p from equation (18); hence
6. the flow intercepted by the gulley $= Q - (Q_o + Q_p)$.

2.3 Results for BS Road Gulleys

The road gulleys to BS 497:1967 may be broadly classified into four categories which include one kerb type and three that are set into the gutter. The latter comprise the heavy–duty grade D10 and D11 gratings; and the medium–duty E12 and E13 types. Whereas the E12 have openings at right angles to the kerb, those of the E13 are inclined at an angle of 60 degrees to the line of the gutter. The results quoted by Hamlin and Hobbs (1970) are confined to the D10 heavy–duty gulleys and relate to the Manning roughness, n, and the constants, k_1, k_2 and k_3, from equations (15), (18) and (25), as displayed in Table 4.1.

TABLE 4.1: **Experimentally–determined values of the Manning roughness coefficient, n, and the constants, k_1, k_2 and k_3 for concrete and asphalt chipping surfaces (after Hamlin and Hobbs, 1970).**

Surface	n	k_1	k_2	k_3
Concrete	0.0134	2.825	0.192	0.0404
Asphalt Chippings	0.021	2.488	0.069	0.0409

Hamlin and Hobbs employed the values of the constants in Table 4.1 to compile estimates of the drained area per gulley for different combinations of crossfall and endfall and dimensions of grating, subject to a maximum width of flow in the gutter and carry–over and carry–past flows being less than or equal to 10 per cent of the gutter flow. Unfortunately, when compared to the drained areas provided by Russam (1969a,b) for the same type of grating, the Hamlin and Hobbs figures are found to be between 25 and 37 per cent lower than the TRRL results. The discrepancy can be attributed partly to assumptions of different values for Manning roughness and maximum widths of flow in the two studies. In addition, the TRRL data relate to gutter flow only, whereas the Birmingham experiments allowed for an additional component of crossflow from the carriageway. In reality, neither series of tests allowed for rainfall on to the sheet flow across the road surface, the effect of which may be to increase the apparent roughness for flows whose Reynolds numbers are within the transition range between laminar and turbulent flow (see Yu and McNown, 1964, for example). The omission of this effect may result in the over–estimation of the areas that can be drained with a particular type of gulley. While the effect may be small for turbulent sheet flows, its neglect in the tests that have been performed tend to suggest that further work remains to be done on the hydraulic testing of road gulleys. In the meantime, the TRRL results continue to be the more comprehensive and should be employed with due allowance for higher surface roughnesses than are indicated by uniform flow tests with gutter flow alone.

3. CONCEPTUAL MODELLING OF INLET AREAS

When attention is turned to the determination of pipe sizes for the sewers serving large numbers of stormwater inlets, hydraulic considerations, such as those outlined in Section 2 relating to the spacing of road gulleys, give way to broader aspects of hydrological behaviour. In modelling the response of such areas to storm rainfall, modern methods for the design and analysis of sewerage systems invariably rely on a conceptual model for the transformation of rainfall into runoff. This conceptual model may take a wide variety of forms from a dimensionless hydrograph, the shape of whose rising limb and recession depend upon both catchment and rainfall characteristics, to groupings of linear reservoirs and linear channels. A further possibility is the division of the catchment into a network of idealised elements, such as planes or converging surfaces, for which the equations of motion, the Saint Venant equations (or their approximation), may be solved numerically.

The Inlet Hydrograph Method outlined by Viessman and Geyer (1962) provides a ready example of a dimensionless hydrograph approach. In this method, the rising limb and the first 200 seconds of the recession are assumed to conform to the shape of the Gaussian distribution, and the remainder of the recession to an exponential decay function. The parameters governing the shape of each portion are estimated from equations derived from multiple linear regression analysis (MLRA) using site and storm characteristics as independent variables. A similar approach was successfully applied by Swinnerton et al (1972) in a design method for the drainage systems of motorways running in cutting.

The linear channel and the linear reservoir are conceptual elements which with an appropriate choice of parameter values may be used to synthesise the response of the inlet area. A linear channel is an element that provides pure translation of the input hydrograph without change in its shape. The time of entry which is added to the time of flow in a drainage system to estimate the time of concentration when applying the well–known Rational Method and its derivatives (see Hall, 1984) may be regarded in conceptual terms as the time lag introduced by a linear channel. In contrast, the linear reservoir introduces both a delay and an attenuation of the peak rate of inflow. More specifically, the storage, S, of a linear reservoir is directly proportional to its outflow, Q:

$$S = K_c.Q \tag{28}$$

where K_c is the storage constant of the reservoir and its only parameter. In more rigorous terms, the linear reservoir may be regarded as a special case of the non–linear reservoir:

$$S = K_c.Q^m \tag{29}$$

where the power m, which provides the second parameter, is usually less than unity. Since the non–linear reservoir has two parameters, this model offers more flexibility in fitting a measured inlet hydrograph, but at the expense of additional computation, compared with the linear reservoir. However, the latter model is easily elaborated to provide a second parameter. Table 4.2 lists a selection of conceptual models that have been evaluated in the context of small, urban catchments.

For models that employ idealised elements, such as cascades of planes or converging surfaces, to represent the areas draining to an inlet, the Saint Venant equations have frequently been approximated by the kinematic wave equations. The WITWAT model described by Green and Stephenson (1986) provides a ready example of such a model. In version III of the Storm Water Management Model, SWMM (see Bedient and Huber, 1988), the inlet areas are approximated by planes of equivalent area, but the routing of the rainfall excess is accomplished by the use of a non–linear reservoir. The Hydrograph and Simulation Methods within the Wallingford Procedure (National Water Council and Department of the Environment, 1981), originally employed a non–linear reservoir model with a power of 2/3 (see Kidd, 1978), but more recent versions of the computer software have adopted a linear reservoir (Osborne, 1988).

TABLE 4.2: A summary of conceptual models that have been employed to describe the response of small, urban drainage areas.

Model	Number of Parameters	Reference
Single linear reservoir	1	Willeke (1966); Viessman (1966, 1968); Watt and Kidd (1975); Pedersen et al 1980)
Single non–linear reservoir	2	Kidd and Helliwell (1977) Kidd (1978)
Cascade of linear reservoirs	2	Nash (1957, 1960); Rao et al (1972)
Cascade of two linear reservoirs with unequal storage constants	2	Sarginson and Bourne (1969)
Single linear reservoir with different storage constants with and without rain	2	Swinnerton et al (1973)
Single linear reservoir with linear channel	2	Viessman et al (1970)

These examples are sufficient to illustrate the variety of approaches that have been invoked to describe the routing of the rainfall excess through storage in the inlet area to provide the hydrograph of flow into the nearest road gulley. For the current range of software packages in the urban drainage field, such as WASSP and its latest version, WALLRUS, SWMM and WITWAT, the variety of simple, conceptual elements is notable. Nevertheless, each model provides the flow field within which the water quality changes are enumerated. Attention must therefore be turned to the qualitative aspects of stormwater runoff and the processes that are involved therein.

4. WATER QUALITY SIMULATION MODELLING OF INLET AREAS

4.1 Introduction to Water Quality Modelling

Whereas the response of inlet areas to storm rainfall and the generation of the runoff hydrograph can be simulated with tolerable accuracy by simple conceptual models with regionalised estimates of parameter–values, the available simulation models for urban runoff quality are by no means as robust. If absolute values of pollutant loads and concentrations are required, then local, site–specific data must be collected for the purposes of model calibration and verification. Even in one of the simplest of situations in which constant concentrations of pollutants are assumed and loadings are obtained from their product with runoff volume, the question immediately arises as to the choice of values. Alternative model formulations may be considered, and these are reviewed in Section 5 of this chapter. Although the results from many urban water quality studies are available in the form of event mean concentrations (EMCs), their variation with climate, land use and flow regime is far from clear (Jewell and Adrian, 1982; Desbordes and Servat, 1984; Ellis et al, 1986). Recourse to the theories of sediment transport and the application of sediment rating curves may bypass the parameter estimation problems associated with the more complex water quality models, but depend upon local data for their successful implementation. In these circumstances, a physically–based model of urban water quality is an obvious improvement, although the requirement for calibration data is ever present. As noted in a recent major review by Huber (1986), this type of model, which focuses on the processes by which solids and pollutants build up over the surface of an inlet area and are subsequently washed off during rainfall, has a remarkable longevity. The modelling of these processes is therefore considered below in more detail.

4.2 Dry–weather Build–up of Pollutants

The accumulation of dust and dirt on the impervious surfaces of an urban catchment may be attributed to numerous sources, of which Fletcher et al (1978) listed the following as the more significant:

1. open land contributes dust and soil particles which accumulate in roadside gutters and act as a source of nutrients and organisms;

2. vegetation and gardens are a source of leaves and grass cuttings, and animal activity gives rise to deposits of faecal material;
3. roofs and roads are themselves sources of inorganic solids, salt and the by–products of surface degredation;
4. vehicles are a source of oil, petrol, and eroded material from car bodies and tyres;
5. industry and transport are sources of dust and gaseous material which is emitted to the atmosphere and subsequently settles out or is washed out by rainfall; and
6. human activity which may contribute litter, garden fertilisers and detergents from vehicle washing.

During dry weather, this material may be removed by street sweeping, or simply redistributed by wind action and the slipstream of passing traffic or (in some countries) by the runoff of water from street spraying. Since few measurements have been taken from the urban surfaces draining to individual road gulleys and the majority of data relate to within–pipe measurements downstream of the tributary inlet area, the modelling of the accumulation or 'build–up' process has invariably been based upon semi–empirical formulations in which time is the principal independent variable. Huber (1986) quotes four functional forms that have been employed to describe the build–up process, including:

$$P = a_1.t \tag{30}$$

where P is the mass of pollutant on the surface at time t following the last rainfall event or the last street sweeping operation, and a_1 is a coefficient;

$$P = a_2.t^{b_1} \tag{31}$$

where a_2 is another coefficient and b_1 is an exponent;

$$P = P_L.[1 - \exp(-b_2.t)] \tag{32}$$

where P_L is an upper limit to the accumulated mass of pollutants and b_2 is another exponent: and the Michaelis–Melton function:

$$P = P_L.[t/(a_3 + t)] \tag{33}$$

where a_3 is a third coefficient.

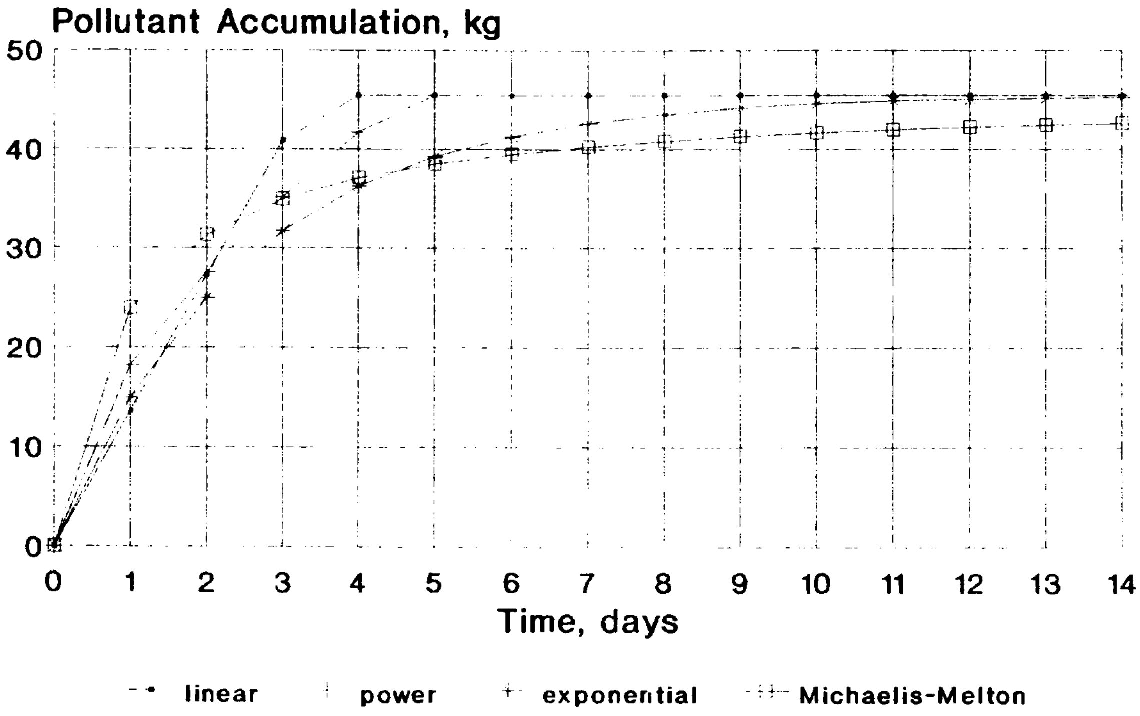


FIGURE 4.5 *COMPARISON OF ACCUMULATION FUNCTIONS*

An assumption common to the application of all these models is the complete clearance of all the dust and dirt from the impervious surfaces of the inlet area by the last street sweeping or rainfall event. However, Alley and Smith (1981), in presenting a systematic methodology for estimating the parameters of equation (32), found that allowing for residual loads gave better results than assuming complete removal.

Figure 4.5 shows a plot of these four functions with arbitrary values assigned to the constants and exponents. As noted by Huber (1986), users of SWMM have the choice of any one of these four functions, with or without an arbitrary upper limit to the linear and power functions. Since the linear and power functions of equations (30) and (31) do not possess an upper bound, an arbitrary limit can be imposed to provide some approximation to the behaviour of equations (32) and (33).

Equations 31 and 33 are purely empirical, the former being an approximation (when $b_2t << 1$) to equation (32) with the coefficient a_l being equal to the product of P_L and b_2. The physical basis of equation (32) involves a more detailed consideration of deposition, removal and resuspension processes, as given below.

4.3 Resuspension and surface Accumulation

The re–entry of deposited particulate material back into the atmosphere has long been of interest to air quality scientists. Much attention has been focussed on erosion, solid transport and fugitive emissions. In recent years, the importance of resuspension processes has been recognised in fields as diverse as industrial spills, the transport of pesticides, the spreading of crop disease by fungal spores, the transmission of human disease and the translocation of radioactive dust. The Chernobyl accident provided a ready example falling in the last category. The processes involves in resuspension have been reviewed by Sehmel (1980) and Nicholson (1988).

The basic theory for dust deposition, resuspension and accumulation on a highway surface assumes that the dust accumulates by dry deposition according to the expression

$$\frac{dP_1}{dt} = D_d \tag{34}$$

where D_d is the dry deposition rate, computed as the product of the airborne concentration and the dry deposition velocity, v_g. In addition, dust is removed by vehicle–induced resuspension according to

$$\frac{dP_2}{dt} = -\Lambda.N.P \tag{35}$$

where Λ is the resuspension rate per vehicle and N is the rate of traffic flow. Combining equations (34) and (35), and integrating leads to an expression similar in form to equation (32) above with $b_2 = \Lambda.N$ and $P_L = D_d/\Lambda.N$. Using these values in equation (32), the accumulated mass is seen to reach half its maximum value after a time T given by

$$T = 0.3/\Lambda.N \tag{36}$$

Values for Λ have been obtained from tracer studies (Sehmel, 1980; Nicholson, 1988), and have also been inferred from P_L, D_d and H at a highway site by Hamilton et al (1985). A value of 2×10^5 is typical, although variations of an order of magnitude are not uncommon. Substituting this value into equation (36) results in the predicted time, T, for resuspension to become a dominant removal mechanism of $7 \times 10^{-4}/N$. For a major highway site with 2000 vehicles/h, T is computed as 35 h, i.e. resuspension becomes a dominant mechanism after about a day–and–a–half. However, for a residential side street carrying 50 vehicles/h, T = 60 days, so that in practice resuspension is negligible compared to other removal processes.

4.4 Removal of Pollutants

Dust and dirt which accumulates on urban surfaces may also be removed either by routine street sweeping during dry weather or the erosive action of stormwater runoff following rainfall. Street sweeping is generally assumed to remove only a fraction of the accumulated material. However, according to Huber (1986), recent studies performed in the United States as part of the National Urban Runoff Programme (NURP) have indicated that sweeping is not very effective as a method for improving urban water quality. The removal of material during wet weather may occur either by erosion or by constituents being taken into solution. Erosion and the subsequent transporting of the products could be described by the theories of sediment transport. Indeed, a model which includes the disturbance of particles by raindrop impact and their entrainment by sheet flow as well as the deposition of material has been described by Price and Mance (1978). Such formulations are, however, limited by the ability of the modeller to determine appropriate values of the threshold shear stresses and other parameters for the complex of surfaces that comprise even the smallest of inlet areas. Operational models of urban water quality therefore tend to rely on semi–empirical descriptions of the wash–off process, thereby mirroring the development of the functions used to describe build–up discussed in Sections 4.2 and 4.3. The model which has generally been adopted to describe the flushing of contaminants from road surfaces was clearly evident in the results of experiments performed by Sartor et al (1974) with a portable rainfall simulator which indicated that the wash–off rate is directly proportional to the amount of material remaining, ie,

$$dP/dt = -C.P \qquad (37)$$

where P is the mass of pollutant remaining on the surface, t is time and C is a rate constant. Integrating equation (37) and assuming that C varies with the rate of runoff such that $C = K_w.Q$, the amount of pollutant removed during time step, Δt, is given by (Alley, 1981)

$$P_o - P = P_o.[1 - \exp(-K_w.Q.\Delta t)] \qquad (38)$$

where P_o is the mass of pollutant present at the beginning of the time step; K_w is the rate coefficient; and Q is the average rate of runoff during the time step Δt.

Application of equation (38) requires the estimation of the mass of pollutant present at the beginning of the storm and the rate coefficient, K_w. However, the model is subject to an anomaly in that positive values of K_w lead to predictions of decreasing concentrations of contaminants with increasing time from the beginning of a storm, as demonstrated by Alley (1981). This problem can be circumvented by multiplying the right–hand side of equation (35) by an 'availability factor', K_a, which is a function of runoff rate:

$$K_a = c_1 + c_2.Q^{c_3} \tag{39}$$

where c_1, c_2 and c_3 are further constants to be determined. The model fitting is further complicated in many urban water quality models by the increase of the amount of the pollutant washed off by fractions of the suspended and settleable solids removed.

The applicability of equations (38) and (39) for the modelling of the wash–off process has been the focus of considerable attention. For example, Jewell et al (1980) and Jewell and Adrian (1981) have concluded that equation (38) cannot be verified statistically using available field data. and have proposed new algorithms for describing the wash–off process. Unfortunately, no single model has been found applicable to all pollutants and all catchment areas. The efficacy of a linear relation between the availability factor and the rate of runoff, ie $c_3 = 1$ in equation (39), was investigated by Alley (1981), who concluded that its inclusion showed no significant advantage over the use of equation (38) alone. Dissatisfaction with the performance of the exponential wash–off model has led several investigators (eg, Litwin and Donigian, 1978) to adopt algorithms based upon the theories of sediment transport. The removal of other pollutants is then estimated as a fraction of the sediment discharge through the application of 'potency factors'. Huber (1986) has noted that this approach may lend itself to easier calibration than exponential wash–off equations with separate rate constants for individual pollutants.

4.5 The Effect of Gulleypots

The gulleypot or catchbasin represents the connecting point between the above–ground and the below–ground phases of runoff on urban catchment areas. Their principal purpose may be seen as the trapping of solids and coarse debris before they can enter the pipe system. However, since the exit from the gulleypot is usually above its base level, a water seal is provided that prevents the emission of odours from the downstream sewers.

The efficiency of gulleypots is heavily dependent upon the frequency with which they are cleared by the local authority. The consequences of not maintaining the cleaning programme vary with season, but the inevitable result is that the gulleypot becomes a source of pollutants. Fletcher et al (1978) have shown that the dissolved oxygen in a typical gulleypot tends to fall to zero after some 4–7 days of dry weather. The development of septic conditions is followed by the onset of anaerobic digestion. During hot weather, hydrogen sulphide is produced. Summer dry periods result in sharp increases in biochemical oxygen demand, chemical oxygen demand, nitrate, ammonia and suspended solids. At the end of the dry period, the septic supernatant is flushed away early in the storm and contributes to the 'first foul flush' of pollutants. With regard to the mass of particulate matter passing through the gulleypot, results can be extremely variable. Mance and Harman (1978) have noted that gulleypots may trap up to half the total mass discharged during one event, but contribute up to 20 per cent of the mass discharged for another.

SWMM is one of the few urban water quality models that includes the effects of the gulleypots explicitly. Assuming a volume, V, of liquid is retained in the gulleypot before the beginning of the storm, the percentage of the pollutant removed, R_m, is given by:

$$R_m = 100\,[1 - \exp(-Q_c/1.6\ V)] \qquad (40)$$

where Q_c is the cumulative inflow volume. This algorithm was also adopted by Fletcher and Pratt (1981).

5. ALTERNATIVE APPROACHES TO WATER QUALITY SIMULATION MODELLING

The hydrological and hydraulic simulation models reviewed in earlier sections of this chapter provide a framework for relating pollutant inputs to surface dust contamination levels, the erosive action of runoff on that dust and consequently the level of pollutant in runoff. Suspended solids remain the only water quality parameter to which this approach has been satisfactorily applied. Problems with applying this technique to the prediction of minor and trace pollutants in runoff include

i) uncertainties associated with each stage of the simulation, leading to a large and generally unacceptable error in the predicted water quality parameter,

ii) lack of chemical theory to describe the partitioning of pollutants between the dissolved and particulate–associated phases of runoff,

iii) lack of modelled input for pollutants in rainfall contributing to runoff.

Developments in hydrological, hydraulic and water chemistry models may result in simulation models capable of predicting trace pollutant levels in urban runoff, but these are some years away. In the meantime, alternatives based on a systems approach have been considered. In this approach, the system contains the hydrological, hydraulic and chemical processes and these are not described explicitly. Instead, the output (water quality parameters) is linked directly to the input which includes air, pollutant source and catchment characteristics. The link is formulated in terms of probability theory (probabilistic models) or mass balance consideration (mass balance models).

5.1 Probabilistic Modelling

The probabilistic approach was developed by the U.S. Federal Highways Administration, and has been summarised by Gaboury et al (1987) and Strecker et al (1990). The methodology assumes that all inputs can be described by a lognormal distribution, and that the resulting water quality distribution is also lognormal. The analysis requires input data about the site, such as the drainage area, the percentage of impervious cover and the nature of the site, i.e., whether rural or urban, and long–term rainfall records for the area.

In the United States, the Environmental Protection Agency has established a national data base from the results obtained a the NURP sites. Flow and concentration data at each site were analysed to determine event mean concentrations (EMCs), defined as the total mass of pollutant discharged during each storm divided by the total volume of runoff (see Chapter 5, Section 4). The EMC–values for each site were represented by a 2–parameter lognormal distribution, which is completely specified by its mean and variance. These distributional parameters were stored on the data base for a range of pollutants, including TSS, COD, nitrate, phosphate and heavy metals. The model predicts the runoff volume from the rainfall data and an EMC from the site data. The method has been extended to prediction of the effect of highway runoff on receiving water quality (see Chapter 5).

However, the approach is heavily dependent upon the distributional assumptions, which have recently been further reviewed by Brizio et al (1989) and Hall et al (1990).

5.2 Mass Balance Modelling

Mass balance models use the emission inventory approach described in Chapter 1 to quantify the mass of pollutants introduced into the site. An empirical, site–specific expression then realtes water quality to the input parameters. As with the probabilistic models, only EMC–values can be estimated. A study in an outer London urban catchment (Revitt et al, 1990) yielded the relationship

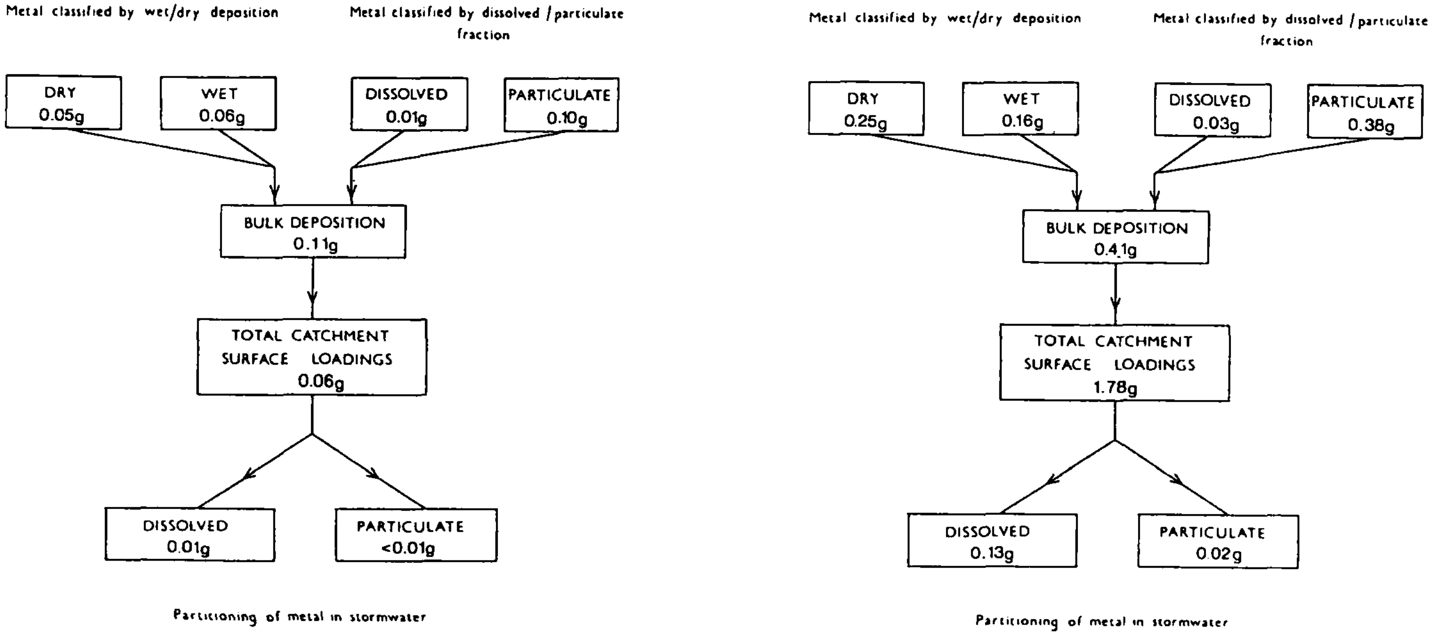
$$EMC = (1/R).(A_{rd}\, f_{rd} + A_{rf}\, f_{rf})\,(C_a V_d\, t + C_r\, r) \qquad (41)$$

where R is the total runoff volume, A_{rd} is the area of the road surface in m^2, A_{rf} is the roof area in m^2, f_{rd} is the fractional removal efficiency of the road, f_{rf} is the fractional removal efficiency of the roof area, C_a is the airborne pollutant concentration, C_r is the fractional concentration of pollutants in the rainfall, r is the depth of precipitation in m and t_a is the length of the antecedent dry period in compatible units.

The parameters, f_{rd} and f_{rf}, are site, storm and pollutant–specific. For example, at the outer London site referred to above, f_{rd} ranged from 0.07 for Pb to 0.3 for Zn, while f_{rf} will always be less than unity because, as described in Section 4.4, the erosive effect of stormwater runoff is only one of several removal mechanisms. Metal mass balances at the urban site are presented in Figure 4.6, which show that the percentage of metal present in deposition that appears in runoff varies between 10 per cent for Cd and Pb to 40 per cent for Cu. The metals are associated mainly with the particulate fraction. A similar study at a motorway site also showed that less than 10 per cent of the emitted Pb passed into the runoff system (Hewitt and Rashed, 1990).

6. CONCLUSIONS

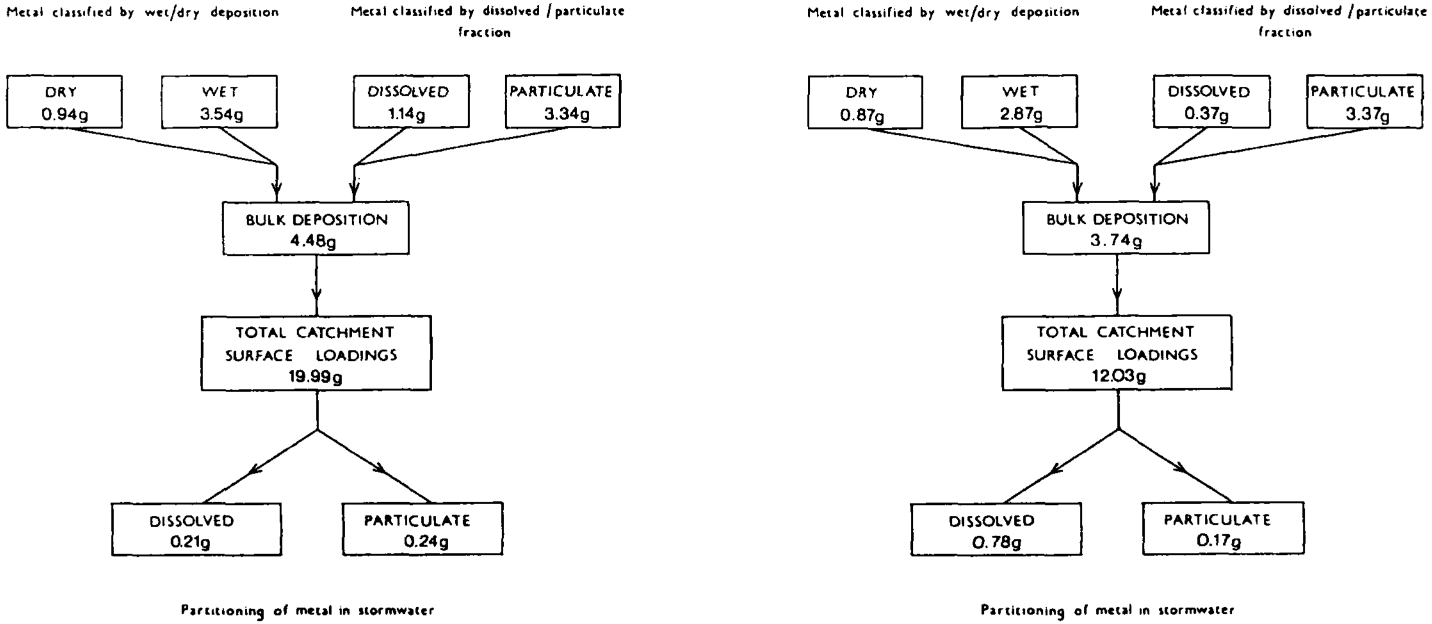
The hydrology and hydraulics of highway runoff are intrinsic to two distinct problems of engineering design: the drainage of the carriageways themselves; and the contribution of the roads and other impervious surfaces that drain to individual road gulleys to the design of the sewerage system of the wider urban area. The former is principally a problem of civil engineering hydraulics. However, analytical description of the three–dimensional flows over the surfaces of pavements and roads into the gutter, and the complex flow patterns around different types of road gulley has largely been avoided in favour of laboratory



a) cadmium

b) copper

FIGURE 4.6 (1 of 2) *TRANSPORT OF HEAVY METALS IN AN URBAN CATCHMENT*



c) lead

d) zinc

FIGURE 4.6 (2 of 2) *TRANSPORT OF HEAVY METALS IN AN URBAN CATCHMENT* modelling at both full–size and reduced scales. The analogy of the flow into a wide opening introduced by Li (1954) provides a convenient framework for the results obtained from laboratory tests without introducing too many coefficients that require calibration. Nevertheless, few investigators have explored its utility for engineering design. Furthermore, despite their acknowledged inefficiency relative to gratings set into the gutter, kerb inlets continue to attract attention (eg, Uyumaz, 1988).

When considering the design of the sewerage system, the detailed hydraulics of individual inlet areas becomes less relevant, and the transformation of storm rainfall into runoff is invariably described using a conceptual model. Moreover, this model has 'lumped' parameters in the sense that they apply to the whole inlet area. This somewhat broad brush approach has the advantage of effecting some economy in data collection and the coding of input to computer software. The values of the parameters are often estimated by means of relationships derived by MLRA in which the independent variables are catchment characteristics easily measured from catchment maps or climatic variables that are also tabulated or mapped. Since the conduits and manholes that form the sewerage system are well defined in their geometry and hydraulic characteristics, more powerful hydraulic algorithms can be applied on a routine basis for routing the inlet hydrographs to the catchment outlet.

When attention is turned to the modelling of water quality, the majority of the available models are seen to be semi–empirical. Moreover, since both accumulation and wash–off parameters are invariably chosen to apply to the whole inlet area, these models are also 'lumped'. For calibration purposes, runoffs and quality determinands are measured at the gulley, and the loadings estimated are assumed to emanate from the effective (connected) impervious area. Alternative formulations which borrow from the theories of sediment transport have yet to establish themselves as worthy replacements for these long–serving, semi–empirical accumulation/wash–off models. Should they prove successful in the longer term, the necessity to calibrate water quality models for individual pollutants and specific catchment areas is likely to remain an essential feature of engineering practice.

The simplicity of the models that are applied to describe the quantity and the quality of the runoff from inlet areas is often inadvertently obscured by the user–friendliness of the computer packages into which they are incorporated. The screen input systems and pop–up menus that are a feature of modern microcomputer software undoubtedly widen the use of packages to those without the experience of, or the confidence to apply, programming skills. Nevertheless, the sensitivity of model results to the input parameters or the model assumptions seldom receives more than cursory attention. The default values of key parameters that are available in some packages can be particularly misleading if applied out of context. Although the problem is one of education, perhaps the time has come for the modellers to pay more attention to the manner in which users will apply their results. An expert system for urban water quantity and quality modelling is now long overdue.

NOTATION

a	acceleration
a_1,a_2,a_3	coefficients
b_1,b_2	exponents
c	constant depending on the units of the Manning formula
c_1,c_2,c_3	constants
d	depth of approach flow q
f	gutter slope
g	acceleration due to gravity
i	average rainfall intensity
k_1,k_2,k_3	constants
m	power to which outflow discharge is raised in a non–linear reservoir model
n	roughness coefficient
q	flow towards a wide opening
s	cross slope = tan Θ
t	time
u	mean velocity of flow Q
v	mean velocity of flow on road side of grating
y_k	depth of flow at the kerb
y_s	depth of flow at upslope edge of the gully grating
A	area drained by the gully
C	constant
D	depth of flow approaching the gully grating
D_d	dry deposition rate
Dt	time increment
K	constant
K_a	availability factor
K_c	storage constant of a conceptual reservoir model
K_w	rate constant
L	length of opening to capture flow q
L'	length of opening to capture flow Q
L_p	length of the notional kerb inlet that will just capture flow outside the line of the gully grating
L_y	the gully grating length required to capture a flow depth y
S	storage of a conceptual reservoir model
P	mass of pollutant on the surface of an inlet area
P_L	upper limit to the accumulated mass of pollutants
P_o	mass of pollutant at the beginning of the time step
Q	flow approaching the gully grating; outflow from a conceptual reservoir model
Q_c	cumulative inflow to a gullypot
Q_i	flow intercepted by gully grating
Q_o	carry–over flow
Q_p	carry–past flow
R	total runoff volume
R_m	percentage of pollutant removed
V	volume of liquid retained in the gullypot
W	width of gulley grating
Θ	angle between the kerb and road surface
Λ	resuspension rate

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Chapter 5

HIGHWAY RUNOFF QUALITY, ENVIRONMENTAL IMPACTS AND CONTROL

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Contents **Page**

1. INTRODUCTION

During the last 10–15 years increasing control of discharges from point sources like municipal sewage treatment plants and industrial waste outlets has taken place in most of the industrialized countries of the World. Nevertheless, a corresponding increase in the quality of the surface waters has not always been the result. Therefore, an increasing concern has been devoted to investigate and evaluate these non–point impacts on receiving water bodies and introduce control measures if necessary.

In many ways pollution from non–point sources is more difficult to identify, assess and control than pollution from point sources. Pollution from highway areas related to wet weather discharges is typical in this respect. There are several reasons for this, but the fact that relatively large amounts of runoff water are generated during short wet weather periods randomly distributed over time is a major problem compared with the continuous discharges. Therefore, special methods and techniques must be put into service as far as both research and management are concerned.

Runoff from highway surfaces may take place in separate sewered as well as combined sewered areas. However, runoff in separate sewer systems must be regarded as the general case. Therefore, in this chapter emphasis will be put on the pollutional problems related to runoff in separate sewered areas.

2. SOURCES

Highway contaminants are deposited on roadway surfaces, median areas and right–of–ways from moving vehicles, stationary constructions and atmospheric fallout. The magnitude and pattern of accumulation appear to be a function of the roadway pavement and grade, traffic volume, maintenance activities, seasonal characteristics and adjacent land use. Much of the contaminants are typical of road surface material, fuel combustion products, lubrication system losses, degradation of automobile tyres, transported load losses, road surface cleaning/de–icing, paint and corrosion products. The accumulated mass rate of contaminants is a function of average daily traffic (ADT), air quality and intensity – duration – frequency of rainfall events. Heavy traffic in an industrialized area is expected to produce higher pollutant mass rate than light traffic in rural areas.

Rainwater falling upon highway areas washes out gases and foreign substances such as particulate matter, sulphuric acid, nitrate and nitric acids and other substances introduced into the atmosphere. After surface impaction of rainfall, a variety of substances is transported which include sand, silt, clay, plant debris, leaves, grass clippings, animal waste, refuse, chemicals, pavement material, fuel, oil, grease, and others. Therefore, highway runoff quality is extremely difficult to predict and depends on both traffic and local conditions. In regions with precipitation as snow in the winter period, pollutants may contaminate the snow at or near highways. Falling snow may absorb pollutants to a higher degree than rain, and depending on the length of time the snow has been lying on the ground, the concentration of pollutants in the snowmelt may reach higher values compared to the rainwater runoff. It is true that highway runoff of snowmelt is a special case but in many areas of relatively great importance.

2.1 Pollutant Accumulation

Particulates and other associated pollutants on highway surfaces can be generally attributed to atmospheric deposition, road degradation and traffic activities. Physical principles and models were reviewed in Chapter 4. This section concentrates on results for some specific pollutants. The quantity and quality of particulates vary greatly with land use and geographic location. Smith et al. [48] reported dust fall loads in the USA to approximate 0.23 $g/m^2 \cdot d$ at the northern region, 0.16–1.53 at the central region, 0.07–0.18 at the southern region and 0.06–0.16 at the eastern region. He concluded that the dry areas of mid USA are dustier than the wet areas to the east. The average annual dustfall loads for various water resources regions in the USA may vary between 0.05 and 0.7 $g/m^2 \cdot d$[60].

The ambient concentration of total suspended particles (TSP) is measured at nearly 2000 sites in the USA and it has remained relatively constant at about 60 $\mu g/m^3$ from 1975 through 1981. During the same period, emissions of particles from automobiles decreased about 20% [3]. It is not known whether strict enforcement of air pollution control regulation has any significant effect or not. However, a study reported by Rehm [41] concluded that atmospheric deposition in Milwaukee, Wisconsin showed a decreasing trend starting in 1967.

Vehicle related particulates on highway surfaces originate from exhaust emissions, roadway abrasion, tyre wear, vehicle load and degradation of moving parts in vehicles. The composition of vehicle related particulates studied by PEDCO Environmental Inc. [36] revealed that 37% of the particles were caused by each of the tyre wear and pavement wear, 18.5% by engine and brake wear, and 7.5% due to settleable exhaust. Exhaust emissions vary widely with age, size and condition of the vehicle and driving speeds, etc. [50]. Huntzicker et al. [22] reported exhaust emissions averaging 0.065 kg/1000 vehicle–km and consisted of 45% particulate lead, and 54% other metallic substances or organic hydrocarbons. Reductions in the level of lead in petrol produced reductions in Pb emissions, as described in Chapter 1.

A recent study by Rexnord, Inc. [43] on sources and migration of highway runoff pollution measured solids deposition and their particle size distribution on selected highways in the USA. The sites studied include: I–94, 8 lanes asphalt highway in urban Milwaukee, Wisconsin, HWY–50, 8 lanes concrete highway in urban Sacramento, California, and I–85, 4 lanes asphalt highway in rural Effland, North Carolina. The solids loadings and size distribution of dry solids after sweeping and the remaining solids in water after flushing the highway area are summarized and presented in Table 5.1. The data shows a wide variation in solids loading based on location, pavement material and traffic volume. The average daily traffic volume (ADT) was 116,000 at I–94, 85,900 at HWY–50, and 25,000 at I–85. Also the table shows the size particle analysis of deposits removed from highway surfaces by sweeping and followed by water flushing.

3. QUALITY CHARACTERISTICS

Highway runoff contains contaminants in dissolved and particulate forms. The standard pollutants are: total suspended solids (TSS), biochemical oxygen demand (BOD), chemical oxygen demand (COD), total phosphorus (TP), orthophosphate (OP), total kjeldahl nitrogen (TKN), nitrite plus nitrate nitrogen (NO_2+NO_3–N), copper (Cu), lead (Pb), zinc (Zn), nickel (Ni), chromium (Cr), iron (Fe), and cadmium (Cd), hydrocarbons and coliform bacteria. These pollutants are representative of important categories namely: suspended solids or particulates, oxygen consuming constituents, nutrients, heavy metals, trace organics and microorganisms. Runoff flows and concentrations are highly variable. Considerable variations in flow intensity and pollutant concentration are noticed during a single storm event and between different events at each of the monitoring sites [67, 68]. Data for each pollutant from all storms at a specific location appear to be well represented by a log–normal distribution. It is not known whether a log–normal distribution actually defines the underlying population of pollutant concentrations, or is at the least a satisfactory approximation for most environmental analysis [8].

TABLE 5.1

Solids loadings and particle size distribution on selected highways in USA (43).

Fraction	Size fraction	I–95		HWY–50		I–85	
		kg/km	%	kg/km	%	kg/km	%
Dry solids from sweeping	>3350 μm	119	16.1	6.57	24.9	181	40
	1410–3350	85.7	11.6	3.00	11.4	82.0	18.2
	841–1410	71.9	9.7	1.50	5.7	42.0	9.3
	420–841	127	17.2	1.61	6.1	49.6	11.0
	250–420	123	16.6	0.76	2.9	29.0	6.4
	74–250	158	21.5	0.96	3.6	43.7	9.7
	44–74	20.7	2.8	0.15	0.6	7.41	1.6
	<44	15.6	2.1	0.06	0.2	4.93	1.1
Flushing by water	Flushed solids	18.0	2.4	11.8	44.6	12.3	2.7
Total Solids Loading		739	100	26.4	100	452	100

For over ten years the Federal Highway Administration (FHWA) of the United States Department of Transportation and other state departments of transportation have been engaged in a research program to characterize stormwater runoff from highways, assess its impacts on receiving waters, and determine the effectiveness of various control measures. The highway runoff quality data consist of laboratory analysis of sequential discrete samples that are taken at recorded times throughout the runoff events, plus analyses of flow weighted composite samples taken over the entire runoff event. The event mean concentration (EMC) for a certain pollutant during a stormwater runoff event is calculated as the mass of pollutant discharged divided by the total quantity of water discharged during the event. Shelley and Gaboury [47] presented a preliminary approach to estimate pollutant load from highway runoff for planning purposes. They recommended highway runoff characteristics (Table 5.2) to be used by planners to estimate the pollutant load coming from an ungauged highway site. The values for concentrations and coefficient of variation for each pollutant are the best initial estimates for screening purposes.

TABLE 5.2

Highway runoff median concentrations after Shelley and Gaboury (47).

Parameter	Concentration (mg/ℓ) Urban sites	Rural sites	All sites	Coefficient of variation Cv
SS	220	26	108	0.8 – 1.0
COD	124	41	86	0.5 – 0.8
TKN	2.72	1.4	2.18	0.7 – 0.9
TP	0.19	0.04	0.11	0.6 – 0.9
Pb	0.55	0.09	0.31	0.7 – 1.4
Zn	0.38	0.09	0.24	0.6 – 0.7

A relationship exists between median values and mean values for the log–normal distribution as follows:

$$\frac{\text{Mean}}{\text{Median}} = \sqrt{1 + C_v^2}$$

where C_v = coefficient of variation

The data indicate a coefficient of variation less than 1.0 (Table 5.2) except for Pb. Also, extensive studies were conducted to characterize runoff quality at Maitland Interchange near Orlando, Florida [65]. Statistical analysis of EMC for storm events collected from Maitland Interchange are presented in Table 5.3, which shows lower values if compared with median values presented in Table 5.2. Of course, Maitland site in Florida is washed off more frequently, no de–icing salts are used and no heavy industry exist in the vicinity of the site. The Florida site is expected to produce lower pollutant loads compared to other sites which are heavily urbanized with less rainfall during the year. Furthermore, de–icing salts are used frequently during winter season in areas with a cold climate.

TABLE 5.3

Standard analysis of urban highway runoff at Maitland Interchange near Orlando, Florida [67, 68].

Parameter	Unit	No. of obs.	Statistical analysis			
			Mean	Cov	Median	Range
pH		13	6.6	0.3	7.5	5.9–7.8
Turbidity	NTU	11	9.30	0.60	9.10	2.51–22.5
Alk.	mg/ℓ	13	41	0.26	44	10–70
Sp.Conduct.	μmho/cm	12	107.0	0.49	80	45–175
Organic N	μg/ℓ	6	965	0.64	741	292–1891
NH_3–N	μg/ℓ	13	152	0.63	78	9–972
NO_2–N	μg/ℓ	13	13	0.35	8	1–37
NO_3–N	μg/ℓ	12	306	0.49	246	46–665
OP	μg/ℓ	13	76	0.81	65	26–178
TP	μg/ℓ	13	170	0.66	147	62–346
Total Pb	μg/ℓ	16	163	0.70	119	30–379
Diss. Pb	μg/ℓ	16	34	0.81	25	13–128
Total Zn	μg/ℓ	16	71	0.68	53	13–173
Diss. Zn	μg/ℓ	16	40	0.74	33	13–134
Total Cu	μg/ℓ	16	37	0.63	35	10–101
Diss. Cu	μg/ℓ	16	26	0.59	22	10–64

3.1 Suspended Solids

Total solids accumulation on the highway surface are shown to be correlated to the average daily traffic. Also, highway surface pollutants are highly correlated to suspended solids [14,15]. Existing data indicate that large fractions of heavy metals and other pollutants are attached to solid particles and the concentrations are generally higher in small size particles than large ones. Therefore, the particle size distribution is an important design parameter if one attempts to examine the pollutant fraction in suspension and the effectiveness of best management practices for highway runoff. Settling column data on urban and highway runoff indicate that a large fraction of TSS and other associated pollutants is removed by sedimentation. Driscoll [7] presented best estimates for the distribution of particle. Settling velocities in urban runoff from various study sites in the USA are shown in Table 5.4. The calculated particle size diameters shown in the table are based on settling of discrete particles using Stoke's Law. The water temperature is assumed to be 10°C, water viscosity is 1.31 x 10^{-2} g/cm·s (poise), specific density for particles is 2.65 g/cm³ and for water 1 g/cm³. Therefore Stoke's Law is applied as follows:

$$u = \frac{gd^2(\rho_s - \rho_v)}{18\mu}$$

Where
u = settling velocity of particles, cm/s
g = acceleration due to gravity, cm/s²
d = particle diameter, cm
ρ_s = specific density of solids, g/cm³
ρ_v = specific density of water, g/cm³
μ = water viscosity at 10°C, poise

TABLE 5.4

Particle settling velocity distribution in urban runoff.

Size fraction	particles in urban runoff (% of mass)	Average settling velocity (m/hr)	Calculated particle diameter (μm)
1	00–20	0.01	2
2	20–40	0.09	6
3	40–60	0.48	14
4	60–80	2.2	30
5	80–100	20	90

The calculated particle size in Table 5.4 shows that suspended solids in runoff water are primarily less than 100 μm diameter. This appears to be consistent with published literature (Table 5.5) of particle size distribution for highway runoff composite samples collected from various sites [43]. On the average, approximately 78% of suspended particles in highway runoff are less than 44 μm diameter. This is consistent with Table 5.4 which shows that 60–80% of suspended solids in urban runoff are less than 30 μm diameter. Also, Randall et al. [40] reported the analysis of particle size distribution of suspended solids in seven urban runoff samples. An average of 80% of the total particles were less than 25 μm in diameter and 57% had diameters less than 15 μm. There appears to be no difference between size particle distribution carried by highway runoff and urban runoff.

The distribution of pollutant loads among size fractions of suspended particulate matter in runoff entering stormwater management systems has ecological and practical implications. Wagner and Mitchell [58] reported that a substantial portion of TSS, TP, NO_3–N and TKN loads entering two Massachusetts lakes during one storm event was associated with the smaller size particles less than 10 μm. Failure to remove the small particulate and dissolved fractions of these loads would prevent achievement of target concentrations for these pollutants.

TABLE 5.5

Wet sieving analysis of highway runoff composite samples.

Particle size (μm)	Percent of suspended solids, mean value				
	Sacramento HWY 50	Harrisburg I–81	Milwaukee I–94	Effland I–85	
>250	1.54	6.10	14.56	3.58	6.45
88–250	9.07	6.70	7.00	1.30	6.02
44–88	10.70	11.70	5.84	8.06	9.08
<44	78.69	75.50	72.60	87.06	78.45

Distributions of pollutants loads among particle size was recognised in early studies by Sartor and Boyd [45], who reported varying distributions according to pollutant and geographic location. In general, much of the TSS and TKN loads were associated with the 250–2000 μm fraction, while TP loads were concentrated in the less than 100 μm particles and most nitrate nitrogen was dissolved. It is important to realize that the lowest metal concentrations are associated with the largest particle size fractions, while the highest metal concentrations are associated with the lowest size fraction. Svensson [49] reports that there is a linear relationship between solids associated metal concentrations and surface area of the particles. The particle shape can be approximated by a sphere, thus creating an inverse relationship between solids associated metal concentrations and particle diameter.

The dissolved and colloidal fractions for various pollutants can vary between 20% and 70% of the total concentrations for most pollutants. Some pollutants show higher affinity for suspended solids than others. Table 5.6 presents data on percent dissolved fractions in highway runoff [68] and urban runoff water discharged into Lake Eola, Orlando, Florida [60]. These results suggest that much of the pollutants load remain in suspension or solution after short–term settling.

TABLE 5.6

Dissolved fractions for pollutants in runoff water

Pollutant	Average percent dissolved	
	Highway runoff*	Urban runoff**
TN	34	66
TP	41	49
Pb	21	20
Zn	57	43
Cu	70	78
Ni	76	52
Cr	65	30
Cd	72	55
Fe	27	

* Yousef et al. (68)
** Wanielista et al. (60)

3.2 Heavy metals

Sources of metals in highway runoff include atmospheric deposition, naturally occurring metals in soils, and highway related sources such as paint, corrosion products and others. Metals commonly found in highway runoff include lead, zinc, iron, copper, cadmium, chromium and nickel. However, arsenic and cyanide may also be present in significant quantities in de–icing salts. Pb, Zn and Fe represent the major metal load in highway runoff. Measured concentrations of heavy metals in 150 sequential samples during sixteen individual storm events are reported by Yousef et al. [68] (Table 5.7). The data collected during this study may indicate a "first–flush" effect for total concentrations of Pb, Zn, and Fe. In general, 50 percent of the total mass of these metals was found to be transported during the first quarter of a storm event, 25 percent during the second quarter, and the remaining 25 percent divided between the third and fourth quarters. This trend was not observed for dissolved species of any measured metals.

Probability distribution of mean flow–weighted concentrations (EMC) of heavy metals in highway runoff appears to follow a log–normal distribution as shown in Figure 5.1. A linear fit to a log–normal distribution generally indicates a positively skewed data distribution when plotting the probability of occurrence on the Y–axis versus metal concentration on the X–axis. This suggests that the majority of events are presented by concentrations which are on the lower end of the range of values, with the more

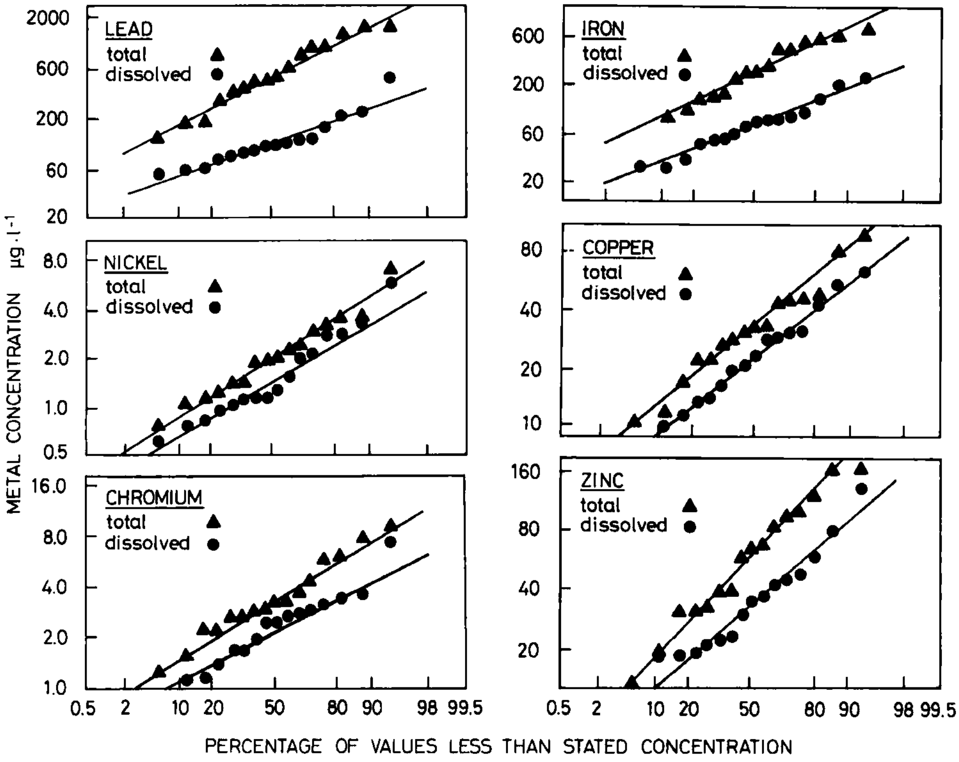


FIGURE 5.1 *DISTRIBUTION OF HEAVY METALS IN HIGHWAY RUNOFF ENTERING MAITLAND POND DURING 1983–84*

concentrated events making up a much smaller percentage. Dissolved concentrations also seem to follow a log–normal distribution although more deviation may be present.

TABLE 5.7

Event mean concentrations of heavy metals in highway runoff at Maitland Interchange, Orlando, Florida, (68).

Metal	Metal concentration ($\mu g/\ell$)					
	Dissolved			Total		
	Min	Max	Mean	Min	Max	Mean
Pb	13.1	128	33.5	29.7	379	163
Zn	12.8	134	40.4	12.8	173	71
Cu	10.3	63.8	26.4	10.4	101	37.5
Ni	0.9	8.3	2.6	1.1	10	3.4
Cr	1.0	7.7	2.6	1.3	9.7	4.0
Cd	0.5	7.2	1.8	0.7	8.9	2.5
Fe	27.7	255	93.4	25.9	796	341

Table 5.7 shows that Cd, Ni, and Cu are all present in dissolved fractions which are about 75% of the total metal measured. On the other extreme, Pb and Fe are predominantly particulate in nature with dissolved fractions of only approximately 20%. Zn and Cr may be divided equally in dissolved and particulate forms. The quantities of heavy metals in highway runoff and the forms in which they exist depend to a large degree on the physical and chemical behaviour of the specific elements. Lead has been shown to exist on the highway in a predominantly insoluble particulate form which is largely inorganic in nature [30]. Most of the lead emitted from automobile exhaust occurs in two distinct particle sizes: < 1 μm and 5–50 μm. Wang et al. [59] found approximately 92% of the total lead present was associated with particles > 20 μm. In most highway runoff investigations, it is found that Zn, Cu, and Cd are considerably more soluble than Pb. Concentrations and loading rates of heavy metals in highway runoff are highly site specific and reflect variations in dustfall, average daily traffic, land use in the surrounding area of the highway as well as many other contributing parameters. It is interesting to notice that significant reductions in Pb were observed at the Milwaukee, Wisconsin site compared to earlier studies. These reductions were directly related to reduction in sale of leaded gasoline [43].

Measuring the total concentration of a particular metal in a water sample can be very misleading because, depending on the chemical forms of the metal, a water with high total metal concentration may be, in fact, less toxic than another water with a lower total metal concentration. The study of a chemical speciation of trace heavy metals and subdividing each metal concentration into several fractions is a task of theoretical and practical difficulty. However, the Differential Pulse Anodic Stripping Voltammetry (DPASV) measures different species of a metal in concentration as low as 10–8 or 10–9 moles/litre. Yousef et al. [66] used DPASV and reported that up to 70% of the soluble Cd and Zn in highway runoff responded directly to measurement by DPASV. Most of the lead exists as $PbCO_3$ and a good fraction of Cu is associated with organic complexes if humic substances are present.

Accumulation of pollutants in snow along highways has been identified as important in regions with snow precipitation as a common phenomenon. The accumulation of heavy metals in snow was studied by Lisper [33] near a highway in Gothenburg, Sweden. Results from this study are presented in Table 5.8. Also, heavy metal concentrations in snow along a highway in the midwestern part of Sweden were reported [34],(Table 5.9). The variations in the results were large, probably affected by alternating snowfall and snowmelt in the sampling period. Significantly higher concentrations in the highway snowmelt runoff were recorded compared to rooftop runoff from the same catchment.

TABLE 5.8

Heavy metals in different types of snow near a highway in Gothenburg, Sweden [33].

Metal	Metal concentration ($\mu g/\ell$)		
	newly fallen snow	composite snow	melting snow
Pb	75	205	400
Zn	85	135	540
Cu	25	35	120

TABLE 5.9

Heavy metal concentrations in snow from a highway grass dividing strip in the midwestern part of Sweden (34).

Metal	Metal concentration ($\mu g/\ell$)	
	mean	max.
Pb	1,700	3,100
Zn	250	490
Cu	170	320
Cd	4.9	12
Fe	45,000	60,000

Also, the bioavailable species in snowmelt may occur in higher concentrations than in stormwater from the same catchment [35]. This investigation also reports that temporal variations of Zn and Cu were strongly dependent on suspended solid organic carbon discharges, while Pb and Cd loadings were controlled by chloride.

3.3 Nutrients

Nutrients in highway runoff, particularly nitrogen and phosphorus generally originate from atmospheric deposition and roadside fertilizer applications. Concentrations of N and P in runoff from highway sites are generally in the range of values for urban runoff. However, if there is any difference the nitrogen tends to be higher and the phosphorus tends to be lower in highway runoff samples than urban runoff water. Median values for TKN and TP concentrations are presented in Table 5.2. Also, analysis of nutrients in urban highway runoff at Maitland Interchange is presented in Table 5.3. The data on concentrations of nutrients presented in Table 5.2 are in agreement with data presented in Table 5.3.

Loadings of nutrients and heavy metals are estimated in four highway sites in the USA [43]. These loadings are runoff normalized, kg/km/cm of runoff, and averaged in Table 5.10. It should be noted that the loadings are produced for one kilometre of highway length regardless of the number of lanes and for both directions. By estimating the average EMC and the quality of runoff produced from a section of highway, an estimate of pollutant mass carried by highway runoff water can be made for planning purposes.

3.4 Oil and grease

These pollutants originate from spills, leaks, blow–by of motor lubricants, antifreeze, hydraulic fluids and asphalt surface leachate. Oil and grease concentrations in highway runoff samples varied between 1 and 27 mg/ℓ and averaged 9 mg/ℓ [43]. These concentrations produced a loading rate varying between 0.32 and 12.5 kg/km/cm and a mean value of 3.46 kg/km/cm.

3.5 Bacteriological analysis

Highway runoff samples generally contain total coliform, fecal coliform and fecal streptococci. Bacterial count is generally less than 1000 organisms per one millilitre of water and the fecal coliform (FC) to fecal streptococcus (FS) ratio can vary between 0.01 and 12.8. When the ratio of fecal coliforms to fecal streptococci is in excess of 4.0, it is indicative of pollution from human sources, while ratios less than 0.7 indicate pollution of animal origin [11, 12]. However, the bacteriological contamination in runoff water are predominantly of animal origin.

TABLE 5.10

Pollutant loading from four highway sites in USA after Rexnord (43).

Parameter	Loading kg/km/cm	
	Range	Mean
TP	0.05 – 1.02	0.15
TKN	0.30 – 4.56	0.93
NO_2+NO_3–N	0.02 – 2.55	0.23
Pb	ND* – 2.93	0.12
Zn	0.01 – 1.35	0.13
Fe	0.10 – 32.6	2.52
Cr	ND – 0.08	0.004
Cd	ND – 0.27	0.03
Ni	ND – 0.10	0.001
g	ND – 0.33 x 10^{-3}	0.05 x 10^{-3}
AS	ND – 0.011	ND
*Not Detectable		

Geldreich et al. [11] analyzed 177 samples from street gutters during a two year period for fecal coliform (FC) and fecal streptococcus (FS) and total coliform (TC). He reported higher counts of TC, FC and FS during summer and fall in comparison to the winter and spring and concluded that fecal contamination in gutter debris is from warm blooded animals other than man. Gupta et al. [14] supported these findings at sites in Wisconsin, Pennsylvania, Tennessee and Colorado. They also reported FC to FS ratio typically below 0.7. The same findings are supported by a more recent study by Rexnord, Inc. [43] on highway runoff from four other sites in the USA.

3.6 Organic toxic matter

Polychlorinated biphenyls (PCBs) and pesticides enter the highway system from spraying highway right–of–ways and atmospheric deposition. PCB has also been used as a catalyst in synthetic tyres. Pesticides originate mostly from the use of weed killer compounds on the highway system. Atmospheric deposition is likely to be the main source of PCBs [10,53]. Detectable concentrations of PCBs were observed in the atmospheric wash–out at these sites studied by Rexnord, Inc. [43], and PCB loading was estimated to be 0.0006 kg/highway km. Concentrations in runoff samples ranged from ND to 1.60 $\mu g/\ell$.

4. LOADINGS

The water quality responses of pollutants from highway runoff are influenced by many factors such as meteorological, hydrological and geological conditions, plus land use practices. The quantity of flow is characterized by amount, frequency, intensity, duration and pattern of precipitation. These parameters are indicative of the spatial and temporal fluctuations of the precipitation within a storm and between storm events. Therefore, grab sampling is of little use since it represents one instantaneous value, which most likely is not representative of the average conditions, and mass changes cannot be described. Sequential samples at equal time intervals or at equal flow volumes discharged during a storm event are much preferred than one grab sample; this was reviewed in Chapter 1.

4.1 Concentrations and mass loadings

It is appropriate and convenient to propose that the event duration be the natural timescale for the analysis and evaluation of runoff concentrations and loads. On this time scale the average pollutant concentration can be represented by the total mass discharged during a storm event divided by the total runoff volume of the event. This average flow–weighted concentration is defined as the event mean concentration (EMC). It may be determined by integration of discrete samples taken during a storm, or less expensively, from composite sampling.

$$\mathrm{EMC} = \frac{M}{C} = \frac{\sum_{i=1}^{n} V_i \cdot C_i}{\sum_{i=1}^{n} V_i}$$

where
M = pollutant loading during a storm event, g.
V = total runoff volume during a storm event, m^3.
Vi = volume discharged during sampling period "i".
Ci = concentration representing sampling period "i".

Some generalisations may be made about EMCs (Huber et al. [21]:

- EMCs are log–normally distributed, Figure 5.1, or closely approximate. This is true both for all EMCs at one site and for site median EMCs for all sites.
- The variation across all sites and for most pollutants is similar. The coefficients of variation (standard deviation divided by the mean) of EMCs fall in the range of 0.5 to 1.0.
- EMCs are for all practical purposes uncorrelated with the runoff volume.
- Runoff volumes also tend to be log–normal, but with a significant bias toward large events. This results in the mean runoff volume being much larger than most storms at a site.

It is important to realize the definition of an event. Available rainfall data for a specified location are generally divided into separate, statistically independent rainfall events based on the time intervals of dry weather period between two successive events. These dry periods should be long enough to ensure independent runoff events and to minimize cumulative effects of successive storm events [25]. If one changes the definition of a storm event based on the minimum length of the inter–event dry period, the total number of events in a known rainfall record, some of the EMCs and the associated statistical analysis will be changed. Resulting values of inter–event dry periods are in the range of 1–72 hours [25, 54].

An annual mass loading from a specific site can be estimated. This mass load is provided by the product of the runoff volume and concentration in the following way:

annual mass load =
mean event mass load x number of events per year.

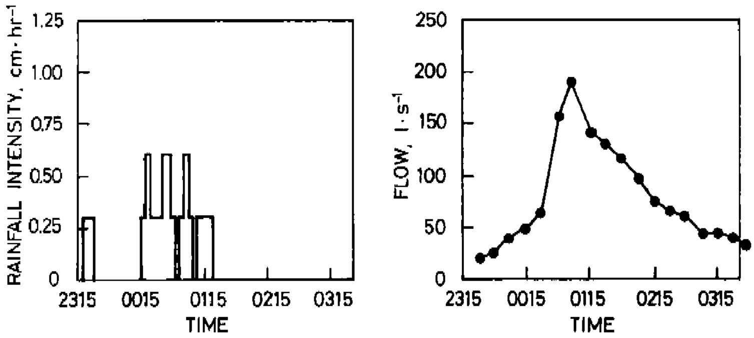
In turn, the mean event load is computed as follows:

mean event mass load = mean event runoff volume x mean EMC

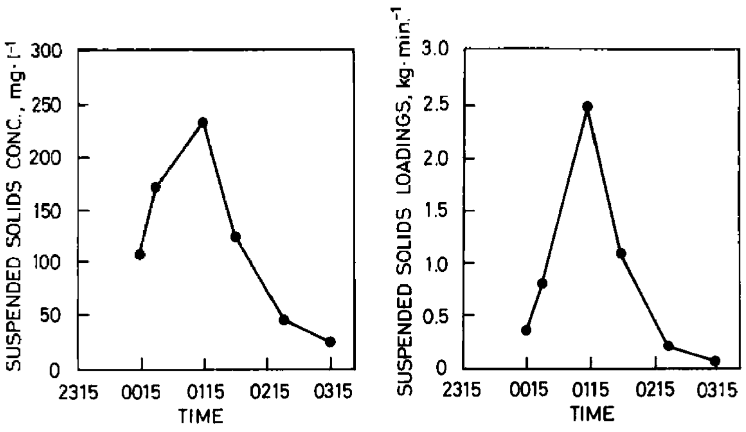
Details in this procedure is given by Driscoll et al. [9]. Mass loading of a constituent in highway runoff can be estimated in different units. Examples are g/km/yr, g/m^2/yr or g/km/cm of runoff.

The concentrations measured in runoff water may be insignificant. However, contributions based on mass loading may be significant. Typical stormwater quantity and quality response curves are shown in Figure 5.2. The hydrograph represents changes in flow rate with time or runoff volume during a storm event, while the pollutograph reflects changes in concentration. The loadograph reflects the changes in cumulative mass of constituents "i" with time. Figure 5.2 illustrates the general shape of hyetograph, pollutograph and loadograph.

The relationship between the hydrograph and the loadograph, i.e. the relative variation in pollutant loading related to the temporal change in stormwater runoff volume throughout a storm event, can be described by introducing the first flush phenomenon. This phenomenon can be illustrated by plotting the cumulated pollutant mass load to the total mass load ratio versus the cumulated stormwater volume to the total runoff volume ratio.



a) HYETOGRAPH b) HYDROGRAPH



c) POLLUTOGRAPH - CONCENTRATION d) POLLUTOGRAPH-LOADINGS

Fig. 5.2 Typical hyetograph, hydrograph, pollutograph and loadograph from highway runoff, adapted from Gupta et al. [14, 15].

Results exemplifying the first flush effect, originating from a highway site in central Copenhagen, Denmark, are shown in Figure 5.3 (63).

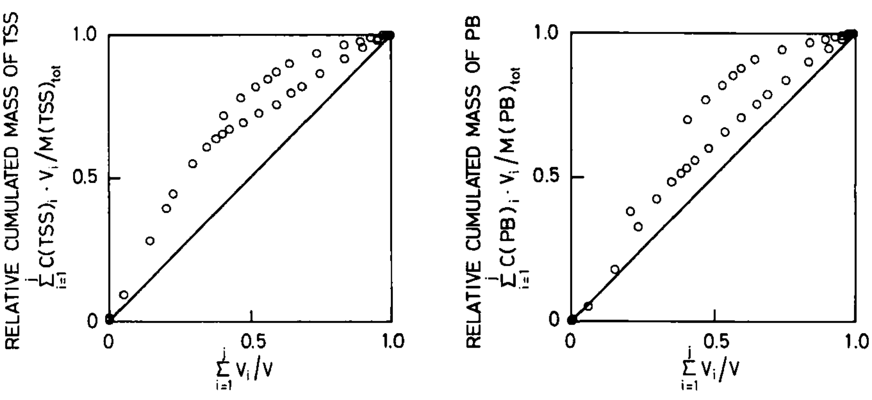


Fig. 5.3 Relative cumulated mass of total suspended solids and lead versus relative cumulated volume indicating a first flush phenomenon for two storm events, adapted from [62].

Total mass of pollutant discharged:

$$M = \sum_{i=1}^{n} V_i \cdot C_i$$

Total volume of stormwater discharged:

$$V = \sum_{i=1}^{n} V_i$$

i" denotes the sampling period.

Previous investigators reported a marked first flush effect in which the concentrations of pollutants initially rise to high values and then fall off to lower, but still significant levels during storm events. This phenomenon is known as the first flush and it is less noticeable during storms with a low, even rate of runoff. This can also be explained by the relatively high rate of pollutant wash–off during the initial phase of the storm. The nature of chemical species and their solubilities in water may also affect the magnitude and extent of the first flush effect. For example total lead and total zinc show a much more pronounced first flush than soluble lead and soluble zinc (Figure 5.4). Also the total lead shows more pronounced first flush than total zinc.

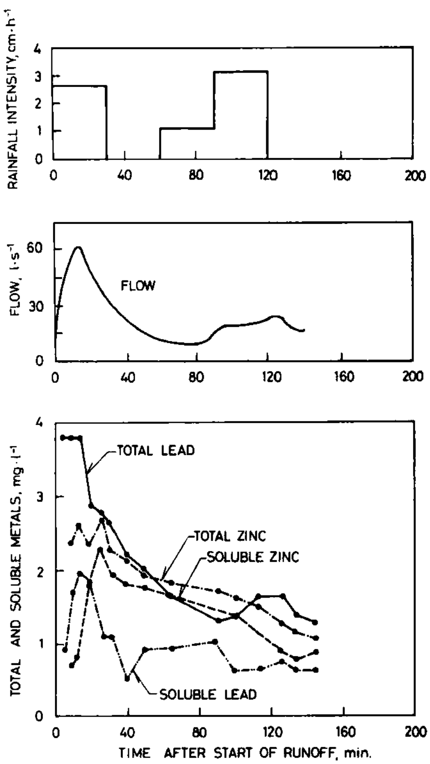


Fig. 5.4 **Typical first flush effect for various metals in runoff water, adapted from Shaheen [46].**

It has been intensively discussed whether first flush is a phenomenon of importance for receiving water impacts. In order to evaluate pollutant impacts it is in this connection important to distinguish between immediate effects, mainly affected by the concentration and its temporal variations in the runoff water, and delayed or cumulative effects resulting from the stormwater load during one or several runoff events.

4.2 Computations and modelling

In order to compute loadings from highway systems a simple predictive procedure has been developed [27]. The procedure is made up of four components corresponding to the following elements:

Rainfall–runoff
Pollutant buildup (accumulation) on highway surfaces
Pollutant wash–off during runoff
Constituent loadings

A simple linear pollutant accumulation equation of the predictive model is, following equation (30) of Chapter 4,

$$P_t = P_o + K_l \cdot \Delta t$$

where P_t = amount of pollutant per unit length of a highway at time t, g/km.
P_o = residual on surface prior to time t, g/km.
K_l = pollutant accumulation rate, g/km·d.
Δt = time of accumulation, d.

Various methods based on monitored data exist for estimating P_o and K_l. These methods are described and discussed in details in Kobriger et al. [27]. Also, from this study the average daily traffic (ADT) appears to show a good relationship with K_l values based on the total solids accumulation rate as shown in Figure 5.5. Regression analyses show dustfall to be a poor estimator of K_l.

As for wash-off of pollutants accumulated on highway surfaces an exponential equation is chosen according to early investigations in the USA [1, 19]. A similar equation is also used in European runoff models, e.g. Lindholm [32]:

$\Delta P = P_t[1 - (\exp{-K_2 \cdot v})]$, corresponding to equation (38) of Chapter 4,

where ΔP = amount of pollutant discharged during a runoff event, g/km.

K_2 = washoff coefficient, hr/m.

v = average runoff rate, m/hr.

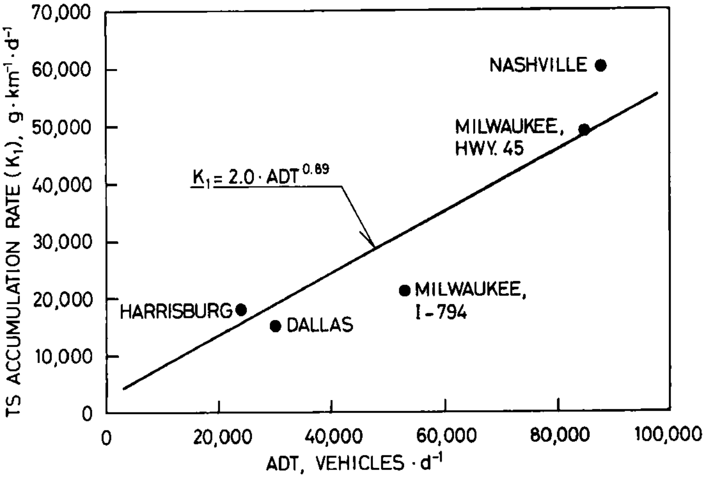


Fig. 5.5 **Selected total solids (TS) accumulation rates versus average daily traffic (ADT). Regression coefficient r = 0.93.**

Determination of K_2-values may be based on a calibration technique. For the monitored runoff events covering the highway systems shown in Figure 5.5, the range of K_2-values that were selected for each site are presented in Table 5.11.

TABLE 5.11

Range of K_2-values for each monitoring site.

Site		K_2(hr/m)
Milwaukee	I-794	5 – 8
Milwaukee	HWY-45	5 – 8
Harrisburg	I-81	4 – 20
Nashville	I-40	5 – 8

Low values of K_2 may be expected for e.g. urban, elevated bridge decks, 100% paved, with impact barriers and highways with kerbs and a paved drainage area. For rural sites with flush shoulders and runoff through ditches higher values of K_2 may be chosen, e.g. 12 hr/m.

In Figure 5.6 the variability in pollutant accumulation and wash-off is indicated.

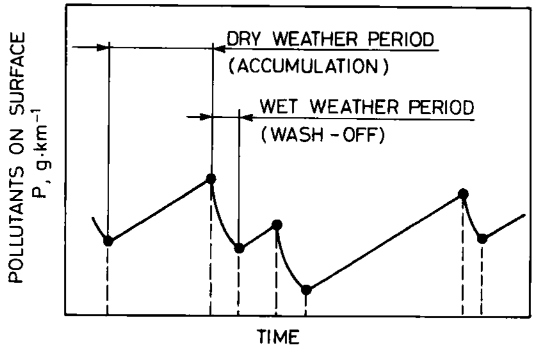


Fig. 5.6 **Sketch showing the variability of pollutants on a highway surface during dry and wet periods according to the pollutant accumulation and washoff model.**

Chapter 4 discussed alternative models for pollutant buildup and washoff, also Huber[20]. However, it must be recognized that there is a need to develop more physically based runoff quality models. The main reason is the evident need for calibration of the simple, empirical models for each application.

A simulation model which is based on the fundamental physical and chemical processes of heavy metal washoff and transport on street surfaces has recently been presented [49]. The solids transport model is based on sediment transport theories and a metal transport model which includes solids associated metals concentrations in the runoff. The concept of including the fundamental processes in runoff quality models seems correct and sound. However, the model in question needs further development before appropriate use in management is possible.

5. RECEIVING WATER IMPACTS

The constituents in highway runoff have been studied by many investigators who reported high loads of BOD, SS, nutrients, metals, de–icing salts, petroleum and oil compounds. They are concerned over detrimental effects on receiving water impacts which include hydrological, water quality, sediment and biological effects. Hydrological impacts result from the increase in percent imperviousness and changes in water storage and flow direction from the natural hydraulic regime of the drainage basin due to structural features. Additionally, density stratification in lakes may occur due to discharge of large quantities of de–icing salts. Pollutant concentrations in highway runoff are considered comparable to urban runoff from separately sewered areas, however, the overall loadings from highway runoff to surface waters are considerably lower. This is expected since highway right–of–way surface area is generally a small fraction of the total urban watershed for most surface waters. The average pollutant concentrations in highway runoff from six monitoring stations [15] and urban runoff from 28 cities in USA [55] are shown in Table 5.12 for comparison purposes. Because of high concentrations and loading shown in Table 5.12, operating highways have been identified as a potential source of a variety of pollutants to adjacent surface and subsurface waters.

Water quality response to highway runoff discharges may result in an increase of suspended sediment deposition and sediment pollutant concentration in localized areas close to the input source. These increased levels of pollutants can result in bioaccumulation of toxic materials and/or subtle changes in benthic organisms. Other dissolved pollutants may be sufficient to increase concentrations in water to a level higher than acute or chronic water quality criteria established for a particular receiving stream. The increased levels may cause highly visible impacts such as fish kills, taste and odour problems and/or severe alteration of the aquatic biological community.

TABLE 5.12

Summary of highway runoff and urban runoff quality data from study sites in the USA.

Pollutant	Highway runoff				Urban runoff
	Concentration (mg/ℓ)		Loading (kg/hr/cm runoff)		Median EMC (mg/ℓ)
	Average	Range	Average	Range	
Total Solids	1147	145–21,640	115	15–2,170	
Susp.Solids	261	4–1,656	26	0.4–166	100
Volatile SS	77	1–837	8	0.1–84	–
BOD (5 day)	24	2–133	2	0.2–13	9
COD	15	5–1,058	16	0.5–106	65
TKN	3	0.1–14	0.3	0–1.40	1.5
$NO_2 + NO_3$	1.1	0–8.4	0.1	0–0.8	0.7
TP	0.79	0.1–3.6	0.1	0–0.36	0.33
Chloride	386	5–13,300	39	1–1,333	–
Pb	0.96	0.02–13.1	0.1	0–1.3	0.144
Zn	0.41	0.01–3.4	0.04	0–0.34	0.16
Fe	10.3	0.1–45	1.0	0.01–4.5	–
Cu	0.10	0.01–0.88	0.01	0.0–0.09	0.034
Cd	0.04	0.01–0.40	0.004	0.0–0.04	–
Cr	0.04	0.01–0.14	0.004	0.0–0.014	–
Ni	9.9	0.1–49	1.0	0.01–1.2	–
$Hg \times 10^{-3}$	3.2	0.13–67	0.32	0.01–6.72	–
Polychlorinated biphenols $\times 10^{-3}$	0.33	0.02–8.89	0.03	0.0–0.9	–
Oil and Grease	9.5	1–104	0.95	0.1–10.4	–

5.1 **Dissolved oxygen impacts**

Many researchers have investigated the "potential" dissolved oxygen (DO) depletion of receiving waters resulting from discharges of stormwater and combined sewer overflows. Most of the DO sags related to urban runoff discharge result from combined sewer overflow (CSO) systems.

Lager et al. [28], demonstrated the potential for DO depletion as a major impact of urban stormwater on the basis of an average stormwater five–day biochemical oxygen demand (BOD_5) of 20 mg/ℓ. Table 5.12 shows a median value of BOD_5 for urban areas of 9 mg/ℓ and an average value for highway runoff of 24 mg/ℓ. These concentrations are often compared with BOD in wastewater treatment plant effluents (about 10–30 mg/ℓ), but the receiving water impacts are not comparable.

Stormwater BOD loadings are acute, intermittent and associated mainly with particulate matter which tend to settle rapidly [14, 15, 37, 46]. However, one must realize that kinetics of bacterial degradation of stormwater organic material are considerably different compared to the kinetics for scondary effluent [39]. Also, sedimentation of organic matter near the discharge point may result in delayed effects to increase the sediment oxygen demand (SOD) and to reduce the DO concentration in the water column.

Dissoved oxygen deficits due to long–term accumulation of organic deposits from highway runoff flowing into lakes and slow moving streams are difficult to document and to segregate from other sources of pollution. Organic deposits accumulate through direct input of runoff particulates to the sediments or through decaying and sedimentation of debris and biomass from primary producers. Of course nutrients carried in highway runoff can stimulate excessive algal and plant production which settle to the bottom sediments and increase SOD. It is extremely difficult to separate between the relative contribution from each source to the SOD. For example, Lake Eola is located in downtown Orlando, Florida and receives only stormwater runoff from the surrounding urban area. The DO in the top 0.5 metre of the water column fluctuates between 7 and 12 mg/ℓ. Concentrations of dissolved oxygen, although usually at or above saturation near the surface, drop periodically during the spring and summer month to 1 mg/ℓ or less at depths of 4.0 metres or greater. It seems reasonable to assume that areas in Lake Eola below 4–5 meters deep remain anoxic during much of the wet season resulting from storm–water impacts.

5.2 Nutrients impacts

Nutrient concentrations in highway runoff can average 0.79 mg/ℓ for TP and 4.1 mg/ℓ for TN (Table 5.12). These high concentrations may accelerate eutrophication in lakes or slow moving streams. However, the loadings from highway runoff to receiving waters may be quite low if compared with nutrient contribution from all other sources. Eutrophication in lakes depends on the nutrient loadings, the availability of nutrients and their removal mechanisms. Removal of nutrients can be accomplished by settling of runoff particulates, uptake by plant and algal species followed by settling, and flushing nutrients out of the receiving water.

Available nitrogen include ammonia plus nitrate N and available phosphorus include all inorganic soluble phosphorus plus 20–30 percent of the particulate. Cowen et al. [4] reported that algal available N ranged from 4 to 66% of the total nitrogen in 13 urban runoff samples in Madison, Wisconsin and the total nitrogen averaged 0.75 to 3.53 mg/ℓ. They also showed that the maximum percent available nitrogen should not exceed 69%. Bioassay studies are generally conducted to evaluate the effects of highway storm–water runoff on aquatic ecosystems [13, 38]. Portele et al. [38] examined the response to runoff of a green algal, Selenastrum Capricornutum, a zooplankton, Daphnia Magna, and a native salmonid, Salmo Gairdneri (rainbow trout). These organisms presented three trophic levels. Gjessing et al. [13] examined the effect of snowmelt on two algal species, Selenastrum gapricornutum and Synedra Acus. The data generally indicate adverse effects on the growth rate and higher mortality when high fractions (50–100%) of highway runoff is used. However, lower concentration of highway runoff or high dilutions may result in stimulatory effects and increased biomass.

The extent of accumulation and exchange reactions of nutrients across the sediment/water interface will highly affect the eutrophication of a receiving water system. Hvitved–Jacobsen et al. [23] investigated the nutrient mass balance in a shallow pond receiving highway runoff at the Maitland interchange (I–4), Orlando, Florida. The experiments demonstrated the importance of the sediment as a sink for both phosphorus and nitrogen, Figure 5.7. Although the actual specific loadings from highway runoff were as high as 5 gP/m^2·yr and 31 gN/m^2·yr the efficient nutrient removal to the sediments resulted in low nutrient concentrations in the water phase, Table 5.13.

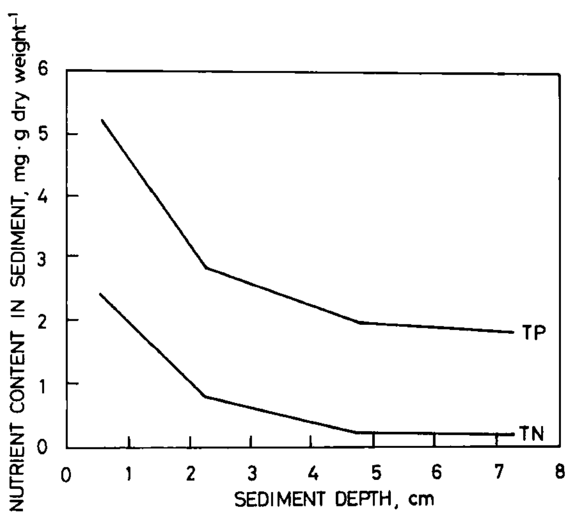


Fig. 5.7 **Average nutrient content versus sediment depth from the West Pond, Mairland Interchange. Results are based on 43 core samples. After Hvitved–Jacobsen et al. [23].**

Eutrophication is a very complex ecological phenomenon involving a large number of microbial and chemical processes in the water and the sediment phase of a receiving water system. It is by no means the purpose of this Chapter to discuss these matters in detail. However, it is important to realize that phosphorus is normally the limiting nutrient for algal growth in lakes and therefore of major interest compared to nitrogen. Furthermore, referring to Table 5.12, an average TN:TP–ratio in highway runoff is 4.1:0.79 = 5.2. This ratio is low compared with the ratio from rural runoff, showing the relative importance of phosphorus from highway runoff on the eutrophication of a lake.

TABLE 5.13

Water quality parameters from West Pond of Maitland Interchange. Data summarized for the period from November 1982 to May 1983. After Hvitved–Jacobsen et al. [23].

			Phosphorus ($\mu g/\ell$)			Nitrogen ($\mu g/\ell$)				
	pH	Alkalinity (mg/ℓ)	Diss. OP	Tot OP	TP	Org.N	NH_3–N	NO_2–N	NO_3–N	TN
Average	7.2	50	11	14	33	556	14	0.3	33	597
Standard deviation	0.4	9	11	11	17	228	14	0.2	28	234
Number of samples	41	40	37	37	37	40	42	42	41	38

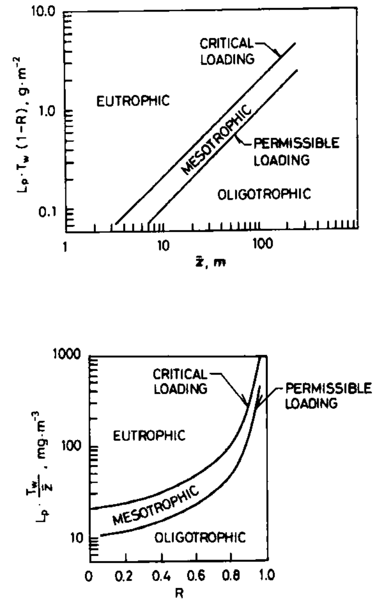
If it is possible to distinguish between loadings from the different sources the relative impact of highway runoff on lake eutrophication can be assessed. Several methodologies exist. The most simple are based on an estimation of the trophic state based on the areal nutrient (phosphorus) loading and lake mean depth [56, 57]. Further development of these types of models also include simple hydraulic properties and empirical estimation of phosphorus retention in the sediments [6, 29]. These models are graphically shown in Figure 5.8.

The phosphorus retention coefficient, R, defined as the fraction of influent phosphorus retained in the lake, can be measured. Under steady state conditions different empirical statistical equations for calculating R are available [26, 29].

Because of the simple relationship between loading and the resulting water quality the phosphorus loading models are valuable tools in a screening process. If the phosphorus loading on a lake can be calculated both with and without highway runoff, the relative impact of highway runoff on eutrophication can be estimated by a simple comparison.

5.3 Metal impacts

Metals such as Pb, Zn, Fe, Cu, Cr, Cd, and Ni in highway runoff are of most concern to environmental agencies and the general public if levels exceed those acceptable for water supplies and stream standards. Also, metals may accumulate in the sediments, phytoplankton, benthic organisms and fish. Toxicity of metals may reduce diversity and abundance of the sensitive aquatic biota and replace them with pollution tolerant species. Metal impacts from highway runoff is a function of the form in which the metal exists. Depending on the physical/chemical properties of receiving water and runoff water, metals can exist as inorganic or organic complexes, chelated, adsorbed onto particulates, or in free ionic forms; the latter of which is generally the most toxic [66].


Dillon

Larsen and Mercier

Fig.5.8 **Dillon's and Larsen and Mercier's phosphorus loading models, where:**
L_p = areal phosphorus loading, g/m^2·yr.
T_w = hydraulic retention time, yr.
R = phosphorus retention coefficient.
$\bar{z}$ = lake mean depth, m.

Iron and lead are formed in mostly insoluble form (5 to 12 percent dissolved), while other metals are much more soluble. The speciation of metals shows that most of dissolved metals to be of labile (reactive) inorganic forms with variable distribution between colloidal and non–colloidal forms.

The effects of highway runoff metals on Lake Padderudvann near Oslo were evaluated by Gjessing et al. [13]. Metal concentrations in the tributary stream draining a paved area were considerably higher than in two other inflowing streams. The lake appeared to retain and/or dilute these metals since the concentrations in the outflow stream were comparable to the inflowing clean streams. A study of highway runoff entering Lake Ivanhoe, Orlando, Florida, through a series of scupper drains running along the entire length of I–4 bridge crossing over the lake has been completed by Yousef et al. [65]. Sediment samples were collected from the lake bottom located directly beneath scupper drains for analysis and compared with sediment samples beneath a bridge section without scupper drains. It is concluded that concentrations in sediments associated with scupper drains are at least twice as high as those under a bridge without scupper drains (Table 5.10). Similarly, phytoplankton (Spirogyra and Hydrilla) exposed to direct bridge runoff contained about twice as much Zn, Pb, Ni, Cd, Cr, and Fe concentrations compared to concentrations detected in samples beneath bridges without scuppers. Also, the crustaceans and the Tubifex worms exhibited high metal concentrations. Therefore, it is concluded that the Tubifex worms could be used as indicator organisms for the detection of heavy metals (Pb, Zn, Cu, Ni, and Cr) enrichment from pollutional sources [2].

TABLE 5.14

Significance of differences in heavy metal concentrations of bottom sediments from Lake Ivanhoe, Orlando, Florida.

Pollutant	of observations*	Mean value (μg/g dry weight)		Percent probability
		with scuppers	without scuppers	
Zn	8/7	96.9	42.0	99.6
Pb	8/7	423	132.0	99.9
Cr	8/7	239	11.0	97.1
Ni	8/7	7.2	2.8	99.6
Cu	8/7	80.1	29.2	98.7
Fe	8/7	1,689	643	99.9
Cd	8/7	0.5	0.3	91.4

* *Eight samples were collected from sediments beneath bridges with scuppers and seven samples from beneath bridges without scuppers.*

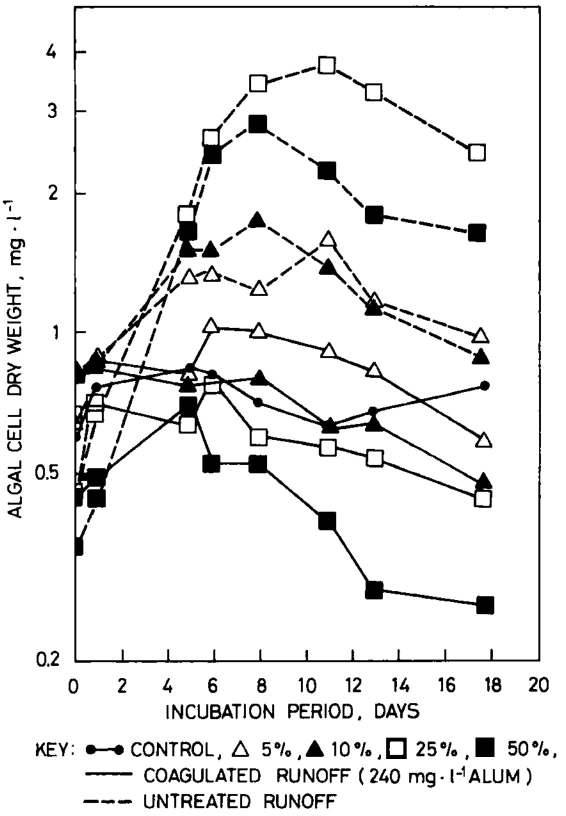


Fig. 5.9 Responses of indigenous algal species in Lake Eola to various concentrations of stormwater runoff and coagulated runoff. [17].

Winters and Gidley [61] found severe inhibition of mixed algal populations when exposed to low percentages of runoff (1, 5, and 19 percent) from a high traffic urban freeway (185,000 ADT) in California after 2 weeks of antecedent dry period. However, runoff from lower ADT rural highways, or from urban freeways with short antecedent dry days, was generally stimulatory to the algal populations. Also the amphipod, Gammarus, did exhibit acute toxicity to undiluted runoff from an urban highway (120,000 ADT) and a rural highway (7,400 ADT) in Milwaukee, Wisconsin [44]. It is interesting to notice that runoff water coagulated with alum in order to remove bioavailable phosphorus, metals and particulate matters has stimulatory effects as shown in Figure 5.9.

An investigation on the removal from the water phase of heavy metals originating from highway runoff and subsequent accumulation in the sediments of a receiving water system, a highway runoff retention pond at the Maitland interchange (I–4), Orlando, Florida, was reported by Yousef et al. [65]. The portions in the sediments of Cu, Pb, Cd, Zn, Ni, Cr and Fe varied from 95.1 for Cu to 99.9% for Fe compared to the total mass in the pond system. Also, results presented in Table 5.15 demonstrate that the heavy metals are efficiently removed from the water phase to the sediments.

TABLE 5.15

Average heavy metal concentrations in highway runoff and in the pond at Maitland interchange and I–4. After Yousef et al. [65].

			Average concentration*						
Sample	No. of obs.	Form	Zn	Pb	Cr	Ni	Cu	Fe	Cd
Runoff water	15	Diss.	50	39	3	3	22	47	1
		total	292	617	10	6	48	988	2
Pond water	34	Diss.	5.8	16	2.3	1.8	14	20	<1
		total	6.4	22	3.4	2.3	16	61	1
Pond sediment	43	Extract ($6N\ HNO_2$)	0.84	1.77	1.39	0.40	0.44	104	0.09

**Concentration in water is mg/m^3 and in sediment is g/m^3.*

5.4 De–icing salt impacts

In areas and countries with a cold climate the use of de–icing salts on highways may give rise to high salt concentrations in the runoff water. Especially NaCl has been widely used as a de–icing agent.

In Denmark NaCl levels of 1,000–1,500 g/m^3 and peak values of about 4,000 g/m^3 were measured during periods of snow melting in a detention pond receiving highway runoff [63].

The environmental mobility and bioavailability of heavy metals may be influenced by high concentrations of chloride. Investigations related to metal speciation analyses i.e. studies on the size spectrum of heavy metal containing particles and the chemical form in which the metals occur have contributed to obtained results [16, 18, 42]. Sequential extraction of heavy metals following the procedure of Tessier et al. [51] was a key element in these analyses. However, the results are ambiguous: high concentrations of chloride may increase the solubility of some heavy metals. e.g. Cd, but parameters like redox potential, pH and suspended solids concentrations seem to be more important.

Furthermore, great variations during a year in the chloride concentration of a receiving water system may influence the ecosystem considerably. However, a clear documentation of the specific impacts on the biota of chloride seems to be missing.

6. MANAGEMENT AND CONTROL

In recognition of the potentially toxic and degrading properties of highway runoff waters, a number of management practices have been proposed to either prevent the discharge of these waters into receiving bodies or to treat them prior to release. Some of the management practices in current use include:

(1) vegetative controls such as drainage canals, swales, and overland flow;
(2) wet detention/retention ponds with a permanent pool of water;
(3) infiltration systems such as shallow infiltration pits, tranches and/or basins;
(4) wetland treatment utilizing ecosystems to assimilate large quantities of highway runoff contaminants;
(5) other management and control practices which include litter controls, street–cleaning, catch basins, dry ponds, and cloth or gravel filters are considered less effective or ineffective.

Highway contaminants are removed in these systems by physical, chemical, and biological processes. Particulates may be removed by sedimentation, retention and/or filtration. Dissolved fractions are removed by adsorption, chemical interaction and precipitation and biological uptake. Many researchers report relatively higher concentrations of lead, chromium and zinc in the soils adjacent to highways than the background concentrations of these metals. In light of this ability of soils to retain or immobilize heavy metals, it is likely that management practices will promote maximum exposure of highway runoff to soils through overland flow, infiltration systems, wet ponds and others. Effectiveness of selected management practices will follow.

6.1 Swales

Roadside swales are commonly used along highways to convey rainfall excess and to infiltrate runoff water flowing to selected locations for retention, detention, storage, or discharge into adjacent receiving waters. Contaminants in runoff, such as heavy metals and nutrients, may be reduced and retained on sites. However, regeneration and relocation of loosely–bound contaminants may occur at intermediate location. It is reasonable to assume that particulate contaminants are filtered out by the grassy cover on swales and settle down to the bottom sediments. These contaminants are exposed to various dry and wet periods and slowly decay which may result in release and relocation. Dissolved contaminants will interact with surrounding water, soil and biota.

A comprehensive study on effectiveness of swales has been completed by Yousef et al. [66]. The minimum observed infiltration rates were found to be 0.013–0.036 $m^3/m^2 \cdot hr$. These rates are three to four times lower than measured values. Also calculated Manning's friction coefficient "n" for flow calculations through swales were found to vary between 0.035 and 0.059 with an average of 0.053 for most of cases. The swale areas were covered with 5–10 cm high grasses, predominantly bahia. Also swales built on dry soils with good drainage and high infiltration rates show better mass removal efficiencies for highway contaminants than swales built on soil close to the water table which are constantly wet.

Concentrations of pollutants in highway runoff flowing over roadside swales may decline, increase or remain constant depending on contaminant species. Charged species are retained by sorption processes and the removal efficiencies are governed by the predominant ionic species and complexes. Dissolved species of Zn, Cd, Ni, Pb, and Cr are more effectively removed than Cu and Fe. Generally, metals are more effectively removed then nitrogen and phosphorus. Also, swales filter out heavy metals with large particulate fractions showing high removal efficiencies. Average mass removal rates of dissolved metals and nutrients flowing over experimental swales are presented in Table 5.16 [69]. It is important to note that these rates are highly site and condition specific and are influenced by the total mass input, velocity of flow and percent infiltration. Slower moving runoff water in a well maintained swale with high infiltration rates will result in higher mass removal rates.

6.2 Detention/retention ponds

The construction of retention/detention basins or ponds to reduce stormwater pollutant loadings into receiving lakes and streams, is the most common practice in USA, Europe and other parts of the world. Retention basins with a fairly long retention time (weeks and months), concentrate most of the metals and phosphorus in the bottom sediments and the nitrogen is reduced by nitrification/denitrification process (68). Detention basins are used for temporary storage (hours and days) are classified as dry ponds or wet ponds. Wet ponds maintain a permanent pool and offer greater performance capabilities than dry basins which detain portions of flow from storms for a limited period of time.

TABLE 5.16

Average mass removal rates of dissolved pollutants in runoff water flowing over experimental swales.

Parameter	Mass removal rates (mg/m^2·hr)			
	Maitland site		Epcot site	
	Range	Average	Range	Average
Pb	0.2–2.8	1.14	1.3–3.9	2.61
Zn	0.7–3.1	1.85	0.8–10.7	5.76
Cu	0.1–0.8	0.42	0.5–0.6	0.60
Fe	4.3–30.0	15.23	1.5–9.2	5.33
Ni	–	–	0.9–4.4	2.63
Cd	–	–	0.2–0.4	0.26
Cr	0.3–3.4	1.85	0.3–1.5	0.90
OP–P	5.1–56.0	23.80	1.4–25.8	13.60
TP–P	6.9–61	17.10	1.4–26.4	13.96
Inorganic N	32–104	62.50	4.6–12.9	8.75
Organic N	9–51	22.80	33.9–38.7	36.30
TN	41–122	85.30	42.8–46.7	44.80

When wet detention ponds are adequately sized, removals from the water phase in excess of 90 percent for total suspended solids and lead can be obtained while pollutants with relatively high soluble fractions show lower reductions; approximately 65% for total phosphorus, 50% for BOD, COD, TKN, Cu, and Zn [55]. Figure 5.10 depicts the relationship between the fraction of mass removed from total SS and total P and the ratio between detention basin volume (VB) to runoff volume from a mean storm (VR). It is interesting to notice that a detention pond in Viborg, Denmark, followed the same general removal efficiencies as sites in the USA, Figure 5–10. Also maximum removal efficiencies appeared to coincide with VB/VR ratio between 6 and 8. Of course, an approximate pond sizing rule should consider the trade–off between the degree of removal efficiency and the cost of achieving the desired efficiency.

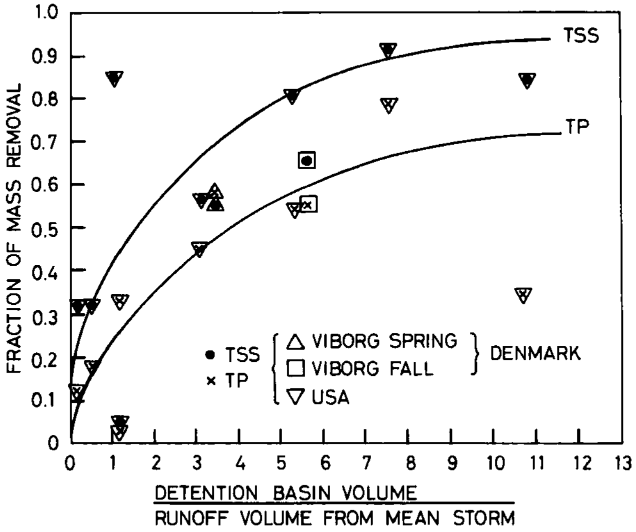


Fig. 5.10 **Removal of suspended solids and total phosphorus in detention ponds[25].**

Little is known about the design and operation guidelines and criteria needed to maximize their cost effectiveness for pollutant removal. However, the pond surface area relative to the area of the drainage basin or the pond volume relative to the rainfall pattern of the drainage area appear to be a main design parameter for pollutant removal. The underlying reason for this is probably related to the detention time for stormwater, determining the extent to which chemical, biological and physical processes, important for pollutant removal, may take place. A criterion which takes into account the inter–event dry period between successive storms as a measure for the stormwater detention time and relates exceedence statistics for a series of historical storm events with rainfall depth of a single event may be a sound practical approach [25].

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Chapter 6

QUANTITATIVE DETERMINATION OF HIGHWAY EMISSIONS IN THE AIR USING RECEPTOR MODELS

P.K. Hopke

Contents **Page**

1. INTRODUCTION

The advent of a U.S. national ambient air quality standard for total suspended particles (TSP) in the early 1970s created the need to identify particle sources so that effective control strategies could be designed and implemented. The initial efforts at identification of particle sources focused on dispersion models of point sources and in most cases, resulted in substantial reductions in TSP levels. However, as the increment of additional control needed to reach standard levels became smaller, the model uncertainties lead to difficulties in identifying the actual sources of continuing problems. In addition, fugitive and other non–ducted emissions are generally not treated, or are poorly handled, in these models. Thus, additional methods were required to identify and quantitatively apportion particle mass to sources.

These new methods are called receptor models. In them, the measured properties of the collected ambient samples are used to infer the contributions of the sources to the ambient pollutant concentration. These methods therefore require that samples be obtained at locations of interest, receptor sites and that the samples so collected be analyzed for the properties that are characteristic of the pollutant sources.

These requirements have arisen at a time when new analytical methods have been developed that permit multielemental analysis of large numbers of airborne particle samples or microscopic characterization of large numbers of individual particles (see Chapter 2). Thus, large data bases on the composition of airborne particles are available for use in these receptor models. Although much of the thrust of the model developments have been aimed at identification of sources of particle mass, they also can be used to elucidate the origins of the various measured species observed in the samples. It then becomes possible to quantitatively apportion the observed airborne concentrations such as airborne lead among the various source types including highway emissions.

The importance of receptor models as air quality management tools in the U.S. has recently been increased substantially by the promulgation of a new ambient air quality standard for particulate matter. This new standard requires all of the state and local air quality planning agencies to revise their plans for improving air quality and reducing the particulate level concentrations where they are expected to exceed the prescribed levels. In the associated guidance documents provided by the U.S. Environmental Protection Agency [1], receptor models are explicitly approved for use in this planning process along with the traditional dispersion models. Thus, receptor models have now become an accepted tool for air quality management.

This chapter will outline several of the applicable models, provide examples of their use in apportioning materials in a number of airsheds, and demonstrate how they can identify the influence of highway emissions on the overall airborne concentrations.

2. APPROACHES BASED ON BULK SAMPLE ANALYSIS

2.1. Principle of Mass Conservation

All of the currently used receptor models are based on the concept of conservation of mass and the use of a mass balance analysis. For example, let us assume that the total airborne particulate lead concentration (ng/m^3) measured at a site can be considered to be the sum of contributions from independent source types such as motor vehicles, incinerators, smelters, etc.

$$Pb_T = Pb_{auto} + Pb_{incin.} + Pb_{smelter} + \ldots \qquad (1)$$

However, a motor vehicle burning leaded gasoline emits particles containing materials other than lead. Therefore, the atmospheric concentration of lead from automobiles in ng/m^3, Pb_{auto}, can be considered to be the product of two cofactors; the gravimetric concentration (ng/mg) of lead in automotive particulate emissions, $a_{Pb,auto}$, and the mass concentration (mg/m^3) of automotive particles in the atmosphere, f_{auto}.

$$Pb_{auto} = a_{Pb,auto} f_{auto} \qquad (2)$$

The normal approach to obtaining a data set for receptor modelling is to determine a large number of chemical constituents such as elemental concentrations in a number of samples. The mass balance equation can thus be extended to account for all m elements in the n samples as contributions from p independent sources

$$x_{ij} = \sum_{k=1}^{p} a_{ik} f_{kj} \qquad \begin{matrix} i=1,m \\ j=1,n \end{matrix} \qquad (3)$$

where x_{ij} is the ith elemental concentration measured in the jth sample, a_{ik} is the gravimetric concentration of the ith element in material from the kth source, and f_{kj} is the airborne mass concentration of material from the kth source contributing to the jth sample.

There are several different approaches to receptor model analysis that have been successfully applied including chemical mass balance (CMB), multivariate receptor models including principal components analysis and target transformation factor analysis (TTFA),and computer–controlled scanning electron microscopy (CCSEM) combined with a particle class balance analysis (PCB). These models can be applied to both particulate and gaseous species. The basis for each of these methods will be presented in subsequent sections of this chapter along with examples of their application to the identification of highway pollution in the atmosphere.

2.2 Chemical Mass Balance

The chemical mass balance (CMB) sometimes called the chemical element balance solves equation 3 directly for each sample by assuming that the number of sources and their compositions at the receptor site are known. This approach was first independently suggested by Winchester and Nifong [2] and by Miller, Friedlander, and Hidy [3]. The composition of an ambient sample is then used in a multiple linear regression against source compositions to derive the mass contribution of each source to that particular sample. Miller, Friedlander, and Hidy modified equation 3 to explicitly include changes in composition of the source material while in transit to the receptor

$$x_{ij} = \sum_{k=1}^{p} \alpha_{ik} a'_{ik} f_{kj} \tag{4}$$

where α_{ik} is the coefficient of fractionation so that if a'_{ik} were the composition of the particles as emitted by the source, a_{ik} is the composition of the particles at the receptor site ($a_{ik} = \alpha_{ik} a'_{ik}$). In practice, it is generally impossible to determine the α_{ik} values and they are assumed to be unity ($a_{ik} = a'_{ik}$).

Early applications of this approach to urban aerosol mass apportionment included Pasadena, CA [4], Heidelberg, Germany [5], Ghent, Belgium [6], and Chicago, IL [7]. In all of these analyses, the quality of available source compositions severely limited the precision to which the ambient compositions could be reproduced.

Several major research efforts have subsequently resulted in substantially better source data. Studies at the University of Maryland have provided data on coal–fired power plants [8,9], oil–fired power plants [10], incinerators [11–13], motor vehicle traffic [14,15], soil [16], and assorted industrial sources [17]. These studies led to much improved resolution of the particle sources in Washington, D.C. [18,19]. In the first report of these studies, Kowalczyk, Choquette, and Gordon [18] used a weighted least–squares regression analysis to fit six sources with eight elements for ten ambient samples. In these analyses, the ambient elemental concentrations are weighted by the inverse of the square of the analytical uncertainty in that measurement.

Subsequently, Kowalczyk, Gordon, and Rheingrover [19] examined 130 samples using 7 sources with 28 elements included in the fit. The average results for this extended data set are presented in Table 1 taken from Kowalczyk, Gordon, and Rheingrover [19]. Although 28 elements were used in the fitting process, the fit did not change appreciably with varying numbers of elements included with the exception of some of the key tracer elements such as Na, Pb, and V. The values of larger/smaller ratio of the observed and predicted values is an indication of the amount of fluctuation in the fit. This elemental balance sheet allows the identification of the major sources of metals in the air. For example, vanadium and nickel primarily arise from oil–fired power plant emissions; 23 of 25 ng/m^3 for V and 4.0 of 17 ng/m^3 for Ni with most of the nickel unexplained. Zinc is mainly released by incinerator sources but also comes from motor vehicles (51 ng/m^3 from refuse incinerations and 7.3 ng/m^3 from motor vehicles). The reverse is true for lead with motor vehicles as the primary source and refuse incineration as a lesser but important source; 428 ng/m^3 from motor vehicles and 34 ng/m^3 from incineration. In this way sources of both particulate mass and specific elements can be identified.

Mayrsohn and Crabtree [20] presented the use of an iterative least– squares approach to solving six sources of airborne hydrocarbon compounds. The sources were automotive exhaust, volatilization of gasoline and release of gasoline vapor, commercial natural gas, geological natural gas, and liquified petroleum gas. They performed the least–squares fit to the hydrocarbon compound concentrations using gas chromatography to determine the concentrations of eight compounds. Their ordinary least–squares source reconciliation algorithm recognized that not all sources may contribute to every sample, and, if negative contributions were obtained, a different configuration of sources was employed with certain qualifying assumptions [21]. Each possible configuration with positive coefficients was considered and the one with the lowest standard error was chosen as the optimum solution. On the average, automotive exhaust was the source of almost 50% of observed hydrocarbons. Gasoline and its vapour contributed 30% by weight and the balance resulted from commercial and geological natural gas. Thus, automobiles and other highway related sources were responsible for the majority of these hydrocarbons.

Table 6.1 Average Results of Chemical Element Balances of 130 Samples from Washington, D.C. Area for Summer 1976.

element	predicted contributions[a], ng/m^3 soil	limestone	coal	oil	refuse	motor vehicle	marine	predicted	observed[d]	larger/ smaller[b]	missing values[c]
Na[e]	43	0.83	8.3	12	35	---	201	300	300 ± 20	1.00[f]	0
Mg[e]	74	101	27	3.8	5.4	32	26	270	440 ± 30	1.63[f]	3
Al[e]	812	9	517	0.4	6.1	—	<0.01	1340	1350 ± 110	1.16[f]	0
K[e]	154	6	67	0.4	47	13	7	295	400 ± 20	1.38[f]	0
Ca[e]	66	635	47	8.2	7.1	47	7.6	820	860 ± 40	1.09	0
Sc[e]	0.15	0.002	0.18	0.0002	0.0006	—	<0.0001	0.33	0.33 ± 0.03	1.19[f]	0
Ti[e]	52	0.83	31	0.03	1.0	—	<0.0001	85	110 ± 10	1.35[f]	0
V[e]	1.06	0.042	1.6	23	0.013	—	<0.0001	26	25 ± 2	1.07	0
Cr	0.81	0.023	0.84	0.062	0.21	—	<0.0001	2.0	14 ± 2	7.07[f]	5
Mn[e,g]	13	2.3	1.6	0.10	0.31	1.3	<0.0001	18	17 ± 2[g]	1.26[f]	0
Fe[e]	511	8.3	362	2.8	2.8	34	0.0002	920	1000 ± 60	1.15[f]	0
Co[e]	0.22	0.0002	0.25	0.16	0.003	0.05	<0.0001	0.68	0.83 ± 0.08	1.19[f]	0
Ni	0.46	0.042	1.04	4.0	0.07	0.34	<0.0001	6.0	17 ± 2	4.35[f]	1
Cu	0.23	0.009	2.2	0.89	0.71	3.0	0.0001	7.1	17 ± 2	2.68[f]	23
Zn[e]	1.14	0.04	2.6	1.6	51	7.3	<0.0001	64	85 ± 6	1.42[f]	0
Ga[e]	0.38	0.008	0.46	0.0001	—	—	<0.0001	0.85	1.29 ± 0.17	1.89[f]	42
As[e]	0.061	0.002	3.1	0.028	0.10	—	0.0001	3.32	3.25 ± 0.2	1.58[f]	0
Se	0.0009	0.0002	0.78	0.035	0.016	0.035	0.0001	0.87	2.5 ± 0.2	4.10	0
Br	0.097	0.013	2.1	0.054	0.66	167	1.25	171	139 ± 9	1.84	0
Rb[e]	1.19	0.0064	0.60	0.0001	—	—	0.0023	1.8	2.1 ± 0.2	1.54[f]	17
Sr[e]	3.65	1.29	3.5	0.09	0.027	—	<0.0001	8.6	10 ± 1	1.40[f]	32
Ag[e]	0.0007	0.0001	—	0.006	0.23	—	<0.0001	0.24	0.20 ± 0.01	1.44[f]	26
Cd[e]	0.0011	0.0001	0.13	0.0028	0.64	1.03	<0.0001	1.80	2.4 ± 0.02	1.89[f]	0
In	0.0007	0.0001	0.0023	<0.0001	0.0024	—	0.0004	0.0059	0.020 ± 0.001	5.12[f]	3
Sb[e]	0.0081	0.0004	0.13	0.007	0.89	0.60	<0.0001	1.6	2.1 ± 0.2	1.39[f]	0
I	0.058	0.0026	2.1	—	—	—	1.14	3.3	2.0 ± 0.1	2.41	12
Cs[e]	0.028	0.0011	0.039	0.0005	0.0025	—	<0.0001	0.07	0.17 ± 0.05	3.36[f]	2
Ba	7.1	0.02	4.7	2.0	0.30	6.4	0.0006	21	19 ± 2	1.42[f]	1
La[e]	0.75	0.014	0.31	0.016	0.0016	—	<0.0001	1.1	1.5 ± 0.1	1.42[f]	0
Ce[e]	0.98	0.024	0.62	0.014	0.007	—	<0.0001	1.6	2.0 ± 0.2	1.30[f]	1
Sm[e]	0.068	0.002	0.057	0.0012	0.0003	—	<0.0001	0.13	0.20 ± 0.02	1.59[f]	0
Eu[e,h]	0.015	0.0004	0.014	0.0005	0.0002	—	<0.0001	0.030	0.030 ± 0.003	1.32[f]	5
Yb	0.037	0.0011	0.030	0.0004	0.0009	—	<0.0001	0.070	0.034 ± 0.003	2.60[f]	19
Lu	0.0067	0.0004	0.0080	0.0001	0.0004	—	<0.0001	0.016	0.0056 ± 0.0006	3.34[f]	1
Hf[e]	0.031	0.0006	0.022	0.0008	0.0004	—	<0.0001	0.055	0.10 ± 0.01	1.80[f]	1
Ta[e,h]	0.041	0.0008	0.0077	0.0011	0.0018	—	<0.0001	0.052	0.036 ± 0.004	1.84[f]	39
W	0.014	0.0013	0.038	0.0004	0.0072	—	<0.0001	0.061	0.24 ± 0.02	4.76	23
Pb[e]	0.15	0.019	2.1	0.39	34	428	<0.0001	465	440 ± 20	1.28[f]	0
Th[e]	0.11	0.0036	0.10	0.0016	0.0008	—	<0.0001	0.22	0.25 ± 0.02	1.19[f]	1

a. Contributions designated by "-" indicate that concentration of the element in particles from the source is not known.

b. larger/smaller is the avearge of the ratio of predicted/observed or vice versa, whichever is larger over all samples.

c. Number of samples for which no value is available because peak is not strong enough in spectra for determination or concentration is not above filter blank.

d. Uncertainty is standard deviation of mean value.

e. Element fitted by least-squares procedure.

f. larger/smaller value included in average given below.

g. For reasons described in text, all observed values reduced by factor of 0.69 prior to fitting. Actual average observed values was 26 + 3 ng/m.

h. As noted in the text, three observed values were eliminated from fits.

A similar study utilizing this mass balance approach for resolving hydrocarbon sources has been made. Nelson, Quigley, and Smith [22] have examined the atmospheric hydrocarbons in Sydney, Australia. They have used much more extensive hydrocarbon profiles for their sources and have obtained good agreement between the mass balance approach and a resolution based on an emissions inventory. They also find that the major hydrocarbon sources are direct automobile exhaust (36 ± 4%) and evaporative emissions of gasoline (32 ± 4%). Thus, it is possible to identify the impact of highway emissions on gaseous as well as particulate pollutants.

In 1979, Watson [23] and Dunker [24] independently suggested a mathematical formalism called effective variance weighting that includes the uncertainty in the measurement of the source composition profiles as well as the uncertainties in the ambient concentrations. As part of this analysis, a method has also been developed to permit the calculation of the uncertainties in the mass contributions.

The most extensive use of effective–variance fitting has been made by Watson and colleagues in their work on data from Portland, OR [23,25]. Since that study, a number of other applications of this approach have been made including Medford, OR [26], Philadelphia, PA [27,28], and at a number of locations in the U.S. Environmental Protection Agency's Inhalable Particulate Network [29].

One of the important uses of the mass balance results is that it can be used to calibrate the dispersion models being used for making air quality management decisions [30,31]. For example, the Portland airshed was modelled using GRID [32], a conservation of mass, advection–diffusion code that has been designed to model the complex terrain of the Portland area. The area is divided into 2 by 2 km grid cells. For each of the 5000 cells, it is necessary to know the topography, the wind field flows for each of the eight meteorological regimes, and the spatially and seasonally resolved point and area source emissions as well as the stack parameters for each point source.

The results of the dispersion model can be separated into the same source groups as the mass balance results so that direct comparisons can be made. The results of the GRID and chemical mass balance models given by Core *et al*. [31] are compared for road dust in Figure 6.1. Road dust was substantially underpredicted by the dispersion model compared to the CMB results. For the auto exhaust, three sites agree within the uncertainty in the mass balance results. However, for one location, there was an underprediction by the GRID model. Core *et al*. [31] indicated that these discrepancies can be readily understood.

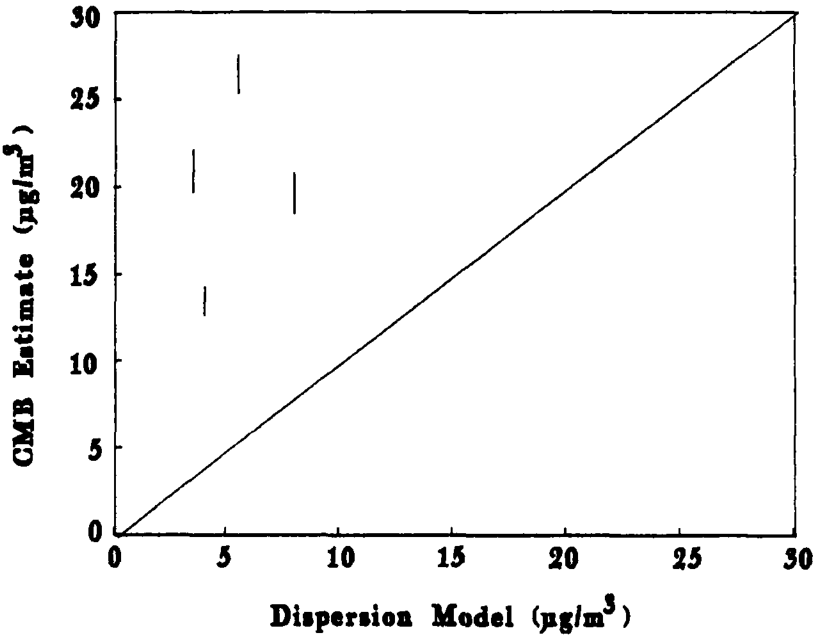

FIGURE 6.1 *Comparison of CMB Receptor Model to initial GRID Dispersion Model for Road Dust and 1:1 agreement line. Drawn from values given in Reference [31]*

For the auto exhaust, a heavily travelled road was incorrectly assigned to an adjacent grid. When properly placed, the dispersion model value increased by 10% and provided good agreement for the auto exhaust.

However, the road dust was underpredicted because of the low value of the EPA generalized paved road emission factor. There were only a few unpaved roads in the region so it was decided that the paved road emission factor should be increased. Since the automotive tailpipe emissions appeared to be correct because of the generally good agreement between the receptor and dispersion model results, the road dust emission factor was scaled by the ratio of the mass balance result for road dust to that for auto exhaust. Also, the road dust emission was suggested to be larger in heavily industrial areas of Seattle, WA [33] compared to commercial land use areas. This adjustment substantially increased the road dust contribution to the airborne particle loading. This adjustment brings the receptor and dispersion model results into good agreement for road dust as shown in Figure 6.2.

The test of these adjustments is then to compare dispersion model prediction of total suspended particulate matter (TSP) with the measured values. Figure 6.3 shows these comparisons before and after making the adjustments described above. Clearly, the use of the mass balance results to calibrate the dispersion model has made a noticeable improvement in the quality of the dispersion model predictions and hence its value as an air quality management tool. The U.S.E.P.A. is encouraging this kind of reconciliation between dispersion and receptor modelling results [1]. It provides a mechanism for calibration of dispersion models and for greater reliability in the use of the dispersion model for planning the pollutant control strategy.

It must be made clear, however, that the CMB analysis works well in these examples because both the source and ambient samples were collected and analyzed during the same time period. A much less detailed resolution of lead sources was all that was possible in Kellogg, Idaho [34] when on–site samples could not be obtained. In an intercomparison study organized by the U.S. Environmental Protection Agency [35] to examine receptor models, a set of ambient particulate elemental compositional data sets were analyzed by a number of investigators using similar CMB methods. The compositions of particles from sources in Houston were not available and were not measured during this program so that source composition profiles had to be obtained from literature sources. The lack of source data immediately raised problems in the use of the mass balance methods and comparison of results from different investigators [36]. It is not always certain exactly which sources

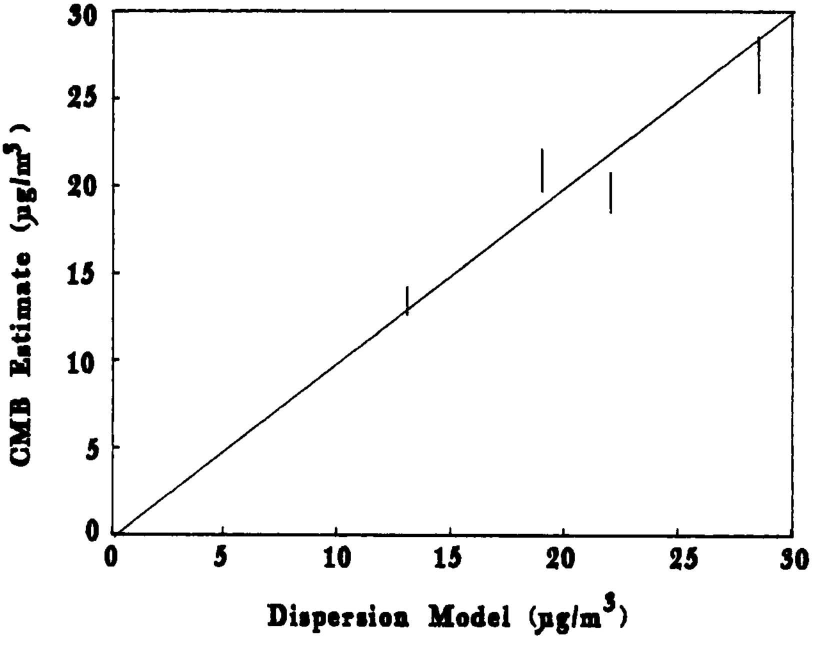

FIGURE 6.2 *Comparison of CMB Receptor Model to corrected GRID Dispersion Model for Road Dust and 1:1 agreement line. Drawn from values given in Reference [31].*

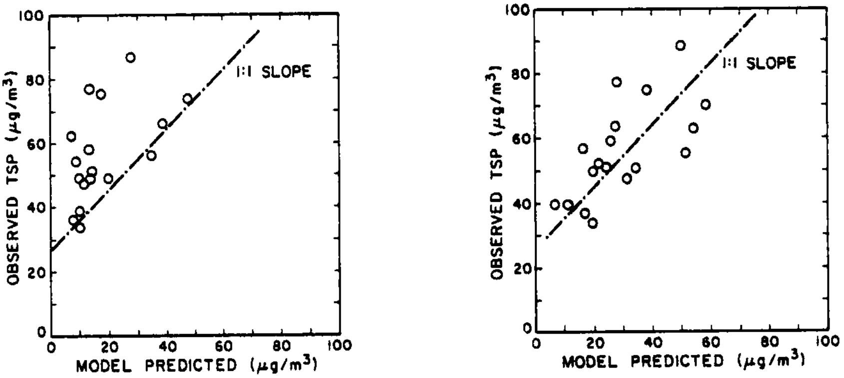

FIGURE 6.3 Comparison between measured TSP and GRID model predictions before (left) and after (right) model adjustments developed from receptor model results (figure redrawn from reference 31 and used with permission of the Journal of the Air Pollution Control Association).

should be included in the analysis. Although emission inventories may be available for the region, it may be that the measured source composition for a coal–fired power plant in Maryland burning eastern bituminous coal is not a particularly good representation for a lignite–burning plant in Texas.

An additional problem for receptor modelling is that the motor vehicle profile in the United States is undergoing rapid changes in lead and bromine concentrations with time as the mix of new, catalyst–equipped and diesel cars and leaded–fuel burning vehicles change. An interesting solution to the problem of the changing lead concentration in motor vehicle emissions was recently provided by Dzubay et al. [28]. They obtained particle samples in the summer of 1982 in Philadelphia, Pennsylvania and vicinity, in the size ranges of $<2.5\mu m$ and 2.5 to 10 μm using a dichotomous sampler. The samples were analyzed using ion chromatography for sulfate and nitrate, XRF and INAA for elemental composition and a thermo–optical method for organic and elemental carbon. Because there is also a non–ferrous metal smelter in the airshed, lead in the air comes from incinerators, the smelter, and tailpipe emissions. Using the other measured species in the data set, they derived the amounts of lead that could be attributed to all sources other than motor vehicles. They then used a second multiple regression analysis to relate the amount of unaccounted lead, total lead minus all sources other than vehicles, to the motor vehicle source and obtained a lead value of 6% lead in motor vehicle emissions. It appears that as long as sufficient leaded fuel is still in use, it will be possible to employ an approach such as this one to obtain the current fleet–weighted average. With leaded fuel being phased out entirely, the lead and bromine may cease to be useful tracers for motor vehicles in a few years. A similar trend exists in Europe.

There may be other tracers appearing for automobiles. As part of the Philadelphia study discussed above, Olmez and Gordon [37] identified unusually high values of the rare earth elements lanthanum, cerium, and samarium arising from the catalysis support material from an oil refinery. It is likely that similar materials arise from the catalytic converters in automobiles and could serve as new markers for tailpipe emissions.

The results from Mayrsohn and Crabtree [20] and Nelson et al. [22] suggest that a mass balance is applicable for the gaseous aliphatic hydrocarbons. These species along with CO could possibly provide good tracers for particulate emissions from highways. Such a result is less likely to be obtained for more reactive species like olefins. There will be problems for semivolatile species like polycyclic aromatic hydrocarbons (PAHs) because of the partitioning of the species between the gaseous and particulate phases. This problem has been recently reviewed by Pankow [38]. The sampling and analysis problems of reactive hydrocarbons and the modelling needed to account for their reactions in transit from source to receptor makes it very difficult to perform accurate receptor modelling and is an area of study that requires considerable additional effort.

Another interesting application of a mass balance analysis has been reported by Lewis et al. [39] where they apportion the observed mutagenicity of airborne particulate matter between wood smoke and motor vehicle emissions. Samples were obtained in a residential area of Albuquerque, New Mexico and analyzed for a similar suite of XRF species as in the Philadelphia study. In addition, parallel samples collected on quartz fibre filters were Soxhlet extracted for 24 h with dichloromethane. The filtered extracts were concentrated and transferred to dimethyl sulphoxide. The samples were assayed for mutagenicity using Salmonella typhimurium strain TA98 with a microsomal oxidase mixture to activate promutagens to mutagens.

They then used Pb as a tracer for motor vehicles and K as a tracer for wood smoke after subtracting a correction for soil K derived using the other measured crustal elements. They were able to relate all of the observed mutagenicity to these two sources. It was found that although highway emissions were three times more effective in inducing mutagenesis than wood smoke on a per mass of organic matter basis, the majority of the mutagenicity came from wood smoke. Their results certainly suggest the need for further study in areas of higher traffic volume as to the potential importance of highway emissions of genotoxic material.

2.3. Multivariate Receptor Models

Alternative approaches have been developed for identifying and quantitatively apportioning sources of airborne particles using multivariate statistical analysis. Eigenvector analysis has been the principal method that has been applied to airborne particle composition data. An eigenvector analysis tries to simplify the description of a system by determining the minimum number of new variables necessary to reproduce the measured attributes of the system. The mathematical basis of these methods has been described by Hopke [40].

Principal components and factor analysis are names given to several of the variety of forms of eigenvector analysis. It was originally developed and used in psychology to provide mathematical models of psychological theories of human ability and behaviour [41]. However, eigenvector analysis has found wide application throughout the physical and life sciences. Unfortunately, a great deal of confusion exists in the literature in regard to the terminology of eigenvector analysis. Various changes in the way the method is applied has resulted in it being called factor analysis, principal components analysis, principal components factor analysis, empirical orthogonal function analysis, Karhunen–Loeve transform, etc., depending on the way the data are scaled before analysis or how the resulting vectors are treated after the eigenvector analysis is completed. All of the methods have the same basic objective; the compression of data into fewer dimensions and the identification of the structure of interrelationships that exist between the variables measured or the cases studied.

The first step in the eigenvector analysis is the calculation of a dispersion matrix, the matrix that contains quantitative information on the relative variation of pairs of variables or pairs of samples (cases). There are two basic types of dispersion matrices. They are covariance matrices and correlation matrices. For a correlation matrix, the data are scaled such that each variable or each case has an equal weight. The data are not scaled before calculating covariance. In both instances, the data may be centered by subtracting a mean value before scaling and the calculation of the matrix elements. The choice of dispersion matrix depends on the nature of the data set to be analyzed. For many types of chemical spectroscopic data, the covariance matrix is the choice because each variable has the same measurement scale. For many geochemical problems, the difference in scale between major, minor, and trace components requires scaling to avoid domination of the analysis by the major components.

The dispersion matrix is then decomposed into a series of orthogonal vectors by the process outlined by Joreskog, Klovan, and Reyment [42] so that

$$U'DU = \Lambda \qquad (5)$$

where U is the matrix of eigenvectors, U' is its transpose, D is the dispersion matrix, and Λ is a diagonal matrix of eigenvalues where the trace of Λ is equal to the trace of D. If there were no errors in the data from which D is calculated, the number of non–zero eigenvalues would be the dimensionality of the problem called the rank of D. The rank for the original data matrix is the same as that for the dispersion matrix. However, experimental error generally results in a number of small but non–zero eigenvalues. The determination of the number of vectors containing significant information relative to those dominated by noise is often a difficult one. The lack of universally applicable criteria for determining the dimensionality of the data is a major problem in the application of factor analysis.

In the most commonly used approach to calculating the eigenvectors, the maximum amount of variance is packed into the first eigenvalue. The maximum possible amount of the remaining variance goes into the second and so forth. This compression of the information into a few components permits much of the variation in the data set to be displayed in a two or three dimensional plot. For many classification problems, the first few factors are able to reproduce most of the data structure and to remove some of the noise. The objects can then be plotted using the components axes and thus display the features of high dimensional data in a few dimensions [43].

The compression of variance into the first factors will improve the ease with which the number of factors can be determined. However, their nature has now been mixed by the calculational method. Thus, once the number of factors has been determined, it is often useful to rotate the axes in order to provide a more interpretable structure.

The axis rotation can retain the orthogonality of the eigenvectors or cause them to be oblique. Depending on the initial data treatment, the axes rotations may be in the scaled and/or centered space or in the original variable scale space. The latter approach has proved quite useful in a number of chemical applications described by Malinowski and Howery [44] and in environmental systems as described by Hopke [40].

The first receptor modeling applications of classical factor analysis were by Prinz and Stratmann [45] and Blifford and Meeker [46]. Prinz and Stratmann examined both the aromatic hydrocarbon content of the air in 12 West German cities and data from Colucci and Begeman [47] on the air quality of Detroit. In both cases they found three factor solutions and used an orthogonal varimax rotation to give more readily interpretable results. Blifford and Meeker used a principal component analysis with both varimax and a non–orthogonal rotation to examine particle composition data collected by the National Air Sampling Network (NASN) during 1957–61 in 30 U.S. cities. They were generally not able to extract much interpretable information from their data. Since there is a very wide variety of particle sources among these 30 cities and only 13 elements were measured, it is not surprising that they were not able to provide much specificity to their factors.

The factor analysis approach was then reintroduced by Hopke et al. [48] and Gaarenstroom, Perone, and Moyers [49] for their analysis of particle composition data from Boston, MA and Tucson, AZ, respectively. In the Boston data for 90 samples at a variety of sites, six common factors were identified that were interpreted as soil, sea salt, oil–fired power plants, motor vehicles, refuse incineration and an unknown manganese–selenium source. The six factors accounted for about 78% of the system variance. There was also a high unique factor for bromine that was interpreted to be fresh automobile exhaust. Since lead was not determined, these motor vehicle related factor loading assignments remain uncertain. Large unique factors for antimony and selenium were found. These factors represent emission of volatile species whose concentrations do not covary with other elements emitted by the same source. Subsequent studies by Thurston and Spengler [50] where other elements including sulphur and lead were measured showed a similar result. They found that the selenium was strongly correlated with sulphur for the warm season (May 6 to Nov. 5). This result is in agreement with the Whiteface Mountain results [51] and suggests that selenium is an indicator of long range transport of coal–fired power plant effluents to the northeastern U.S. They found lead to be strongly correlated with bromine and readily interpreted as motor vehicle emissions.

In the study of Tucson [49], whole filter data were analyzed separately at each site. They found factors that were identified as soil, automotive, several secondary aerosols such as $(NH_4)_2SO_4$ and several unknown factors. They also discovered a factor that represented the variation of elemental composition in their aliquots of their neutron activation standard containing Na, Ca, K, Fe, Zn, and Mg. This finding illustrates one of the important uses of factor analysis; screening the data for noisy variables or analytical artifacts.

One of the valuable uses of this type of analysis is in screening large data sets to identify errors [52]. With the use of atomic and nuclear methods to analyze environmental samples for a multitude of elements, very large data sets have been generated. Because of the ease in obtaining these results with computerized systems, the elemental data acquired are not always as thoroughly checked as they should be, leading to some, if not many, bad data points. It is advantageous to have an efficient and effective method to identify problems with a data set before it is used for further studies. Principal component factor analysis can provide useful insight into several possible problems that may exist in a data set including incorrect single values and some types of systematic errors. The quality control procedures described in Chapter 2 are also important for these reasons.

In other receptor modelling applications, Gatz [53] used a principal components analysis of aerosol composition data for St. Louis, MO taken as part of project METROMEX [54,55]. Nearly 400 filters collected at 12 sites were analyzed for up to 20 elements by ion–induced X–ray fluorescence. Gatz used additional parameters in his analysis including day of the week, mean wind speed, percent of time with the wind from NE, SE, SW, or NW quadrants or variable, ventilation rate, rain amount and duration. At several sites the inclusion of wind data permitted the extraction of additional factors that allowed identification of motor vehicle emissions in the presence of specific point sources of lead such as a secondary copper smelter. An important advantage of this form of factor analysis is the ability to include parameters such as wind speed and direction or particle size in the analysis.

In the early applications of factor analysis to particulate compositional data, it was generally easy to identify a fine particle mode lead/bromine factor that could be assigned as motor vehicle emissions. In many cases, a calcium factor sometimes associated with lead could be found in the coarse mode analysis and could be assigned as road dust. However, the problem of diminishing lead concentrations in gasoline discussed earlier for the CMB analysis also applies here. As the lead and related bromine concentrations diminish, the clearly distinguishable covariance of these two elements is disappearing. In a study of particle sources in southeast Chicago based on samples from 1985 and 1986, much lower lead levels are observed and the lead/bromine correlation is quite weak [56]. Thus, the identification of highway emissions through factor analysis based on lead or lead and bromine is becoming more and more difficult and other analytic species are going to be needed in the future.

A problem that exists with these forms of factor analysis is that they do not permit quantitative source apportionment of particle mass or of specific elemental concentrations. In an effort to find an alternative method that would provide information on source contributions when only the ambient particulate analytical results are available, Hopke and coworkers [57–61] have developed target transformation factor analysis (TTFA). In this analysis, resolution similar to that obtained from a CMB analysis can be obtained. However, a CMB analysis can be made on a single sample if the source data is known while TTFA requires a series of samples with varying impacts by the same sources, but does not require a priori knowledge of the source characteristics.

In matrix notation equation 3 can be rewritten as

$$X = AF$$

where X is the matrix of ambient aerosol compositions, A is the matrix of source composition profiles, and F is the matrix of mass contributions of the sources to the samples. The objectives of TTFA are

1) to determine p, the number of independent sources that contribute to the system,
2) to identify the components of matrix A, the elemental source profiles, and
3) to calculate F, the contribution of each source to each sample.

One of the first applications of TTFA was to the source identification of urban street dust [57]. The sample of street dust was vacuumed from an intersection in Urbana, Illinois. The sample was physically fractionated by particle size, density, and magnetic susceptibility to produce 30 subsamples. Each subsample was analyzed by instrumental neutron activation analysis and atomic absorption spectroscopy to yield analytical results for 35 elements.

The number of sources is determined by performing an eigenvalue analysis on the matrix of correlations between the samples. A target transformation determines the degree of overlap between an input source profile and one of the calculated factor axes. The input source profiles, called test vectors, are developed from existing knowledge of the emission profiles of various sources or by an iterative technique from simple test vectors [62]. The identified source profiles are then used in a simple weighted least–squares determination of the mass contributions of the sources [60].

In the analysis of the street dust, six sources were identified including soil, cement, tyre wear, direct automobile exhaust, salt and iron particles. The lead concentration of the motor vehicle source was found to be 15% with a lead to bromine ratio of 0.39. This ratio is in good agreement with the values obtained by Dzubay et al. [63] for Los Angeles freeways and in the range presented by Harrison and Sturges [64] in their extensive review of the literature. A comparison of the actual mass fractions with those calculated from the TTFA results are presented in Table 6.2. TTFA provided a good reproduction of the mass distribution and source apportionments of the street dust that suggest that a substantial fraction of the urban roadway dust is anthropogenic in origin.

One of the principal advantages of TTFA is that it can identify the source composition profiles as they exist at the receptor site. There can be changes in the composition of the particles in transit from the source to the receptor and approaches that provide these modified source profiles should improve the receptor model results. Chang et al. [61] have applied TTFA to an extensive set of data from St. Louis, MO, to develop source composition profiles based on a subset selection process developed by Rheingrover and Gordon [65].

Rheingrover and Gordon [65] identify samples strongly affected by single point sources using wind–trajectory analysis. They select samples from a data base such as the one obtained in the Regional Air Pollution Study (RAPS) of St. Louis, MO, that were heavily

Table 6.2. **Comparison of Actual and Calculated Mass Distributions for Urban Street Dust Sample**[a]

Sample No.	Actual Mass, g	Calcd/mass, g	% Error
1	8.040	8.3	–2.7
2	2.760	2.3	15.0
3	200.210	143.0	29.0
4	40.190	46.0	–14.7
5	5.070	5.4	–7.1
6	3.320	2.9	14.0
7	144.890	176.0	–21.0
8	41.730	20.0	51.0
9	1.690	1.8	–6.4
10	3.270	4.6	–39.0
11	61.420	85.0	–33.0
12	46.320	29.0	37.0
13	1.740	2.7	–57.0
14	0.960	1.3	–35.0
15	26.800	27.0	–2.3
16	14.010	8.6	39.0
17	0.340	0.53	–57.0
18	0.900	1.7	–92.0
19	15.300	32.0	–112.0
20	1.760	1.6	9.4
21	1.400	2.2	–61.0
22	7.400	8.4	–14.0
23	1.000	1.5	–51.0
24	5.000	6.7	–34.0
25	3.300	3.3	0.92
26	6.000	7.8	–30.0
27	1.100	0.55	48.0
28	1.900	2.2	–18.0
29	0.095	0.085	10.0
30	0.105	0.12	–19.0

[a] Taken from Hopke et al [57].

influenced by major sources of each element. These samples were identified according to the following criteria:

1. Concentration of the element in question $> X + Z_{cr}$ where X is the average concentration of that particular element for each station and size fraction (coarse or fine particle size fraction), Z_{cr} is typically set at about three for most elements, and is the standard deviation of the concentration of that element.

2. The standard deviation of the 6 or 12 hourly average wind directions for most samples, or minute averages for 2–hour samples, taken during intensive periods is less than 20 degrees.

Samples that are strongly affected by emissions from a source were identified through observation of clustering of mean wind directions for the sampling periods selected with angles pointing toward the source.

The RAPS data of about 35,000 individual ambient aerosol samples were collected at 10 selected sampling sites in the vicinity of St. Louis, MO, was screened according to the criteria stated above. With wind trajectory analysis, specific emission sources could be identified even in cases where the sources were located very close together [65]. A compilation of the selected impacted samples was made so that target transformation factor analysis could be employed to obtain elemental profiles for these sources at the various receptor sites.

A subset of 31 fine particle mode samples from four of the sampling sites were identified that had a high lead concentration, a strong bromine/lead correlation, and trajectories that did not converge on a known point source. The TTFA results showed the profile given in Table 6.3. This profile is compared to ones obtained by Rheingrover and Gordon [65] from a simple regression analysis and with earlier TTFA results of Alpert and Hopke [59].

The motor–vehicle emissions are characterized by Pb (14.7 ± 0.6%), Br (6.5 ± 0.2%), and Al (3.0 ± 0.2%). Bromine and Pb are strongly correlated with each other as expected for leaded–gasoline combustion. The high Al concentration is surprising, as aluminum is

Table 6.3. Comparison of Compositions (Relative to Pb) of Fine Particles Released by Motor Vehicles

Element	Chang et al.[a]	Rheingrover & Gordon[b]	Alpert and Hopke[c]	
			Station 112 July–Aug 1976	All Stations 1 week
Al	0.20 ± 0.02	0.21 ± 0.01	0.05	0.10
Si	0.11 ± 0.03	0.14 ± 0.02	0.0	0.084
P	0.006 ± 0.004	0.013 ± 0.004	d	d
Cl	d	0.14 ± 0.03	0.02	0.08
K	0.06 ± 0.008	0.030 ± 0.005	0.013	0.05
Ca	0.08 ± 0.02	0.11 ± 0.01	0.10	0.07
Ti	0.006 ± 0.004	–	0.0	0.05
V	d	0.0051 ± 0.0006[e]	d	d
Cr	d	0.0036 ± 0.0005[e]	d	d
Mn	0.01 ± 0.004	0.008 ± 0.005[f]	0.0	0.003
Fe	0.09 ± 0.01	0.085 ± 0.010	0.0	0.0
Ni	0.004 ± 0.001	0.0051 ± 0.0006	0.0007	d
Zn	0.012 ± 0.010	0.06 ± 0.02	0.007	0.028
Br	0.44 ± 0.02	0.39 ± 0.02	0.28	0.19
Cd	0.001 ± 0.0009	–	d	d
Ba	d	–	0.007	0.021
Pb[g]	(1.00 ± 0.04)	(1.00)	(1.00)	(1.00)

[a] Chang et al. [61].

[b] Rheingrover and Gordon [65].

[c] Alpert and Hopke [58].

[d] Too many missing values to include in TTFA.

[e] Below formal detection limit on more than half of samples.

[f] Correlation weak, see Rheingrover and Gordon [65].

[g] Absolute concentrations of Pb in components are: 14 ± 4, 14.7 ± 0.6, 10.7 and 22.7%, respectively.

generally present in coarse particles from abrasional and mechanical sources (such as wind blown dust). Rheingrover and Gordon [65] also obtained a high correlation between Al and Pb and suspected that it arose from an analytical artifact, the L X–rays of Br interfering with K X–rays of Al to create a false correlation. Goulding et al. [66] indicate that they have made a full set of interelemental corrections, but an incomplete subtraction of the Br L X–rays could produce an apparent correlation of Al with Pb and Br. A Br/Pb ratio of 0.44 is characteristic of fresh exhaust particles for which Dzubay et al. [63] reported a range from 0.39 to 0.5.

Except for some elements present in very small amounts (P, K, and Zn), the agreement between Chang et al. [61] and that of Rheingrover and Gordon [65] is excellent for elements included in both components. Agreement is expected because motor–vehicle emissions made up 24% of the mass of samples treated. These trajectory samples appear to represent very fresh motor–vehicle exhaust. By contrast, components obtained by Alpert and Hopke [59] from samples collected at Station 112 and at All Stations without regard to wind direction represent more aged particles, as shown by much lower Br/Pb ratios, especially for All Stations, which include several rural sites. The reduced Al concentrations in these components with lower Br concentration supports, but does not confirm, the suggestion that Al X–ray peaks contain Br interference. Disagreements for many minor elements are not considered serious as the Station 112 and All Station samples contained only 15 and 6% mass contributions from motor vehicles, respectively.

It can be seen that TTFA may be very useful in determining the concentration of lead in motor vehicle emission as the mix of leaded fuel continues to change. Multivariate methods can thus provide considerable information regarding the sources of particles including highway emissions from only the ambient data matrix. The TTFA method represents a useful approach when source information for the area is lacking or suspect and if there is uncertainty as to the identification of all of the sources contributing to the measured concentrations at the receptor site. It provides a useful parallel analysis with CMB to help insure that the profiles used are reasonable representations of the sources contributing to a given set of samples.

3. MICROSCOPIC METHODS

Optical microscopy has been an important tool for particle source identification for a long time. A variety of particles have unique morphological, crystallographic, or other features that make it possible to identify their source with a high degree of specificity [40,67]. However, with this specificity there is a problem because of the small number of individual particles that can be scanned and interpreted by a skilled observer in a reasonable amount of time. The typically small number of particles limits the precision with which analyses can be made. There is also the problem in optical microscopy of the physical limits of visual resolution that limits detailed studies only to particles larger than 1 μm. Thus, although optical microscopy has been widely used as a qualitative tool to identify sources, it has not been practical to use it as a quantitative tool.

An alternative approach that has shown the ability to provide quantitative source apportionment is computer–controlled scanning electron microscopy (CCSEM). With an associated X–ray fluorescence analysis system, CCSEM can perform elemental analysis from carbon through uranium on extremely small quantities of matter. In addition, scanning and image processing of the electron microscope can be done under computer control such that both chemical composition and size/shape information can be estimated in as little as two seconds for each particle. The use of CCSEM can greatly increase the information available on the physical and chemical characteristics of ambient or emitted particles. Figures 6.4 and 6.5 show the several kinds of information that can be obtained for a single particle. The secondary electron image provides a three dimensional image showing the particle morphology and surface texture. The image data have not yet been fully exploited in receptor modelling because of the lack of quantitative methods to interpret them in an automated process. The other data shown in these figures is the fluoresced X–ray spectra. The bombardment of the particles by the electrons from the microscope induces inner shell electron vacancies that result in the emission of X–rays whose energies are characteristic of the elements present. Because of the high vacuum in an electron microscope and new ultrathin vacuum windowed detectors, it is possible to observe the X–rays from carbon to uranium. However, the cross–sections for vacancy production are low for electrons and the total excited volume is small. It is generally only possible to observe elements present at a few tenths of atom percent levels or higher. Trace elements are generally not observable. Thus, information on the size, shape, and composition are directly available from the microscopic examination. These properties can then be used to group the particles into particle classes. The mass fractions associated

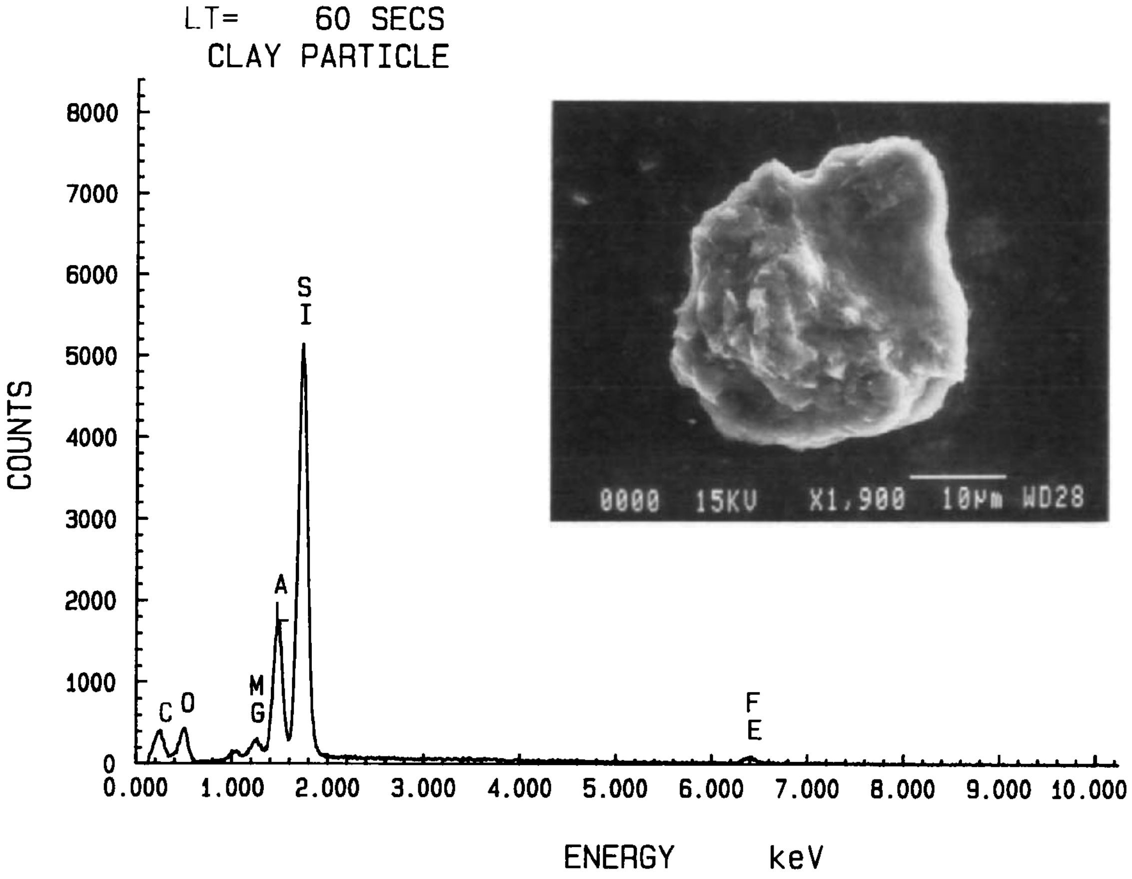

FIGURE 6.4 *Electron micrograph and fluoresced X–ray spectrum of a clay particle. Figure courtesy of the R. J. Lee Group, Monroeville, PA.*

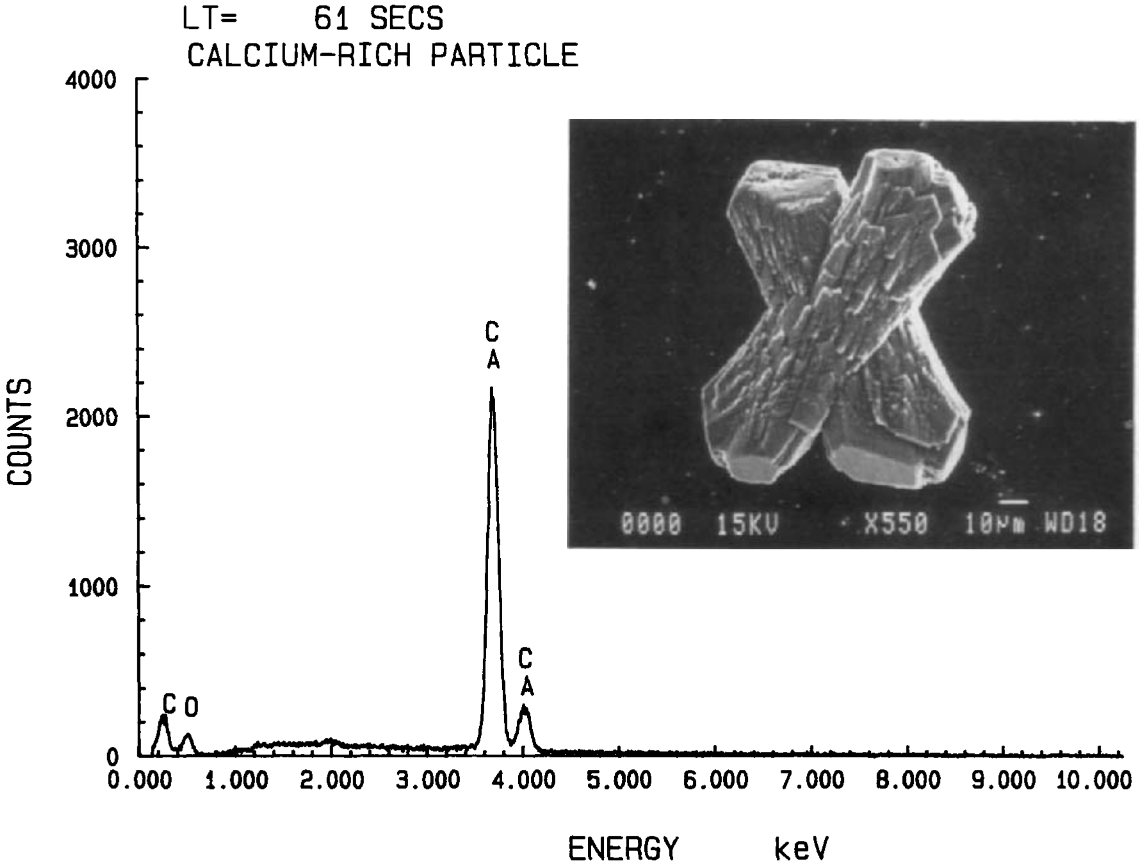

FIGURE 6.5 *Electron micrograph and fluoresced X–ray spectrum of a calcium–rich particle. Figure courtesy of the R.J. Lee Group, Monroeville, PA.*

with each of these classes become the variables on which the source apportionment will be made. A particle class balance assumes a linearly additive sum of class mass fractions times a fractional contribution of aerosol mass by the particulate sources analogous to the chemical mass balance used for bulk sample composition data.

Johnson and McIntyre [68] applied this technique for aerosol apportionment study in Syracuse, N.Y. They used twelve source types from 47 possible source signatures to fit 23 measured ambient samples. In this analysis, they created only 25 separate classes of particles based on the observed relative net X–ray intensities. As they mentioned, their classification had a substantial possibility of overlapping among 25 particle classes when comparing signatures of similar sources. Casuccio and Janocko and coworkers [69–71] also suggested a particle balance model based on the segregation of the source and the ambient samples into aerodynamic size categories. However, they also use empirically derived class definitions that permit the possibility of misclassification of particles. The ability to assign particles unequivocally to well defined particle classes will provide a better set of variables for the apportionment study since there will be more specificity in these better defined classes and thus less noise in the mass balance fitting process [72].

The basic approach to the particle classification problem was to obtain possible class members from various cluster analysis methods [73]. Thus, potential homogeneous classes were obtained from the cluster patterns after deleting obvious outliers (particles belonging to no class). The particles in these classes were then used as examples in a rule–building expert system. The system could be used to develop a universal classification rule based on the representative examples of the homogeneous particle classes. Since such an expert system is highly dependent on the examples, all the examples from each source sample have been carefully examined. This process will now be reviewed in more detail.

The study identified homogeneous particle classes by cluster analysis of a set of source samples [72]. A classification rule was then based on examples from the homogeneous particle classes in order to identify ambient aerosol particles. Figure 6.6 shows the flow chart for the study. The methods were developed using data from the El Paso Quantitative Microscopy Study to identify and quantify the major sources of total suspended particulates (TSP) and particulate lead. These improved class assignments were then used to apportion TSP.

As an example of the use of these techniques to examine sources of airborne particles and lead, a study made in El Paso, TX [71] will be presented with this new analysis of the data made using improved classification methods for the individual particle class assignments. In El Paso, TX an air quality problem existed as shown by an annual geometric mean value of total suspended particle (TSP) concentration and quarterly ambient lead concentrations significantly above the corresponding U.S. ambient air quality standards. In an effort to identify the sources of TSP and lead, the Texas Air Control Board initiated a study whose primary approach utilized computer–controlled scanning electron microscopy. Both ambient filter samples and a large number of samples from suspected sources were taken in the El Paso area particularly in the vicinity of a large smelter that was thought to be the major contributor to both the observed TSP and lead levels.

Since there were other major sources of lead including a lead and copper smelter in the El Paso area, the auto emission source sample was collected at the side of a major highway near San Antonio, Texas, using a standard TSP sampler and a cellulose filter. The particles were removed from the filter and redeposited onto a power–type filter for CCSEM analysis. The results of the sample analysis were 735 particles with each particle characterized by 25 variables including 6 physical descriptive variables (maximum and minimum diameter, its aspect ratio, mass, aerodynamic diameter, and effective mass) and 19 elemental X–ray intensities concentrations (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Cu, Ni, Zn, Pb, and Br). Since each particle type was assigned a density based on the most likely chemical compound, an aerodynamic diameter and a mass can be calculated from the measured physical diameter and the assumption that the particle is spheroid of revolution. By employing a segregated particle counting scheme, the effective mass contribution of each particle to the total sample mass can be calculated. CCSEM results of this sample indicated that Ca–rich, Ca–Si and Si– rich particle types accounted for the majority of the particulate matter [74].

To begin the analysis of a large data set having unknown distribution, data description and data management by simple univariate analysis will be helpful to look at the probable distribution of values and statistics of the raw data set. These analyses are needed since many statistical analyses require the data distribution to meet an assumption of normality or at least symmetry with respect to the mean value.

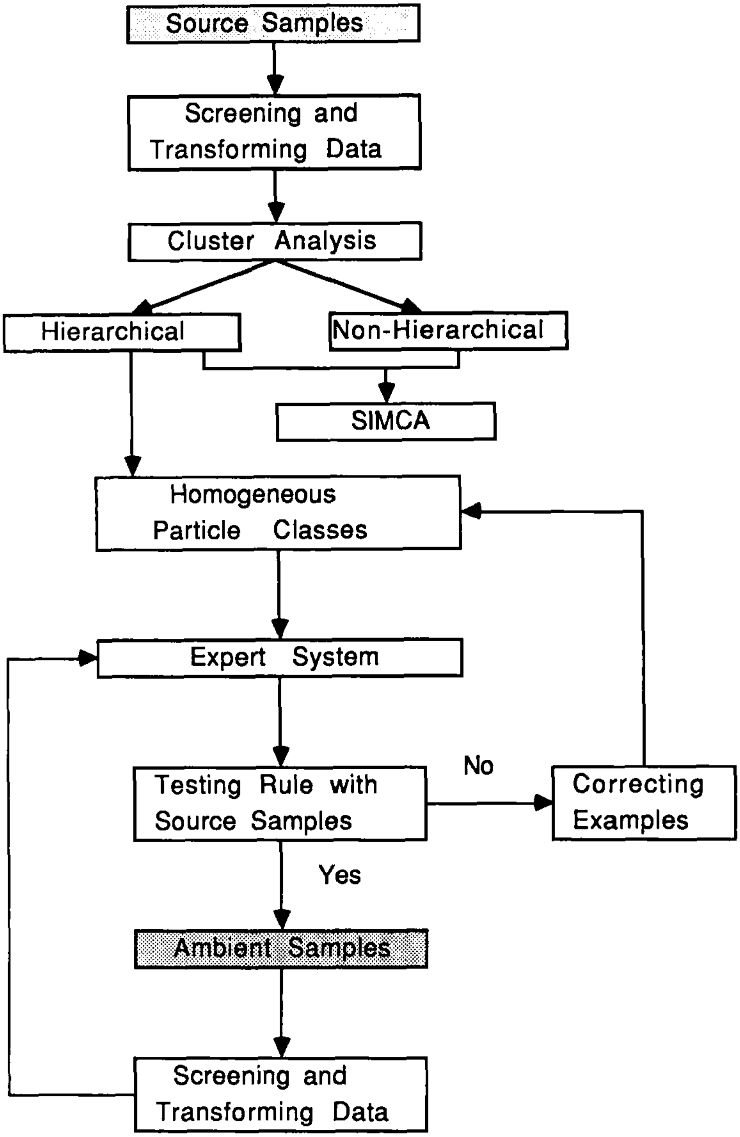

FIGURE 6.6 *Flow chart for the classification of individual particles based on CCSEM data.*

Since the ability to determine the particle size distribution is one of the advantages of CCSEM, the frequency distribution for aerodynamic diameter can be directly deduced. The distribution was highly positively skewed and bimodal in shape, demonstrating both fine and coarse modes, with a minimum occurring around 3 μm. Figure 6.7 shows the frequency distribution of aerodynamic diameter in the auto emission sample. In this bimodal distribution, there are quite noticeable differences in composition between the two modes: most of Pb– or Cu–rich particles are observed to exist in the fine mode, whereas most Si–, Ca–, Fe–, Mg–, Cr–, and Ni–rich particles are in the coarse mode. Because Figure 6.7 is a particle number distribution, the accumulation mode contains the larger fraction of particles while the coarse mode would represent the greater mass.

Figure 6.8 shows the frequency distribution of lead X–rays. This distribution is also bimodal: one mode where the X–ray intensity is zero and a second, positively skewed distribution for those particles that actually contain lead. Such strongly skewed distributions will yield significant problems for most classification methods. It is therefore necessary to perform a transformation on these data and a logarithmic transformation is suggested. A simple logarithmic transformation cannot be made directly because of the zero values. The empirical transformation of log (1+x) can avoid this difficulty. The addition of single count for the non–zero values makes a very small perturbation to the non–zero values since total X–ray counts are being used.

However, another problem arises with the X–ray intensities that needs to be solved before applying the transformation. This problem is the presence of false peaks. Because the X–ray analysis is a statistical counting based method, there can be spurious counts in the region where an X–ray peak would be found if the element were present. The presence of noise in the spectrum causes weak peaks to be identified when they are really not statistically significant. Thus, a noise reduction step was included where it is assumed that any peak whose area was less than twice the uncertainty in the total counts in the spectrum would be set to zero. Then the spectrum was subjected to the log (1+x) transformation resulting in the distribution shown in Figure 6.9.

Finally since the calculation of distance depends on the scale of the data, the data are transformed to have zero means and unit variance (z– transformation). This transformation insures that all variables will have equal weight in determining the similarity between any given pair of objects. These rescaled log transformed X–ray intensities after cleaning–up were then used as a basis for classification.

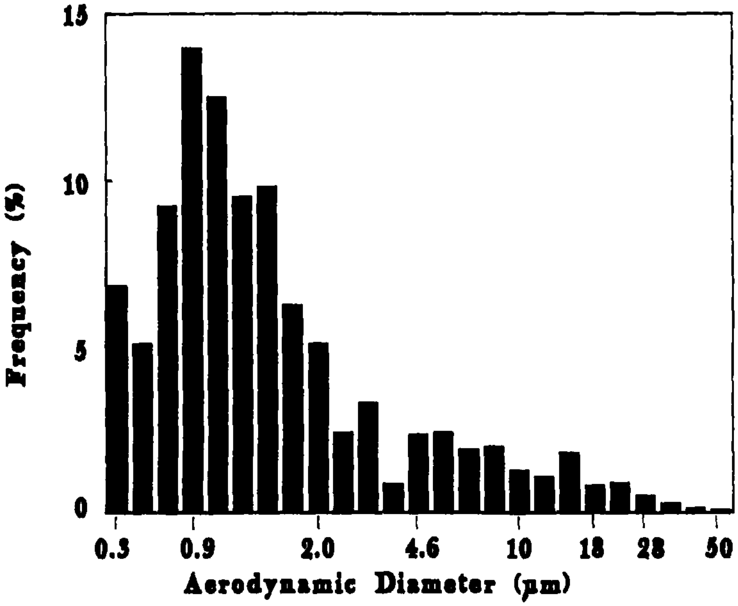

FIGURE 6.7 *Frequency distribution for particle aerodynamic diameters in Auto Emission sample as determined by CCSEM.*

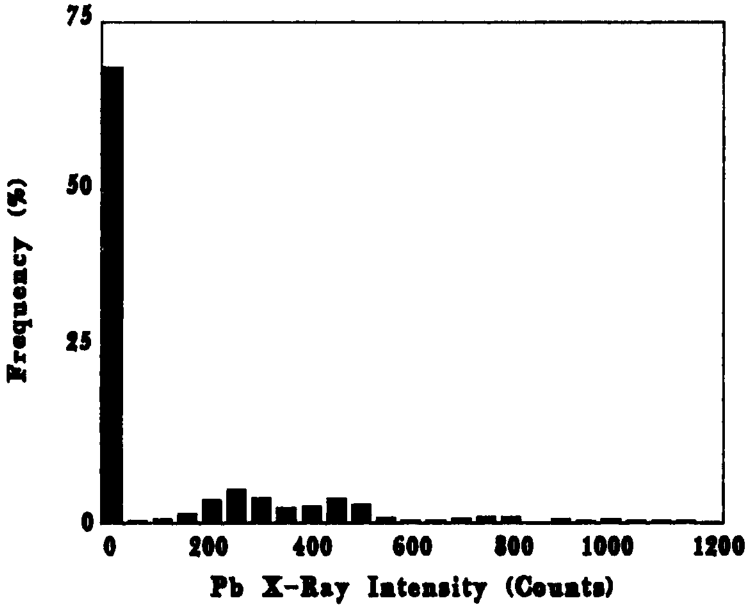

FIGURE 6.8 *Frequency distribution for lead X-ray intensities in Auto Emission sample as determined by CCSEM.*

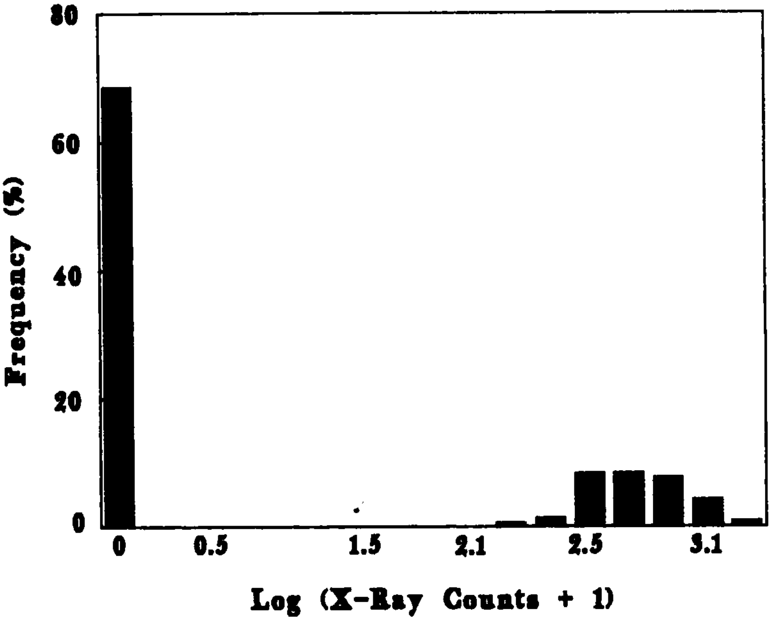

FIGURE 6.9 *Frequency distribution for log (1 + x) transformed X–ray intensities after noise reduction in Auto Emission sample as determined by CCSEM.*

There are two general methods in clustering: hierarchical and non–hierarchical (or *partitioning*) algorithms. Hierarchical algorithms are most widely employed in natural science handling large amounts of data. These techniques are efficient in terms of computational scheme, but have the drawback that a sequence of decision is made and once objects are assigned to groups, it cannot correct early mistakes [73] because there are no reallocation procedures included.

The computer program AGCLUS is implemented in a neutral dialect of FORTRAN IV for various criteria of agglomerative hierarchical clustering [75]. Seven popular hierarchical methods based on measures of within–class–distance or between–class–distance can be chosen by determining a measure of dissimilarity from observed data.

MASLOC, based on the k–median model, was developed at the Vrije Universiteit Brussel, Belgium and was written in transportable FORTRAN [76]. This program separates the objects using k objects that are representative of each cluster. These objects are called medoids or centrotypes. It finds these objects as the ones that minimize the sum of the distances from each object to its nearest centrotype object. The clustering obtained at k clusters is totally independent of the ones obtained for k–1 or k+1 clusters. MASLOC can determine "robust" clusters where no objects aggregate again with objects of other clusters in going to higher k. It can also determine isolated clusters where the within–class-distances are all small compared to the between–class–distances.

A k–means algorithm is performed by using the BMDP (Biomedical Computer Programs) statistical package. The PKM program partitions a set of particles into a given number of clusters, which is based on the Euclidean distance between the individual particles and the centers of the clusters. Strong points of the BMDP package are that the control language is English oriented and easily implemented and the data and the cluster member indicator variable can be saved on a BMDP file for future use. BMDPKM requires relatively short computational time, comparable to the hierarchical algorithm.

The agglomerative hierarchical clustering program AGCLUS, was used to see the initial systematic behavior of the large data set. All 735 objects with 19 X–ray intensity variables were studied. Squared Euclidean distance is used as measure of distance. Figure 6.10 shows the first and the last parts of the auto emission dendrogram. It can be observed that in the part shown on the left side, there are large groups obtained at a relatively low level of clustering. However, there are a number of small groups that appear near the end of the dendrogram as shown on the right side of the figure.

The results of the MASLOC k–median program and the BMDPKM k–means program with 60 seedpoints ($k = 60$) have identified well isolated clusters containing up to 77 particles. The same data treatment as AGCLUS was used for these non–hierarchical clustering processes. Before the logarithmic transformation, both non–hierarchical algorithms find many singletons (outliers) so that the analyses mostly find extreme valued particles. The log transformation yields a clustering with well separated clusters. The 26 isolated clusters of MASLOC and 18 clusters of BMDPKM having more than 10 particles are considered to be the main groups in auto emission source. The results of cluster analyses shown in Table 6.4 indicate that AGCLUS separates crustal clusters fairly well. However, MASLOC performs better with the heavy metal clusters.

Main particle classes deduced from three different algorithms explain almost 80% of total mass. From the results of MASLOC clustering, a representative crustal particle type, Ca–Si–Al, and the lead particle type Pb–Br–Al–Si were selected and their size distribution histograms are shown in Figure 6.11. As expected, most of Ca–Si–Al particles exist in the coarse mode and Pb–Br–Al–Si particles in the fine mode.

In order to use the cluster analysis results for the particle class balance analysis, a particle classifier program must be established that uses the X–ray intensities as input and yields a particle class assignment. The cluster analysis provides potentially homogeneous groups of particles that can be designated as a particle class. However, an efficient sorting procedure is needed to apply the classification rules to the source and ambient samples. Such an approach has been presented by Kim and Hopke [72] using a rule–building expert system.

In the study of classification for particle classes based on their X–ray intensities, an important consideration is the definition of the "homogeneous particle classes." A homogeneous class is one in which all of the objects have similar characteristics. In this case a similar pattern of x–ray intensities. When implementing various clustering methods to classify the objects into particle classes, the clustering methods only provide tentatively homogeneous classes. A possible group of particles chosen from cluster analysis can often have chemically different particles assigned to it. These particles are considered to be outliers to homogeneous classes. Thus, a class is determined to be homogeneous only when

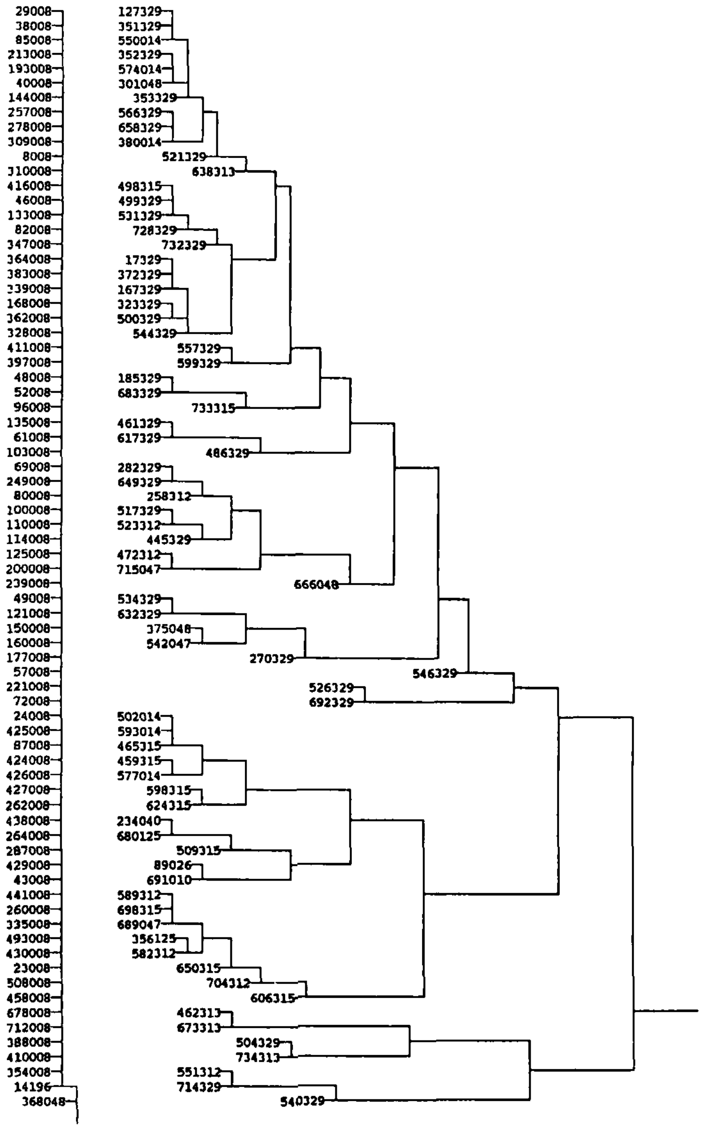

FIGURE 6.10 Two portions of the agglomerative hierarchical dendrogram for the auto emission CCSEM data showing a cluster of essentially identical particles on the left and many more diverse clusters on the right.

Table 6.4. **The mass fraction of main groups of auto emission source sample based on various clustering methods**

	Clustering Method		
Particle Type	MASLOC	BMDPKM	AGCLUS
Ca	0.1436 ± 0.0193	0.1417 ± 0.0190	0.1430 ± 0.0191
Ca–Si	0.0745 ± 0.0107	0.0746 ± 0.0107	0.0759 ± 0.0105
Ca–Si–Al	0.1422 ± 0.0167	0.1426 ± 0.0169	0.1403 ± 0.0169
Ca–Na–Si	0.0311 ± 0.0075	0.0314 ± 0.0077	0.0245 ± 0.0064
Ca–Si–Al–S	*	0.0605 ± 0.0211	0.0569 ± 0.0224
Si	0.0430 ± 0.0201	0.0563 ± 0.0205	0.0432 ± 0.0203
Si–Al–Fe–Ca	0.0567 ± 0.0064	0.0569 ± 0.0063	0.0596 ± 0.0069
Si–Al–K–Ca	*	0.0303 ± 0.0094	0.0310 ± 0.0101
Si–Na–S–Al–Ca	0.0097 ± 0.0038	*	0.0104 ± 0.0039
Pb–Br	*	0.0266 ± 0.0082	*
Pb–Si	0.0088 ± 0.0065	*	0.0015 ± 0.0007
Pb–Br–Si	0.0101 ± 0.0004	*	*
Pb–Si–Br	*	0.0171 ± 0.0073	*
Pb–Br–Cl	0.0185 ± 0.0051	0.0159 ± 0.0047	0.0173 ± 0.0051
Pb–Br–Cr–Si	0.0048 ± 0.0036	0.0045 ± 0.0035	*
Pb–Br–Al–Si	0.0202 ± 0.0084	*	0.0303 ± 0.0094
Cu–Si	0.0111 ± 0.0004	*	*
Zn–Si–S	0.0214 ± 0.0151	*	*
Cr–S–Si–Al	0.0180 ± 0.0079	*	*
Mn–Si–S–Fe	0.0124 ± 0.0059	*	*

* *indicates no group generated or minor group which have less than 10 particles in a given particle class type.*

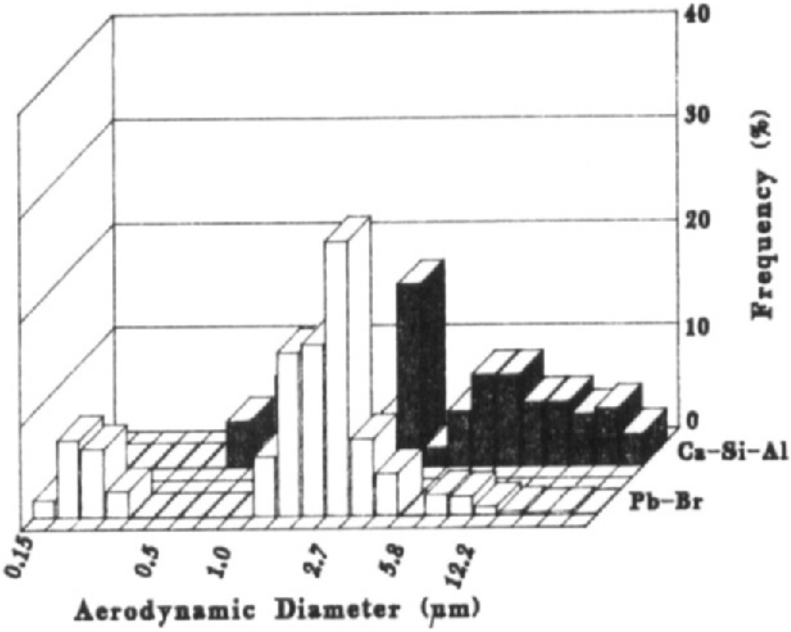

FIGURE 6.11 *Aerodynamic size distributions for the Ca–Si–Al and Pb–Br–Si particle classes derived from the Auto Emission sample.*

all particles in a class show only the same elements. A particle in Al–Si–Fe class will show only Al, Si, and Fe X–ray intensities. In the sense of this concept, all particles initially clustered from cluster analysis can be examined, and thus it is possible to remove outlier particles for each class. Those particles in the class after deleting outliers can be potential "representative" examples for the expert system.

With the 19 chemical compositions as variables, the $2^{19} - 1$ combinations of particle classes can occur without replacement of the elements. For example, when there are only three elements, Al, Si, and Fe, the possible particle classes can be observed as one of the seven particle classes (Al, Si, Fe, Al–Si, Al–Fe, Si–Fe, and Al–Si–Fe). However, because of possible isomorphous substitutions of elements in terms of geochemistry of formation of the material, the sequential order of elements in the class name is not related to the relative X–ray intensities for particle classes having more than two elements. With two elements, it is likely that a change from one being the major X–ray intensity to the other being dominant reflects a change from one particle type to another. Thus, Si–Al and Al–Si classes are defined as different from each other. However, since there is the possibility of composite particles as well as substitution, the Si–Al–Fe, Al–Si–Fe, and Fe–Si–Al classes are considered to be of the same homogeneous class.

The total number of possible particle classes is then $(2^{19} - 1) + C(19,2)$, where the second term includes the number of combinations with replacement of the element when considering only two elements. The objective is to have a precise enough classification method to obtain sufficient resolution of one source material from another. There are practical limitations to how fine the classification can be. As the class definitions become more specific, more particles must be examined so that the mass fraction that class represents can be determined with reasonable precision. Thus, it is possible to provide more classes if needed at the cost of analyzing enough particles to insure each class mass fraction is well determined.

Expert systems, a class of high performance computer programs in the area of artificial intelligence, are applied as knowledge–engineering tools in any field to interpret, predict, diagnose, design, plan, monitor, and control systems. An expert system is dependent on obtaining the knowledge in fully concrete terms. Mittal and Dym [77] warned of potential mismatch because a single domain expert in complex and varied domains like applied science can only provide information on a small subset of the tasks in the domain. Thus, a system attempting to expand beyond that subset may provide poor or misleading guidance.

In general, to draw a conclusion about scientific evidence it is necessary to collect all the possible evidence or examples characterized by various variables and then analyze them systematically. Recent trends to reach this task are to analyze data statistically and develop experimental design to collect data easily analyzed by expert systems. Gale and Pregibon [78] reviewed a project applying artificial intelligence methods in statistics.

EX–TRAN 7 [79] is a series of programs designed to generate a set of rules in the form of a decision tree based on examples for which various attributes (variables) are known and which have known outcomes (classes). It then produces a self–contained FORTRAN program that can implement these rules. The rule–generator program is called the Analog Concept Learning Translator (ACLTRAN). It can use numerical or logical variables as attributes for the objects in the known classes. ACLTRAN uses an extension of Quinlan's ID3 algorithm [80].

The program searches the features one at a time to identify the one for which it can "best" separate one class from the others. The choice of attribute is based on the maximum decrease in the entropy derived from information theory between the undivided and the split classes. The information theoretical approach used to determine the entropy of each state is described by Hunt et al. [81]. For a given variable, the entropy of the whole and divided states are calculated, and the separation is made based on the maximum entropy decrease obtained. The decision rule is then formulated for the attribute with the largest entropy change. The rule splits the data such that an attribute value greater than or equal to the value midway between the closest points of two data sets are assigned to one group and those less than that critical value are assigned to the other class. The program systematically divides the data set until all of the objects are separated into single class subsets. Thus, EX–TRAN is a sequential univariate classification system.

This rule–building expert system was used to build a decision tree from examples of particle classes following classification studies. To perform this study, it was necessary to find a set of "representative" examples of the "homogeneous" particle classes from all of the source samples to be included in the particle class balance.

The agglomerative hierarchical clustering algorithm AGCLUS was used to obtain the potential number of the homogeneous particle classes. Distinct particle classes at the lowest level of the dendrogram in cluster analysis were selected when the class contained more than four particles. These initial classes were not perfectly homogeneous and, therefore, the outlier particles were removed. Some classes were split into smaller classes to obtain homogeneity. Each data set representing one of twenty–one source samples was split into 23 to 67 homogeneous particle classes. A total of 11,294 (73%) out of 15,499 particles (80% of total mass) were placed into one of the homogeneous classes so that they could be used as examples for expert system implementation.

It is important to note that the remaining particles (27%), including both particles not selected from the cluster analysis and outlier particles removed from the potential homogeneous classes, were placed into a miscellaneous class that was not active in the EX–TRAN algorithm. As described above, since there are many possible combinations for homogeneous classes, in order for the miscellaneous class to be active in the EX–TRAN, a corresponding number of examples would be needed to build a rule for the universal particle classification. This limitation could be overcome by simple set theory.

Let the totality of particles be the universal set, S; the homogeneous classes be a subset, H; and the miscellaneous class be a subset, M. Further, let P be the probability of belonging to any given set. Then P(H) is the probability of the event H, so that the following properties are satisfied:

i) $P(H) > 0$ and $P(M) > 0$

ii) $P(H) + P(M) = P(S) = 1$

iii) $P(H \cap M) = 0$

iv) If $H_1, H_2, \ldots H_k$ are the sub–sub sets of sub set H, then

$$P(H_1 \cup H_2, \ldots \cup Hk) = P(H_1) + P(H_2) + \ldots + P(H_k)$$
$$= P(H)$$

for each positive integer **k**.

H1, H2,...Hk are independent from one another; therefore $Hi \cap Hj = 0$, $i \neq j$. In particular, if $k = (2^{19} - 1) + C(19,2)$, then $P(M) = 0$ and $P(H) = P(S) = 1$. Since H is the complement of M, even though subset M is not active in the expert system study, this problem can be solved after obtaining a perfect subset H.

In order to create a classification rule from the expert system, it is necessary to find a set of "representative" examples. For this study, 283 homogeneous particle classes from the 21 source data sets were created after deleting outlier particles.

It must be noted that all of 11,294 particles assigned to one of the 283 classes cannot be used directly as the representative examples since the maximum number of examples that can be used by EX–TRAN is 300. To select 300 representative particles from the 11,294, various techniques have been tested to provide the best set from which to induce an accurate rule tree. EX–TRAN sets its decision value at the midpoint value between the variable values of the two closest points between the groups of particles being separated [82]. Thus, an example which has minimum or maximum value for an attribute (variable) in a given class can be considered as a representative example. For instance, the Si–Al–K particle class observed from the 17 sources consisted of 434 particles. A particular particle is then selected as a representative example which has the maximum value of the Si X–ray intensity among all the particles in the class. Next, a particle with the minimum value of Si is selected. The Al and the K X–ray intensities are also considered in the same manner. Then 6 particles can be chosen to represent this class among the 434. If a class consists of 10 attributes, at most 20 particles are needed. For each of the 283 classes, the same approach was used. However, 283 classes are more than enough to exceed the program limitation of the 300 examples. The next technique used to solve this problem was to make subsets such as H'_1, H'_2,....H'_j, and then $S > H > H'_j > H'_i$, where i,j = integer. To do this, the first subset H'_1 was defined as a set of all the Na–containing classes; the second subset, H'_2 as a set of the Mg–containing classes except objects in H'_1; and the third subset, H'_3 as a set of Al–containing classes except objects in H'_1 and H'_2, and so on.

An example file for subset H'_1 was prepared for ACLTRAN. Each example is characterized by the 19 elemental X–ray intensities as attributes and labelled with a homogeneous class type. ACLTRAN generates induced classification rules in several different forms such as a decision tree, and a standard FORTRAN–77 code. All the source samples were used to build the rule tree, test the rules and correct the input set of examples.

Figure 6.12 shows an example of a tree from the decision rule. The decision tree shows the assignment of particles containing sodium into classes. For example, if the transformed Ca value is less than 1.0205, the particle will go into classes, C1, C4, C8, or C10 depending on the variables S, Si, and Al. For particles with Ca greater or equal to 1.0205, they will be assigned to classes C2, C3, C5, C6, C7, or C9 depending on the values of Al, Si, and K. The names given to each class are listed in the box below the figure.

The decision rules for all the subsets were determined in an identical manner, and then each FORTRAN code was incorporated into a complete universal classification code so that the rules could be executed as a program on an IBM or compatible microcomputer.

```
[ca      ] :
   < 1.0205 : [s       ] :
                  < 1.00 : [si      ] :
                             < 0.8265 : c10
                             >=0.8265 : c8
                  >=1.00 : [al      ] :
                             < 1.145 : c4
                             >=1.145 : c1
   >=1.0205 : [al      ] :
                  < 1.00 : [si      ] :
                             < 2.1125 : [si      ] :
                                          < 1.0395 : c9
                                          >=1.0395 : c7
                             >=2.1125 : [k       ] :
                                          < 1.057 : c6
                                          >=1.057 : c5
                  >=1.00 : [k       ] :
                             < 1.0395 : c3
                             >=1.0395 : c2
```

```
Class c1......Na-Mg-Al-Si- S-Fe        Particle Type
Class c2......Na-Mg-Al-Si- K-Ca-Fe     Particle Type
Class c3......Na-Mg-Al-Si-Ca           Particle Type
Class c4......Na-Mg-Si- S- K           Particle Type
Class c5......Na-Mg-Si- K-Ca           Particle Type
Class c6......Na-Mg-Si-Ca              Particle Type
Class c7......Na-Mg-Si-Ca-Pb           Particle Type
Class c8......Na-Mg-Si-Pb              Particle Type
Class c9......Na-Mg-Ca                 Particle Type
Class c10.....Na-Mg-Pb                 Particle Type
```

FIGURE 6.12 *Example of a tree from decision rule from the El Paso, Texas, study.*

In the beginning of the test step, a decision rule obtained from the 21 source samples was examined sequentially for each source data set for all 15,499 particles. For this test, each raw source data set was screened and transformed as described above and its particles were labelled with one of the 284 classes, including the miscellaneous class. Thus, the test was performed like a supervised pattern recognition analysis. Only two (0.01%) out of the previously assigned particles were misclassified by the final routine. These misclassifications occurred when the number of representative examples in a class was much less than that of the active attributes. Thus, the frequency of the misclassification was small and acceptable. From the results of testing all particles, it was found that the universal decision rule could also detect the misclassification by the hierarchical cluster analysis. For example in one source sample from a lead blast furnace, 501 of 735 particles were assigned to one of the identified homogeneous classes. However, three events in the unclassified miscellaneous set were found to belong to one of the homogeneous classes defined by examples from other sources. In this manner, a total 48 particles from the 21 source samples had been misclassified. The misclassified examples were reassigned to the appropriate homogeneous particle class, and the mass fraction values for each homogeneous class were corrected. It is an important step to correct the mass fractions of each class since they will be used as source profiles in a subsequent source apportionment study.

The final decision rule for El Paso data was then applied to the classification of ambient samples for the source apportionment study. To perform this study, it is not necessary to start from cluster or principal component analysis. Nineteen ambient samples were examined by Kim and Hopke [83]. Each ambient data set has almost the same number of particles as that of source samples and has the same data formats. Initially the data for each ambient particle was screened and transformed in the same manner as the source data. Classification was performed by the execution of the decision rule. A number of classes were found in each sample. Of the total of 12,805 particles analyzed in the 19 ambient samples, 10,347 particles were assigned to one of the homogeneous classes. In terms of mass fraction, over 83% of the total mass was partitioned into the homogeneous classes.

The source apportionments for two ambient samples taken at the site nearest the non–ferrous metal smelter are shown in Figure 6.13. The analyses of the remainder of the nineteen samples are given by Kim and Hopke [83]. They find that the larger number of variables (particle class mass fractions) obtained in the particle class balance provides much greater specificity in the identification of particle sources. They found that various process effluents from the smelter could be distinguished from one another. However, they also found a substantial decrease in the generality of the source profiles so that the quality of fit decreased noticeably for ambient sampling locations far from the location where most of the source samples were obtained suggesting the presence of unsampled sources or substantial changes in the source profiles in transit. Certainly it is clear that CCSEM and particle class balance can provide very detailed source resolutions including highway emissions and are likely to be more widely used in the future.

SUMMARY

In this chapter several of the active areas of receptor modelling have been introduced. Their use as indicators of the sources of metals in the air can be very helpful in developing air quality management strategies and can potentially become enforcement tools as well. Since receptor models must of necessity be retrospective in nature, another important use can be in the calibration and testing of the prognostic dispersion models so that prediction of changes in air quality can serve as a more reliable basis for management decisions. The field of receptor modelling has grown and developed rapidly during the last several years and can be expected to continue to do so as methods are improved and new applications discovered.

ACKNOWLEDGEMENTS

Professor Philip Hopke, author of this chapter, would like to acknowledge the assistance of Gary Casuccio of the R.J. Lee Group for the material for Figures 6.4 and 6.5. Much of the studies reported here have been performed by students or post–doctoral associates in his group and their substantial contributions to the results presented here must be acknowledged. The work could not have been conducted without the support of the U.S. Department of Energy under contract DE AC02–80EV10403, the U.S. Environmental Protection Agency under Grant No. R808229 and Cooperative Agreement R806236 and the National Science Foundation under Grant ATM 85–20533.

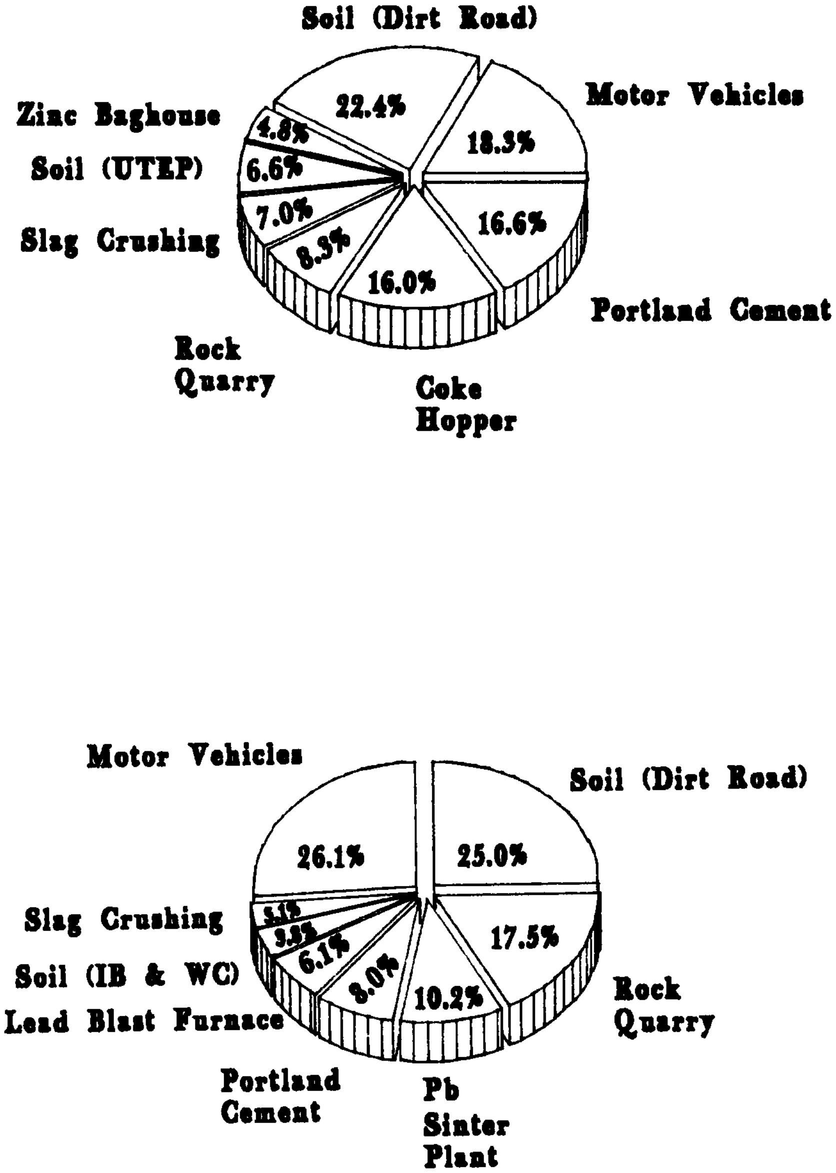

FIGURE 6.13 *Pie charts for course apportionment of ambient samples A222 (top) and A223 (bottom).*

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Chapter 7

HUMAN HEALTH EFFECTS OF HIGHWAY–RELATED POLLUTANTS

D.R. Lynam and G.D. Pfeifer

Contents **Page**

1. INTRODUCTION

Automobile and truck exhausts contain substances which can adversely affect human health if encountered in sufficiently high concentrations. Included are carbon monoxide, oxides of nitrogen, oxides of sulphur, uncombusted and partially combusted hydrocarbons (including aliphatics, olefins, benzene and other simple aromatic compounds, aldehydes, ketones, and polynuclear aromatic hydrocarbons), and particulate matter of varying composition. In addition, exhaust product mixtures may react in the presence of sunlight to form other potentially toxic materials like ozone and peroxyacetyl nitrate (PAN), as described in Chapter 1.

A substance must reach a target organ at levels above a certain concentration, called the threshold level, to be detrimental to health. The target organ may be non–specific (as when corrosives are involved) or may be specific (as when hydrocarbons affect the liver). The respiratory tract is the most common route of entry for highway related pollutants, both gases and particulates. Particulates with an aerodynamic diameter above 10 microns are trapped in the upper respiratory tract. They are then removed from the upper respiratory tract by the mucociliary clearance mechanism and enter the body by ingestion.

A large percentage of particles in the 5–10 micron range are also trapped in the upper respiratory tract. Particles not deposited in the upper respiratory tract may enter the deep lung areas along with smaller particles. Those that reach the deep lung areas may be deposited by the action of gravity, or in the case of very small particles, by diffusion processes. A portion of small particles are trapped in the upper respiratory tract. These are removed by the mucociliary clearance mechanism. Some deposited materials are absorbed into the bloodstream while others may remain in the lung tissue.

Threshold levels are a function of both the specific toxic material and individual differences. Some materials are inherently more toxic than others and have harmful effects at lower concentrations. Toxicity depends on the specific action of the material on the target organ, the rate it is excreted or transformed, and the rate it is absorbed. In addition, other genetic, environmental and past history factors may affect the reserve capacity of the body. For instance, exposure to a hepatotoxin not harmful to the normal individual could be fatal for an individual already suffering from liver disease.

The concentration of toxic material that reaches the target organ depends on factors such as concentration in the ambient environment and the frequency and length of exposure.

Usually, concentrations of highway–related pollutants remain well below hazardous levels. However, photochemical smog may occur in some high traffic areas. When this occurs, toxic substances may reach levels which can cause adverse health effects for some individuals.

Understanding how highway pollutants affect human health requires knowledge about potential health effects of the individual components and synergistic effects that might occur from exposure to multiple toxins. Knowledge that individual components may react to produce new toxic substances is also important. In most cases, little is known of toxicological properties of pollutants at the low levels likely to be encountered. Effects are often non–clinical or occur only after prolonged low–level exposure. It is therefore necessary to extrapolate from studies on animals or from exposure of humans at higher concentrations (occupational exposures).

Epidemiological studies may be unsuccessful because the effects are often quite subtle, the concentration is variable, and multiple pollutants are present. The following sections discuss toxic effects of individual components and describe some of the interactions which arise from photochemical smog.

2. EFFECTS OF GASEOUS EMISSIONS

2.1 Carbon Monoxide

Carbon monoxide is found in relatively high concentrations in the exhausts of diesel and petrol powered engines. The effects of carbon monoxide exposure at high levels are well known. When carbon monoxide is inhaled, it enters the blood stream and binds to hemoglobin. The resultant compound is carboxyhemoglobin. Because the binding affinity of carbon monoxide to hemoglobin is so much stronger than that of oxygen, about 210 times as strong, [1], it blocks the reaction between haemoglobin and oxygen. The blood is then unable to supply sufficient oxygen to the cells. The ultimate effect is similar to that seen in severe anaemia.

Carboxyhaemoglobin formation effectively lowers the available haemoglobin. In the normal individual, no effect is noted until at least 5–10% of the haemoglobin is converted to carboxyhaemoglobin. The reserve capacity of the blood prevents effects at lower carboxyhaemoglobin levels. Effects may be noted at lower levels in individuals suffering from cardiovascular disease or anaemia. As carboxyhaemoglobin levels increase, other signs and symptoms occur. These may include headaches, visual disturbances, impairment of judgment, nausea and vomiting, coma and convulsion. Death may occur if the carboxyhaemoglobin levels reach about 70%.[2].

Exposures to carbon monoxide from automobile exhausts are low except when vehicles are run in confined areas (garages, tunnels, enclosed car parks and parking garages). Carboxyhaemoglobin levels remain low, well below 5% in most cases. If other factors are present, an increase in carboxyhaemoglobin of a few percent may be important. For instance, inhaled cigarette smoke may contain well over 400 ppm carbon monoxide. Heavy smokers may have up to 9% carboxyhaemoglobin, even in the absence of other exposures. [2].

An example of a synergistic effect involves the widely used solvent dichloromethane (methylene chloride). This compound is metabolized to carbon monoxide. Exposures to this solvent may lead to increased levels of carboxyhemoglobin. Individuals having occupational exposures to carbon monoxide who are heavy smokers and who have exposures to dichloromethane are at particular risk from effects of carbon monoxide from auto exhaust.

The effects of chronic exposure to low levels of carbon monoxide are not well understood and are subject to some controversy. It has been hypothesized that low level exposures to carbon monoxide contribute to cardiovascular disease. Conflicting results have been obtained from animal studies. Weir and Fabiano reviewed the evidence and concluded that chronic exposure to low levels of carbon monoxide neither contributed to the atherogenic process nor adversely affected cardiac rhythm in man.[3].

Gates and Goldmuntz hypothesized that a decrease in automotive emissions of carbon monoxide was accompanied by a decrease in ambient levels. They claimed that this was the cause of the decrease in levels of carboxyhaemoglobin noted in the general population in Chicago during the same time period.[4].

Between 1970 and 1974 in the USA, the weighted average carbon monoxide emission decreased from 86.87 to 67.06 g/mile, a decline of 22.8%. During the same time period, the mean carboxyhaemoglobin levels in non–smokers declined from 2.04% to 1.53%, a 25.0% reduction. This reduction was attributed to the decline in auto emissions. Several studies have shown that exposures to persons riding in automobiles are several times the levels found at fixed site monitors.[5,6]. The heaviest exposures to motorists occurred in heavy, stop–go traffic.

Altitude is frequently overlooked when considering the possible health effects of carbon monoxide. Because barometric pressure decreases rapidly as altitude increases, there is less oxygen available to the body.[1]. People who live in higher altitudes adapt and often have a higher concentration of haemoglobin in their blood. This compensates for the decrease in the partial pressure of oxygen. The amount of carboxyhaemoglobin formed is a function of the oxygen:carbon monoxide ratio. Therefore, carboxyhaemoglobin levels increase with altitude given a constant concentration (mass/volume) of carbon monoxide. Because displacement of CO from the haemoglobin is at a lower rate due to the decreased partial pressure of oxygen, cumulative exposures are important. For cities at high elevation with significant pollution problems such as Denver or Mexico City, these effects may be important. In Denver, for instance, the barometric pressure, (and therefore the partial pressure of oxygen), is only about 80% of that at sea level. Carbon monoxide concentrations which would have no effect at sea levels could have a significant effect at higher altitudes on individuals with certain health problems, such as anaemia or cardiovascular disease.

2.2 Nitrogen Oxides

There are several species of nitrogen oxides (nitrous oxide, N_2O; nitric oxide, NO; nitrogen dioxide, NO_2; dinitrogen trioxide, N_2O_3; and dinitrogen pentoxide, N_2O_5). Only nitric oxide and nitrogen dioxide will be considered in this discussion. The other forms are found only in very low concentrations and are relatively non–toxic. Nitric oxide is the nitrogen oxide produced in greatest quantity during combustion. It has virtually no direct effect on health because of its low toxicity and relatively rapid disappearance from the atmosphere. However, it is converted to NO_2 which plays a role in photochemical smog. Although nitrogen dioxide is formed in small quantities during combustion, significant ambient levels of NO_2 may be found because nitric oxide is converted to nitrogen dioxide. These processes were in Chapter 1.

Exposure of experimental animals to high concentrations of nitrogen dioxide for long periods results in a number of pathological changes in various species.[7]. These changes include: (1) ciliary loss in the upper respiratory tract in mice and rats, (2) emphysematous changes in dogs and guinea pigs but not in rabbits, and (3) edema in squirrel monkeys. In addition, several species showed a decreased ability to resist bacterial and viral infection when exposed to nitrogen dioxide.

Research on human volunteers, exposed to nitrogen dioxide at 4–5 ppm, has also been conducted. The most important health effect noted was an increase in expiratory flow resistance. This effect had a delayed onset.[7]. Immediately after exposure, no effect was noted. Measurements at 10, 20 and 30 minutes showed an increasing effect. Delayed effects of very high occupational exposures (250 ppm or higher) have also been noted. In some cases, there was a latent period of 8 hours or more before rapid onset of pulmonary edema with severe consequences occurred. In other cases, only slight effects were present immediately after exposure. After a 2–3 week latency period there was rapid onset of fever, chills and difficulty in breathing.[2]. These severe effects have only been noted as a result of very high acute exposures which would never be encountered in ambient air.

Epidemiological researchers have attempted to link nitrogen dioxide exposures to various health parameters. Such studies have major flaws, including questionable measurements of nitrogen dioxide concentrations and the presence of multiple air pollutants. Therefore, no effect as yet can be definitively ascribed to nitrogen dioxide at ambient levels.[7].

2.3 Volatile Organic Compounds

As described in Chapter 1, volatile organic compounds (VOCs) are a general class of compounds found in ambient air as a result of highway–related activities. They are a mixture of compounds making up the lower boiling fractions of fuels and lubricants, as well as partially combusted fuels. These compounds are emitted into the atmosphere during refuelling, through leaks in the crankcase, and from the tailpipe.

VOCs are complex mixtures consisting of aliphatics, olefins, aldehydes, ketones and aromatics. Many of these compounds are recognized as being potentially hazardous to human health. For the most part, they are unlikely to be present in sufficiently high concentrations in ambient air to cause acute effects. Because these compounds contribute to photochemical smog, they may have indirect effects on health. These effects will be discussed later.

From a toxicological standpoint, benzene is probably the most important of the VOCs. Literature reports on the toxicological properties of benzene date from the 1800's.[8,9]. Prolonged elevated exposures to benzene, either through the respiratory tract or cutaneous contact, can result in aplastic anaemia or acute myelogenous leukaemia.[10]. Bone marrow is the target organ for benzene. As the marrow becomes increasingly affected, there is a decrease in circulating erythrocytes, platelets and leukocytes. In the most severe stages of benzene poisoning, all three cell types are depressed. This condition is called pancytopaenia. When there is no longer any functional marrow , the condition is called aplastic anaemia.[10]. Benzene interferes with both DNA and RNA synthesis in bone marrow cells. In addition, chromosomal aberrations have been noted as a result of benzene exposure.[11].

Leukaemia is another blood disorder associated with benzene. Epidemiological studies have revealed that the rate of acute myelogenous leukaemia is elevated among benzene–exposed workers. A risk–assessment for benzene–related leukaemia, carried out in the U.S. for the Occupational Safety and Health Administration [12], showed there may be a significant risk for workers exposed for 40 years.

Aromatic fractions with high benzene content have been added to petrol to increase octane in recent years. In general, the benzene content of fuels in the European Community is somewhat higher than those in the U.S. The total benzene emissions increase incrementally with both the benzene and total aromatic content of the fuel.[13]. For each 1% volume increase in benzene, the total emission rate of benzene increases by about 4 mg/mile. About 70% of the emissions are from the tailpipe, 20% are evaporative and 10% are from refuelling. Increasing the amount of non–benzene aromatics in the fuel also results in an increase in tailpipe emissions of benzene.[13].

A major fraction of VOCs consists of aliphatic hydrocarbons. Inhalation of these vapours may be harmful because, at high concentrations, they depress the central nervous system causing dizziness and incoordination. Lower molecular weight compounds are the most important of these compounds because of their higher vapour pressure and hence higher ambient concentrations. Of special interest are hexane and 2–hexanone (methyl n–butyl ketone). These compounds have very low acute toxicities. However, prolonged exposure may result in peripheral neuropathies characterized by sensory loss and motor weakness in the hands, arms, legs and feet.[10]. These two compounds have a common metabolite, 2,5–hexanedione, which produces identical toxic effects in experimental animals as the parent compounds.[14].

Little is known of the toxicological consequences of exposure to these hydrocarbons at the very low levels expected as a result of vehicular traffic. Most individual hydrocarbons would be below measurable levels in the ambient atmosphere except in special circumstances. It is generally accepted that exposures to hydrocarbons at these low levels have little or no effect on human health. Because of the major role they play in photochemical smog, they are important in the overall health–effect picture.

Alcohols, particularly methanol and ethanol, have recently been used as fuel additives. In general, they are thought to be "clean burn" fuels that result in a decrease in overall emissions. However, they are relatively volatile compounds and will contribute to the overall VOC load. Methanol fueled vehicles tend to emit formaldehyde as well as methanol.[15]. Formaldehyde is recognized as a probable human carcinogen.[16]. Formaldehyde has also been implicated as a key initiator of photochemical smog.[17].

3. PHOTOCHEMICAL SMOG

There are two types of smog. The first results from incomplete combustion of coal which produces sulfur dioxide and smoke. In cool weather the sulphur dioxide and smoke combine with fog to form "smog." This condition has been known for a long time, Dicken's "London Particular", (Chapter 1 and [18]). In the early 1940s, a second type of pollution was noted in the Los Angeles area. Automobile exhausts were the source of the oxidative pollutants which led to this condition, called photochemical smog. Complaints of eye irritation have been associated with this smog which results from a complex series of reactions involving nitrogen oxides, VOCs, oxygen and sunlight, (Chapter 1 and [17–20]). These reactions lead to the formation of ozone, aldehydes, peroxyacetyl nitrate (PAN) and other compounds that may adversely affect human health. The chemistry of photochemical smog production is described in Chapter 1.

Ozone is the substance most likely to reach hazardous concentrations during smog episodes. The health effects of ozone have been extensively studied, both in experimental animals and in humans. Stockinger and Coffin, after reviewing acute toxicity studies, reported that exposures of about 6 ppm for 4 hours resulted in a 50% mortality rate in rats and mice.[21]. Damage to pulmonary structures in small animals has been reported after exposures as low as 1 ppm.[22]. Pulmonary oedema, swelling of lung tissues and haemorrhage were noted. Stockinger[23] and a number of other researchers have reported that experimental animals exposed to low concentrations develop an immunity against the toxic effects of ozone. This immunity allowed experimental animals to withstand exposures to levels lethal to animals not previously exposed. The immunity lasted for a month or more.

Chronic exposures to ozone below 1 ppm have been shown to cause a variety of changes in lung structure, biochemistry and function in a number of species. Last et al.[24] found a significant increase in the rate of collagen synthesis in lungs of rats exposed to 0.96 ppm O_3. There was a non–significant increase in the rate of collagen synthesis in lungs of rats exposed at 0.64 ppm. Wright et al.[25] were unable to show an increase in collagen synthesis in lungs of rats chronically exposed to ozone at 0.50 ppm or less. An increase in collagen formation could be important because a certain type of collagen is involved in pulmonary fibrosis. Reiser et al.[26] reported an increase in synthesis of collagen characteristic of fibrotic material in lungs of monkeys exposed 8 hours a day for one year to 0.61 ppm ozone.

Gross and White noted changes in several lung function measurements (inspiratory capacity, forced vital capacity, functional residual capacity) in rats after a 4–week exposure period at 0.7 ppm. [27]. The same researchers found no change in inspiratory capacity and forced vital capacity in rats throughout 52 weeks of exposure at 0.5 ppm.[28]. The changes in lung function appeared to be at least partially reversible in these studies. The microscopic morphologic changes reported were also reversible.

Human volunteers have also been used to determine the effects of ozone. Though these studies involve relatively short–term exposures at low concentrations, several conclusions can be drawn.

- The adaptation to ozone exposures seen in animal studies appeared to be present in humans [29,30].
- Asthmatics did not suffer more severe effects from ozone exposure than did other individuals, with or without light exercise [31,32,33].
- There was little response in lung function upon exposure to 0.16, 0.14, 0.12, 0.10, 0.08 ppm ozone for 2 hours other than irritation at 0.16 ppm [34].
- There was irritation at 0.12 ppm with high exercise levels [35].
- The effect at high exercise levels was a product of ozone concentration, ventilation rate, and exposure time [36].

In some cities, photochemical smog episodes may result in ozone concentrations similar to those known to produce biochemical or lung function changes in humans. Because photochemical smog is dependent on sunlight, these episodes are intermittent and relatively short–term. The long–term health effects of these episodes are relatively unknown.

4. PARTICULATE EMISSIONS

4.1 Lead

The antiknock properties of tetraethyl lead (TEL) were discovered in 1921. Within two years it was used in commercial petrol. The use of TEL made possible the early development of high compression, high powered internal combustion engines. Environmental concerns, particularly in the more technologically advanced countries, have resulted in a decrease in the amount of lead used in petrol in recent years. In the U.S., reduction of lead in petrol usage has resulted from two major actions: (1) the requirement for lead–free petrol needed by catalyst equipped cars (catalysts were introduced to control gaseous emissions), and (2) the phasedown of allowable lead content in leaded petrol.

The toxic effects of lead at high concentrations have been known since antiquity. However, there is much disagreement over whether low exposures have demonstrable health effects in humans. At high concentrations lead causes neurotoxic, renal, and reproductive effects. Some researchers have postulated subtle effects at low exposure levels. These effects include decrements in intelligence, hyperactivity, hypertension, auditory deficiencies and reduced nerve conduction velocity. Many researchers are unable to detect these changes if confounding variables are taken into account.

The direct effect of lowering petrol lead on human health is the subject of a great deal of controversy. Lowering lead exposures by any route will result in a decreased body burden, measured as blood lead levels. Petrol lead contributes to airborne lead levels. Therefore, lowering the amount of lead in petrol should cause a decrease in blood lead in the exposed population. Indeed, Annest et al. attributed the decrease in blood lead levels seen in the U.S. during the second National Health and Nutrition Evaluation Survey (NHANES II) solely to the decrease in petrol lead usage [37]. Their conclusions have been questioned because of methodological weaknesses and because they failed to take into account that other exposure sources (i.e. water, food, paint) were declining during the study period [38]. Declines in blood lead levels have been reported in other countries in the absence of a decrease in the usage of petrol lead during a similar time frame [38–43]. In addition, all individuals residing near major motorways would be expected to have blood lead concentrations well above those of the general population if petrol lead is a major contributor. This is not the case [44].

An opportunity for measuring the impact on blood lead by lowering the allowable level of lead in petrol became available in the U.K. In 1983, regulations lowering the allowable level of lead in petrol from 0.4 to 0.15 grams per litre were promulgated. The regulations were to take effect on January, 1986. The Department of Environment in the U.K. carried out a study to measure blood lead levels before and after the decrease. Blood lead levels were lower in 1986 as compared to 1985. However, the magnitude of this decline was about the same as that found in the 1984 to 1985 comparison, before the decrease in lead in petrol usage. The researchers concluded: "In the U.K., exposure to lead from a number of sources is being reduced simultaneously; blood lead concentrations probably fell in both 1985 and in 1986 for reasons other than the reduction in the lead content of petrol."[45].

Based on the best available evidence, it appears that petrol lead in most cases contributes, at most, a few μg Pb dl^{-1} of blood. This is confirmed by looking at studies of the relationship between air lead and blood lead. Snee et al. reviewed 10 such studies [46]. The blood lead—air lead slope ranged from 0.5 to 3.0 μg Pb dl^{-1} per μg Pb m^{-3}. They concluded that the single best estimate was 1.2 μg Pb dl^{-1} per μg Pb m^{-3}. This means that for every increase in airborne concentration of 1.0 μg Pb m^{-3}, blood lead concentrations should increase by about 1.2 μg Pb dl^{-3}. Few cities have airborne lead levels greater than 1 μg m^{-3} in the absence of point sources such as smelters or secondary lead plants [47]. Even if 90% of the airborne lead is associated with lead in petrol,[48] the total contribution to blood lead could not be more than about 1–2 μg Pb dl^{-1}.

It has been postulated that children are more susceptible to effects of lead toxicity because of their rapid development. Relatively low lead exposures have been implicated as causing, among other things, a decrement in cognitive skills [49–54] and hyperactivity [55–57] in children. Others have reported, after reviewing the same or similar data, that these effects are not evident if confounding factors are properly accounted for. If the effects are present, they are extremely small unless blood lead levels are highly elevated [58–64].

Another controversial issue is whether lead at low exposure levels causes hypertension. Pirkle et al. reported an increase in both the diastolic and systolic measurements in men aged 40 [59–65]. These conclusions were based on data from the Second National Health and Nutrition Examination Survey (NHANES II) in the U.S. Others, using the same data, could not confirm this relationship [66,67]. Results of studies on two separate police populations suggest that there may be a relationship between elevated systolic levels and lead [68,69]. On the other hand, Pocock et al. found no association between blood pressure and blood lead levels in a large cohort in 24 towns in Britain [70]. Cramer and Dahlberg reported there was no difference in occurrence of hypertension between workers with occupational exposure to lead and those without occupational exposures [71]. Rogan et al. reported they found no association between blood lead and blood pressure in children [72]. Urinary lead levels, another measure of body burden of lead, was also shown not to be associated with blood pressure variability [73]. An international symposium was held in 1987 to examine the issue of a possible relationship between lead and blood pressure. The data presented indicated that if there is an association between lead and blood pressure, it is extremely weak.

4.2 Diesel Emissions

Emissions of diesel engines are similar to those of petrol–powered, spark–fired engines except for an increase in particulate emissions of several orders of magnitude [74,75]. The risk associated with the increased particulate emissions is disputed. There have been concerns that these emissions may have carcinogenic potential because extracts have been shown to have mutagenic activity [76]. Mauderly et al. reported that high exposures to diesel particulates resulted in lung inflammation, accumulation of soot and chronic lung disease in rats [77]. They also reported an increase in lung tumors at high concentrations but not at low concentrations. In a review of the literature up to 1986 on the carcinogenic potential of diesel emissions, Steenland found, of four animal studies reported, two were negative and two were positive. Positive results were obtained only at very high exposure levels [78]. He also reported that the epidemiological studies reviewed gave equivocal results. Negative studies often lacked a sufficient latency time and positive results often failed to correct for smoking and other confounding factors. Hall and Wynder concluded there was no association between exposure to diesel exhaust and lung cancer [79]. Howe et al. found a slight increase in risk among Canadian railway workers exposed to diesel emissions [80]. However, they were unable to rule out smoking as the cause of the association. Schenker et al. found no significant increase for any specific neoplasm among U.S. railway workers [81]. The same group reported an increase in lung cancer in a larger cohort in a follow–up study [82]. McClellan et al [83] concluded that:

> "Epidemiologic data and laboratory studies appear to indicate that the human lung cancer risk from exposure to diesel exhaust would be quite low, even if use of diesel vehicles increased substantially."

McClellan's group estimated that even if the light–duty vehicle population in the U.S. became 20% diesel, the increase in lung cancer from exposure to diesel emissions would be less than 200 per year [84]. An increase of zero could not be ruled out based on available evidence. This compares with an estimated 100,000 cases per year among smokers and

10,000 cases per year among non–smokers in the U.S. presently. In support of this view, airborne particulates outside a New York City bus terminal had greater mutagenic activity than particulates inside the terminal even though there was an increase in airborne fine particulates, apparently from diesel emissions, inside the terminal [85].

The risk of noncarcinogenic effects at expected ambient levels is also small. Ames et al. reported they found no difference in lung function and other chronic respiratory effects in coal miners who worked in dieselized mines as compared to those who worked in non–dieselized mines [86]. Other researchers reported there was no obstructive lung disease after exposure for two years to diesel exhaust in rats or monkeys [87].

In summary, potential for health effects from diesel exhaust appears to be about the same as from petrol powered engines.

4.3 Manganese

Methylcyclopentadienyl manganese tricarbonyl (MMT) is another metal containing antiknock additive. This compound has been used in the U.S. in leaded petrol since the early 1970s. It has been used to replace octane numbers lost during the phasedown in allowable lead concentrations. It has been used in Canada in both leaded and unleaded petrol since 1978. The concentration of manganese in petrol to which MMT has been added is very low. The levels of airborne manganese have not risen measurably in the U.S. or in Ontario, Canada as a result of MMT usage [89]. In the U.S., airborne manganese levels decreased by approximately 50% between 1971 and 1982. These reductions occurred as a result of greater controls of particulates from point sources. Ter Haar et al. showed that at least 98.7% of the combusted manganese is inorganic [90]. They also demonstrated that MMT was photochemically unstable, decomposing rapidly in sunlight to give inorganic manganese products. The authors concluded that organic manganese from MMT usage would not normally be present at detectable levels in ambient air. This was confirmed by Coe et al. who found that MMT was below the detection limit of 0.05 ng/m^3 of air along Toronto streets [91]. Using lead as a surrogate model and the relative concentrations of lead and manganese in petrol, Ter Haar et al. calculated that auto exhaust would raise air levels of manganese only slightly, even in urban areas [90]. These calculations have been confirmed by actual air measurement data [88,89].

Chronic exposure to high levels of manganese in occupational settings have resulted in manganism. This disease is characterized by psychotic behaviour with hallucinations, delusions and compulsions [92]. Other neurological signs and symptoms include an expressionless face, sudden laughter or crying, tremors, and staggering or stumbling. A condition resembling Parkinsonism and eventually death may occur in severe cases.

On the other hand, manganese is an essential element. It is required in the diet and is present in virtually all food. The average adult in the U.S. ingests approximately 2,000–9,000 μg Mn/day [88] in food and water. The level needed to cause the neurological toxic effects noted above is many orders of magnitude higher than the average dietary intake.

Two reviews of health issues related to MMT use in petrol have been published. Cooper [93] concluded:

> "There is thus a wide margin of safety between the intakes of Mn essential to health and the high concentrations that have been associated with toxic effects. The small amounts of manganese added to the environment by the combustion of MMT used as a fuel additive would be comparable to the normal background and should not create health problems."

Abbott [94] came to similar conclusions:

> "The small increase in airborne manganese from the use of MMT in petrol is 3–4 orders of magnitude lower than the level required to produce toxic symptoms of manganese exposure, even in areas of high traffic density, and no health risk from the use of MMT is likely."

As the use of lead antiknocks decreases worldwide, it is likely that MMT usage will increase to help make up for lost octane numbers. There should be no adverse health effects from this action.

CONCLUSIONS

The concentrations of most highway related pollutants in ambient air normally remain well below the threshold level for manifest health effects. A number of these pollutants, however, may cause more subtle health effects, i.e. subclinical effects. Subclinical effects may only become apparent after years of exposure. Many of the adverse subclinical health effects postulated to be caused by highway related pollutants occur at a significant rate in the general population and have multiple causes. This makes it extremely difficult to show that a specific pollutant is involved. For instance, there is the possibility that low–level carbon monoxide exposures contribute to cardiovascular disease. Cardiovascular disease is found in a high percentage of the adult population and many factors contribute to the development of cardiovascular disease (genetics, diet, life–style). It is, therefore, unlikely that epidemiological studies will be useful in proving whether low level carbon monoxide exposures are a contributing factor. Experiments on animal models may be the only way to investigate this type of possible effect. Even these types of studies are not completely satisfactory because of uncertainties in extrapolating results to humans. It is also somewhat difficult to interpret the meaning of changes seen at the molecular or cellular level and relate them to whole body effects.

A number of highway related pollutants have sources other than automobiles. Carbon monoxide is again a good example. Numerous industrial processes result in carbon monoxide release into the ambient environment. Residential usage of gas–fired stoves and water heaters and burning of coal or wood also contributes to the total carbon monoxide load in ambient air. Even if future research shows that low level carbon monoxide exposures have adverse health effects, the contribution from the transportation sector will be difficult to determine.

There are many research projects presently being carried out to elucidate what adverse health effects may be caused by highway related pollutants. With the possible exception of photochemical smog, especially ozone, most of these pollutants remain at low levels. Even ascribing specific health effects to ozone is subject to dispute. Clarification of health effects of specific pollutants will be a slow process and will no doubt continue to cause a great deal of controversy.

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Chapter 8

EFFECTS OF HIGHWAY POLLUTANTS UPON TERRESTRIAL ECOSYSTEMS

P.F. Scanlon

Contents **Page**

1. GENERAL EFFECTS OF HIGHWAYS ON ANIMALS LIVING IN ROADSIDE RIGHTS–OF–WAY

1.1. General Considerations

Increases in human populations have meant great increases in needs for transportation of people and goods. To facilitate such transportation, numbers and size of roadways have been increased. Traffic volumes have increased in line with transportation needs. (see Figure 1.2 in Chapter 1). Accordingly, lands in the vicinity of roads and the animals species associated with these lands are influenced by the activities and by–products of transportation vehicles. Wild animals living in roadside habitats are subjected to a variety of risks from vehicles. Collisions with vehicles inevitably occur. They are at risk from contamination and sublethal poisoning by exhaust by–products of gasoline and diesel combustion. Products of combustion of lubrication oils, materials which wear from tyres, brake linings, engine moving parts, and other vehicle body parts can cause contamination. Materials spilled from vehicles, especially following accidents can damage habitats and animals.

1.2 The Extent of Highways

Some sense of the potential influence of roads and highways on wildlife habitats may be gauged from the extent of roads and highways in the United States. Total mileage of roads in the United States in 1982 was 6222000 km [1]. Of this, 62500 km are in the federal interstate system, and have controlled access with central divisions. All roads have rights–of–way with lands which are occupied by some fauna. The rights–of–way are typically distinguished by being managed by different means than the surrounding lands which may be urban, suburban, or rural. They are long and relatively narrow and, particularly with the interstate system, can contain very extensive lands due to the use of wide rights–of–way and the frequent inclusion of significant amounts of land in the central medians. Some sense of the amount of potential wildlife habitat within interstate highway rights–of–way may be gleaned from the following. A minimum right–of–way width for interstate highways is 33.6 m. Total pavement and shoulder widths for a 4–lane highway within this right–of–way is 23 m. Accordingly, at a minimum, 32.7% of the land in the right–of–way is unpaved and can produce vegetation and support animal life. While land in median strips typically is less supportive of wild species than lands on the outer edges, it is readily apparent that the land associated with the 62500 km of interstate highways is particularly extensive and represents a tremendous quantity of wildlife habitat. While the allocation of lands to highway development in the United States likely is more generous than in other regions of the world, consideration of the land areas involved in rights–of–way is illustrative of the extent of wildlife habitat influenced by roads and highways. It is also worth considering that animals living in highway rights–of–way are models for pollution, particularly as pollution may influence humans living in the immediate vicinity of roads.

1.3 Highway Construction Effects

The construction of roads can have impacts on animals living in the immediate environments. Road construction influences are clearly manifest through exposure of subsoils or their use in filling roadbeds and roadsides. Road building materials, particularly hydrocarbon paving materials, impact the immediate environments. Altered drainage patterns effect soil moisture, vegetation growth, and microhabitats of animals. Compaction by construction vehicles effects soil conditions for both plants and animals species.

1.4 Management of Road Surfaces

Roads and roadside rights–of–way are subjected to a wide range of management practices which may have direct or indirect effects on animal dwellers in the rights–of–way. Roads must be managed to minimize the effects of adverse weather on the safety of vehicular traffic. Snow, ice, rainfall, and wind are all factors which require management and which may impact animals and their habitats depending on the management actions taken. Vegetation may be managed to minimize adverse effects of wind and/or blowing snow and this in turn can influence the desirability of the rights–of–way to birds including predatory birds. Use of chemicals, principally salt, for ice removal from roads can result in saline wet areas which are attractive to some wild species. Fraser [2] and Fraser and Thomas [3] have documented that salt used for snow and ice removal attracts moose (Alces, alces), black bears (Ursus americana), and white–tailed deer (Odocoileus virginianus) to roadsides, especially in spring. In particular, roadside ponds made saline by road salt are particularly attractive to moose and are frequently the points where moose–vehicle collisions occur in Ontario. Water removal can influence soil moisture patterns in the vicinity of roads thereby influencing vegetation, earthworm populations, and the variety of small mammals utilizing the roadside environments. Management of road surfaces includes resurfacing, grading of unpaved roads, and chemical treatment of unpaved roads for dust suppression. The hydrocarbons associated with repaving operations can influence roadside habitats. Grading of roads has minimal effects unless ditches are graded, thereby greatly disturbing roadside habitats. The use of chemicals to suppress dust may be relatively unimportant unless oil, and especially waste engine lubricant engine lubricant oil, is spread on road surfaces. There are very high concentrations of lead in waste crankcase oil [4, 5]. Use of such oil for dust control on unpaved highways could have significant impacts on animals in the rights–of–way.

1.5 Management of Rights–of–Way Vegetation

Management of rights–of–way vegetation is done for a variety of reasons including safety, aesthetics, and fire control. Removal of vegetation, be it grass, shrubs, or trees, necessarily influences habitats of wild species. When herbicides are used to reduce vegetation, seed sources used by mammals and birds may be contaminated or made unavailable to these species. Repellent or poisonous chemicals, such as Endrin, have been used to lessen girdling of roadside shrubbery by small rodents. The compounds frequently were quite toxic and persistent and had potential for major adverse effects on rodent populations and predators which used them.

1.6 Types of Animals in Rights–of–Way

In large part, the size and mobility of animals dictate the extent of dependence of animals on rights–of–way habitats. The mobility of birds generally indicates minimal dependence on roadside habitats with certain exceptions. The maintenance of territories during breeding seasons may ensure relatively long residence in rights–of–way. The availability of cover, especially for ground–nesting birds, also may involve substantial length of residence by some species in rights–of–way. Readily available food supplies may be a major attractant keeping birds in rights–of–way. For example, high populations of small rodents frequently attract raptor species to roadsides.

Those animals which are most likely to be permanent residents of rights–of–way are a wide range of small mammals, species which have small home ranges, are fossorial (i.e., burrowing) in habits, or are willing to restrict their activities to the vicinity of roadways in response to attractive conditions in the rights–of–way. Seasonal residents are likely to be birds using vegetation as nesting cover, or predatory birds using the small mammal residents as food bases. For example, many Buteo hawk species frequent highway rights–of-way during the winter in the United States following their southernly migrations. In urban, and indeed some rural environments, pigeons (Columbia livia) utilize bridges as roosting and nesting spaces and essentially are permanent residents of rights–of–way and indeed have been studied as monitors of lead pollution [6–10].

1.7 Roadside Vegetation and Animal Associations

The nature of vegetation at roadsides contributes to whether these areas are attractive or not to wild species. Also, the type of vegetation influences the total fauna attracted to rights–of–way. Management of vegetation may consciously influence the animal associations from the standpoint of decreasing the attractiveness to large animals. By and large management decisions concerning vegetation relate to aesthetics, fire–hazards, safety considerations, and what are essentially farming type operations such as harvesting hay. These decisions usually are made without reference to the fauna and may have inadvertent effects on the majority of animals inhabiting rights–of–way. However, many states in the United States such as prairie regions, consider highway rights–of–way to be significant nesting areas for duck species, pheasants, and for rabbits as the grass ground cover provides good nesting habitat [11–15]. In several states, mowing of grass in rights–of–way is delayed to accommodate the relatively long nesting period of ducks and pheasants. On the other hand, when significant droughts occur as in 1987 and 1988, farmers may be allowed to mow hay in rights–of–way in spite of needs of ground nesting species.

Rights–of–way vegetation frequently provide an edge–effect with vegetation in the areas outside the right–of–way, thereby enhancing the attractiveness to animal species. Some simple examples of vegetation cover – animal associations are: dense grass and forbs favour small mammals that are herbivorous, granivorous and insectivorous; large trees adjacent to grassy covered areas provide perches for predators of small mammals; fences and overhanging trees provide perches for insectivorous birds; low woody vegetation provides nesting cover for a wide range of birds and any thick ground cover favours ground–nesting birds. Appropriate reptile predators may exploit all of these habitats and prey on the inhabitants. In short, any given type of ground cover will attract its appropriate faunal association and the richness of species attracted will be further enhanced if edge effects are created between rights–of–way vegetation and contrasting vegetation beyond the rights–of–way. Moreover, the ecosystems associated with rights–of–way may be somewhat unique within the surrounding areas.

1.8 Roads as Barriers to Animal Movements

Roads, especially busy ones, appear to provide significant barriers to movement of small mammals such as mice, voles and shrews [6–18]. Adams and Geis [18] speculate that roadways are barriers to natural gene flow in small mammal populations. The inability to cross roads would tend to compromise the shape of home range area of individuals of the species, and consequently, would increase their exposure to roadside contamination. Highways may be a relatively absolute barrier for small mammal species provided traffic volume is high enough. With lower traffic volumes probably all animals cross roads and medium and larger mammals cross roads regardless of traffic volumes. Many animals are killed crossing roads. Adams and Geis [18] document frequency of mammal, bird, and reptile mortality in relation to traffic volume, and review pertinent literature. From the standpoint of contaminants, a question exists as to whether exposure to contaminants alters behaviour to an extent which increases the probability of accidental mortality.

2. CONTAMINATION OF ROADSIDE ECOSYSTEMS BY HIGHWAY POLLUTANTS

2.1 The Contaminants

The highway pollutants most frequently studied in roadside ecosystems have been the heavy metals lead, cadmium, nickel, and zinc. The effects of salt used for deicing highways on roadside ecosystems have been studied to a limited extent. Rather little, if any, emphasis seems to have been placed on the effects of by–products of fuel combustion on terrestrial ecosystems. A large body of literature exists on effects of crude oils and refined oils on vertebrates – particularly waterfowl and sea birds. These types of oils typically are not polluters of roadside ecosystems, except as accidental spills and the literature on effects of crude or "clean" oils will not be reviewed here. Used crankcase oil typically is heavily contaminated with combustion by–products and also with lead. It has been used in the past as a dust suppression material on unpaved roads and a limited literature exists on effects of used oil on birds. Reports of the effects of materials worn from vehicles such as brake linings, worn parts, and trash lost from vehicles tends to be anecdotal. Wear of tyres contributes to contamination, but studies of this contribution centre mostly around the contribution to zinc and cadmium contamination.

2.2 Effects of Heavy Metals

Gasoline treated with alkyl lead anti–knock compounds results in lead contaminated exhaust. The forms of lead in exhaust are inorganic lead halides because of the use of halogenated scavenger compounds in gasoline. Lead is not an essential element for animals. Excess lead in animals accumulates in bones, kidneys, livers, and brains. Lead can bind calcium sites in the body. It causes reductions in enzymes concerned with synthesis of the heme component of red blood cells. This can result in anaemia. Other effects include destruction of the central nervous system, impaired energy metabolism, decreased fertility, foetal growth impairment, kidney damage, and impaired learning ability. Cadmium is also a non–essential element. It is believed to occur in roadside ecosystems as a result of release from tyre–wear. The adverse effects of cadmium includes growth retardation, anaemia, weight loss, destruction of kidney tissues, and destruction of esticular tissue. The toxic effects of cadmium are similar to those of zinc deficiency. Cadmium can interfere with metabolism of copper and zinc. Of special interest is the effect of cadmium of increasing hypertension in animals as this effect may lead to changes in normal behavior of species contaminated with cadmium.

Nickel is a component of fuels and lubricating oils. It is not fully regarded as an essential element in animal nutrition, but may be essential for some species. Adverse effects of excess nickel are influences on liver and muscle glycogen metabolism, on growth rates, and on reproduction.

Zinc is an essential nutrient. Excess zinc interferes with gastrointestinal tract function, with liver enzyme function, and with bone metabolism. It interferes with copper metabolism and causes anaemia.

2.3 General Considerations of Metals in Roadside Ecosystems

Consider lead as the prime example. Lead is introduced into the roadside ecosystem primarily as a component of exhaust emissions. It settles on soils and plants within the ecosystem though this settling may be greatly complicated by air disturbance due to traffic, as described in Chapter 3. Lead settling on soil varies as the vegetative cover varies which in turn is influenced by seasonal effects on plant growth stages. Dying parts of plants contribute to soil lead. The incorporation of lead into soil below the immediate surface is accomplished by being carried downwards by drainage water, by the actions of invertebrates in bringing dead plant material below the surface and by the soil mixing effects of burrowing animals. Evidence exists that lead is bound by organic matter in soil and tends to be retained in the upper soil profile.

The animal component of ecosystems is exposed to lead as an aerosol, as a contaminant of vegetative foods, as a contaminant of their physical surroundings, and as a contaminant of the animal components of foods eaten by omnivorous, insectivorous, and carnivorous species. Lead excreted by animals and lead released by death and decay of animal bodies is returned to the system which continues to be replenished by vehicular exhaust. Lead is recycled within the system by uptake by plants from soil, by earthworms consuming humus and then being victims of predation, by animals being physically contaminated by burrowing in soil, and by the normal patterns of consumption of components of the ecosystem by animals within the system. The primary means of export of lead out of the roadside ecosystem is through surface water, ground water, consumption of plants and animals by animals outside the ecosystem, and by animals leaving the system.

Impact to the system is governed by traffic volume and the amount of lead in gasoline and age of vehicle and engine load [19–21]. Within the roadside ecosystem there is a gradient of lead which is governed by distance from the highway (Chapter 3 and [19]) and within the plant component of the ecosystem the lead content varies with season, more probably with growth stages of the plants [19].

Essentially similar comments can be made concerning the behaviour of cadmium, nickel, zinc, and copper in the system with one caveat. Much of each of these metals is inserted into the ecosystem by wear of parts, especially tyres, rather than as components of exhaust emissions. Their distribution within the ecosystem will tend to be relatively higher at the edge of roadsides than will be that of lead and relatively lower further from the edge of the roadside relative to the lead distribution [19].

2.4 Heavy Metals in Soil, Vegetation, and Earthworms

A substantial body of literature now exists on the extent of heavy metal, particularly lead, contamination of roadside soils [19, 22–45]. Smith [46] and Hiller [47] have provided excellent reviews of the dynamics of heavy metals in soils, plants and earthworms prior to the late 1970s. Up to that time much of the literature was concerned with identifying the types of metals, polluting roadside ecosystems (Pb, Cd, Zn, Ni, Fe), with attribution to source (vehicular traffic) and measurement of accumulation with less than full consideration of the possible effects of traffic volume, distance from highways, depth of soil, soil organic matter, or effects of season on concentrations of contaminants on vegetation.
Since those reviews [46, 47] the addition of new data and commentary to the literature [48–51] allow a reasonable summation of the effects of heavy metal contamination on soil, vegetation and earthworm components of roadside ecosystems. (Tables 8. 1–3, 5–6, 8–11) Lead, cadmium, nickel, and zinc are all contributed to roadside ecosystems in relation to traffic volume. Traffic volumes in excess of 25,000 vehicles per day cause accumulations of lead, cadmium, nickel, and zinc worthy of concern about the well–being of higher animals inhabiting roadside ecosystems. Soil organic matter plays an important role in returning metals in the upper soil horizons. Earthworms as consumers of soil organic matter may be particularly badly contaminated and allow ready contamination of birds and mammals which prey on them. Distance from highway edges affects the degree of contamination of soil and vegetation but the effect varies with element. Those elements deposited by exhaust are more widely distributed than those generated by wear of parts which are skewed towards high concentrations near the road edges. Significant contamination exists up to 50m from the highway edge for all elements for traffic volumes of 25,000 vehicles per day and upwards.

The effects of season on concentrations of polluting metals on plants is very substantial. Relatively dormant plant materials tend to have higher concentrations of metals than rapidly growing samples. There is probably an overemphasis on trying to interpret results of metal contamination in terms of biomagnification. Evidence for biomagnification is relatively meager, however, the more relevant facts are the concentrations of elements available to the higher animals in the polluted ecosystems.

The impact of lead removal from gasoline on future concentrations of lead in roadside soils is a factor for speculation. Byrd et al. [50] provide evidence for a decrease in soil lead since mandatory use of unleaded gasoline with the decreasing lead being removed slowly by surface water. Over the long term, lead removal from soil will be a relatively slow process, and lead will be continually recycled by earthworms and by fossorial mammals.

2.5 Heavy Metals in Other Invertebrates in Roadside Ecosystems

What work exists is largely confined to insects and arachnids.(Tables 8. 3, 6, 9 and 11). There is evidence that heavy metals, particularly lead, increase in insects in roadside environments [52–57]. The preponderance of evidence is that lead concentration tends to be quite low in insect caterpillers and imagos despite close proximity to highways of high traffic volumes. Woodlice and millipedes may accumulate high lead concentrations. Other metal concentrations have not been reported for invertebrates for roadside settings. Elevated lead concentrations in insects at roadsides may be inferred from effects on insect feeding animals living in roadway environments. Higher lead concentrations in bats and bat guano were noted near highways [58] in barn swallows [59], and in bullfrogs [60] though the frogs may reflect further contamination from the drainage ditches they occupied.

2.6 Heavy Metals in Small Mammals in Roadside Habitats

A considerable body of literature (Tables 8. 4, 7, 9 and 11) on the accumulation of heavy metals (Pb, Cd, Zn, Ni) by small mammals in roadside ecosystems is available [19, 44, 56, 58, 61–64]. Blair [65] provides a comprehensive review. Clear patterns emerge from the data sets. Concentrations of all polluting metals in small mammals increase as traffic volumes increase. Claims that nearness to roadsides influence metal concentrations should be interpreted carefully as small mammals are quite mobile. Insectivores accumulate higher concentrations of lead, zinc, and cadmium than do herbivores, which in turn tend to have concentrations exceeding those of seed eating mammals. Insectivores which also consume earthworms, such as shrews tend to have similar concentrations of lead as bats from the same areas. The bat species concerned are totally insectivorous. Concentrations in earthworms greatly exceed those in insects, but differences in longevity in favour of bats may influence concentrations seen in bats. It should be noted that concentrations of heavy metals found in mammals in roadside ecosystems, while of serious proportions, may be considerably below those found in mammals at smelter sites. Most studies to date have only addressed concentrations in individuals. Little consideration has been given to effects on populations of small mammals.

2.7 Heavy Metals in Birds in Roadside Ecosystems

Because of their mobility, it is usually difficult to ascribe contamination in birds to their relatively tenuous associations with roadside ecosystems. Nevertheless, some interesting patterns have been noted. Lead increased in barn swallows nesting near highways [59]. Swallows are insectivorous. Ground feeding birds had greatly increased lead concentrations when feeding near roads or in urban areas [6–10, 66] though one report did not fit the pattern of higher lead in ground feeders [67]. It should be noted that birds use grit in their gizzards and many species select grit from roadsides increasing possibilities of lead contamination.

One group of birds which have received no study are those species which prey on mammals in rights–of–way. As noted earlier, large numbers of individual raptors seem to use roadsides as hunting grounds almost exclusively for several months during their winter migrations. The effects on them are unknown and because of their protected status, it will be very difficult to study any problems with them.

2.8 Possible Effects of Exhaust Gases and Particulates on Ecosystems

Little has been done to apply knowledge of toxic effects of exhaust gases and particulates to problems of ecosystems. Newman [68] in his recent review of air pollution and wildlife toxicology lists known sensitive species, some of which could be found in highway ecosystems. However, none of the literature he reviewed would allow conclusions to be drawn concerning risks to animals in rights–of–way.

2.9 Highway De–icing Compounds as Toxins

While salt is an essential nutrient for animals and salt attracts wildlife to roadways, it has been documented as a poison of both birds and mammals. Trainer and Karstad [69] reported deaths in rabbits, pheasants, quail, and a pigeon. The central nervous systems of the victims were adversely affected. Addition of ferrocyanide compounds to salt to prevent caking has led to speculation on the possible toxic effects on wild animals though no problems have been documented. Rust inhibitors added to salt which contain phosphorus could contribute to fertility of rights–of–way soil and so change vegetation. Alternate deicing compounds are probably too costly to substitute for salt. One possible substitute, ethylene glycol, should be considered with caution as it is known to be toxic to some mammal species.

2.10 Litter Contamination of Roadside Ecosystems

While no real research is apparent on litter effects on animals in roadside ecosystem, one important detrimental effect has been recently recorded. Disposal on roadsides of bottles have created a new hazard for small mammals, and especially shrew species. Animals which enter these bottles are unable to exit them and are trapped and die. Pagels and French [70] report as many as 570 bottles per kilometre of road and as many as 62 animals trapped per kilometre of road. It is not possible to assign a trapping period to these observations other than "since the bottle was thrown there".

3. FUTURE PROSPECTS AND RESEARCH NEEDS

Most contaminants presently being introduced into roadside ecosystems will probably continue to be a problem with one exception. The increasing use of lead free gasoline should have the effect of diminishing the input. Based on experiences of orchard soils contaminated with lead arsenate, lead is still a significant factor for some mammals 35 years after cessation of lead input to the system. Fossorial species still have considerable lead concentrations due to their close involvement with soil [71]. However, shrew species now have lower lead concentrations than fossorial herbivorous species from the same orchards [72].

The single most pressing need for future research will be on the effects of pollutants on populations of mammals rather than merely on individuals. Further research is needed to clarify whether in fact, biomagnification of metals in food chains in roadside ecosystems is indeed occurring. In addition to the commentary of Beyer [51], work with fishes of different trophic levels in streams influenced by highways indicated that biomagnification of metals may not be taking place [73–75]. Needs to assess the effects of multiple contaminants on mammals are clear. Roadsides contain a variety of contaminants and it is difficult to predict combined effects of several contaminants from concentrations of single contaminants. On pertinent example is the work of Yocum [76] which showed that numbers of young produced by mice fed cadmium and lead in combination were greatly reduced.

Table 8.1.

Lead concentrations in soils in relation to traffic volumes and distance from roads. (Depths of soil sampled and distance from the road pavement edge are given where appropriate. All concentrations are on a dry weight basis).

	ADT[a]	Depth (cm)	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Soil						
	64,180	0–6.25	700	1.5		24
	1,900	0–6.25	128	1.5		24
	56,000	0–5	122	7.6		27
	56,000	0–5	75	15		27
	56,000	0–5	63	30		27
	56,000	5–10	13	7.6		27
	56,000	10–15	15	7.6		27
	24,000	0–5	403	7.6		27
	24,000	5–10	252	7.6		27
	24,000	10–15	121	7.6		27
	control	0–10	(15–50)	N.S.[c]		30
	47,800 – 44,500	0–2.5	700	3	geometric mean	31
	47,800 – 44,500	0–2.5	204	6.1	geometric mean	31
	47,800 – 44,500	0–2.5	94	12.2	geometric mean	31
	47,800 – 44,500	0–2.5	60	24.4	geometric mean	31
	47,800 – 44,500	0–2.5	82	48.8	geometric mean	31
	25,100 – 500	0–2.5	313	3	geometric mean	31
	25,100 – 500	0–2.5	90	6.1	geometric mean	31
	25,100 – 500	0–2.5	54	12.2	geometric mean	31
	25,100 – 500	0–2.5	39	24.4	geometric mean	31
	25,100 – 500	0–2.5	35	48.8	geometric mean	31
	control	0–2.5	14	N.S.	geometric mean	31
	54,700 – 35,200	0–15	(47–266)	7.6		33
	54,700 – 35,200	0–15	(30–171)	22.7		33
	54,700 – 35,200	0–15	(66–109)	37.9		33
	54,700 – 35,200	0–15	(48–129)	53		33
	54,700 –					

Table 8.1 (continued) (1-2)

	ADT[a]	Depth (cm)	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Soil						
	35,200	0–15	(49–228)	68.2		33
	54,700 – 35,200	15–31	(42–178)	7.6		33
	54,700 – 35,200	15–31	(23–86)	22.7		33
	54,700 – 35,200	15–31	(18–48)	37.9		33
	54,700 – 35,200	15–31	(20–61)	53		33
	54,700 – 35,200	15–31	(26–229)	68.2		33
	19,800 – 12,800	0–15	(48–442)	7.6		33
	19,800 – 12,800	0–15	(39–95)	22.7		33
	19,800 – 12,800	0–15	(39–129)	37.9		33
	19,800 – 12,800	0–15	(30–332)	53		33
	19,800 – 12,800	0–15	(23–193)	68.2		33
	19,800 – 12,800	15–31	(26–616)	7.6		33
	19,800 – 12,800	15–31	(17–64)	22.7		33
	19,800 – 12,800	15–31	(24–86)	37.9		33
	19,800 – 12,800	15–31	(24–76)	53		33
	19,800 – 12,800	15–31	(27–247)	68.2		33
	1,200	0–0.5	160	10		36
	1,200	0–0.5	90	30		36
	1,200	0–0.5	55	100		36
	1,200	3	60	10		36
	1,200	6	50	10		36
	16,000	0–5	145	2		38
	16,000	0–5	30	125		38
	16,000	0–5	25	250		38
	16,000	12	60	2		38
	16,000	25	40	2		38
	3,700	0–5	130	2		38
	3,700	0–5	20	125		38

Table 8.1 (continued) ($_{1-3}$)

	ADT[a]	Depth (cm)	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Soil						
	3,700	0–5	20	250		38
	3,700	12	40	2		38
	3,700	25	30	2		38
	48,000	0–5	540	8		39
	48,000	0–5	220	16		39
	48,000	0–5	140	32		39
	48,000	5–10	300	8		39
	48,000	5–10	105	16		39
	48,000	5–10	60	32		39
	48,000	10–15	98	8		39
	48,000	10–15	60	16		39
	48,000	10–15	38	32		39
	23,000	0–5	150	8		39
	23,000	0–5	101	16		39
	23,000	0–5	55	32		39
	23,000	5–10	29	8		39
	23,000	5–10	14	16		39
	23,000	5–10	10	32		39
	23,000	10–15	11	8		39
	23,000	10–15	8	16		39
	23,000	10–15	6	32		39
	20,000	0–5	522	8		39
	20,000	0–5	378	16		39
	20,000	0–5	164	32		39
	20,000	5–10	460	8		39
	20,000	5–10	260	16		39
	20,000	5–10	108	32		39
	20,000	10–15	416	8		39
	20,000	10–15	104	16		39
	20,000	10–15	69	32		39
	7,500	0–5	242	8		39
	7,500	0–5	140	16		39
	7,500	0–5	61	32		39
	7,500	5–10	112	8		39
	7,500	5–10	104	16		39
	7,500	5–10	55	32		39
	7,500	10–15	95	8		39
	7,500	10–15	66	16		39
	7,500	10–15	60	32		39
	70,000	0–7.5	118	15		40
	70,000	0–7.5	76	76		40
	70,000	0–7.5	85	197		40
	70,000	0–7.5	81	364		40

Table 8.1 (continued) ($_{1\text{-}4}$)

	ADT[a]	Depth (cm)	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Soil						
	70,000	25–40	83	76		40
	70,000	>65	<1	76		40
	58,000	0–7.5	118	15		40
	58,000	0–7.5	74	198		40
	58,000	0–7.5	85	362		40
	17,500	0–10	130	1		41
	17,500	0–10	80	25		41
	12,470	0–2	(543–190)	0		44
	12,470	0–2	(50–47)	10		44
	control		18	>345		44
	21,040	0–2.5	87	6		45
	21,040	0–2.5	80	12		45
	21,040	0–2.5	47	18		45
	11,905	0–2.5	127	6		45
	11,905	0–2.5	42	12		45
	11,905	0–2.5	32	18		45
	11,905	0–2.5	22	50		45
	8,120	0–2.5	110	3		45
	8,120	0–2.5	47	6		45
	8,120	0–2.5	27	12		45
	8,120	0–2.5	20	18		45
	1,085	0–2.5	22	3		45
	1,085	0–2.5	26	6		45
	1,085	0–2.5	20	12		45
	1,085	0–2.5	27	18		45
	control	0–2.5	8	>50		45
	92,400	0–2.5	736	6		47
	92,400	0–2.5	273	12		47
	92,400	0–2.5	140	24		47
	92,400	0–2.5	81	48		47
	24,095	0–2.5	140	6		47
	24,095	0–2.5	265	12		47
	24,095	0–2.5	87	24		47
	24,095	0–2.5	56	48		47
	7,510	0–2.5	79	6		47
	7,510	0–2.5	28	12		47
	7,510	0–2.5	20	24		47
	7,510	0–2.5	15	48		47
	525	0–2.5	23	6		47
	525	0–2.5	21	12		47
	525	0–2.5	19	24		47
	525	0–2.5	20	48		47
	control	0–2.5	(20–22)	>500		47

Table 8.1 (continued) (1-5)

	ADT[a]	Depth (cm)	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Dust						
	37,180 – 21,140		2,002	3,876– roadside	wide standard deviation	48
	18,710 – 1,550		423	2,461 – roadside	wide standard deviation	48
	980 – 510		617 – 386	roadside	wide standard deviation	48
Sediment						
	15,000		4 (2–6)	0	median	74
	6,500		4 (2–5)	10	median	74
	<500		3 (2–4)	100	median	74
	15,473	0–3	7	34.8	median	75
	10,468	0–3	19	21.4	median	75
	10,468	0–3	4	47.3	median	75
	10,468	0–3	2	55.4	median	75
	10,468	0–3	2	58.9	median	75
	5,806	0–3	25	26.2	median	75

[a] ADT = average daily traffic

[b] Lead concentrations represent arithmetic means (dry weight basis) unless stated in "notes" column.

[c] N.S. = Distance from roads not stated.

Table 8.2. **Lead concentrations in vegetation in relation to traffic volumes and distance from roads. (Samples are on a dry–weight and unwashed basis unless otherwise stated.)**

Species	ADT[a]	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Cauliflower	58,000	0.3	15	flower	22
Cauliflower	58,000	0.02	198	flower	22
Tomato	58,000	0.6	15	fruit	22
Tomato	58,000	0.04	198	fruit	22
Strawberry	48,000	0.05	5	fruit washed	22
Strawberry	48,000	0.04	187	fruit washed	22
E.white pine	24,000	212	6	needles and twigs	23
E.white pine	24,000	170	12	needles and twigs	23
E.white pine	24,000	125	24	needles and twigs	23
Grasses	56,000	48	7.6	blades	27
Grasses	56,000	41	15	blades	27
Grasses	56,000	24	30	blades	27
Grasses	24,000	60	7.6	blades	27
Grasses	24,000	56	15	blades	27
Grasses	24,000	40	30	blades	27
Grasses	12,800	63	7.6	blades	33
Grasses	12,800	76	22.7	blades	33
Grasses	14,700	35	68.2	blades	33
Grasses	17,700	31	37.9	blades	33
Grasses	19,700	133	0	blades	33
Grasses	19,700	84	7.6	blades	33
Grasses	19,700	65	22.7	blades	33
Grasses	19,700	41	37.9	blades	33
Grasses	19,700	41	53	blades	33
Grasses	19,700	34	68.2	blades	33
Grasses	41,000	141	0	blades	33
Grasses	41,000	66	7.6	blades	33
Grasses	41,000	103	22.7	blades	33
Grasses	41,000	60	53	blades	33
Grasses	41,000	56	68.2	blades	33
Grasses	45,600	118	0	blades	33
Grasses	45,600	192	7.6	blades	33
Grasses	45,600	66	37.9	blades	33
Grasses	45,600	46	53	blades	33
Grasses	45,600	41	68.2	blades	33
Grasses	48,600	664	0	blades	33
Grasses	48,600	(154–454)	7.6	blades	33
Grasses	48,600	(66–198)	22.7	blades	33
Grasses	48,600	(45–139)	37.9	blades	33
Grasses	48,600	66	53	blades	33
Grasses	48,600	(48–68)	68.2	blades	33
Grasses	54,700	219	0	blades	33
Grasses	54,700	139	7.6	blades	33
Grasses	54,700	83	22.7	blades	33

Table 8.2. (continued) (2-2)

Species	ADT[a]	PPM[b] (range)	Distance from road (meter)	Notes		Ref.
Grasses	54,700	78	37.9	blades		33
Grasses	54,700	61	53	blades		33
Grasses	54,700	59	68.2	blades		33
Grasses	12,800	40	0	blades	washed	33
Grasses	12,800	37	7.6	blades	washed	33
Grasses	12,800	64	22.7	blades	washed	33
Grasses	12,800	58	37.9	blades	washed	33
Grasses	12,800	47	53	blades	washed	33
Grasses	12,800	50	68.2	blades	washed	33
Grasses	14,700	40	0	blades	washed	33
Grasses	14,700	23	7.6	blades	washed	33
Grasses	14,700	34	22.7	blades	washed	33
Grasses	14,700	43	37.9	blades	washed	33
Grasses	14,700	35	53	blades	washed	33
Grasses	14,700	31	68.2	blades	washed	33
Grasses	17,700	91	0	blades	washed	33
Grasses	17,700	64	7.6	blades	washed	33
Grasses	17,700	46	22.7	blades	washed	33
Grasses	17,700	26	37.9	blades	washed	33
Grasses	19,700	133	0	blades	washed	33
Grasses	19,700	58	7.6	blades	washed	33
Grasses	19,700	59	22.7	blades	washed	33
Grasses	19,700	58	37.9	blades	washed	33
Grasses	19,700	18	53	blades	washed	33
Grasses	19,700	36	68.2	blades	washed	33
Grasses	41,000	136	0	blades	washed	33
Grasses	41,000	85	7.6	blades	washed	33
Grasses	41,000	80	22.7	blades	washed	33
Grasses	41,000	55	37.9	blades	washed	33
Grasses	41,000	46	53	blades	washed	33
Grasses	41,000	58	68.2	blades	washed	33
Grasses	45,600	71	0	blades	washed	33
Grasses	45,600	80	7.6	blades	washed	33
Grasses	45,600	46	37.9	blades	washed	33
Grasses	45,600	31	53	blades	washed	33
Grasses	45,600	32	68.2	blades	washed	33
Grasses	48,600	(128–492)	0	blades	washed	33
Grasses	48,600	(62–262)	7.6	blades	washed	33
Grasses	48,600	(50–77)	22.7	blades	washed	33
Grasses	48,600	(36–59)	37.9	blades	washed	33
Grasses	48,600	45	53	blades	washed	33
Grasses	48,600	(31–44)	68.2	blades	washed	33
Grasses	54,700	98	0	blades	washed	33
Grasses	54,700	83	7.6	blades	washed	33
Grasses	54,700	60	22.7	blades	washed	33
Grasses	54,700	45	37.9	blades	washed	33

Table 8.2. (continued) (2-3)

Species	ADT[a]	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Grasses	54,700	62	53	blades washed	33
Grasses	54,700	43	68.2	blades washed	33
Carrots	49,000	53	9.1	tops washed	33
Carrots	49,000	22	30.3	tops washed	33
Carrots	49,000	17	75.8	tops washed	33
Carrots	47,100	37	9.1	tops washed	33
Carrots	47,100	26	30.3	tops washed	33
Carrots	47,100	21	75.8	tops washed	33
Carrots	12,500	18	9.1	tops washed	33
Carrots	12,500	11	30.3	tops washed	33
Carrots	12,500	14	75.8	tops washed	33
Maize	49,000	88	9.1	leaves washed	33
Maize	49,000	51	30.3	leaves washed	33
Maize	49,000	40	75.8	leaves washed	33
Maize	47,100	86	9.1	leaves washed	33
Maize	47,100	47	30.3	leaves washed	33
Maize	47,100	36	30.3	leaves washed	33
Maize	12,500	19	9.1	leaves washed	33
Maize	12,500	17	30.3	leaves washed	33
Maize	12,500	14	75.8	leaves washed	33
Lettuce	49,000	56	9.1	leaves washed	33
Lettuce	49,000	35	30.3	leaves washed	33
Lettuce	47,100	24	9.1	leaves washed	33
Lettuce	47,100	21	30.3	leaves washed	33
Lettuce	47,100	14	75.8	leaves washed	33
Lettuce	12,500	12	9.1	leaves washed	33
Lettuce	12,500	13	30.3	leaves washed	33
Grass	48,000	51	8	blades lightly rinsed	39
Grass	48,000	30	16	blades lightly rinsed	39
Grass	48,000	19	32	blades lightly rinsed	39
Grass	23,000	31	8	blades lightly rinsed	39
Grass	23,000	26	16	blades lightly rinsed	39
Grass	23,000	8	32	blades lightly rinsed	39
Grass	20,000	68	8	blades lightly rinsed	39
Grass	20,000	47	16	blades lightly rinsed	39
Grass	20,000	26	32	blades lightly rinsed	39
Grass	7,500	21	8	blades lightly rinsed	39
Grass	7,500	13	16	blades lightly rinsed	39
Grass	7,500	7	32	blades lightly rinsed	39
Cassinia vauvillinersii	1,200	*826	1–6	leaves *ash wt. basis	36
Cassinia vauvilliersii	1,200	*358	7–12	leaves *ash wt. basis	36
Hebe odora	1,200	*308	1–6	leaves *ash wt. basis	36
Hebe odora	1,200	*250	7–12	leaves *ash wt. basis	36

Table 8.2. (continued) (2-4)

Species	ADT[a]	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Alfalfa	37,000	30	18	top 15 cm	40
Alfalfa	37,000	19	50	top 15 cm	40
Alfalfa	37,000	15	80	top 15 cm	40
Strawberry	48,000	6	5	leaves	40
Strawberry	48,000	6	97	leaves	40
Strawberry	48,000	5	189	leaves	40
Tomato	70,000	4	15	fruit	40
Tomato	70,000	0.3	197	fruit	40
Lima beans	70,000	99	23	leaves	40
Lima beans	70,000	83	53	leaves	40
Lima beans	70,000	35	84	leaves	40
Grass	19,267	*583	1	blades *ash wt. basis	41
Tree of heaven	13,000	50	6	washed	43
Tree of heaven	13,000	30	24	washed	43
Mixed	12,470	(158–84)	0	mixed	44
Mixed	12,470	(78–33)	30	mixed	44
Mixed	control	7	>345	mixed	44
Grasses	12,040	75	6	blades	45
Grasses	12,040	42	12	blades	45
Grasses	12,040	27	18	blades	45
Grasses	11,905	70	6	blades	45
Grasses	11,905	30	12	blades	45
Grasses	11,905	20	18	blades	45
Grasses	11,905	25	50	blades	45
Grasses	8,120	21	3	blades	45
Grasses	8,120	20	6	blades	45
Grasses	8,120	18	18	blades	45
Grasses	1,285	12	3	blades	45
Grasses	1,285	15	12	blades	45
Grasses	1,285	10	18	blades	45
Forbs & Herbs	12,040	75	6	blades	45
Forbs & Herbs	12,040	(42–30)	12	blades	45
Forbs & Herbs	12,040	(24–40)	18	blades	45
Forbs & Herbs	1,285	32	3	blades	45
Forbs & Herbs	1,285	20	6	mixed washed	45
Forbs & Herbs	1,285	20	12	mixed washed	45
Forbs & Herbs	1,285	20	18	mixed washed	45
Shrub	12,040	72	12	mixed washed	45
Shrub	1,285	(17–32)	6	mixed washed	45
Deciduous	12,040	(30–75)	6	mixed washed	45
Deciduous	11,905	155	6	mixed washed	45
Deciduous	1,285	35	6	mixed washed	45

Table 8.2. (continued) (2-5)

Species	ADT[a] Ref.	PPM[b] (range)	Distance from road (meter)		Notes
Mixed	92,400	287(99–614)	6	mixed leaves	47
Mixed	92,400	167(86–309)	12	leaves seasonal variation noted	47
Mixed	92,400	92(29–150)	24	leaves seasonal variation noted	47
Mixed	92,400	85(16–194)	48	leaves seasonal variation noted	47
Mixed	24,095	147(47–287)	6	leaves seasonal variation noted	47
Mixed	24,095	101(30–207)	12	leaves seasonal variation noted	47
Mixed	24,095	57(29–221)	24	leaves seasonal variation noted	47
Mixed	24,095	57(15–136)	48	leaves seasonal variation noted	47
Mixed	7,510	33(9–190)	3	leaves seasonal variation noted	47
Mixed	7,510	39(15–66)	6	leaves seasonal variation noted	47
Mixed	7,510	17(10–42)	12	leaves seasonal variation noted	47
Mixed	7,510	15(5–27)	24	leaves seasonal variation noted	47
Mixed	7,510	11(4–23)	48	leaves seasonal variation noted	47
Mixed	525	14(8–30)	3	leaves seasonal variation noted	47
Mixed	525	8(4–12)	6	leaves seasonal variation noted	47
Mixed	525	10(3–16)	12	leaves seasonal variation noted	47
Mixed	525	8(3–16)	24	leaves seasonal variation noted	47
Mixed	525	7(2–13)	48	leaves seasonal variation noted	47
Mixed	control	10(4–15)	>500	leaves seasonal variation noted	47
Mixed	control	7(2–13)	>500	leaves seasonal variation noted	47
Prunus serotina	40,000	(9–14)	<30	leaves	55
Prunus serotina	40,000	(5–6)	>30	leaves	55
Prunus serotina	control	(2–5)	>1,000	leaves	55

[a] ADT – Average daily traffic

[b] – Lead concentrations represent arithmetic means unless otherwise stated in "notes" column.

Table 8.3. **Lead concentration in invertebrates in relation to traffic volumes and distance from roads. (Lead concentrations represent whole body on a dry weight basis. Earthworms were allowed to cast alimentary to tract soil.)**

Species	ADT[a]	PPM[b] (range)	Distance from road (meter)	±SE[d]	Notes	Ref.
EARTHWORMS						
Alabophera, Lumbricus & Octolasium	control	(4–6)	–			30
Lumbricus & Allolobophora	47,800 44,500	331	3		geometric mean	31
	47,800 44,500	168	6.1		geometric mean	31
	47,800 44,500	102	12.2		geometric mean	31
	47,800 44,500	49	24.4		geometric mean	31
	47,800 44,500	67	48.8		geometric mean	31
	25,100 500	220	3		geometric mean	31
	25,100 500	76	6.1		geometric mean	31
	25,100 500	64	12.2		geometric mean	31
	25,100 500	39	24.4		geometric mean	31
	25,100 500	41	48.8		geometric mean	31
	control	12	N.S.[c]		geometric mean	31
Oligchaeta	21,040	51	6		pooled	45
	21,040	50	12		pooled	45
	21,040	32	18		pooled	45
	11,905	24	18		pooled	45
	11,905	14	50		pooled	45
	1,085	9	12		pooled	45
	1,085	12	18		pooled	45
Lumbricus terrestris	control	1	roadside	±0.2	rinsed	49
	high use	130	roadside	±4	rinsed	49
	recreation use	32	roadside	±0.8	rinsed	49
	high use	274	roadside	±30	rinsed	49
	service road	38	roadside	±5	rinsed	49
Allolobophora chlorotica	control	0.3	roadside	±0.09	rinsed	49
	high use	341	roadside	±31	rinsed	49
	recreation use	77	roadside	±13	rinsed	49
	high use	500	roadside	±44	rinsed	49
	service road	26	roadside	±9	rinsed	49

Table 8.3 (continued)(3-2)

Species	ADT[a]	PPM[b] (range)	Distance from road (meter)	±SE[d]	Notes	Ref.
<u>Lumbricus rubellus</u>	recreation	37	roadside	±8	rinsed	49
Oligchaeta	21,040	51	6			56
	21,040	50	12			56
	21,040	32	18			56
	11,905	24	18			56
	11,905	14	50			56
	1,085	9	12			56
	1,085	12	18			56
"Earthworm"	92,400	273	6	±14		65
	92,400	275	12	±207		65
	92,400	198	24	N.S.		65
	92,400	99	48	±23		65
	24,095	99	6	±26		65
	24,095	168	12	±38		65
	24,095	54	24	±16		65
	24,095	25	48	±3		65
	7,510	28	6	±4		65
	7,510	12	12	±1		65
	7,510	7	24	±3		65
	7,510	7	48	±2		65
	525	17	3	±4		65
"Earthworm"	525	9	6	±2		65
	525	9	12	±1		65
	525	8	24	±0.1		65
	525	14	48	±7		65

Table 8.3 (continued) (3-3)

Species	ADT[a]	PPM[b] (range)	Distance from road (meter)	±SE[d]	Notes	Ref.
INSECTS						
Malacosoma americanum	40,000	(7.1–7.4)	<10		tent caterpillar	55
	40,000	(2.8–5.3)	>10		tent caterpillar	55
Caelifera (grasshopper)	21,040	4	NS		imago (adult)stage	45
	1,905	4	NS		imago (adult)stage	45
	11,085	3	NS		imago (adult)stage	45
Tipulidae	15,000	17(6–44)	0		median	74
	6,500	12(0.3–2–26)	10		median	74
	500	7(0.3–37)	100		median	74
Perlidae	15,000	28(6–57)	0		median	74
	6,500	20(3–36)	10		median	74
	500	18(2–29)	100		median	74
Pteronarcidae	15,000	20(0.3–77)	0		median	74
	6,500	16(0.5–27)	10		median	74
	500	7(0.3–60)	100		median	74

[a] ADT – Average daily traffic

[b] – Lead concentrations represent arithmetic means unless otherwise stated in "notes" column.

[c] – Distance from road not stated.

[d] – Standard error

Table 8.4. **Lead concentrations in vertebrates in relation to traffic volumes and distance from roads. Lead concentrations represent arithmetic means of whole body counts on a dry weight basis unless otherwise stated in "notes" column.**

Species	ADT^a	PPM^b	(SD)/±SE^c	Distance from road (meters)	Body part	Notes	Ref.
MAMMALS – HERBIVORES							
Microtus pennsylvanicus		12,470	16 (7–29)	(5.4)	0–10		44
	12,470	6 (5–6)	(0.6)	25–45			44
	12,470	4 (0.6–5)	(1)	135–185			44
	Control	5 (.09–8)	(2)	NS			44
	11,095	10	±0.8	30–70			56
	8,120	12	±0.2	0–20			56
	1,085	7	±0.6	0–2			56
	Control	0.8(0.5–1.4)		<610		geometric mean, W.W.d	58
	35,000	1 (0.2–5)		<18		geometric mean, W.W.	58
M. agrestis	Arterial road	3		<3		W.W.	61
	Minor road	2		<3		W.W.	61
Clethrionomys glareolus	Arterial road	0.7–2		<8		W.W.	61
	Minor road	2–3		<3		W.W.	61
M. pennsylvanicus	92,400	23	±3	<24			65
	24,095	17	±1	<24			65
	7,510	7	±0.8	<24			65
	525	2	±0.2	<24			65
	Control	1–4	±0.2–1	>500			65
M. ochrogaster	19,600	5	(3)	<10			66
	1,360	6	(5)	<10			66
	340	2	(0.3)	<10			66
	19,600	11	(4)	<10	gut		66
	1,360	18	(18)	<10	gut		66
	340	3	(0.8)	<10	gut		66
	19,600	5	(5)	<10	spleen		66
	1,360	2	(0.8)	<10	spleen		66
	340	2	(2)	<10	spleen		66
	19,600	2	(0.6)	<10	liver		66

Table 8.4 *(continued)* (4-2)

Species	ADT^a	PPM^b	(SD)/±SE^c	Distance from road (meters)	Body part	Notes	Ref.
	1,360	1	(0.7)	<10	liver		66
	340	1	(0.5)	<10	liver		66
	19,600	3	(2)	<10	lung		66
	1,360	2	(1)	<10	lung		66
	340	1	(0.8)	<10	lung		66
	19,600	8	(3)	<10	kidney		66
	1,360	8	(5)	<10	kidney		66
	340	3	(0.9)	<10	kidney		66
	19,600	17	(7)	<10	bone		66
	1,360	23	(30)	<10	bone		66
	340	5	(1)	<10	bone		66
	19,600	8	(14)	<10	muscle (thigh)		66
	1,360	3	(4)	<10	muscle (thigh)		66
	340	2	(1)	<10	muscle (thigh)		66
MAMMALS – GRANIVORES							
Peromyscus leucopus	12,470	7 (5–12)	(3)	0–10			44
	12,470	4 (2–8)	(3)	10–55			44
	12,470	3 (0–7)	(2)	115–195			44
	Control	3 (0.5–4)	(1)				44
	21,040	16	±1	0–20			56
	8,120	10	±0.9	0–20			56
	Control	5–8					56
	35,000	5 (2–11)		<18		geometric mean, W.W.	58
	Control	1 (0.3–13)		>610		geometric mean, W.W.	58
P. maniculatus	19,800	52	(34)	15	bone		63
	Control	5	(6)	800	bone		63
	19,800	8	(3)	15	kidney		63
	Control	3	(2)	800	kidney		63
	19,800	3	(2)	15	liver		63
	Control	1	(0.6)	800	liver		63
	19,800	0.8	(0.3)	15	brain		63
	Control	0.1	(0.06)	800	brain		63

Table 8.4 (continued) (4-3)

Species	ADT[a]	PPM[b]	(SD)/±SE[c]	Distance from road (meters)	Body part	Notes	Ref.
P. leucopus	92,400	22	±8	<24			65
	24,095	13	±2	<24			65
	7,510	5	±1	<24			65
	525	2	±0.3	<24			65
	Control	2–3		>500			65
P. maniculatus	19,600	6	(5)	<10			66
	1,360	4	(3)	<10			66
	340	3	(6)	<10			66
	Control	3	(3)	>50			66
	19,600	19	(16)	<10	gut		66
	1,360	6	(4)	<10	gut		66
	340	5	(5)	<10	gut		66
	Control	4	(3)	>50	gut		66
	19,600	19	(25)	<10	spleen		66
	1,360	3	(2)	<10	spleen		66
	340	7	(7)	<10	spleen		66
	Control	4	(2)	>50	spleen		66
	19,600	4	(2)	<10	liver		66
	1,360	2	(0.6)	<10	liver		66
	340	2	(2)	<10	liver		66
	Control	1	(0.6)	>50	liver		66
	19,600	6	(5)	<10	lung		66
	1,360	2	(2)	<10	lung		66
	340	6	(8)	<10	lung		66
	Control	1	(0.5)	>50	lung		66
	19,600	8	(3)	<10	kidney		66
	1,360	9	(8)	<10	kidney		66
	340	3	(2)	<10	kidney		66
	Control	2	(4)	>50	kidney		66
	19,600	25	(26)	<10	bone		66
	1,360	8	(5)	<10	bone		66
	340	6	(8)	<10	bone		66

Table 8.4 (continued) (4-4)

Species	ADT^a	PPM^b	(SD)/±SE^c	Distance from road (meters)	Body part	Notes	Ref.
	Control	6	(11)	>50	bone		66
	19,600	7	(5)	<10	muscle (thigh)		66
	1,360	7	(7)	<10	muscle (thigh)		66
	340	2	(1)	<10	muscle (thigh)		66
	Control	2	(2)	>50	muscle (thigh)		66
Reithrodontomys megalotis	19,600	12	(3)	<10			66
	1,360	3		<10			66
	340	3	(1)	<10			66
	19,600	18	(17)	<10	gut		66
	1,360	6		<10	gut		66
	340	4	(1)	<10	gut		66
	1,360	9		<10	spleen		66
	340	6	(2)	<10	spleen		66
	19,600	5	(0.5)	<10	liver		66
	1,360	1		<10	liver		66
	340	2	(0.9)	<10	liver		66
	19,600	21	(14)	<10	lung		66
	1,360	4		<10	lung		66
	340	5	(2)	<10	lung		66
	1,360	2		<10	kidney		66
	340	5	(1)	<10	kidney		66
	19,600	109	(29)	<10	bone		66
	340	18	(7)	<10	bone		66
	19,600	27	(18)	<10	muscle (thigh)		66
	340	4	(1)	<10	muscle (thigh)		66
MAMMALS – OMNIVORES							
Mus musculus	92,400	64	±17	<24			65
	24,095	22	±15	<24			65
	7,510	11		<24			65
	19,600	7	(6)	<10			66
	1,360	6	(12)	<10			66

Table 8.4 (continued) (4-5)

Species	ADT[a]	PPM[b]	(SD)/±SE[c]	Distance from road (meters)	Body part	Notes	Ref.
	340	7	(10)	<10			66
	Control	2	(3)	>50			66
	19,600	19	(18)	<10	gut		66
	1,360	9	(14)	<10	gut		66
	340	5	(4)	<10	gut		66
	Control	3	(0.9)	>50	gut		66
	19,600	12	(37)	<10	spleen		66
	1,360	3	(2)	<10	spleen		66
	340	5	(7)	<10	spleen		66
	Control	2	(1)	>50	spleen		66
	19,600	3	(5)	<10	liver		66
	1,360	2	(1)	<10	liver		66
	340	2	(2)	<10	liver		66
	Control	2	(3)	>50	liver		66
	19,600	3	(5)	<10	lung		66
	1,360	3	(3)	<10	lung		66
	340	2	(2)	<10	lung		66
	Control	3	(6)	>50	lung		66
	19,600	8	(5)	<10	kidney		66
	1,360	7	(3)	<10	kidney		66
	340	3	(3)	<10	kidney		66
	Control	3	(2)	>50	kidney		66
	19,600	19	(24)	<10	bone femur		66
	1,360	21	(59)	<10	bone femur		66
	340	24	(47)	<10	bone femur		66
	Control	9	(9)	>50	bone femur		66
Rattus norvegicus	24,095	28	±2	≤24			65
Mus musculus	19,600	6	(9)	<10	muscle (thigh)		66
	1,360	4	(4)	<10	muscle (thigh)		66
	340	3	(6)	<10	muscle (thigh)		66
	Control	3	(5)	>50	muscle (thigh)		66

Table 8.4 (continued) (4-6)

Species	ADT^{a}	PPM^{b}	(SD)/±SE^{c}	Distance from road (meters)	Body part	Notes	Ref.
MAMMALS – INSECTIVORES							
Blarina brevicauda	12,470	23 (8–64)		0–10			44
	12,470	5 (3–9)		25–345			44
	Control	5 (0–10)					44
	Smelter	18–109					51
	21,040	35	±10	0–20			56
	11,095	22	±5	30–70			56
	8,120	16	±3	0–20			56
	1,085	12	±1	0–20			56
	Control	14–15	±1				56
	35,000	26 (6–130)		≤18		geometric mean, W.W.	58
	Control	2 (0.2–8)		610		geometric mean, W.W.	58
	92,400	73	±22	≤24			65
	24,095	35	±12	≤24			65
	7,510	12	±2	≤24			65
	525	4	±0.9	≤24			65
	Control	3–7		≥500			65
	19,600	18	(11)	≤10			66
	1,360	7	(7)	≤10			66
	340	6	(6)	≤10			66
	19,600	24	(18)	≤10	gut		66
	1,360	7	(6)	≤10	gut		66
	340	3	(1)	≤10	gut		66
	19,600	5	(5)	≤10	spleen		66
	1,360	4	(2)	≤10	spleen		66
	340	2	(2)	≤10	spleen		66
	19,600	5	(3)	≤10	liver		66
	1,360	2	(0.7)	≤10	liver		66
	340	1	(0.5)	≤10	liver		66
	19,600	17	(20)	≤10	lung		66
	1,360	6	(6)	≤10	lung		66
	340	8	(13)	≤10	lung		66
	19,600	12	(6)	≤10	kidney		66

Table 8.4 (continued) (4–7)

Species	ADT^{a}	PPM^{b}	(SD)/±SE^{c}	Distance from road (meters)	Body part	Notes	Ref.
	1,360	6	(4)	≤10	kidney		66
	340	4	(4)	≤10	kidney		66
	19,600	67	(53)	≤10	bone femur		66
	1,360	20	(21)	≤10	bone femur		66
	340	12	(8)	≤10	bone femur		66
	19,600	10	(10)	≤10	muscle (thigh)		66
	1,360	6	(3)	≤10	muscle (thigh)		66
	340	5	(5)	≤10	muscle (thigh)		66
	Control	3		>50			66
Cryptotis parva	11,095	13	±0.8	30–70			56
	8,120	10	±0.4	0–20			56
	1,085	7	±1	0–20			56
Sorex cinereus	8,120	14	±1	0–20			56
	Control	17	±3				56
Cryptotis parva	24,095	17	±2	≤24			65
	7,510	7	±1	≤24			65
	525	3	±1	≤24			65
	Control	3–5		≥500			65
Sorex cinereus	7,510	10		≤24			65
	Control	3–4	±0.21	≥500			65
Eptesicus fuscus							
Male	(35,000)	47 (20–90)	610			geometric mean, W.W.	58
Female	(35,000)	31 (20–56)	610			geometric mean, W.W.	58
Myotis lucifugus							
Female	(35,000)	17 (11–29)	610			geometric mean, W.W.	58
BIRDS – INSECTIVORES^{f}							
Hirundo rustica	37,925	5		roadside	stomach^{e}	males, geometric mean	59
	37,925	7		roadside	stomach^{e}	females, geometric mean	59
	37,925	3		roadside	stomach^{e}	nestlings, geometric mean	59
	330	0.2		400	stomach^{e}	males, geometric mean	59
	330	2		400	stomach^{e}	females, geometric mean	59
	330	2		400	stomach^{e}	nestlings, geometric mean	59
	37,925	5 (4–6)		Roadside	carcasses	males, geometric mean	59

Table 8.4 (continued) (4-8)

Species	ADT^a	PPM^b	(SD)/±SE^c	Distance from road (meters)	Body part	Notes	Ref.
	37,925	9 (6–12)		Roadside	carcasses	females, geometric mean	59
	37,925	2 (1–2)		Roadside	carcasses	nestlings, geometric mean	59
	330	4 (3–5)		400	carcasses	males, geometric mean	59
	330	5 (3–7)		400	carcasses	females, geometric mean	59
	330	0.7(0.7–1.4)		400	carcasses	nestlings, geometric mean	59
	37,925	67 (55–82)		Roadside	feathers	males, geometric mean	59
	37,925	54 (43–68)		Roadside	feathers	females, geometric mean	59
	37,925	2.5(2.3–2.7)		Roadside	feathers	nestlings, geometric mean	59
	330	24 (21–28)		400	feathers	males, geometric mean	59
	330	19 (16–22)		400	feathers	females, geometric mean	59
	330	2.3(2.0–2.6)		400	feathers	nestlings, geometric mean	59
AMPHIBIANS							
Rana catesbeiana	Control 0	(3–6)		≥4,000		washed rinsed	60
(Bull frog) tadpole	4,272 – 108,800	(0.7–270)				washed rinsed	60
R. clamitans	Control	(0.9–9)		≥4,000		washed rinsed	60
(Green frog) tadpole	4,272 – 108,800	(5–240)				washed rinsed	60
FISH							
Bluehead chub	15,000	8 (2–61)				median	74
	6,500	7 (2–19)		10		median	74
	500	5 (3–7)		100		median	74
Blacknose dace	15,000	16 (10–26)				median	74
	6,500	11 (5–36)		10		median	74
	500	7 (2–18)		100		median	74
Fantail darter	15,000	20 (12–29)				median	74
	6,500	14 (0.2–42)		10		median	74
	500	10 (0.2–15)		100		median	74

Table 8.4 (continued) (4-9)

[a]ADT = Average Daily Traffic.

[b] = Lead concentrations represent arithmetic means (dry weight basis) unless otherwise sated in the note column.

[c] = Parentheses indicate standard deviation; + indicates standard error.

[d] = W. W. indicates wet weight basis.

[e] = Pooled samples.

[f] = Data from known nesting species only included; references 6–10, 66–67 not included.

Table 8.5. **Cadium concentrations in soils and vegetation in relation to traffic volumes and distance from roads. (Depth of soil sampled and distance from pavement edge are given where appropriate. All concentrations are on a dry weight basis).**

	ADT[a]	Depth	PPM[b] (range)	(SD)/± SE	Distance from road (meter)	Notes	Ref.
Soil	control	0–10	(0.2–0.8)				30
	47,800–44,500	0–2.5	2		3.0	geometric mean	31
	47,800–44,500	0–2.5	0.8		6.1	geometric mean	31
	47,800–44,500	0–2.5	0.7		12.2	geometric mean	31
	47,800–44,500	0–2.5	0.7		24.4	geometric mean	31
	47,800–44,500	0–2.5	0.7		48.8	geometric mean	31
	25,100–500	0–2.5	0.9		3	geometric mean	31
	25,100–500	0–2.5	0.7		6.1	geometric mean	31
	25,100–500	0–2.5	0.8		12.2	geometric mean	31
	25,100–500	0–2.5	0.6		24.4	geometric mean	31
	25,100–500	0–2.5	0.7		48.8	geometric mean	31
	control	0–2.5	0.7			geometric mean	31
	48,000	0–5	0.9		8		39
	48,000	0–5	0.7		16		39
	48,000	0–5	0.2		32		39
	48,000	5–10	0.7		8		39
	48,000	5–10	0.4		16		39
	48,000	5–10	0.2		32		39
	48,000	10–15	0.3		8		39
	48,000	10–15	0.2		16		39
	48,000	10–15	0.1		32		39
	23,000	0–5	2		8		39
	23,000	0–5	2		16		39
	23,000	0–5	1		32		39
	23,000	5–10	0.4		8		39
	23,000	5–10	0.3		16		39
	23,000	5–10	0.2		32		39
	23,000	10–15	0.3		8		39
	23,000	10–15	0.3		16		39
	23,000	10–15	0.2		32		39
	20,000	0–5	1		8		39

Table 8.5. (continued) (5-2)

	ADT[a]	Depth	PPM[b] (range)	(SD)/± SE	Distance from road (meter)	Notes	Ref.
Soil	20,000	0–5	0.4		16		39
	20,000	0–5	0.2		32		39
	20,000	5–10	0.8		8		39
	20,000	5–10	0.4		16		39
	20,000	5–10	0.2		32		39
	20,000	10–15	0.5		8		39
	20,000	10–15	0.3		16		39
	20,000	10–15	0.2		32		39
	7,500	0–5	0.9		8		39
	7,500	0–5	0.8		16		39
	7,500	0–5	0.7		32		39
	7,500	5–10	0.7		8		39
	7,500	5–10	0.7		16		39
	7,500	5–10	0.5		32		39
	7,500	10–15	0.6		8		39
	7,500	10–15	0.6		16		39
	7,500	10–15	0.5		32		39
	92,400	0–2.5	0.7		6	seasonal variations noted	47
	92,400	0–2.5	0.5		12	seasonal variations noted	47
	92,400	0–2.5	0.4		24	seasonal variations noted	47
	92,400	0–2.5	0.3		48	seasonal variations noted	47
	24,095	0–2.5	0.4		6	seasonal variations noted	47
	24,095	0–2.5	0.9		12	seasonal variations noted	47
	24,095	–2.5	0.3		24	seasonal variations noted	47
	24,095	0–2.5	1		48	seasonal variations noted	47
	7,510	0–2.5	1		6	seasonal variations noted	47
	7,510	0–2.5	0.5		12	seasonal variations noted	47
	7,510	0–2.5	0.4		24	seasonal variations noted	47
	7,510	0–2.5	0.5		48	seasonal variations noted	47
	525	0–2.5	0.7		3	seasonal variations noted	47
	525	0–2.5	0.5		6	seasonal variations noted	47
	525	0–2.5	0.5		12	seasonal variations noted	47
	525	0–2.5	0.4		24	seasonal variations noted	47

Table 8.5. (continued) (5-3)

	ADT[a]	Depth	PPM[b] (range)	(SD)/± SE	Distance from road (meter)	Notes	Ref.
Soil	525	0–2.5	0.3		48	seasonal variations noted	47
	control	0–2.5	(0.3–0.4)		≥500	seasonal variations noted	47
Sediment	15,000		0.04(0.02–0.2)		0	median	74
	6,500		0.02(0.03–0.24)		10	median	74
	<500		0.03(0.04–0.14)		100	median	74
Dust	37,180		28	(15)	NS		48
	23,730		22	(11)	NS		48
	21,140		23	(5)	NS		48
	18,710		17	(8)	NS		48
	17,970		11	(10)	NS		48
	9,220		28	(17)	NS		48
	7,180		10	(7)	NS		48
	4,190		12	(8)			48
	3,480		12	(10)			48
	1,590		12	(8)			48
	1,550		7	(9)			48
	980		Not detectable				48
	510		15	(17)		48	
	20		6	–			48
Grasses	48,000		0.8		8	blades, lightly rinsed	39
	48,000		0.6		16	blades, lightly rinsed	39
	48,000		0.5		32	blades, lightly rinsed	39
	23,000		0.7		8	blades, lightly rinsed	39
	23,000		0.5		16	blades, lightly rinsed	39
	23,000		0.3		32	blades, lightly rinsed	39
	20,000		0.9		8	blades, lightly rinsed	39
	20,000		0.7		16	blades, lightly rinsed	39
	20,000		0.5		32	blades, lightly rinsed	39
	7,500		0.5		8	blades, lightly rinsed	39
	7,500		0.4		16	blades, lightly rinsed	39
	7,500		0.2		32	blades, lightly rinsed	39

Table 8.5. (continued) (5-4)

	ADT[a]	Depth	PPM[b] (range)	(SD)/± SE	Distance from road (meter)	Notes	Ref.
Mixed	92,400		1	5		mixed leaves	47
	92,400		0.4	12		mixed leaves	47
	92,400		0.4	24		mixed leaves	47
	92,400		0.3	48		mixed leaves	47
	24,095		0.5	6		mixed leaves	47
	24,095		0.2	12		mixed leaves	47
	24,095		0.2	24		mixed leaves	47
	24,095		0.2	48		mixed leaves	47
	7,510		0.3	3		mixed leaves	47
	7,510		0.3	6		mixed leaves	47
	7,510		0.3	12		mixed leaves	47
	7,510		0.3	24		mixed leaves	47
	7,510		0.6	48		mixed leaves	47
	525		0.5	3		mixed leaves	47
	525		0.3	6		mixed leaves	47
	525		0.3	12		mixed leaves	47
	525		0.3	24		mixed leaves	47
	525		0.2	48		mixed leaves	47
	control		(0.2–0.4)	≥500		mixed leaves	47

[a] ADT = average daily traffic

[b] Cadmium concentrations represent arithmetic means (dry weight basis) unless stated in "notes" column.

NS = not stated

Table 8.6. **Cadmium concentrations in invertebrates in relation to traffic volumes and distance from roads. Cadmium concentrations represent whole body counts on a dry weight basis. Earthworms were allowed to cast alimentary tract soil.**

Species	ADT[a]	PPM[b] (range)	(SD)/+SE	Distance from road (meters)	Notes	Ref.
EARTHWORMS						
Alabophera, Lumbricus, & Octolasium	Control	(3–9)				
Lumbricus & Allolobophora	47,800–44,500	11		3	geometric mean	31
	47,800–44,500	10		6.1	geometric mean	31
	47,800–44,500	9		12.2	geometric mean	31
	47,800–44,500	8		24.4	geometric mean	31
	47,800–44,500	9		48.8	geometric mean	31
	25,100–500	14		3	geometric mean	31
	25,100–500	8		6.1	geometric mean	31
	25,100–500	8		12.2	geometric mean	31
	25,100–500	6		24.4	geometric mean	31
	25,100–500	6		48.8	geometric mean	31
Lumbricus terrestris	High use	4	±0.8	roadside		49
	High use	12	±0.3	roadside		49
	Service road	4	±1	roadside		49
	Recreational use	4	±0.6	roadside		49
	Control	0.6	±0.09	roadside		49
Lumbricus rubellus	Recreational use	4	±1	roadside		49
Allolobophora chlorotica	High use	4	±0.8	roadside		49
	High use	9	±0.2	roadside		49
	Service road	1	±0.2	roadside		49
	Recreational use	4	±0.6	roadside		49
	Control	0.2	±0.02	roadside		49
"Earthworm"	92,400	8	±0.8	6		49
	92,400	10	±1	12		65
	92,400	13		24		65
	92,400	6	±0.3	48		65
	24,095	8	±3	6		65
	24,095	7	±1	12		65
	24,095	5	±1.4	24		65

Table 8.6. (continued) (6-2)

Species	ADT[a]	PPM[b] (range)	(SD)/+SE	Distance from road (meters)	Notes		Ref.
	24,095	3	±0.3	48			65
	7,510	4	±0.5	6			65
	7,510	3	±0.2	12			65
	7,510	3	±0.3	24			65
	7,510	4	±0.7	48			65
	525	3	±0.5	3			65
	525	2	±0.4	6			65
	525	2	±0.4	12			65
	525	3	±0.6	24			
	525	4	±0.5	48			
	Control	(5–11)	±(1–5)	≥500			65
BENTHIC INSECTS							
Tipulidae	15,000	1 (0.07–2)		0	median	stage larvae	74
	6,500	0.9(0.03–2)		10	median	stage larvae	74
	500	0.8(0.03–3)		100	median	stage larvae	74
Perlidae	15,000	0.9(0.03–3)		0	median	stage larvae	74
	6,500	0.8(0.06–2)		10	median	stage larvae	74
	500	0.7(0.03–3)		100	median	stage larvae	74
Pteronarcidae	15,000	1 (0.5–3)		0	median	stage larvae	74
	6,500	1 (0.2–2)		10	median	stage larvae	74
	500	0.6(0.03–2)		100	median	stage larvae	74

[a] – ADT = Average daily traffic.

[b] – Cadmium concentrations are arithmetic means and are expressed as ppm dry weight unless otherwise stated in "notes" column.

Table 8.7. **Cadmium concentrations in vertebrates in relation to traffic volumes and distance from roads. (Cadmium concentrations represent whole body counts on a dry weight basis.)**

Species	ADT^{a}	PPM^{b} (range)	(SD)/±SE	Distance to road (meters)	Notes	Ref.
MAMMALS						
Microtus pennsylvanicus	92,400	0.4	±0.2	≤24		65
	24,095	0.2	±0.03	≤24		65
	7,510	0.2	±0.03	≤24		65
	525	0.2	±0.02	≤24		65
	Control	(0.1–0.2)	±0.02	≥500		65
Peromyscus						65
leucopus	92,400	0.8	±0.4	≤24		65
	24,095	0.3	±0.2	≤24		65
	7,510	0.2	±0.4	≤24		65
	525	0.2	±0.08	≤24		65
	Control	(0.2–0.4)		≥500		65
Mus musculus	92,400	0.2	±0.02	≤24		65
	24,095	0.06	±0.02	≤24		65
	7,510	0.1		≤24		65
Rattus norvegicus	24,095	0.5	±0.3	≤24		65
Blarina brevicauda	92,400	2	±0.4	≤24		65
	24,095	0.5	±0.07	≤24		65
	7,510	0.5	±0.08	≤24		65
	525	0.3	±0.11	≤24		65
	Control	(0.7–0.5)	±0.09	≥500		65
Sorex cinereus	7,510	1		≤24		65
	Control	(0.4–0.9)		≥500		65
Cryptotis parva	24,095	0.5	±0.2	≤24		65
	7,510	0.8	±0.4	≤24		65
	525	0.2	±0.02	≤24		65
	Control	0.3	±0.1	≥500		65

Table 8.7.(continued) (7-2)

Species	ADT[a]	PPM[b] (range)	(SD)/±SE	Distance to road (meters)	Notes	Ref.
FISH						
Bluehead chub	15,000	0.4(0.08–13)		0	median	74
	6,500	0.4(0.02–0.7)		10	median	74
	500	0.3(0.09–0.5)		100	median	74
Blacknose dace	15,000	0.8(0.4–1)		0	median	74
	6,500	0.3(0.03–0.6)		10	median	74
	500	0.7(0.4–6)		100	median	74
Fantail darter	15,000	0.6(0.2–1)		0	median	74
	6,500	0.7(0.02–3)		10	median	74
	500	0.6(0.02–2)		100	median	74

[a] – ADT = Average daily traffic.

[b] – Cadmium concentrations are arithmetic means and are expressed as ppm dry weight unless otherwise stated in "notes" column.

Table 8.8. **Nickel concentrations in soils and vegetation in relation to traffic volumes and distance from roads. (Depths of soils sampled and distance from the road pavement edge are given where appropriate. All concentrations are on a dry weight basis).**

	ADT[a]	Depth	PPM[b] (range)	Distance from road (meter)	Notes	Ref.
Soil	47,800–44,500	0–2.5	1.6	3.0	geometric mean	31
Soil	47,800–44,500	0–2.5	0.8	6.1	geometric mean	31
Soil	47,800–44,500	0–2.5	0.7	12.2	geometric mean	31
Soil	47,800–44,500	0–2.5	0.7	24.4	geometric mean	31
Soil	47,800–44,500	0–2.5	0.7	48.8	geometric mean	31
Soil	25,100–500	0–2.5	0.9	3.0	geometric mean	31
Soil	25,100–500	0–2.5	0.7	6.1	geometric mean	31
Soil	25,100–500	0–2.5	0.8	12.2	geometric mean	31
Soil	25,100–500	0–2.5	0.7	24.4	geometric mean	31
Soil	25,100–500	0–2.5	0.7	48.8	geometric mean	31
Soil	Control	0–2.5	0.7		geometric mean	31
Soil	48,000	0–5	7	8		39
Soil	48,000	0–5	4	16		39
Soil	48,000	0–5	2	32		39
Soil	48,000	5–10	6	8		39
Soil	48,000	5–10	2	16		39
Soil	48,000	5–10	1	32		39
Soil	48,000	10–15	1	8		39
Soil	48,000	10–15	0.8	16		39
Soil	48,000	10–15	0.6	32		39
Soil	20,000	0–5	5	8		39
Soil	20,000	0–5	2	16		39
Soil	20,000	0–5	2	32		39
Soil	20,000	5–10	1	8		39
Soil	20,000	5–10	0.9	16		39
Soil	20,000	5–10	0.6	32		39
Soil	20,000	10–15	0.8	8		39
Soil	20,000	10–15	0.6	16		39
Soil	20,000	10–15	0.6	32		39

Table 8.8. (continued) (8-2)

	ADTa	Depth	PPMb (range)	Distance from road (meter)	Notes		Ref.
Soil	92,400	0–2.5	5	6			47
Soil	92,400	0–2.5	2	12			47
Soil	92,400	0–2.5	2	24			47
Soil	92,400	0–2.5	2	48			47
Soil	24,095	0–2.5	5	6			47
Soil	24,095	0–2.5	4	12			47
Soil	24,095	0–2.5	3	24			47
Soil	24,095	0–2.5	2	48			47
Soil	7,510	0–2.5	9	3			47
Soil	7,510	0–2.5	2	6			47
Soil	7,510	0–2.5	2	12			47
Soil	7,510	0–2.5	2	24			47
Soil	7,510	0–2.5	1	48			47
Soil	525	0–2.5	4	3			47
Soil	525	0–2.5	4	6			47
Soil	525	0–2.5	4	12			47
Soil	525	0–2.5	5	24			47
Soil	525	0–2.5	4	48			47
Soil	Control	0–2.5	2	≥500			47
Sediment	15,000	·	2(0.9–4)	0	median		74
Sediment	6,500	·	1(0.4–4)	10	median		74
Sediment	< 500	·	0.9(0.2–2)	100	median		74
Grasses	48,000		4	8	blades	rinsed lightly	39
Grasses	48,000		3	16	blades	rinsed lightly	39
Grasses	48,000		1	32	blades	rinsed lightly	39
Grasses	20,000		5	8	blades	rinsed lightly	39
Grasses	20,000		4	16	blades	rinsed lightly	39
Grasses	20,000		3	32	blades	rinsed lightly	39
Mixed	92,400		3	6	mixed	unwashed	47
Mixed	92,400		2	12	mixed	unwashed	47
Mixed	92,400		2	24	mixed	unwashed	47
Mixed	92,400		3	48	mixed	unwashed	47
Mixed	24,095		2	6	mixed	unwashed	47

Table 8.8. (continued) (8-3)

	ADT^a	Depth	PPM^b (range)	Distance from road (meter)	Notes		Ref.
Mixed	24,095		1	12	mixed	unwashed	47
Mixed	24,095		1	24	mixed	unwashed	47
Mixed	24,095		2	48	mixed	unwashed	47
Mixed	7,510		1	6	mixed	unwashed	47
Mixed	7,510		2	12	mixed	unwashed	47
Mixed	7,510		1	24	mixed	unwashed	47
Mixed	7,510		1	48	mixed	unwashed	47
Mixed	525		1	3	mixed	unwashed	47
Mixed	525		1	6	mixed	unwashed	47
Mixed	525		1	12	mixed	unwashed	47
Mixed	525		1	24	mixed	unwashed	47
Mixed	525		1	48	mixed	unwashed	47
Mixed	Control		1–2	≥500	mixed	unwashed	47

[a] ADT = Average Daily Traffic

[b] Nickel concentrations represent arithemetic means (dry weight basis) unless stated in "Notes" column.

Table 8.9. **Nickel concentrations in invertebrates and vertebrates in relation to traffic volumes and distance from roads. (Nickel concentrations represent whole body on a dry weight basis. Earthworms were allowed to cast alimentary tract soil)**

Species	ADT[a]	PPM[b] (range)	(SD)/±SE	Distance from road (meter)	Notes	Ref.
EARTHWORMS						
Lumbricus & Allobophora	47,800–44,500	32		3	geometric mean	31
Lumbricus & Allobophora	47,800–44,500	31		6.1	geometric mean	31
Lumbricus & Allobophora	47,800–44,500	29		12.2	geometric mean	31
Lumbricus & Allobophora	47,800–44,500	23		24.4	geometric mean	31
Lumbricus & Allobophora	47,800–44,500	32		48.8	geometric mean	31
Lumbricus & Allobophora	25,100– 500	38		3	geometric mean	31
Lumbricus & Allobophora	25,100– 500	25		6.1	geometric mean	31
Lumbricus & Allobophora	25,100– 500	18		12.2	geometric mean	31
Lumbricus & Allobophora	25,100– 500	13		24.4	geometric mean	31
Lumbricus & Allobophora	25,100– 500	19		48.8	geometric mean	31
"earthworms"	92,400	5	±1	6		65
"earthworms"	92,400	7	±3	12		65
"earthworms"	92,400	2		24		65
"earthworms"	92,400	2	±1	48		65
"earthworms"	24,095	3	±7	6		65
"earthworms"	24,095	10	±6	12		65
"earthworms"	24,095	4	±1	24		65
"earthworms"	24,095	4	±0.2	48		65
"earthworms"	7,510	3	±0.6	6		65
"earthworms"	7,510	2	±0.2	12		65
"earthworms"	7,510	2	±0.6	24		65
"earthworms"	7,510	1	±0.1	48		65
"earthworms"	525	2	±0.6	3		65
"earthworms"	525	3	±0.3	6		65
"earthworms"	525	3	±0.3	12		65
"earthworms"	525	2	±0.6	24		65
"earthworms"	525	2	±0.1	48		65
"earthworms"	Control	(4–5)	±1	≥500		65

Table 8.9. (continued) (9-2)

Species	ADT[a]	PPM[b] (range)	(SD)/±SE	Distance from road (meter)	Notes	Ref.
BENTHIC INSECTS						
Tipulidae	15,000	7(0.7–10)		0	median	74
Tipulidae	6,500	4(0.3–9)		10	median	74
Tipulidae	500	2(0.3–16)		100	median	74
Perlidae	15,000	11(1–17)		0	median	74
Perlidae	6,500	6(0.5–13)		10	median	74
Perlidae	500	3(0.4–15)		100	median	74
MAMMALS						
Pteronarcidae	15,000	2(0.7–15)		0	median	74
Pteronarcidae	6,500	3(0.4–10)		10	median	74
Pteronarcidae	500	2(0.1–14)		100	median	74
Microtus pennsylvanicus	92,400	2	±0.8	≤24		65
Microtus pennsylvanicus	24,095	2	±0.2	≤24		65
Microtus pennsylvanicus	7,510	2	±0.7	≤24		65
Microtus pennsylvanicus	525	1	±0.2	≤24		65
Microtus pennsylvanicus	Control 0	2	±0.4	≥500		65
Peromyscus leucopus	92,400	1	±0.2	≤24		65
Peromyscus leucopus	24,095	1	±0.1	≤24		65
Peromyscus leucopus	7,510	1	±0.3	≤24		65
Peromuscus leucopus	525	1	±0.8	≤24		65
Peromyscus leucopus	Control	(1–3)	±(0.4–2)	≥500		65
Mus. musculus	92,400	2	±0.3	≤24		65
Mus. musculus	24,095	0.5	±0.04	≤24		65
Mus. muscolus	7,510	0.2		≤24		65
Rattus norvegicus	24,095	2	±0.3	≤24		65
Blarina brevicauda	92,400	2	±0.3	≤24		65
Blarina brevicauda	24,095	2	±0.3	≤24		65
Blarina brevicauda	7,510	1	±0.2	≤24		65
Blarina brevicauda	525	2	±0.4	≤24		65
Blarina brevicauda	Control	1	±0.2	≥500		65

Table 8.9. (continued) (9-3)

Species	ADT[a]	PPM[b] (range)	(SD)/±SE	Distance from road (meter)	Notes	Ref.
Sorex cinerus	7,510	0.7		≤24		65
Sorex cinerus	Control	(0.9–4.0)	±2	≥500		65
Cryptotis parva	24,095	1	±0.3	≤24		65
Cryptotis parva	7,510	1	±0.4	≤24		65
Cryptotis parva	525	1	±0.1	≤24		65
Cryptotis parva	Control	(0.8–1)	±0.1–0.3	≥500		65
FISH						
Bluehead chub	15,000	3(2–15)		0	median	74
Bluehead chub	6,500	3(0.2–7)		10	median	74
Bluehead chub	500	2(0.8–4)		100	median	74
Blacknose dace	15,000	3(0.4–9)		0	median	74
Blacknose dace	6,500	3(0.4–9)		10	median	74
Blacknose dace	500	2(0.8–4)		100	median	74
Fantail darter	15,000	6(0.2–15)		0	median	74
Fantail darter	6,500	3(0.2–10)		10	median	74
Fantail darter	500	3(0.2–12)		100	median	74

[a] – ADT = average daily traffic

[b] – Nickel concentrations are arithmetic means (dry weight basis) unless stated otherwise in "Notes" column.

Table 8.10. Zinc concentrations in soils and vegetation in relation to traffic volumes and distance from roads. (Depths of soil sampled and distance from pavement edge are given where appropriate. All concentrations are on a dry weight basis).

Soil	ADT[a]	Depth (cm)	PPM[b] (range)	(SD)/±SE	Distance from road (meter)	Notes	Ref.
Soil	Control	0–10	(93–498)				30
Soil	47,800–44,500	0–2.5	229		3	geometric mean	31
Soil	47,800–44,500		81		6.1	geometric mean	31
Soil	47,800–44,500		71		12.2	geometric mean	31
Soil	47,800–44,500		49		24.4	geometric mean	31
Soil	47,800–44,500		73		48.8	geometric mean	31
Soil	25,100–500		141		3	geometric mean	31
Soil	25,100–500		70		6.1	geometric mean	31
Soil	25,100–500		58		12.2	geometric mean	31
Soil	25,100–500		54		24.4	geometric mean	31
Soil	25,100–500		48		48.8	geometric mean	31
Soil	Control		42			geometric mean	31
Soil	48,000	0–5	162		8		39
Soil	48,000	0–5	110		16		39
Soil	48,000	0–5	44		32		39
Soil	48,000	5–10	86		8		39
Soil	48,000	5–10	28		16		39
Soil	48,000	5–10	20		32		39
Soil	48,000	10–15	36		8		39
Soil	48,000	10–15	20		16		39
Soil	48,000	10–15	18		32		39
Soil	23,000	0–5	72		8		39
Soil	23,000	0–5	60		16		39
Soil	23,000	0–5	34		32		39
Soil	23,000	5–10	24		8		39
Soil	23,000	5–10	16		16		39
Soil	23,000	5–10	11		32		39
Soil	23,000	10–15	11		8		39
Soil	23,000	10–15	10		16		39
Soil	23,000	10–15	8		32		39
Soil	20,000	0–5	172		8		39
Soil	20,000	0–5	66		16		39

Table 8.10. (continued) (10-2)

Soil	ADT[a]	Depth (cm)	PPM[b] (range)	(SD)/±SE	Distance from road (meter)	Notes	Ref.
Soil	20,000	0–5	54		32		39
Soil	20,000	5–10	94		8		39
Soil	20,000	5–10	48		16		39
Soil	20,000	5–10	46		32		39
Soil	20,000	10–15	72		8		39
Soil	20,000	10–15	42		16		39
Soil	20,000	10–15	42		32		39
Soil	7,500	0–5	54		8		39
Soil	7,500	0–5	60		16		39
Soil	7,500	0–5	15		32		39
Soil	7,500	5–10	24		8		39
Soil	7,500	5–10	21		16		39
Soil	7,500	5–10	11		32		39
Soil	7,500	10–15	16		8		39
Soil	7,500	10–15	16		16		39
Soil	7,500	10–15	14		32		39
Soil	92,400	0–2.5	152		6		47
Soil	92,400	0–2.5	47		12		47
Soil	92,400	0–2.5	29		24		47
Soil	92,400	0–2.5	19		48		47
Soil	24,095	0–2.5	72		6		47
Soil	24,095	0–2.5	76		12		47
Soil	24,095	0–2.5	28		24		47
Soil	24,095	0–2.5	25		48		47
Soil	7,510	0–2.5	32		6		47
Soil	7,510	0–2.5	22		12		47
Soil	7,510	0–2.5	15		24		47
Soil	7,510	0–2.5	15		48		47
Soil	525	0–2.5	36		3		47
Soil	525	0–2.5	51		6		47
Soil	525	0–2.5	22		12		47
Soil	525	0–2.5	49		24		47
Soil	525	0–2.5	30		48		47

Table 8.10. (continued) (10-2)

Soil	ADT[a]	Depth (cm)	PPM[b] (range)	(SD)/±SE	Distance from road (meter)	Notes	Ref.
Soil	Control	0–2.5	(15–18)		>500		47
Sediment	15,000		13(6–21)		0	median	74
Sediment	6,500		10(4–16)		10	median	74
Sediment	< 500		8(3–15)		100	median	74
Dust	37,180		8,160				48
Dust	23,730		3,633				48
Dust	21,140		4,883				48
Dust	18,710		4,930				48
Dust	17,970		1,583				48
Dust	9,220		722				48
Dust	7,180		1,083				48
Dust	4,190		423				48
Dust	3,480		980				48
Dust	1,590		1,823				48
Dust	1,550		2,870				48
Dust	980		720				48
Dust	510		2,257				48
Dust	20		220				48
Grass	48,000		40		8	light rinse	39
Grass	48,000		35		16	light rinse	39
Grass	48,000		30		32	light rinse	39
Grass	23,000		85		8	light rinse	39
Grass	23,000		72		16	light rinse	39
Grass	23,000		67		32	light rinse	39
Grass	20,000		32		8	light rinse	39
Grass	20,000		29		16	light rinse	39
Grass	20,000		27		32	light rinse	39
Grass	7,500		36		8	light rinse	39
Grass	7,500		33		16	light rinse	39
Grass	7,500		29		32	light rinse	39
Mixed	92,400		219		6	mixed leaves unwashed	47
Mixed	92,400		98		12	mixed leaves unwashed	47
Mixed	92,400		86		24	mixed leaves unwashed	47

Table 8.10. (continued) (10-3)

Soil	ADT[a]	Depth (cm)	PPM[b] (range)	(SD)/±SE	Distance from road (meter)	Notes	Ref.
Mixed	92,400		63		48	mixed leaves unwashed	47
Mixed	24,095		81		6	mixed leaves unwashed	47
Mixed	24,095		71		12	mixed leaves unwashed	47
Mixed	24,095		68		24	mixed leaves unwashed	47
Mixed	24,095		52		48	mixed leaves unwashed	47
Mixed	7,510		55		3	mixed leaves unwashed	47
Mixed	7,510		58		6	mixed leaves unwashed	47
Mixed	7,510		71		12	mixed leaves unwashed	47
Mixed	7,510		70		24	mixed leaves unwashed	47
Mixed	7,510		39		48	mixed leaves unwashed	47
Mixed	525		45		3	mixed leaves unwashed	47
Mixed	525		54		6	mixed leaves unwashed	47
Mixed	525		39		12	mixed leaves unwashed	47
Mixed	525		34		24	mixed leaves unwashed	47
Mixed	525		33		48	mixed leaves unwashed	47
Mixed	Control		(38–54)		≥500	mixed leaves unwashed	47

[a] – ADT = average daily traffic
[b] – Zinc concentrations are arithmetic means (dry weight basis) unless stated otherwise in "Notes" column.

Table 8.11. **Zinc concentrations in invertebrates and vertebrates in relation to traffic volumes and distance from roads. (Zinc concentrations represent whole body on a dry weight basis. Earthworms were allowed to cast alimentary tract soil.)**

Species	ADT[a]	PPM[b] (range)	(SD)/± SE	Distance from road (meter)	Notes	Ref.
EARTHWORMS						
Alabophera, Lumbricus, & Octolasium	control	(93–502)				30
Lumbricus & Allobophora	47,800 – 44,500	495		3	geometric mean	31
	47,800 – 44,500	473		6.1	geometric mean	31
	47,800 – 44,500	509		12.2	geometric mean	31
	47,800 – 44,500	400		24.4	geometric mean	31
	47,800 – 44,500	408		48.8	geometric mean	31
	25,100 – 500	670		3	geometric mean	31
	25,100 – 500	343		6.1	geometric mean	31
	25,100 – 500	333		12.2	geometric mean	31
	25,100 – 500	258		24.4	geometric mean	31
	25,100 – 500	146		48.8	geometric mean	31
"Earthworm"	92,400	654	±148	6		65
	92,400	594	±36	12		65
	92,400	1475	±21	24		65
	92,400	646	±210	48		65
	24,095	785	±79	6		65
	24,095	894	±88	12		65
	24,095	537	±48	24		65
	24,095	462	±46	48		65

Table 8.11. (continued) (11-2)

Species	ADT[a]	PPM[b] (range)	(SD)/± SE	Distance from road (meter)	Notes	Ref.
"Earthworm"	7,510	514	±53	6		65
	7,510	518	±43	12		65
	7,510	417	±18	24		65
	7,510	427	±46	48		65
	525	759	±108	3		65
	525	479	±69	6		65
	525	227	±18	12		65
	525	432	±49	24		65
	525	391	±63	48		65
	control	(223–362)	±(40–39)	≥500		65
BENTHIC INSECTS						
Tipulidae	15,000	106(64–217)		0	median	74
Tipulidae	6,500	92(50–486)		10	median	74
Tipulidae	500	83(61–170)		100	median	74
Perlidae	15,000	204(154–287)		0	median	74
Perlidae	6,500	227(155–527)		10	median	74
Perlidae	500	235(112–454)		100	median	74
Pteronarcidae	15,000	240(217–341)		0	median	74
Pternonarcidae	6,500	232(135–413)		10	median	74
Pteronarcidae	500	254(41–757)		100	median	74
MAMMALS						
Microtus pennsylvanicus	92,400	119	±10	≤24		65
M. pennsylvanicus	24,095	103	±3	≤24		65
M. pennsylvanicus	7,510	105	±4	≤24		65
M. pennsylvanicus	525	97	±3	≤24		65
M. pennsylvanicus	control	(92–107)	±(14–3)	≥500		65
Peromyscus leucopus	92,400	110	±14	≤24		65
P. leucopus	24,095	104	±10	≤24		65
P. leucopus	7,510	90	±6	≤24		65
P. leucopus	525	105	±3	≤24		65

Table 8.11. (continued) (11-3)

Species	ADT[a]	PPM[b] (range)	(SD)/± SE	Distance from road (meter)	Notes	Ref.
P. leucopus	control	(83–107)	±(5–10)	≥500		65
Mus musculus	92,400	99	±7	24		65
Mus musculus	24,095	85	±19	24		65
M. musculus	7,510	51		24		65
Rattus norvegicus	24,095	135	±16	24		65
Blarina brevicauda	92,400	139	±9	24		65
B. brevicauda	24,095	122	±7	24		65
B. brevicauda	7,510	123	±7	24		65
B. Brevicauda	525	100	±7	24		65
B. brevicauda	control	(106–112)		≥500		65
Sorex cinereus	7,510	146		≤24		65
S. cinereus	control	106		≥500		65
Cryptotis parva	24,095	111	±9	≤24		65
C. parva	7,510	123	±9	≤24		65
C. parva	525	109	±6	≤24		65
C. parva	control	(109–118)		≥500		65
FISH						
Bluehead chub	15,000	98(82–171)		0	median	74
Bluehead chub	6,500	94(84–288)		10	median	74
Bluehead chub	500	76(42–88)		100	median	74
Blacknose dace	15,000	331(187–388)		0	median	74
Blacknose dace	6,500	248(103–588)		10	median	74
Blacknose dace	500	151(111–257)		100	median	74
Fantail darter	15,000	147(91–224)		0	median	74
Fantail darter	6,500	115(92–168)		10	median	74
Fantail darter	500	112(87–128)		100	median	74

[a] – ADT = Average daily traffic

[b] – Zinc concentrations are arithmetic means (dry weight basis) unless stated otherwise in "notes" column.

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Chapter 9

NOISE POLLUTION

K. Attenborough

Contents **Page**

1. INTRODUCTION

More people experience nuisance from road traffic noise than from any other noise. That there is need to take action on traffic noise in industrialised Europe is shown by a recent publication [1] of the Organisation for Economic Cooperation and Development. This indicates that in the early 1980's approximately 120 million people (about 15% of the total population) were exposed already to noise levels that would qualify for home insulation in the U.K. Although recent action on noise sources both at source and through urban design is likely to reduce the highest levels, the spread of exposure to intermediate levels which are sufficient to cause annoyance and discomfort warrants further efforts. The problem is worst in the industrially developed countries but traffic noise is beginning to have a significant affect in newly industrialised parts of the world. The climate in such places as Korea and Malaysia and the relatively small proportion of air conditioned buildings require open windows thus allowing particularly high levels of noise in schools, offices and homes. The relatively high proportion of noisy motorcycles exacerbates the traffic noise problem.

Early awareness of the need for noise control and of the options for noise control can lead to measures that have lasting and widespread effectiveness rather than being merely remedial and local in their impact. In this chapter after reviewing basic descriptions of sound and subjective response to sound, methods of assessing traffic noise impact in the community are outlined. The sources of traffic noise are discussed and brief details of methods of control at source are given.Methods of measuring and monitoring traffic noise and highway –induced vibrations are described. Factors to be considered in forecasting the propagation of sound outdoors and in prediction schemes for noise both from free flowing traffic and from stop–start traffic are introduced. Finally methods of traffic noise control are detailed.

2. BASIC ACOUSTICS AND SUBJECTIVE RESPONSES

2.1 Sound pressure, frequency and wavelength [2]

Noise is unwanted sound. Sound is a form of energy that is an inevitable consequence of mechanical processes. On the other hand it represents only a small fraction of the total energy involved in a mechanical process.

Sound may be characterised by frequency or wavelength and pressure. An example of a single frequency sound is that from a tuning fork. Frequency is a physical measure or assessment of the subjective sensation of pitch. The separation between frequencies is often described in terms of octaves. A separation of one octave means that the higher frequency is twice the lower. Traffic noise contains sounds of many different frequencies extending over several octaves from 50 Hz to 6400 Hz. In air at sea level and 20 deg C,the velocity of sound is approximately 340 m/s.

The pressure of a single–frequency sound is defined as the root mean square(rms) of the pressure difference from atmospheric pressure over one cycle of the pressure variation (in time). The accepted MKS unit for pressure is the Newton per square metre (N/m^2) or Pascal(Pa). Rather than expressing sound pressure in Pa it is customary to use a logarithmic scale that is better suited to the non–linear response of the ear and will give a more convenient range of numbers for the pressures of interest. A reference level(P_o) of $2x10^{-5}$ Pa is adopted and corresponds approximately to the lowest sound pressure detectable to normal hearing at 1000 Hz. The sound pressure level in Decibels(dB) equivalent to a sound pressure (P) may then be calculated from

$$20 \log(P/P_o).$$

If any one frequency in a noise could be filtered out and its time history studied it would be found to vary randomly with time. The only way of describing the level at this frequency would be to break the frequency range of the total noise signal into a series of contiguous frequency bands and to measure the signal level in each frequency band over a sufficiently long time to obtain a meaningful average.

The resulting frequency information is known as a frequency spectrum. If the chosen width of the contiguous frequency bands is an octave then the result is known as an octave band spectrum and is a suitable form of analysis for a broad–band sound that does not contain any dominant frequencies. One of the standard octave bands in audio–frequency analysis is that centred at 1000 Hz.

The other octave band centre frequencies are related to 1000 Hz by the factor $10^{-0.3N}$ where N is any integer. The upper and lower limits of the octave bands are given by $10^{-0.15}$ multiplied by the centre frequency. More detailed information about a noise spectrum is given by third–octave analysis. The preferred centre frequencies for both octave and third octave analysis are given in Table 9.1.

Table 9.1. **Preferred centre frequencies for octave and third octave bands**

Preferred Centre Frequency in Hz

25	* 250	2500
* 31.5	315	3150
40	400	* 4000
50	* 500	5000
* 63	630	6300
80	800	* 8000
100	* 1000	10000
* 125	1250	12500
160	1600	* 16000
200	* 2000	20000

* indicates octave band values

Figures 9.1 and 9.2 show typical spectra from individual vehicles. It is noticeable that both large and small vehicles generate considerable sound energy near 500 Hz and that the larger vehicle produces considerable sound energy below 250 Hz.

Human hearing is not equally sensitive at all frequencies. Furthermore the variation of sensitivity with frequency is a function of level. The loudness of a sound is a subjective indication of its level and human judgement of loudness has been investigated widely. The loudness of a sound in *phons* is defined as the sound pressure level of a 1000 Hz tone that is judged to be as loud as the sound of interest. This is found to be a function of frequency and of level. Contours of equal loudness have been obtained between zero and 100 phons. A frequency–weighting that is adopted in most noise measurements is one that corresponds approximately to the 40 phon contour and hence to the frequency characteristic of the ear at low levels of sound.

This characteristic is known as 'A' weighting and the resulting measurements of sound level are in dBA. Figure 9.3 shows a graph of the 'A' weighting curve. It is noticeable that the curve emphasises frequencies in the range 1000 Hz to 3000 Hz at the expense particularly of frequencies below 500 Hz.

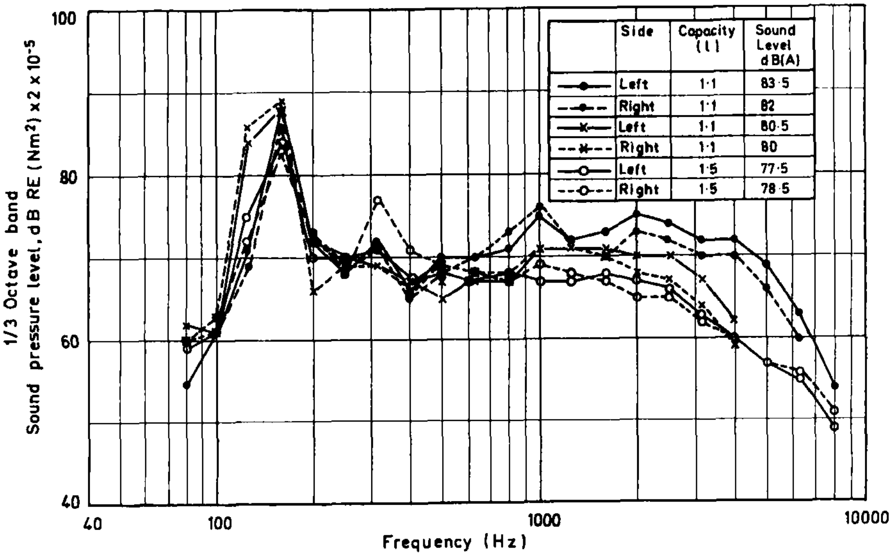

FIGURE 9.1 *SPECTRUM OF NOISE EMITTED BY SMALL CARS*

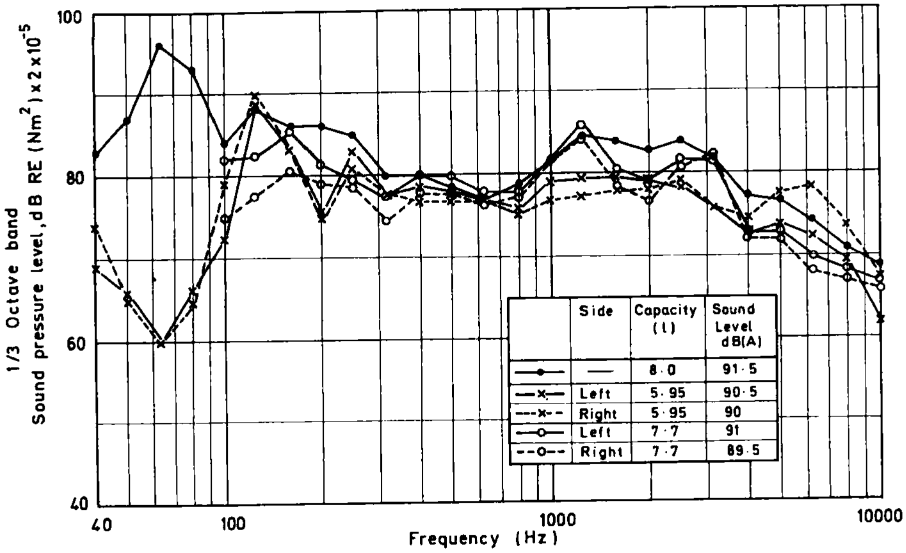

FIGURE 9.2 *SPECTRUM OF NOISE EMITTED BY HEAVY GOODS VEHICLE*

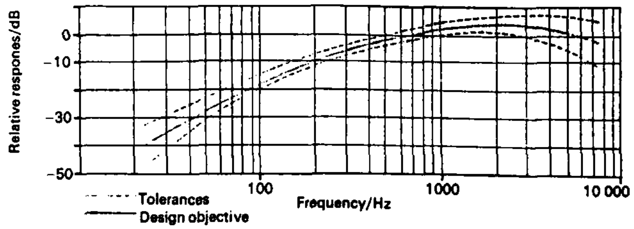

FIGURE 9.3 *A–weighting Values and Tolerances for a Sound Level Meter*

2.2 Absorption and propagation

When sound encounters a solid surface then part of the sound energy is reflected and the remainder is absorbed. If the surface is that of a barrier or a building then there will also be diffraction of the sound around the obstacle and transmission through it. The fraction of energy that is absorbed is defined as the *absorption coefficient*. The difference in sound pressure levels either side of a barrier in the absence of any other paths than that directly through the panel is defined as the transmission loss. Absorption,transmission and diffraction are all frequency dependant. Usually absorption tends to increase with frequency; whereas diffraction and transmission tend to be greatest at low frequencies. By definition a material that offers good absorption will have a poor transmission loss.

An environment in which there is no reflection is called an *anechoic* environmment. Such an environment has to created artificially by lining an enclosure with highly absorbing material or is approximated by open space well above the ground. Under these circumstances the way in which sound travels or *propagates* and decays with distance depends upon the size and form of the source.

For distances large compared with the dimensions of the source the sound waves expand with spherical wave fronts as though from a point; rather like the ripples expanding from the impact of a small stone thrown into still water. Under these circumstances,and in the absence of any nearby reflecting surfaces, the sound pressure level will decrease at a rate of 6 dB per doubling of distance. This will be the case for an individual vehicle at distances of more than twice its largest dimension. A continuous stream of traffic moving along a highway behaves more like a line source at large distances. The resulting rate of decay of the sound pressure level is 3 dB per doubling of distance. The presence of porous ground, various obstacles and the influences of meteorological conditions complicate this simple rule. These complications are discussed in a later section.

2.3. Noise units and indices

Time varying noise,such as traffic noise,may be described in terms of the levels exceeded for certain fractions of the time. These levels are called percentile levels. Frequently used percentile levels are denoted by $L_{A_{10}}$,$L_{A_{50}}$ and $L_{A_{90}}$ representing respectively the A weighted sound pressure levels exceeded for 10%, 50% and 90% of the time. In particular the 18 hour $L_{A_{10}}$ measured during the period 0600 to 2400 GMT, known as the L_{10} index has been used as the noise index for assessing and predicting noise from free flowing road traffic.

The 10% level may, of course, be measured over any period. Typically 5 to 15 minute samples are measured in any one hour. A minimum sampling period may be calculated from

$$t_{min} = \left\{ \frac{4000}{q} + \frac{120}{s} \right\}$$

where q is the vehicle flow rate in vehicles/hour and s is the sampling rate in samples/min. This is only valid for periods between 5 and 55 minutes. A sample $L_{A_{10}}$ measured during a peak flow hour will overestimate the 18 hour value by approximately 3 dBA.

An alternative noise index that is used more widely than L_A is the period $L_{A_{eq}}$. This is the equivalent continuous 'A' weighted sound pressure level and is defined as that constant level resulting in the same total sound energy over the given period as the actual varying sound level over the same period.
Thus

$$L_{Aeq} = 10 \log \left[\frac{1}{T} \int_0^T 10^{\frac{L_A}{10}} dt \right]$$

which represents a logarithmic form of the linear rms value of the signal over the period

T.A common period for the L_{Aeq} measurement is 24 hours. The Day/Night equivalent sound level,which originated in the U.S.A., represents a 24 hour L_{Aeq} in which the noise during the night time period, 2200 to 0700 hours is penalised by the addition of 10 dBA. There is an empirical relationship between the L_{Aeq} and $L_{A_{10}}$ values for traffic noise measured over 18 hours. This is valid for traffic flows greater than 100 vehicles per hour and is

$$L_{A_{10}} = L_{Aeq} + 3 \text{ dBA}$$

A noise unit that may be used to describe a single vehicle pass by is called the *single event noise exposure level* and represents the corresponding L_{Aeq} normalised to a time interval of 1 second. It is variously denoted by SEL, L_{AX} or L_{AE}.

The L_{Aeq} for the period T including the passage of several vehicles for each of which the L_{AX} is known may be calculated from

$$L_{Aeq} = 10 \log \left[\frac{1}{T} \sum_{i=1}^{n} 10^{\frac{L_{AX_i}}{10}} \right] \text{ dBA.}$$

2.4 Social and laboratory surveys

The standard definition of noise as unwanted sound implies a subjective component to the assessment of noise. As might be expected, research shows that the effects of highway noise are modified but not eliminated by a variety of personal and community characteristics. The final result is that individuals' responses to the same sound vary considerably even though the averages of these responses are systematically related to noise level.

Two types of study methods provide most of the evidence about the effects of noise on people and the characteristics which mediate those effects. In laboratory studies, people are asked to rate controlled presentations of sounds. In field studies, people are asked to report feelings that have formed from years of exposure to noise in their everyday setting. Laboratory studies provide most of the information about the importance of physical and measurement characteristics, such as the presence of tonal components, duration and different weighting networks. Furthermore laboratory studies have investigated speech interference effects and task interference effects.

Social surveys measure annoyance by soliciting ratings on various scales from a carefully selected sample of people and by carrying out simultaneous objective measurements. For example the survey that provided the basis for the use of the 18 hr $L_{A_{10}}$ index for highway noise in the U.K. used a seven point scale of dissatisfaction and sampled the noise level in the relevant neighbourhoods for 2 minutes in every hour over 24 hours.

It was concluded from associated questions on activity disturbance and attitudes to noise that the label 'dissatisfaction' would produce responses that would be a reliable measure of annoyance. Average (median) dissatisfaction scores were correlated with various physical measures of exposure including 24 hr $L_{A_{10}}$, $L_{A_{90}}$, and $L_{A_{50}}$. These quantities were calculated by taking the average of the appropriate 24 hr samples. The best correlation was found with a 24 hr composite index, called the Traffic Noise Index(TNI) viz.

$$\mathrm{TNI} = L_{A_{10}} + 4(L_{A_{10}} - L_{A_{90}}) - 30 .$$

The 18 hr LA_{10} index was found to have a worse correlation with median dissatisfaction (figure 9.4) but has been preferred because it may be measured and predicted in a relatively straightforward manner. Outside the UK, indices based on 24 hr LAeq are used widely and do not offer a significantly worse correlation with median annoyance than does the LA_{10} index. Altogether some 200 combined social and physical noise surveys have been carried out with regard to transportation noise throughout the world. Any new survey would have to be extensive indeed to add significantly to the existing body of knowledge[3,4].

3. VEHICLE NOISE SOURCES

The sources of airborne noise from a road vehicle are shown schematically in Figure 9.5. This figure shows also the relative contributions to overall noise levels to be expected in different types of vehicle. The engine–related noises include combustion noise, piston slap, timing drive noise and noises due to bearing forces and rotation in addition to those mentioned explicitly on tHe figure. The relative contributions depend on road speed and conditions. Most noise from present day petrol and diesel powered vehicles moving at low road speeds and in low gears comes from the engine and exhaust. At high speeds and in top gear, tyre noise is greater than power train noise. At speeds of 44 kph(30 mph), tyre noise contributes between 10 and 20 dBA less than the engine but on wet roads the noise levels from both sources are comparable.

3.1 Noise control at source

Combustion noise is produced by the varying loads applied to the engine structure by the

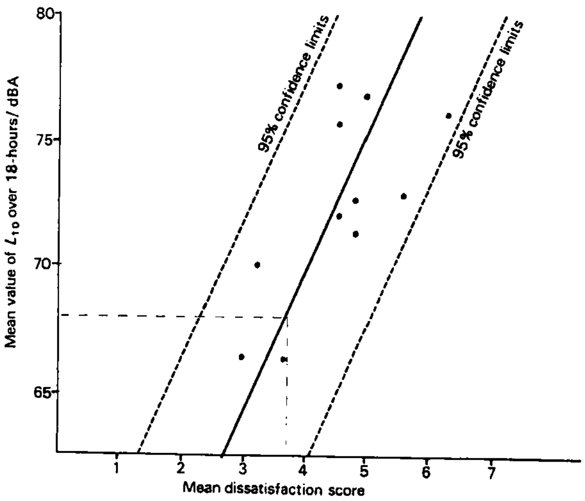

FIGURE 9.4 *Relationship between L_{10} Index and Mean Dissatisfaction Score The dotted lines represent 95% confidence limits.*

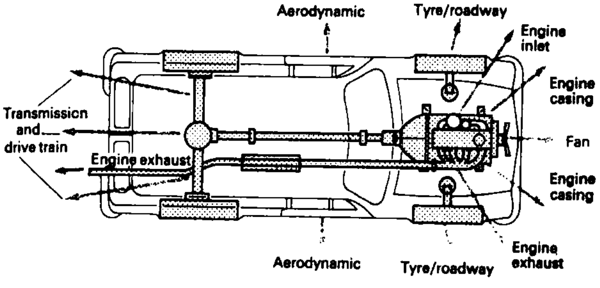

FIGURE 9.5 *Sources of Airborne Sound from a Road Vehicle*

combustion process. Mechanical noise is the general term for vibration applied to the vehicle structure by impacts from the pistons (piston slap), by the timing gears driving the camshaft and fuel pump and by the bearings and valve gear.

Combustion noise is the primary problem with diesel engines and is a significant component with modern petrol engines which are designed with lean mixtures, advanced ignition timing and high compression ratios. An effective means of control requires some form of retardation of the firing. Fuel is injected into the cylinder near the top of the ignition stroke. When mixed with air at a high enough temperature and pressure the fuel ignites. The period between the start of the injection and ignition, known as ignition delay period has an important bearing on combustion noise. Turbocharging, by which air is forced into the intake at pressures higher than atmospheric pressure, results in a shorter delay period and a more gradual rise in pressure with,consequently, lower noise generation, as well as greater power output. It represents a way of obtaining greater timing retard without sacrificing performance.

Piston slap may be controlled by a positively injected air film that cushions the impact, by additional scraper rings, Teflon inserts, differential expansion of piston and cylinder bore material and offset gudgeon pins. Designs that place the gear train at the rear of the engine rather than the front will reduce noise at the front of the engine by up to 12 dBA. Further mechanical noise reduction at source may be obtained by use of toothed belts instead of chain drives and by crankcase and cylinder block restructuring.

Enclosures and shielding have also been used to great effect in the reduction of mechanical noise.Tunnel–type enclosures, open at the front and rear of the engine compartment to permit cooling, are common. Cooling fan design may also assist noise reduction. Development of the package concept system including radiators and cowling is notable in this regard.

Exhaust noise is tackled by use of silencers. However consideration of silencer design is beyond the scope of this chapter.

3.2 Noise from road/tyre interaction

There are several possible sources of tyre noise. These include aerodynamic noise from the rotation of the wheel and the tyre, noise from the vibration of the tyre surface, and pressure fluctuations caused by pumping of air in the tread grooves in the area of contact with the road surface.

The main factors affecting noise from the interaction between the tyres of a Heavy Goods Vehicle and the road are (1) the speed of the vehicle (2) the weight of the vehicle (3) the tread pattern (4) the tyre structure (5) tread materials (6) tyre wear and (7) road surface texture.

The peak noise level in dBA measured at 7.5m along the normal to the centre line of the path of a coasting vehicle is related to the speed such that the noise level increases by between 9 and 13 dBA per doubling of speed, depending on the road texture. The greater rate of increase tends to be associated with regular features in the tyre tread pattern and in the road surface. The greatest effect of vehicle weight is found when the vehicle is fitted with traction tyres. Indeed these tyres are in any case about 1 dBA noisier than equivalent ribbed patterned tyres. Tyres with regular transverse features such as traction block patterns can generate tonal characteristics associated with the impact of the block patterns on the road surface.

On most road surfaces, radial ply tyres in which component rayon or nylon chords cross the circumferential centre line almost at right angles are between 1 and 2 dBA quieter than cross ply tyres, where the ply chords are at an oblique angle to the centre line. As tyre wear increases so does the noise level; the maximum increase being reached when the tyre is 50 to 70% worn.

Apart from regular features in the road surface such as transverse grooves or brush marks which often are added to improve skidding resistance, the principal road surface characteristics that affect tyre noise generation are the presence of surface water and the acoustical absorption of the surface material. The increase in noise levels due to surface water ranges from 1 to 10 dBA, depending upon the amount of water present, and mainly occurs in the part of the noise spectrum above 2000 Hz.

Open textured pervious macadam originally designed to allow water to drain rapidly through the surface matrix to an impervious substrate has been found to reduce tyre/road noise levels by an average of 3 dBA when compared to conventional smooth concrete or other standard surfaces with the same skidding resistance. The effect may be attributed to the increase in the absorption coefficient in the frequency range 630 to 1000 Hz associated with the higher porosity of the surface. Maximum reductions of up to 9dBA have been observed but such performance has been found to degrade as the pores of the road surface become clogged with debris.

4. MEASURING AND MONITORING TRAFFIC NOISE

4.1 Instrumentation

The development of transistors and microprocessors has enabled the manufacture of many,reasonably portable, instruments, called sound level meters,capable of direct measurement of many of the noise units and indices described in section 2. A minimum assembly of parts simply for measuring A weighted sound pressure level is shown in Figure 9.6.

The sound wave is detected by the microphone, usually a condenser microphone, which converts the incident sound pressure fluctuation into a varying voltage. This electrical signal is amplified and rectified before passing to a read–out meter giving a deflection proportional to the electrical signal. After amplification the signal is passed through a series of filters giving the required frequency weighting.

The speed with which the read out meter movement responds may be adjustable. As a sound level meter is the basis for many measurements required by noise standards it is important that it is reliable and accurate. Furthermore it is important that the difference between readings of the same noise level made with different makes of sound level meter is reduced to the practical minimum. For these reasons there are British (BSI), American(ANSI), West German(DIN), East German(GDR), Belgian, Dutch, French, South African, Australian and many other nationalities' standards that specify the performance to be expected of sound level meters. In addition there are several international standards published by the International Electrotechnical Commission(IEC).

Typically these specify

(a) the directionality
(b) the frequency response
(c) the characteristics of the read out indication
(d) the electrical (self) noise floor and
(e) the method of checking the sensitivity adjustment and calibration of the instrument.

Ideally a sound level meter would be equally sensitive to sound from all angles. In practice the sensitivity is greatest to sound that is incident normal to the microphone. The standards lay down the maximum deviations with angle of incidence that are permitted. The frequency range over which a sound level meter is accurate is strongly dependent on the microphone. Figure 9.3 shows a typical permitted tolerance for the accuracy of A weighting. The scale length, graduation density, accuracy and movement characteristics of

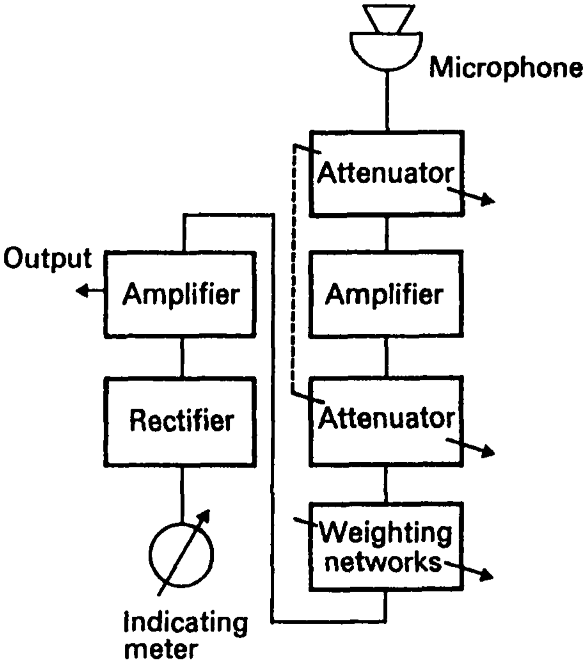

FIGURE 9.6 *Block Diagram of the Concentration of a Sound Level Meter*

the read out meter are specified. Standards for electrical system noise are intended to ensure that this does not influence readings of external noise.

Each of the various nation's standards describe different grades for sound level meters. These are differentiated by the accuracy expected of them. Road traffic noise measurements should be made on the specified appropriate grade of instrument the requirements for which will be less demanding than that those for laboratory measurements.

Moisture and humidity are great enemies of condenser microphones. The microphone capsule must be internally vented to atmosphere to compensate for static atmospheric pressure changes. If moisture can find its way into the gap between the diaphragm and the electrode, it prevents the proper functioning of the microphone. The moisture may be dried out but if it contains impurities these may remain to the eventual detriment of the instrument. For short term outdoor measurements the only precautions that are necessary are to avoid rain and any contact with wet surfaces. For a permanent installation it is important to use a system that vents the microphone through a dehumidifier and provides proection against rain and animals (particularly birds). The other problem encountered outdoors is the pseudo–noise associated with wind turbulence. This is reduced but not eliminated by the use of a windshield. A commonly used and reasonably effective form of windshield is a 90 mm diameter sphere of porous polyurethane foam. However, even with a windshield, measurements should not be taken when the residual wind noise is likely to be within 10 dB of the noise to be measured. In practice this means that measurements should not be taken when the wind speed is more than about 5 m/s.

4.2 Measurement procedures

The International Standard for measuring the noise emitted by individual vehicles is intended for type testing rather than roadside checks. The site is required to be flat and hard within a 50 m radius of the vehicle and there should be no obstructions or nearby reflecting surfaces. The microphone of the measuring instrumentation should be 1.2 m above the ground and 7.5 m from the centre line of the vehicle. The British Standard also requires that the background noise level should be at least 10 dBA below the peak noise level from the vehicle.

In Great Britain the measurement procedure for general traffic noise near dwellings is well defined. The microphone should be located at the appropriate height and 1 m away from the noise affected facade. The road surface must be dry and the wind speed should be less than 2 m/s unless the dominant component is from source to receiver in which case, as long as the residual background criterion is not violated, the wind speed may be as much as 10 m/s.

5. HIGHWAY INDUCED VIBRATION

Two mechanisms for traffic–induced vibration in buildings have been identified. Firstly, low frequency sound waves generated by vehicles, mainly at the exhaust, couple into the structure through the windows and doors causing different parts of the building to vibrate. Secondly, forces generated by vehicles passing over the road surface profile induce vibration of the ground which in turn, at close range, causes vibration of building foundations [5].

Another possible mechanism, as yet unexplored in connection with buildings, is acoustic to seismic coupling; whereby airborne sound couples directly to the ground such that maximum induced displacements occur at seismic resonance frequencies determined by the layering of the ground.

The peaks in the traffic noise spectrum in the frequency range between 40 and 125 Hz are mainly attributable to heavy vehicles and are implicated in vibration due to direct airborne noise coupling. The 1 hr equivalent continous noise level in this frequency range at 1.2m above the ground and at a reference distance of 10m from the nearside kerb may be predicted from

$$L_{eq}\,(40\text{–}125\text{ Hz}) = 53.0 + 9.43\log_{10}(Q_t + 10Q_{MCV} + 40Q_{HCV})\text{dB}$$

where Q is the total traffic flow in vehicles per hour,Q_{MCV} is the hourly flow of medium commercial vehicles and Q_{HCV} is the hourly flow of heavy vehicles. Greatest vibration amplitudes occur when room or floor resonances are excited. Typical resonant frequencies are

	beams	5 – 50 Hz
	floors and slabs	10 – 20 Hz
	window panes	10 –100 Hz
and	plaster ceilings	10 – 20 Hz.

The ground vibration induced directly by the vehicle/road interaction is usually in the form of rapidly damped Rayleigh waves. Consequently the greatest impact is on nearby stuctures.

Subjective response to low frequency noise from vehicles is affected by body resonances. In particular chest cavity resonances occur in the range 64 to 74 Hz. Otherwise, subjective response is governed by perceived nuisance or worry about damage induced by rattling of window panes, radiators and fittings. The amplitude of vibrations induced by traffic is not sufficient over a short period to cause significant structural damage but research is continuing into possible long term fatigue effects.

6. FACTORS AFFECTING PROPAGATION OF TRAFFIC NOISE

6.1 Point sources and inverse square law

Consider sound spreading spherically from a source that may be regarded as a point. This will be true for example at several wavelengths from a vehicle. The surface area of the spherical wave fronts will be increasing with the square of the radius. Consequently the energy density will be decreasing as the inverse square of the radius. For such propagation that is unobstructed the sound pressure will be proportional to the square root of the energy density (or intensity).

This means that for spherical wave propagation, the pressure will decrease as the inverse of the distance from the source.

6.2 Ground effect

If a hard surface is placed beneath a point source, then the volume into which it can radiate is restricted and the wave fronts become hemispherical. The sound pressure at any receiver position around the source may be calculated as the sum of the direct contribution from the source plus a contribution from a mirror–image source below the hard boundary. This means that sound still decays as the inverse of distance but at points close to the boundary the sound pressure will have double the value it would have had, i.e. it will be 6dB more than it would have been, at those positions in the absence of the boundary. Moreover at frequencies where the difference in the lengths of the direct and reflected paths corresponds to an odd number of half wavelengths there will be destructive interference and the sound pressure level has a theoretical value of zero.

For a receiver 1 m above a hard boundary and 20 m away from a sound source which is 0.5m above the boundary, the first of these interferences will occur at 3405 Hz. An example of an acoustically hard ground is an asphalt surface. Grassland, cultivated land and forested areas, however, offer acoustically soft surfaces. The porosity of the surface allows the movement of the air into the ground associated with passage of a sound wave above the ground surface.

The air permeability of the surface has a large influence on the acoustical properties. Since the penetration of air into the ground and the resulting reaction of the air mass within the ground are not instantaneous there is a phase lag between incident and ground–reflected sound. This phase difference adds to that associated with path length difference between direct and reflected waves arriving at a receiver in the presence of ground so it is possible for the first destructive interference to occur at a lower frequency than over hard ground. Indeed, for a range of source–receiver geometries of interest to highway noise studies and for a typical range of ground surfaces, the first destructive interference dip known as the ground effect dip will lie between 250 Hz and 800 Hz. This range of frequencies covers an important part of the traffic noise spectrum.

An empirical formula for predicting the frequency at which the ground effect dip and hence the maximum attenuation occurs is[6]:

$$f_{max} = \frac{1500}{h \log_{10}(r/0.3)}$$

and the amplitude of the maximum attenuation is

$$A_g = 15 \log_{10}(0.065r/h)$$

where h is the mean path height and r is the distance from the source to the receiver. The amplitude of the attenuation in adjacent octave bands may be taken to be Ag/2. Strictly, these equations are valid only for a certain kind of grass surface. As remarked above, the ground effect is dependent upon the properties of the surface. The more permeable the surface the lower the frequency at which the first interference dip occurs and the smaller the magnitude of the dip. Typically, both snow and the ground surfaces beneath mature forest stands offer relatively high porosity and high air permeability compared with grassland or cultivated soils. At long ranges and for the source and receiver heights of typical interest in noise control, the above formulae would overestimate the frequency and the amplitude of maximum attenuation by approximately 300 Hz and 15 dB respectively. Table 9.2 gives values of typical flow resistivities, which are related to the inverse of air permeabilities, for a range of ground materials. Table 9.3 shows expected total attenuations of percentile noise levels in dBA per doubling of distance over four different types of ground surface.

Table 9.2 Typical relative effective flow resistivities for ground surfaces

Surface	Relative Effective Flow Resistivity
newly fallen snow	0.1
packed snow	0.3
mature forests	0.5
grass	1.0
dirt road	4.0
asphalt	> 100

Table 9.3 Expected total attenuations of percentile levels in dBA per doubling of distance over various ground surfaces.

	Concrete	Grass	Cultivated	Cornfield
L_{10}	3.2	4.5	5.4	6.6
L_{50}	2.5	3.3	4.3	5.0
L_{90}	1.8	2.5	3.2	3.4

6.3 Wind and temperature gradients

As discussed in Chapter 3, wind speed increases with height above the ground. The vector wind adds to the sound speed; upwind the net speed is lower and downwind the net speed is higher than in still air relative to fixed ground coordinates. So, ignoring temperature effects for the moment, upwind of a sound source,the higher the sound travels,the lower the effective sound speed and sound rays are refracted away from the ground. This means that a receiver sufficiently far from a source and close to the ground will be in a sound shadow. Downwind of the source there will be downward refraction and the sound will not decay as rapidly as it would in a still, homogeneous atmosphere. These situations are illustrated in Figure 9.7.

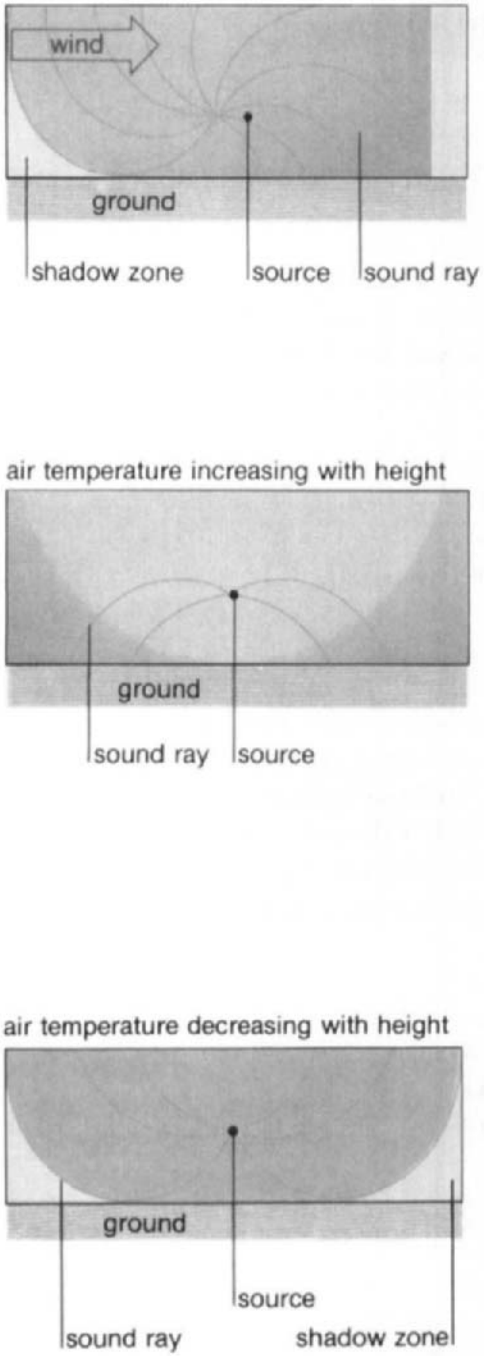

FIGURE 9.7 ***Sound–Ray Bending Due to Velocity Gradients in the Atmosphere***

To a first approximation the rate of change of sound speed with height is proportional to the temperature gradient. Typically during the day, when the ground is warmest, the temperature decreases with height. Under this condition the sound speed also decreases with height and sound is refracted upwards, creating a shadow zone all around the source unlike the shadow created by wind which only occurs upwind of the source.

In late evening when skies are clear and conditions are calm,the ground cools rapidly and it is possible that a relatively cool layer of air may be trapped by warmer air above. This is an inversion condition.Under such a condition sound from a source below the inversion layer is refracted towards the ground and similarly trapped giving rise to higher levels at receivers close to the ground than would be expected in a homogeneous atmosphere.

The presence of even a little wind, say greater than 2 m/s at 1m above the ground, results in considerable mixing of the air layers and the break–up of any inversion. Usually the presence of wind means that wind gradients together with ground effects rather than temperature gradient effects dominate sound propagation close to the ground for several hundred metres.

6.4 Reflecting surfaces and barriers

The sound level in the vicinity of a vertical reflecting surface, like a building facade, is the result of both direct and reflected sound waves. Right at the surface both components are in phase and an increase of 6 dB above the level that would exist in the absence of the vertical surface is to be expected. The effect of the surface will become less as the measuring point is moved away from it, becoming negligible in practice at distances greater than 10 m from the surface. For traffic noise the accepted increase in level at 1m from a building facade is 2.5 dBA.

Sound waves tend to bend around the top and the ends of a barrier like water waves around a breakwater. The extent of this diffraction phenomenon is frequency dependent being greater the lower the frequency. The mass of the barrier is also important but diffraction means that no advantage is gained by increasing the mass beyond a certain limit.

For most practical applications the minimum mass per unit area (M) of barrier material may be determined from the required noise reduction and the site geometry according to

$$M = 3\,\mathrm{antilog}\left[\frac{\Delta L - 10}{14}\right]\ \mathrm{kg/m^2}$$

where ΔL is the potential barrier attenuation in dB.

The attenuation of sound from a point source by an infinitely long thin barrier of sufficient mass that direct transmission may be ignored can be determined approximately from

$$A = 10\log_{10}(3 + 20N)\ \mathrm{dB}$$

where

$$N = \pm\frac{\text{path difference}}{\text{wavelength}}$$

For $N = 0$, corresponding to minimum line of sight across the barrier, the attenuation is approximately 5 dBA. Positive values of N correspond to locations in the geometrical shadow zone and negative values correspond to illuminated locations. N can never be less than -0.1.

In practice the effectiveness of barriers may be reduced by meteorological effects. In particular turbulent scattering above and refraction over the barrier may increase the noise levels behind it.Moreover a barrier of finite length gives a reduced attenuation compared with that predicted by the above formula as a result of noise diffracting around the ends. A better estimate of attenuation is obtained by calculating the separate attenuations due to the three possible routes over the top and around each end as though each path was across an infinitely long barrier, and the combining them according to the expression

$$A_T = -10\log_{10}\left(10^{-A_1/10} + 10^{-A_2/10} + 10^{-A_3/10}\right)\mathrm{dB}$$

The difference between the level at a given receiver position before and after the erection of a barrier is called the insertion loss. One reason why the insertion loss is not as large as the predicted attenuation is ground effect. If part of the propagation path is over soft ground then as described previously there will be ground effect attenuation. The insertion of a barrier will preclude this effect. This may lead to situations where a barrier gives little or no net gain over the ground attenuation that might have otherwise occurred.

6.5 Sound attenuation by trees

Trees may affect the passage of a sound wave in several ways. Firstly a mature forest stand will produce a humus covered ground surface as a consequence of decomposing leaf litter. This surface, being highly porous and highly permeable, will offer a rather different ground effect to that obtained over a similar grass covered tract, as discussed earlier. Secondly the trunks, branches and foliage all scatter and dissipate incident sound to some extent. The trunks and branches scatter sound at wavelengths comparable to their diameters out of the path from source to receiver. The foliage presents a large surface area over which viscous friction may act to reduce the sound intensity. Moreover, trees shrubs and plants are of great value in improving the aesthetics of the roadside environment.

Unfortunately in terms of the noise units commonly used to assess road traffic noise, the effectiveness of thin belts of trees is negligible. A wide and extremely dense planting with foliage down to ground level is needed to produce an appreciable reduction in noise level.

7. PREDICTION SCHEMES [3,4,9,10].

7.1 Prediction Schemes for Free–Flow

Traffic noise prediction methods have been produced in many countries. They are used either to assess the degree of exposure to traffic noise for a given or projected road scheme or as part of a legal framework to assess entitlement for some form of compensation payment. Most of the methods are based on a manual containing a series of charts, formulae and nomograms. These are used to arrive at the predicted noise level through a sequence of adjustments to a reference noise level which is calculated from details of the traffic flow.

The procedures include the following steps:–

(i) the road scheme is divided into an appropriate number of (approximately) straight line segments either according to variations in the road alignment, elevation etc., or according to variations in the traffic parameters

(ii) for each segment a reference level is calculated at a given reference distance from the traffic stream as a function of the traffic volume, speed and composition i.e.percentage of heavy goods vehicles

iii) adjustments to the reference level are calculated for road gradient, road surface,distance (including ground effect), screening by obstacles, site layout features and size of source segment and

iv) the contributions from all of the segments are combined to give the predicted noise level at the receiver for the whole road scheme.

Manual methods incorporating these steps exist in the UK, USA, France, Germany, Scandanavia and several Eastern European countries.

In the UK method the 18(0600–2400) hr $L_{A_{10}}$ is predicted by a procedure described in ref.11. The reference distance is 10 m from the edge of the nearside lane. The reference noise level at this location is calculated from

$$L_{10} = 10 \log_{10} Q + 33 \log_{10}(V + 40 + 500/V) + 10 \log_{10}(1 + 5p/V) - 27.6$$

where Q is the volume of traffic in vehicles per hour, V is the mean vehicle speed (km/h) and p is the percentage of heavy vehicles. Significant modifications to the scheme have just been published to take account of recent research into noise levels at low vehicle flows and on road surface effects and to include prediction methods for retained cuts and dual and double–barrier configurations.

At distances greater than 500 m the quality of predictions using the UK scheme is poor except when there is a strong wind blowing from source to receiver. Moreover the influence of gradual reductions in individual vehicle noise limits is to make the scheme over–predict.

It should be noted that the scheme is intended only for situations where traffic flow travels without significant interaction, individual vehicles being operated in top gear and travelling at near constant speeds. Usually this is true only on motorways and urban freeways. Under these conditions the method is expected to be accurate to ± 2.0 dBA.

7.2 Prediction Schemes for Interrupted Flow

In most busy commercial streets vehicles are caused to slow down or stop because of interactions with other vehicles or as the result of intersections, imposed stop lines or traffic controls. These give rise to interrupted flow or stop–start conditions. High levels of noise may be generated by vehicles accelerating away from an intersection for 200–300 m even in the presence of a 30 m.p.h.(44 k.p.h.)speed limit. For decelerating vehicles noise level tends to decrease with decreasing vehicle speed being governed mainly by the falling power output. For vehicles accelerating from a standing position the initial noise output is high but may subsequently decrease as the vehicles' speed develops.

For traffic speeds between 10 and 70 km/h relationships between noise level (A weighted sound pressure level at 7.5 m),speed(V) and acceleration(A) have been found as follows

$$L_{\mathrm{Alight}} = 33.2 + 23.8 \log_{10} V + 10.6A - 0.08A^2 - 5.73A \log_{10} V$$

$$L_{\mathrm{Aheavy}} = 48.5 + 18.9 \log_{10} V + 7.5A - 0.11A^2 - 4.29A \log_{10} V$$

Consequently for a one–way street there is an upstream/downstream contrast on either side of the stopping position, the length and importance of which depends on the vehicle composition.

So far, prediction schemes for these complicated conditions have been developed empirically from statistical analysis of field measurements, from computer simulations and from scale model studies. A common result of the empirical studies and the computer simulations has been prediction that the percentage of heavy vehicles is relatively important. The empirical models may be expressed in terms of equations as follows:–

(i) for vehicle speeds between 10 and 60 km/h

$$L_{A_{10}} = 43.5 + 11.2 \log_{10}(L + 9M + 13H) - 0.42c - 10.2\log_{10}\left\{ \left[\frac{k + 3.5}{4.5}\right] \delta_1 \right\}$$

$$+4.6\log_{10}\left\{1 + \left[\frac{k + 3.5}{k + 3.5 + (n-k)}\right] \delta_2\right\}$$

where L, M,and H are the numbers of light, medium and heavy vehicles, respectively, n is the distance from the kerb to the nearside facade(m), k is the distance from the kerb to the receiver(m), c is the width of the carriageway(m), $\delta_1 = 1+0.52p_1$ and $\delta_2 = 1+0.52p$,where p_1 and p_2 are the proportions of soft ground in the distances k and n–k respectively.

It is noticeable that this model suggests that the noise level is independent of vehicle speed. An alternative model in which speed is an explicit variable may be expressed as follows:–

$$L_{A_{10}} = 58.6 - 5.99 \log_{10} V + 11.4 \log_{10} Q + 0.183P$$
$$- 5.94 \log F - 0.0102J - 2.46 \log N$$

where V is the speed (km/h), Q is the traffic flow (vehicles/h), P is the percentage of heavy vehicles, F is the distance from the receiver positon to the far–side facade, N is the distance from the receiver to the near–side facade and J is the distance from the receiver to the nearest road junction. Rather unexpectedly this model suggests that the noise level will decrease as the vehicle speed increases. A plausible argument for this is that although the noise level from any individual vehicle will increase with speed the gaps between successive vehicles will widen. Nevertheless computer simulations of vehicle noise in congested conditions show an increase of noise level with vehicle speed. An increase in mean speed from 30 to 50 km/h for example causes an increase in $L_{A_{10}}$ of 3 dBA. These simulations also show that the effect of changing the street width between 10 and 40 m on the noise level is negligible. This is in contradiction to the second empirical model but consistent with the first in which street width does not appear as a variable.

Both of the empirical models assume that the buildings on either side of the road form a continuous facade whereas computer simulations show that gaps between buildings can have a significant affect; the higher the percentage of gaps , the higher the reduction in noise level. Neither of the empirical models may be used to predict the effect of changing receiver height on the received noise level. On the other hand it is important to be able to predict the noise level at floors other than the ground. Computer simulation again can remedy this lack and shows that raising the receiver from 2 m to 20 m reduces the received $L_{A_{10}}$ by about 3 dBA. In cases where the height of buildings on one side of the road is less than the receiver height a dramatic reduction in noise levels may be expected.

8. TRAFFIC NOISE CONTROL

8.1 Traffic management

There are three possible approaches to controlling the impact of traffic noise. Reduction at source involves the design of quieter vehicles and road surfaces as discussed earlier. Control between source and receiver involves such considerations as traffic management, road layout, the use of barriers and land use planning. Control at the receiver involves insulation schemes.

The primary purpose of traffic management is to improve traffic flows. But the resulting changes will not necessarily reduce the noise impact.

One feature of traffic noise and,indeed of noise in general, is that where levels are high it is difficult to increase them further, whereas at places where the levels are low it is relatively easy to make them high. This means that a policy of concentrate and contain is an effective one as far as traffic noise impact is concerned.

Consider for example the noise affected area around a major road with an 18 hr flow of 10000 vehicles. If the noise affected area is judged according to an 18 hr LA_{10} of 70 or 68 dBA then an area of considerably less than 0.1 sq.km. is affected per km of road. If the flow along this road is doubled , then according to the criterion noise contour, the noise affected area remains considerably less than 0.1 sq.km. per km of road. However if the consequent total flow of 20000 vehicles per 18 hr day is split between two identical roads then the total noise affected area per km of road is increased beyond 0.1 sq.km. Moreover if the second road previously had a low traffic flow and hence a low noise environment the resulting increase in levels would have a disproportionate impact.

From basic theory a doubling of traffic volume leads to an increase of 3 dBA in noise level if all other factors remain unchanged. This change, which is only just detectable by the human ear could be caused equally by an increase in traffic from 300 to 600 vehicles per hour or by a increase from 3000 to 6000. It requires a ten–fold increase and hence a 10 dBA increase to produce a doubling of perceived loudness. Consequently, in the previous example, if a traffic management scheme makes the existing major road one way and a parallel road previously carrying only light traffic is designated to carry the reverse flow, then the average flow in the major road will be halved, giving a reduction of 3 dBA, while on the parallel road the average traffic volume will rise to the same level as that in the major road, giving rise to a noise level increase of 20 dBA or more. This means that, if the roads are similar, twice as many people will be affected by high noise levels as before the scheme was introduced.

If the road used for diverted traffic is narrower, and not designed to carry high volumes, the effect is accentuated. Furthermore if the scheme does indeed 'improve' traffic management it will lead to increased flows and greater mean speeds thus raising the noise levels even further.

A traffic management scheme that results in noise control is one that concentrates traffic on suitable main roads while reducing 'rat–runs' whereby vehicles attempt to avoid bottle–necks by pursuing unofficial diversions through minor residential roads. Such is the case, of course, with by–passes specifically designed to take high traffic flows and relieve the network of residential streets. For cities and towns where by–passes do not exist, it may be possible to make use of (non–residential) shopping streets by re–routing traffic through these streets during the night.

In London, environmental assessment of proposed traffic management schemes are carried out routinely. The assessments include predictions of changes of noise levels and estimates of numbers of people within criterion noise contours before and after the introduction of the scheme. These are used together with experience based upon many field measurements to design schemes so that adverse effects on the majority of the population are minimised.

8.2 Heavy goods and other vehicle restrictions

As mentioned earlier noise levels in non–free flow conditions are strongly affected by the percentage of heavy goods vehicles. Some movement of such vehicles in urban centres is essential but much through traffic may be diverted and disturbance may reduced by local restrictions. Techniques of control may take the form either of a total ban on heavy vehicles entering a prescribed area or of restrictions at certain times. With the cooperation of local authorities in England a national system of truck routes has been introduced. In the Greater London area trucks are banned from residential streets at night–time and at week–ends.

The ban affects heavy goods vehicles over 16.5 tonnes, operates from 2100 hrs to 0700 hrs each night and from 1300 hrs on Saturday until 0700 hrs on Monday. The ban does not apply to specified trunk roads and access roads and many individual heavy vehicles are exempt because they are used to carry essential goods or because they meet certain noise criteria. Such vehicles are required to carry an Exemption Plate and to display a renewable permit on their windscreens. Physical width restrictions have been set up in some locations designed so that trucks of typical width are unable to pass through, although provision is made for access by emergency vehicles.

These physical restrictions have proved particularly effective in protecting residential roads that provide a link between two main roads. Overnight parking by heavy goods vehicles in residential roads has considerable noise impact as a consequence of early morning starting and revving up of engines. Overnight parking bans are in force in some areas of London and to accomodate the resulting displaced vehicles special parking lots have been set up where they are unlikely to cause disturbance.

Because of the special noise problems associated with individual noisy motorcycles and their riders some cities also subject them to route and time restrictions.

8.3 Barriers and barrier blocks

In many of our urban areas, land is at a premium and it is not possible to use only the attenuation effects of distance to control highway noise.

One solution to the shortage of land is to build factories or industrial plants, or garages, being relatively insensitive to noise between the highway and more sensitive residential or office developments. A two–storey building can reduce noise levels on the side of the building away from noise by 13 dBA. A second row of buildings may produce only a small additional benefit, of the order of 1–2 dBA. Where residential buildings are used as barrier blocks in this way then it is important that the facade contains as little window area and other weak noise links as possible.

Purpose built noise barriers have been very popular means of control over the last few years. To provide most protection such a barrier should be sited either close to the highway or close to the area to be protected. If possible it should obscure the road from the buildings or space to be protected. Although the mass of the barrier need not be high because of the limiting effects of diffraction, discussed earlier, it is important that all the gaps in the barrier be properly sealed. A hole or gap in the fabric may reduce the screening potential substantially. Furthermore resonance effects created by the hole may alter the character of the sound; changing a broad band sound to one with more annoying tonal components. It should also be borne in mind that sound energy generated by traffic will be reflected by the barrier thus increasing noise levels for the vehicle occupants and for other receivers on the source side of the barrier. When there are barriers on both sides of the road a further problem may occur as a result of multiple reflections between the barrier walls. The screening potential of each barrier is reduced as a consequence of additional sound diffracted across each barrier effectively from image sources.

In the U.K. the height of most road side barriers is restricted to 3 m, whereas in Canada, the U.S.A. and some European countries, higher barriers are permitted. Usually barriers with heights greater than 4 m are regarded as visually unacceptable to the public. Apart from height, the shape of the barrier is important. A simple thin wall is less effective than an earth berm of similar height. Theoretical and scale model investigations have shown that double leaf barriers and T–profile barriers are more effective than single walls of the same height.

Wind loading, snow loading and drifting and the possibility of road icing in the shadow of the barrier are important safety considerations. The materials used in the barrier construction should withstand vehicle impacts and not shatter into fragments. The location of barriers over bridges and viaducts needs special consideration with regard to the possibility of falling debris as a result of a collision. An effective barrier material will also be unaffected by sunlight and other weathering. Close boarded wooden barriers are typical. Other materials include vinyl plastic, precast concrete, cellular concrete, aluminium, and caged rock type constructions. Combinations of vegetation and purpose built barriers are aesthetically pleasing and often effective noise controls. Willow woven screens with metal plate infil have been found to be effective as well as visually pleasing as noise barriers.

8.4 Road design

The noise pattern around a road is very sensitive to the type of road constuction that is used. Road elevation produces greater noise problems overall but can lead to useful noise shadowing at short distances and small heights. Figure 9.8 shows examples of $L_{A_{10}}$ contours around an elevated road. Cuttings are similar to barriers in their effectiveness.

However reflection effects limit the noise reduction that may be achieved with the vertical sided cuts that are common in urban areas. For example there is little noise reduction to be gained by increasing the depth of cutting beyond 3 m at either side of a road 20.7 m wide.

The improvement in screening from the increased depth of cut is offset by the increase in noise reflected from the opposite wall. Improvements in the screening provided by this type of construction can be obtained by reducing the reflectivity of the cutting walls or by sloping the walls away from the vertical. Absorptive lining can result in additional noise reduction of up to 3 dBA within 25 m of the cutting and up to 6 dBA at greater distances. Scale model studies have shown that the greater the ratio of height to width of the cutting the greater the effect of the absorbing lining. In practice it is unnecessary to cover the lower 1.5 m of the cutting walls with absorbing material since this region of the wall does not contribute much to the reflected sound. Furthermore it is the area most likely to be affected by water, dirt and chemicals thrown up by the traffic. A slope of only 15 degrees from the vertical is sufficient to ensure a substantial reduction in the reflected sound. Where space is not limited, the cutting may be formed by embankments with slopes in excess of 45 degrees with additional barrier type screening if required. The embankments may be formed using the soil excavated during the construction of the road.

The most effective form of road construction for noise control is a tunnel. These offer considerable advantages in noise screening compared with conventional retained cut structures particularly for high rise buildings close to the road. The disadvantages are the the much higher costs involved, the necessities for artificial lighting and for good ventilation and the much higher noise levels inside the tunnel (between 4 and 10 dBA higher, depending upon the amount of sound absorption), compared with an open road site.

8.5 Sound insulation of buildings [13–15]

Reduction of noise from road traffic at the receivers themselves inside their homes, schools and workplaces should be regarded as a last resort since it achieves little in gardens and recreational spaces and is a remedial rather than a preventative course of action. Table 9.4 lists possible guidelines for background noise levels within buildings. In the same way as the arrangement of buildings themselves may provide some form of noise shielding the arrangement of rooms within dwellings is important with regard to noise control. Rooms may be characterised by their use as noise sensitive or noise insensitive. The latter may be placed near the noise–affected facades, providing additional screening for the former. Figure 9.9 shows some examples. Moreover the need for expensive construction with high sound insulation may be minimised if the shape and orientation of the buildings are planned with due regard to the noise from the road. For example a monolithic structure should have its gable end pointing towards the road. Wings of a building should be orientated away from the road to reduce the amount of sound incident on the rear of the building. A high rise building may be provided with a low rise podium which therefore may bear the brunt of the traffic noise and shield the major portion of the building. Large glass areas which, generally have poor sound insulation, may be faced away from the road. Thin walled courtyards have been shown to provide reductions in LA_{10} of the order of 12 dBA.

Table 9.4 **Guidelines for acceptable noise levels inside buildings**

Type of room	Background Level dBA
Residential Buildings;	
Work Area	35 – 40
Living Room	30 – 40
Bedroom	30 – 35
Commercial Buildings;	
Office Machine Area	45 – 55
Reception and waiting area	40 – 45
General Office	40 – 45
Private Office	40 – 45
Conference room	30 – 35
Educational Buildings;	
Gymnasia	45 – 55
Open space teaching area	40 – 45
Classroom	35 – 40
Assembly Hall, small	30 – 35
Assembly Hall, large	25 – 30
Auditoria;	
Motion Picture Theatre	35 – 40
Lecture Theatre	30 – 35
Concert Hall, Theatre	25 – 35
Commercial Areas requiring speech communication	40 – 60
Restaurants	40 – 50
Department stores	45 – 55

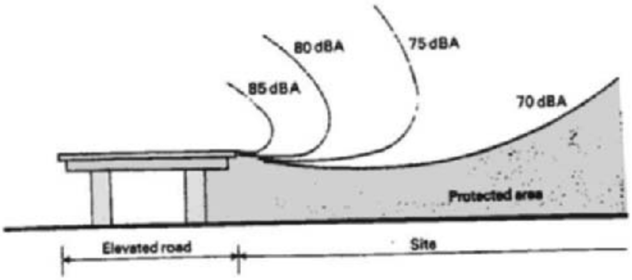

FIGURE 9.8 ***Contours of Equal Attenuation and Equal*** $L_{A_{10}}$ ***Around an Elevated Road***

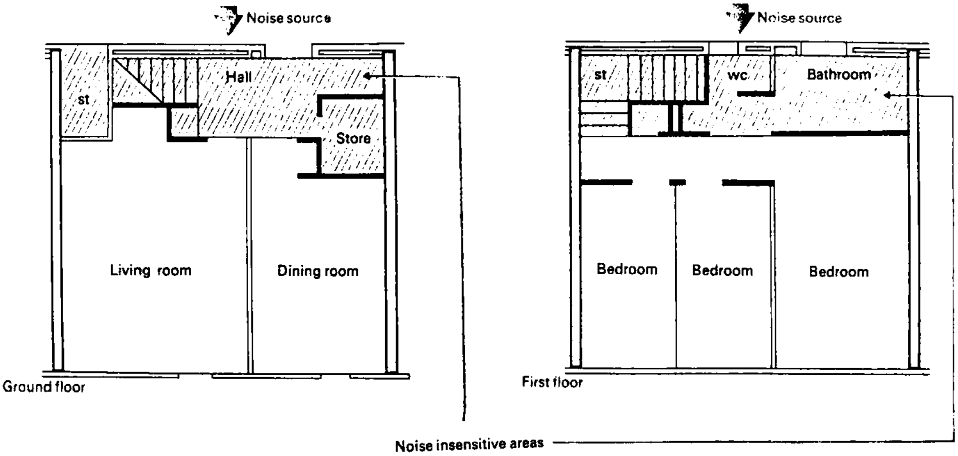

FIGURE 9.9 ***Housing Type Plans Showing Screening of Noise Sensitive Rooms***

The typical facade insulation of the brick built semi detached and short terraced houses included in the surveys leading to the 68 dBA 18 hr L_{10} standard used for the Land Compensation Act: Noise Insulation Regulations in the U.K., was 20 dBA. The implied internal standard therefore for dwellings is 48 dBA on the L_{10} index. This kind of level should be regarded as a minimum standard since it would make normal conversation rather difficult. As indicated in Table 9.5 a more acceptable standard for a residential living room might be 40 dBA (18 hr L_{10}). If a building is deemed to be eligible for insulation against traffic noise under the U.K. scheme the following modifications are provided:–

(i) the replacement by or conversion to double windows of all windows in the facade and venetian blinds between the panes to control solar gain and

(ii) the provision and installation of a mechanical ventilator unit having a sound absorbing lining in each facade–connected room, together with a duct for supplying air from outside the building to the ventilator unit.

These provisions counteract the principal weak points in the sound insulation of a building facade. Doors are another weak link. Many modern houses have glazed doors or hollow core doors which give a maximum of 17 dB sound reduction. Improvements are possible with more solid or heavier doors or with double doors. The need for mechanical ventilation follows since the double glazing is effective only if the windows are kept closed continuously.With the full treatment specified by the regulations, insulations of up to 35 dBA are possible.

Table 9.5 **Typical values for the sound insulation of building components**

	Sound Insulation (dB)
Walls	
100 mm brickwork or concrete blocks	45 – 50
200 mm brickwork or concrete blocks	48 – 52
Cavity Brickwork	50 – 54
100 mm hollow concrete blocks	40 – 42
100 mm brickwork plus 12 mm plasterboard on resilient studs	50 – 52
15 mm timber siding plus fibre board sheathing plus 12 mm plasterboard	36 – 38
as above with fibreglass in cavity	37 – 39
Windows	
single window open	5 – 15
single window 4 mm glass in fixed frame when closed	25 – 30
double window of 4 mm glass in fixed frame with 100 mm airspace when closed	30 – 35
as above when sealed	35 – 40
Doors	
light–weight,hollow core	10 – 15
solid door	15 – 20
double door with well–fitted gaskets	35
Roofs	
flat roof,steel decking	25
pitched roof,concrete tiles	28
as above with 100 mm fibreglass	34

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Chapter 10

THE DEVELOPMENT OF EMISSION CONTROL TECHNOLOGY FOR MOTOR VEHICLES

G.J.K. Acres

Contents **Page**

1. VEHICLE EMISSIONS, EFFECTS AND CONTROL

1.1 A historical perspective

The environmental impact of motor vehicles has been a concern both to the engineer and the general public for as long as the internal combustion engine in its spark and compression ignited forms have been in use. In the early days of the motor vehicle the environmental impact was essentially a local issue concerned with noise and visual emissions. However, as the vehicle population has risen, so has its contribution to environmental problems. These contributions were detailed and quantified in Chapter 1. The result is that today what started as a local issue with individual vehicles, has expanded to become in the 1990s a global issue both geographically and tropospherically [1]. However, despite this and increasingly severe legislation aimed at limiting the effect of engine and exhaust emissions on the environment, the pioneers of the internal combustion engine would have no difficulty in recognising today's engines as the logical development of those which first appeared in the early 1900s.

Engine design and materials developments, as well as the recent introduction of electronics into engine control systems, all contributed to improvements in reliability, durability and fuel economy as well as engine and engine exhaust emissions. However despite these developments and significant work on alternative power systems for motor vehicles including rotary engines, sterling engines and gas turbines, a need for further reductions in exhaust emissions for motor vehicles remains a priority area for the industry.

1.2 Pollutant emissions

As detailed in Chapter 1, the principal emissions of a motor vehicle using petroleum–based fuels fall into three categories. First, and of greatest significance are exhaust emissions of carbon monoxide and hydrocarbons resulting from incomplete combustion of the fuel together with nitrogen oxides formed in the combustion process. Other exhaust emissions generally classified as "particulates" can also arise dependent upon the engine, fuel and fuel additive. Those of particular note are lead, carbon and sulphate materials. Also present in the exhaust gas is carbon dioxide, an inevitable product of the combustion of hydrocarbon based fuels but of increasing concern with respect to the environment.

A second source of emissions from motor vehicles are those gases which may be released from the crankcase of the engine. Such emissions are largely hydrocarbon–based and result from leakages around the pistons and valve system to which may be added any oil losses.

Thirdly, hydrocarbon emissions to atmosphere may occur as a result of fuel evaporation. Sources of these emissions include the fuel tank during filling and normal running, as well as the fuel metering system to the engine. The crank case and evaporative emissions do not represent as much of a problem either to the environment or in terms of providing engineering solutions as do exhaust emissions. However, they would, if not controlled, add a significant hydrocarbon burden to the atmosphere and hence fall within the legislative requirements for the control of emissions to motor vehicles.

The impact of the increasing vehicle population and the cumulative effect of their emission on the environment was first observed in California in the early 1950s with the increasing occurrence and severity in Los Angeles of photochemical smog. With hindsight and today's knowledge of environmental chemistry this is not surprising as California then registered and continued to register a higher density of motor vehicles than any other area in the world. At the time the geographic location was considered to be the key factor in producing photochemicals. However, as the global vehicle population and in particular local vehicle densities have continued to increase, (see Chapter 1, Figure 1.2), so has the occurrence of photochemical smog around the world.

Photochemicals are now recognised to result from the reaction of hydrocarbons with nitrogen oxides and oxygen in the presence of sunlight producing a range of reactive oxygenated hydrocarbon compounds as well as ozone. As the motor vehicle is the biggest single source of hydrocarbon and nitrogen oxide emissions to the environment, control of photochemical smog is largely, but not exclusively, dependent upon reducing emissions from this source. (Table 10.1, also Chapter 1, Section 1.3).

TABLE 10.1 **Motor vehicle contributions to OECD pollutant emissions in Year 1980 (1000 tons)**

Pollutant	Total Amount	Vehicle Emissions	
		Amount	Percentage
Nitrogen oxides	36,019	17,012	47
Hydrocarbons	33,869	13,239	39
Carbon monoxide	119,148	78,227	66

Unfortunately today the problems of atmospheric pollution do not appear to end with photochemical smog. Three further phenomena have now been identified namely acid rain, the greenhouse effect and ozone layer depletion. Although the mechanisms by which these phenomena occur are less well documented than is the case with photochemical smog, similar pollutants are implicated, namely hydrocarbons and nitrogen oxides. In addition carbon monoxide and carbon dioxide from hydrocarbon fuel combustion and chlorofluorocarbons, a significant proportion of which originate from air conditioning units in motor vehicles, are also involved.

As a result of increasing concern about the role of the motor vehicle in contributing to these environmental problems and the possibility of the problem increasing with the projected increase in world vehicle population, the development of engine emissions control technology continues and new approaches are sought to reduce emissions of carbon monoxide, hydrocarbons, nitrogen oxides and carbon dioxide as required by current and projected legislation.

1.3 Exhaust emissions control legislation

Legislation requiring the control of emissions from motor vehicles was first initiated in America in the 1960s and has been progressively revised by incorporating reduced emissions requirements. Of major significance in terms of emissions technology was the introduction in 1970 of the amendments to the United States Clean Air Act requiring a 90% reduction in carbon monoxide, hydrocarbon and nitrogen oxide emissions. Emission trends are shown in Figure 10.1. Similar legislation has been, or is being, introduced in most major industrialised countries including the European Economic Community; see Chapter 2, Table 2.1. Chapter 12 examines legislative approaches in detail.

The American legislation was significant in that it could not be met with engine technology that existed in 1970 and forced new technology to be developed and applied. In the event what was considered in 1970 to be stringent standards were relaxed to allow more time for the new emissions technology to be transferred to production. However today the standards can now be achieved, but in order to maintain and improve air quality, more rigid standards are proposed for hydrocarbon and nitrogen oxide emissions with the initiative again being taken by California.

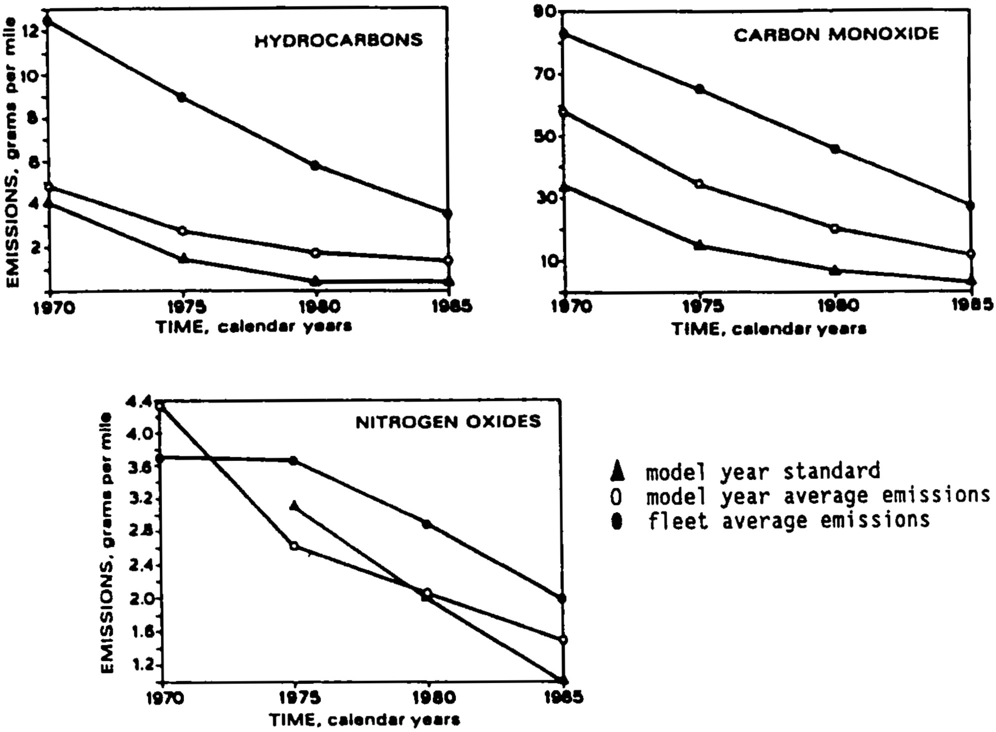

FIGURE 10.1 *Tail Pipe Emission Reductions since the Introduction of the Amendments to the Clean Air Act in America, 1970.*

Carbon dioxide emissions from motor vehicles are currently not subject to legislative standards, other than as an indirect result of American legislation concerning fuel economy of motor vehicles. However for both environmental and economic reasons it is proposed that motor vehicles should be subject to carbon dioxide emissions control legislation.

Emissions control legislation, which in addition to setting emissions standards also defines the means by which they are measured, including the driving cycle to which the vehicle will be submitted, largely dictates the engineering technology which has been utilised and will be needed to meet the future standards.

1.4 Exhaust emissions control technology

While emissions from internal combustion engines are a function of fuel composition, engine type and the power load conditions on the engine, the overriding factor as far as carbon monoxide, hydrocarbons and nitrogen oxides is concerned is the air/fuel ratio under which the engine is operating. (Fig. 10.2). Under no conditions are the three principal emissions at a minimum level and further the maximum power output of the engine corresponds with the maximum nitrogen oxide emissions. It is principally for these reasons that while significant improvements to fuel economy, power output and emissions have been made in recent years by modification and control, none of them have resulted in an engine capable of meeting current American standards while maintaining satisfactory driveability, power output and fuel economy without the use of catalyst units in the exhaust system.

In the case of the diesel engine, fuel composition and the ignition process are different to that of a petrol engine and exhaust emissions differ significantly as shown in Table 10.2. Exhaust emission standards of diesel engines are therefore different. Standards applicable to the American vehicles are shown in Table 10.3.

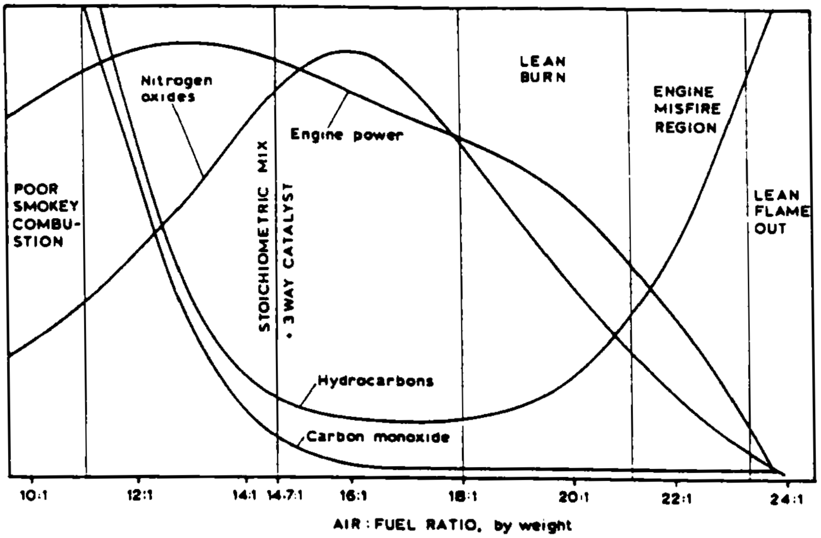

FIGURE 10.2 *The Effect of Air/Fuel Ratio on Engine Operation*

TABLE 10.2 **A Comparison of Diesel and Petrol Exhaust Emissions**

	Carbon Monoxide	Hydrocarbons	Nitric Oxide	Sulphur Dioxide	Particulates
	%	ppm	ppm	ppm	g/m^3
Diesel Engine	0.1	300	4000	200	0.5
Petrol Engine	10.0	1000	4000	60	0.01

TABLE 10.3 **Particulate Emissions from Diesel Engines
American Standards**

Automobiles	grams/mile
Before 1990	0.2
1990	0.08
Light Duty Trucks	grams/mile*
Before 1990	0.26
1990	0.08
Heavy Duty Trucks	grams/brake horse power.hour.
1991	0.25
1994	0.10

* EPA proposed relaxation.

2. THE USE OF CATALYSTS FOR EMISSION CONTROL

The concept of using a catalyst to convert carbon monoxide, hydrocarbons and nitrogen oxides to less environmentally active compounds such as nitrogen, water and carbon dioxide was a well established practice, prior to the need arising from motor vehicle emissions.

The principal reactions are shown in Table 10.4. However, the demands put upon catalyst technology by the need to match the performance of the catalyst to the engine as a result of rapid changes in exhaust gas temperature, volume and composition were features not previously encountered in chemical and petroleum industry applications of catalysis. Other unique requirements were the control of emissions such as ammonia, hydrogen sulphide and nitrous oxide which could result from secondary catalytic reactions and for the catalyst system to maintain its performance after high temperature excursions up to 1000°C and in the presence of trace catalyst poisons such as lead and phosphorous [2].

TABLE 10.4 **Reactions Occurring on Automobile Exhaust Catalysts**

Oxidation Reactions

$$2CO + O_2 \rightarrow 2CO_2$$

$$4HC + 5O_2 \rightarrow 4CO_2 + 2H_2O$$

Reduction/Three–way Reactions

$$2CO + 2NO \rightarrow 2CO_2 + N_2$$

$$4HC + 10NO \rightarrow 4CO_2 + 2H_2O + 5N_2$$

By the nature of the oxidation and reduction reactions which are involved in the removal of carbon monoxide, hydrocarbons and nitrogen oxides and the operating characteristics of the preferred catalyst (Fig. 10.2), several combinations of engine/catalyst system have been used since catalysts were introduced on American cars in 1975.

2.1 The carbon monoxide/hydrocarbon oxidation catalyst concept

As shown in Figure 10.3 which illustrates the best available technology to date, only a limited region of the air/fuel ratio span exists in which carbon monoxide, hydrocarbons and nitrogen oxides can be reacted on the catalyst with greater than 90% conversion. Hence in situations where carbon monoxide and hydrocarbons but not high nitrogen oxide emission control is required, eg, the European "Euronorm" standards, oxidation catalysts are used. A schematic of this system is shown in Figure 10.4.

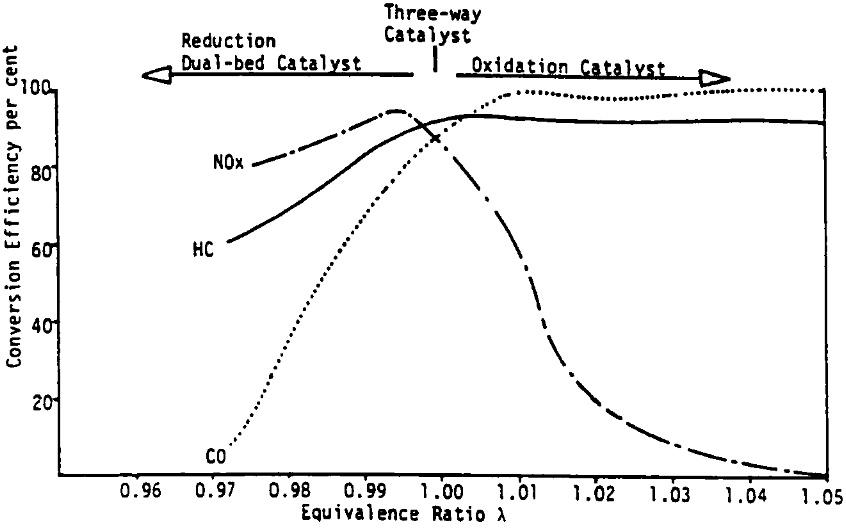

FIGURE 10.3 *The Effect of Exhaust Gas Stoichiometry on the Conversion Efficiency of a Rhodium/Platinum Catalyst.*

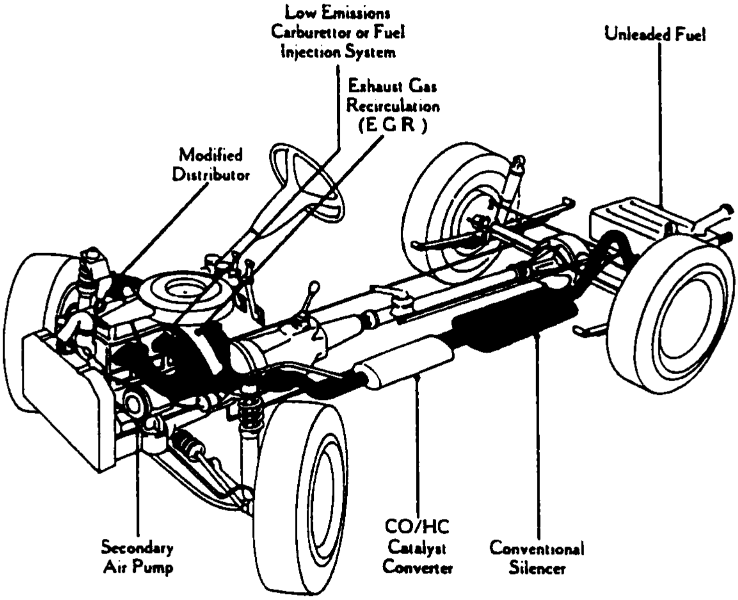

FIGURE 10.4 *The Oxidation Catalyst*

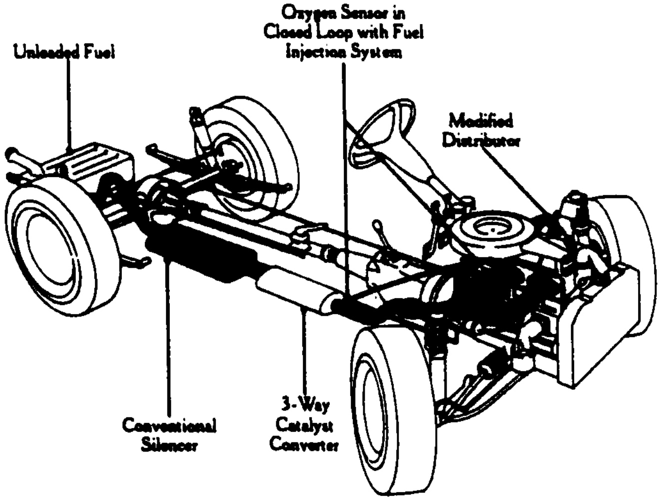

FIGURE 10.5 *The "three–way" catalyst*

Key features of this system are the use of a secondary air supply to the exhaust gas stream to ensure oxidizing conditions under all engine operating loads and the use of exhaust gas recirculation (EGR) to limit nitrogen oxide emissions from the engine.

This system was used initially in America to meet interim emission standards and is likely to be adopted to meet similar standards on medium and smaller engined cars (less than 2 litres) in Europe.

2.2 Dual bed and threeway catalyst concepts

In order to overcome the limitations imposed by the use of EGR and to meet more rigid nitrogen oxide standards, such as those imposed in America in 1981, catalysts capable of reducing nitrogen oxide emissions are necessary. Initially, as a result of the difficulty of controlling air/fuel ratios to the tolerances required by a single catalyst unit (Fig. 10.3), a dual catalyst bed was used. In order to ensure reducing conditions in the first catalyst bed, where nitrogen oxides were reacted, the engine was tuned slightly rich of the stoichiometric ratio. Secondary air was then injected into the exhaust stream ahead of the second catalyst bed, the oxidation bed, to complete the removal of carbon monoxide and hydrocarbons. With developments in engine control and catalyst technology, in the latter case involving widening the air/fuel operating window for 90% removal of hydrocarbons, carbon monoxide and nitrogen oxides, the dual bed system has been replaced with a single threeway catalyst unit. A schematic of this system is shown in Figure 10.5. The performance of the system is summarised in Table 10.5 for regulated pollutants and Table 10.6 for PAH and aldehyde pollution.

Key features of this system, in addition to the catalyst unit, are an electronically controlled air/fuel management system incorporating, in its most advanced form, the use of an oxygen sensor to monitor and control exhaust gas combustion. Systems such as this are now universal on American and Japanese cars and in those countries that have adopted similar emission standards. Cars having engines of greater than 2 litres which are required by EEC legislation to meet standards similar to those in America, used TWC systems from 1989.

2.3 Lean burn catalyst systems

The concept of operating engines under very lean, greater than 20/1 air/fuel ratios is attractive in terms of reducing nitrogen oxide emissions and improving fuel economy. However, with current engine technology, in order to achieve nitrogen oxygen emissions consistent with US legislation requirement, the engine must operate in the very lean region

TABLE 10.5 **Emission levels from small vehicles**
a) **<u>Without Catalysts.</u> Cold ECE15 cycle, g/test.**

	HC + NO_x	NO_x	CO
Peugot 205	18.3	7.8	26.3
Fiat UNO 45	15.2	6.2	26.7
VW Golf C	16.1	5.7	50.5
Rover 213	12.3	3.6	46.7

TABLE 10.5 **Emission levels from small vehicles**
b) **<u>With Catalysts.</u> Cold ECE 15 cycle, g/test.**

	HC + NO_x	NO_x	CO
Peugot 205	8.5	5.8	8.8
Fiat UNO 45	4.1	2.7	9.8
VW Golf C	6.4	2.0	42.7
Rover 213	5.2	1.4	27.5

TABLE 10.6 a) **Polycyclic Aromatic Hydrocarbon Emissions from a Programmed Combustion Engine**

Polycyclic aromatic hydrocarbon	Emissions, micrograms per mile ***Without Catalyst***	***With Catalyst***
phenanthrene	1.85	0.16
anthracene	0.61	0.04
fluoranthrene	2.27	0.23
pyrene	2.91	1.50
perylene	1.21	0.40
benzo(a)pyrene	0.94	0.17
benzo(e)pyrene	2.76	0.41
dibenzopyrenes	0.28	0.23
coronene	0.41	0.27

TABLE 10.6 b) **Aldehyde Emissions**

Vehicle type	**Emissions, grams per mile**
Average of 10 non–catalyst gasoline cars	0.141
Average of 3 catalyst gasoline cars	0.023

where, as shown in Figure 10.2, hydrocarbon emissions increase to levels which may exceed current American standards. In these situations an oxidation catalyst is incorporated into the exhaust system to control hydrocarbon emissions.

A feature of the ECE15 European test cycle was its low average speed as it is intended to be representative of city driving. The emissions that result are therefore typical of low speed, low acceleration conditions. As explained in Chapter 2, a more representative cycle (the EUDC) incorporating higher speeds and accelerations has been introduced so as to assess emissions under other conditions including urban and motorway driving. In order to develop and maintain a higher speed more power is required from the engine which, in the case of the lean burn system, entails decreasing the air/fuel ratio. This in turn increases nitrogen oxide emissions, (Fig. 10.2), to levels where current engine technology is likely to exceed the American 1983 standards and equivalent European standards with a high speed, acceleration requirement incorporated into the test cycle.

It is therefore desirable that catalysts used on lean burn engines should in addition to having a hydrocarbon oxidation capability also have a nitrogen oxide reduction capability when fuel enrichment occurs for increased engine power. The effect on the reduction of hydrocarbons and nitrogen oxide emissions which can be achieved on a lean burn engine using a catalyst with oxidation and reduction capabilities is shown in Table 10.7 for a Volkswagen Jetta Series 1, power with a 1.4 litre Ricardo HRCC (high ratio compact chamber) lean burn engine.

TABLE 10.7 **Lean Burn engine emissions**

ECE 15 cold start cycle. g/test

	Hydrocarbons	*Carbon Monoxide*	*Nitrogen Oxides*
Without Catalyst	11.7	15.9	5.9
With Catalyst	1.7	12.4	4.2

2.4 Advanced emission control technology

While the internal combustion engine is increasingly seen as near to its ultimate development limit, as far as emission technology is concerned, it is likely to remain the principle power unit for motor vehicles up to at least the end of this century. This results in particular from the need for substantial reduction in carbon dioxide emissions of two to three times coupled with significant reductions in hydrocarbon and nitrogen oxide levels. However, further developments are being pursued notably in the use of "clean fuels" such as reformulating gasoline and diesel fuel as well as methanol and natural gas in advanced engine design. Also of interest are high power/weight engines such as that developed by Sarich. Ultimately however, the time scales being dependent on future legislation, new power systems are expected to be required and are currently the subject of increasing development effort in the major vehicle manufacturing centres.

2.5 Diesel exhaust emission control

Although diesel engines emit relatively low concentrations of carbon monoxide and hydrocarbons compared to gasoline engined vehicles, and have a better fuel economy and hence carbon dioxide emissions, particulate emissions are now estimated to account for 40%

of smoke emissions in Britain and 80% of the estimated 140,000 tonnes of carbon particulates emitted in Europe each year (see Chapter 1). However associated with the carbon particulates which are produced during the combustion process are a range of aromatic hydrocarbons having mutagenic activity [3]. It was principally for this reason that the EPA proposals in America for limiting particulate emissions were first proposed.

The carbon and associated organics may be collected on a filter and removed by oxidation so that the filter regenerates and is effective for the life of the vehicle As the particulates are not oxidized at a significant rate below 600°C, and these temperatures are only achieved in the exhaust system when the engine is near or at full power, the more promising systems incorporate a catalyst into the filter which reduces the oxidation temperature to approximately 300°C. A schematic representation of a catalyst trap oxidizer is shown in Figure 10.6. Table 10.8 lists data on the degree of emission control which can be achieved with this system when fitted to a Volkswagen Golf 1.6 litre vehicle [4].

TABLE 10.8 **Catalytic Control of Diesel Exhaust Emissions**

g/mile	*HC*	*CO*	*NO_x*	*Particulate*
Without Catalyst	0.24	1.01	0.90	0.23
With Catalyst	0.05	0.16	0.79	0.11

Catalyst developments aimed at reducing the temperature at which particulate oxidation commences and sulphate emissions are minimized are desirable for the system to be effective in meeting the proposed American standards on motor vehicles, trucks and buses.

2.6 Catalytic combustion

Nitrogen oxide emissions arise principally as a result of the reaction between oxygen and nitrogen at the temperatures arising from the combustion of fuel whether it is initiated by spark, as in the petrol engine, or compression as in the diesel engine. Leanburn operation of a gasoline engine, as described earlier, offers a partial solution to the problem but is limited by hydrocarbon emissions as the non–flammability limit for spark ignition is approached. While the diesel engine does not have these disadvantages it is limited by high particulate emissions and a high cetane fuel requirement.

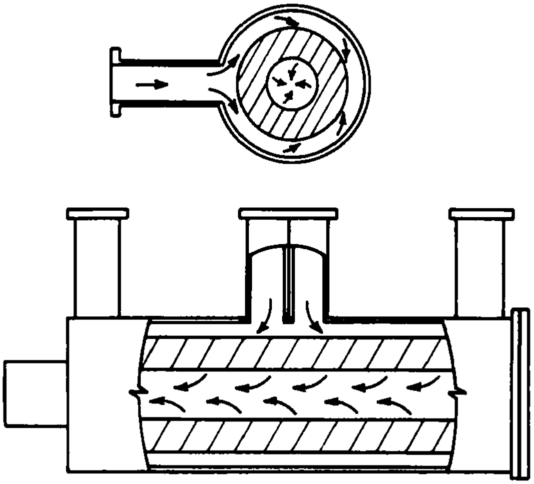

FIGURE 10.6 *A Catalyst Trap Oxidizer.*

An alternative approach is to use a catalyst to ignite the air fuel mixture and in this way overcome the constraining factors of the gasoline and diesel engines. Having removed this constraint a further benefit is offered, namely, the operation of the engine at a compression ratio of 12 to 1 where engine combustion efficiency and mechanical energy losses are optimized and thus fuel economy maximized [5].

The principle of the new catalytic engine is that during the engine operating cycle the fuel is injected into the combustion chamber just before the start of combustion is required. This fuel is then mixed with the air already in the cylinder and then passed through the catalyst, where heat release occurs. The use of this principle overcomes the fundamental problem of the stratfield charge engine, since all the charge is passed through a catalyst and oxidation can occur even at low temperatures and very lean mixtures. Thus all the fuel should be oxidized, and the engine can run unthrottled and hence lean to give good fuel economy.

The formation of nitrogen oxides and carbon monoxide in the combustion chamber is also strongly dependent on the air/fuel ratio, and lean operation gives reduced emissions of these pollutants in the exhaust of the engine. The catalyst enables oxidation of hydrocarbons at much lower temperatures than normally possible, so these emissions are also reduced. Another important advantage of the catalytic engine concept is that it is capable of operating on many different liquid fuels. The engine does not require high octane fuel because: (a) since oxidation occurs on or immediately downstream of a catalyst, rather than in a flame front, there is less tendency for the unburnt gases to be heated above their spontaneous combustion temperature, and (b) since the fuel is injected late into the combustion chamber there is no possibility of pre–ignition; therefore the engine can be operated on fuels like diesel oils and paraffins. It has no requirement for fuels of high cetane number because the start of combustion is controlled by the catalyst instead of by the heat of compression; therefore it can be operated on fuels like petrol or alcohol. The fuel consumption, smoke number and gaseous emissions of the four engine systems using gasoline as fuel are shown in Figures 10.7 and 10.8. Although only limited development of the catalytic Comet engine has been done, of particular note are the low nitrogen oxide and hydrocarbon emissions which are achieved by this system. On alcohol fuels all of these parameters are significantly improved.

3. ELECTRICALLY POWERED VEHICLES

In principle, the emissions problems from motor vehicles can be overcome by using electric power provided by rechargeable batteries. Apart from the range and limited acceleration provided by existing battery technology, recharging of a vehicle's battery pack is dependent upon electricity from nuclear or fossil fuel power plants which, in the latter case, are responsible for a significant part of the environmental carbon dioxide, nitrogen oxides and sulphur oxides burden. Thus while some benefit may be achieved with ground level emissions it is outweighed both environmentally and economically by the other disadvantages. However, there is renewed interest in this field with several major manufacturers announcing electric vehicle developments using new materials technology but conventional electrical storage batteries.

The disadvantages of the battery powered vehicle may be overcome if electric power is provided by a fuel cell since by this means fuel can be converted directly into electric power [6]. The fuel cell, which in addition to emission levels of hydrocarbons, carbon monoxide and nitrogen oxides substantially below those of internal combustion engines, even in their low emissions form, also has the benefit of significantly improved fuel economy. This results from the conversion of fuel directly into electric power by an electrocatalytic process which is not subject to efficiency limitations imposed by the Carnot Cycle. The resulting fuel economy benefits of the fuel cell over the internal combustion engine could be up to a factor of two with a corresponding reduction in carbon dioxide

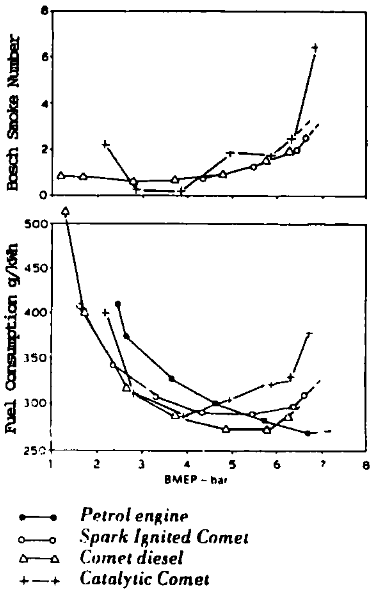

FIGURE 10.7 *The Fuel Consumption and Smoke Number of the Catalytic Comet Engine compared with Conventional Engines.*

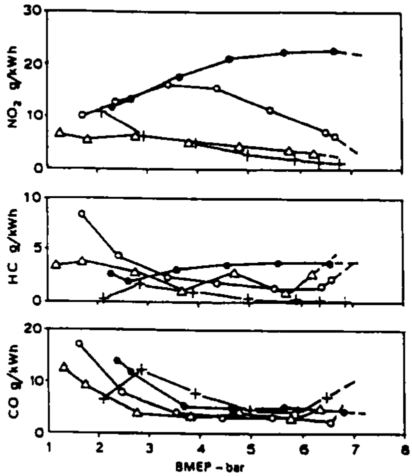

FIGURE 10.8 *Exhaust Emissions from the Caatalytic Comet Engine compared with those from Conventional Engines.*

emissions from transport sources. Current interest in America and Japan in fuel cell powered vehicles centres on the low emissions and fuel economy potential.

The principle of a fuel cell was first demonstrated by Grove in 1849 but it was not until Bacon's pioneering work on alkaline fuel cells in the 1950s that it was shown that significant power could be generated by this means. In the 1960s notably Shell and British Petroleum looked to apply this technology to mobile applications including motor vehicles but were unable at that time to demonstrate a commercially viable system with the resources and materials technology at their disposal. Since then work in America on fuel cell powered system for manned space vehicles and, more recently, power generating plant has significantly advanced the technology.

Fuel cell systems that are potential power units for motor vehicles are listed in Table 10.9. In Table 10.10 is listed key operating parameters for these fuel cell systems based upon a 200 volt 60kWe gross power unit.

CONCLUSIONS

Since the introduction of legislation in America in 1975 requiring substantial reductions in emissions from motor vehicles, catalyst technology has played a major part in maintaining air quality. With the introduction of similar standards in other countries, this application now represents the largest single use of catalyst systems.

While the emission standards for cars set by the 1970 Clean Air Amendments Act was considered adequate at the time, air quality standards have not improved as projected largely as a result of the expanding car population in America and other industralised countries. Hence further developments in internal combustion engine technology and new mobile power systems are sought as highlighted in this Chapter.

TABLE 10.9 **Fuel Cell Systems for Motor Vehicles**

Type	*Electrolyte*	*Electrode*
PAFC	Phosphoric Acid	Platinum
PEM	Solid Polymer	Platinum
MCFC	Potassium Carbonate	Nickel
SOFC	Zirconia	Nickel/Cobalt

TABLE 10.10 **Design Parameters for a 200V, 60 kWe Fuel Cell System**

Parameter	PAFC	PEM	MCFC	SOFC
Cell Voltage, V	0.67	0.63	0.70	0.63
Current Density A/cm^2	0.27	0.75	0.25	0.75
Stack Temperature °C	190.00	80.00	650.00	1000.00
Gross Efficiency %	45.40	40.30	47.40	42.70
Active Stack Volume, L	109.00	38.00	86.00	22.00
Cell Specific Power, W/L	302.00	1575.00	700.00	0.78
System Specific Power, W/L	56.00	62.00	36.00	77.00

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Chapter 11

HIGHWAY RUNOFF QUALITY MANAGEMENT BY REAL TIME CONTROL

W. Schilling

Contents **Page**

1. HIGHWAY RUNOFF AND WATER POLLUTION CONTROL

The problem of highway runoff within urban developments is basically a problem of urban drainage. In cities almost all streets drain into public sewers. Thereby, pollutants from street washoff can find their way to the receiving water body. Only a part is directed to the treatment plant and (partly) removed there.

Highway runoff pollution constitutes a significant fraction of stormwater pollution. Furthermore, it comprises particularly undesirable pollutants such as heavy metals. A comprehensive discussion on the treatment of polluted stormwater in separate sewer systems (SSS) has been presented recently by Grottker [21]. Here in this chapter a relatively new approach is summarized on how to cope with highway runoff pollution in combined sewer systems (CSS) – the technology of real time control (RTC).

1.1 Environmental Problems

1.1.1 **Elements of Urban Drainage Systems.** Urban drainage systems are distinguished between CSS and SSS. In CSS, sanitary sewage and stormwater runoff is collected in a single conduit. SSS employ one conduit for stormwater runoff and a smaller one for sanitary sewage. However, in existing systems these simple definitions almost never apply. Combined sewers often are often "enriched" by additional sewage from upstream sanitary sewers. Sanitary sewage is sometimes discharged into storm sewers through false connections causing continuous "stormwater" runoff and pollution: and, vice versa, sanitary wastewater flow patterns sometimes show a striking similarity with combined flow patterns, i.e. peak flows during rains. In the worst case a separate system might actually consist of two combined sewers. While false connections cause inflows in sanitary sewers jammed tide gates can cause large amounts of backflowing river water into CSS. Sanitary as well as combined sewers usually suffer from the problem of infiltration (through cracks and worn out joints).

Intended deviations from the genuine designs also exist. To reduce stormwater loading, roof leaders might be disconnected from a combined sewer. To reduce the cost of separation sometimes only (public) road runoff is connected to new storm sewers leaving (private) roof drainage hooked up to the old combined sewers. With respect to highway pollution the opposite solution is much more preferable: drain the streets into the combined sewer and the roof runoff into a storm sewer. Rather than allowing sanitary sewers to discharge into a CSS it might also be worthwhile to use parallel combined and sanitary sewer lines or even to install a sanitary sewer within the combined sewer.

1.1.2 **Pollution Problems.** Emphasis in the following sections of this chapter is given to CSS because of the following reasons:

1. Among the different processes in drainage systems the description of CSS is probably the most difficult. Both the quantity and quality spects of SSS are easier to handle and can be viewed as simplified cases of CSS.
2. CSS serve the majority of cities in Europe and many in North America. Most of these systems will remain for the foreseeable future.
3. Combined sewer overflows (CSO) discharge large loads of BOD and suspended solids (SS) into receiving waters. With higher levels of treatment the impacts of these CSO might be even more visible: fish kills might occur where there were formerly no fish since the dry weather pollution has been remedied, but the CSO continues.
4. It is expensive to reduce CSO to acceptable levels using traditional techniques such as additional storage and treatment.
5. There is no water system which has such a large discrepancy between design and operating conditions as CSS. At approximately 90% of the time CSS carry flows of less then 10% of their design capacity. This discrepancy creates the potential for RTC.
6. Controlling CSS means controlling a mixture of everything that possibly fits into the conduits. Besides water this can, among others, include gravel, solid wastes, explosives, toxic and aggressive chemicals, floatables, grease, foam, etc. Whatever equipment works and keeps on working in CSS, will also work in less harsh environments such as wastewater treatment plants, storm sewers, ponds, etc.

1.1.3 **Impacts of Urban Runoff Pollution.** Economics and performance of treatment processes require that only sewage with high enough pollutant concentrations should be treated. Secondary treatment capacity is usually only 1.5 to 4 times dry weather flow. Excess flow may be treated by primary processes alone or diverted directly to the receiving water. During a storm up to 95% of the organic load may bypass the treatment plant. Although this happens only at approximately 6% of the time and between 10 and 100 times per year the annual load discharges from CSO have been estimated for the United States to be 22% of the total urban biochemical oxygen demand (BOD) and 44% of the total suspended solids (SS) discharges [19]. CSS combine the problems of sanitary and storm sewers including

- very low dry weather flows during nights causing sedimentation, septic conditions, and corrosion of pipes,

- very high flows during storms causing CSO and flooding with wastewater,
- high concentrations of organic pollution (50 % of raw sanitary sewage),
- high concentrations of solids, heavy metals, hydrocarbons, and other toxics from street washoff,
- particularly high first flush concentrations due to both street washoff and sewer sediment resuspension,
- good settleability of CSO–SS in receiving waters.

The variety of flow and pollutant concentration patterns in CSS complicates treatment procedures. However, if storage is activated and if the number of CSO locations is reduced, much of the pollution loads can be captured within a CSS. Treatment activities can be concentrated at a few locations and thereby ease monitoring and operation of the entire drainage system. CSO have to be allowed to protect the population against flooding. However, often CSO happen although there is no actual need for this protection since there are still idle capacities. This is a major motivation for the implementation of RTC.

Since CSO only occur during short periods, conclusions about their impact on receiving waters must not only be drawn by looking at annual loads but also at the short periods of occurrence. Features where CSO may produce major disruptions to aquatic ecosystems include

- stagnant or low flow in the receiving water,
- upstream and downstream CSO into small streams or CSO into dead end canals, ("fish traps"),
- low dilution ratio compared to upstream flow, or large catchment area compared to upstream catchment,
- low background pollution allowing for a large variety of aquatic life,
- high water temperatures causing low dissolved oxygen (DO) concentrations in receiving water,
- extended dry periods implying high pollution concentrations from street washoff and combined sewer sediments in cases of CSO,
- high intensity storms producing high flow rates and causing resuspension of receiving water sediments by scour.

Therefore, in the worst but by no means unrealistic case, present pollution (e.g. sanitary sewage) is strengthened with short term sediments (from CSS) and long term sediments (from receiving water), [25, 31, 41].

Other impacts of runoff pollution include fish kills [13] and beach closings [23]. Receiving waters within urban regions mostly have a water quality that do not allow direct body contact uses. Nevertheless, these waters are often important for recreation. Although aquatic life within these waters might not be highly developed the public is usually sensitive to any further deterioration of water quality. Aesthetic disruptions caused by CSO might be floatables or resuspension of septic sediments by hydraulic scour.

1.1.4 **Static Solutions and Passive Operation.** Any approach to solve a runoff pollution problem has to start with upgrading the drainage system to its initially intended purpose. Upgrading techniques include

- better maintenance (flushing, fixing defective regulators, correct diversion settings),
- infiltration/inflow analysis with subsequent fixing of cracks, worn out joints, breakdowns, blocked tide gates,
- sedimentation analysis to identify conduit reaches with cumulative sedimentation caused by low flows, inverse slopes, etc.,
- hydraulic analysis to identify low capacity reaches which are prone to frequent flooding, and high capacity reaches which could be activated for storage,
- reduction of the number of CSO locations,
- expansion of treatment plant capacity for secondary treatment to at least peak dry weather flow.

The most rigorous upgrading technique of CSS is sewer separation where usually a new storm sewer is constructed. Meanwhile, a number of important disadvantages resulted in a more cautious application: e.g.

- Separation is generally an extremely expensive solution.
- It is very disruptive, especially within downtown areas (which are the common CSS catchments).
- Separation because of its cost and technical complexity cannot be achieved within a short period.
- There is increasing evidence that stormwater runoff from urban catchments contributes large amounts of especially undesirable pollutants [29], [30]. It is the largest source of SS and lead [20]; and usually contains significant concentrations of other heavy metals, nutrients, hydrocarbons, phenols, and pesticides [14], [41]. Some pollutant levels were listed in Chapter 1, Table 1.1.
- Stormwater and CSO solids settle better than effluent solids from treatment plants. Therefore, the former accumulate close to outfalls and cause long term hazards to the receiving water [41].

Separation is regarded as being cost–effective in only a few cases. For new developments, however, mostly separate systems are applied.

Alternatives to high–cost separation at the other extreme of best management practices (BMP) are a number of low–cost techniques for pollution abatement [16], [29], [30] such as

- improved street sweeping and catchbasin cleaning,
- improved catchbasins,
- erosion control and porous pavement,
- wastewater source control,
- disconnection of roof leaders,
- rainwater barrels, etc.

Although most BMP are source controls and therefore conceptually superior to end–of–pipe controls, their implementation requires much personal effort, enthusiasm for details, and close collaboration between different departments of municipalities.

A widely used approach to abate runoff pollution is construction of additional storage. Storage functions reliably and serves the two–fold purpose of flood protection and pollution reduction. Whereas off–line storage is only activated if a preset flow level is exceeded, in–line storage is part of the conduit and thereby permanently used. Although expensive, detention is conceptually simple. However, like any other static measure for pollution abatement they suffer from one disadvantage: they only perform optimally for one loading pattern, namely the design hydrograph. For all other loadings, static systems with passive operation necessarily have to operate suboptimally. Therefore, the concept of permanently achieving maximum effectiveness of a drainage system is a driving force for the proliferation of RTC.

1.2 Institutional Problems

1.2.1 Public Opinion, Legislation and Finance. During the 1960s the public in many industrialized countries developed concern about the integrity of the natural environment. Subsequently, different countries released legislation to ensure water quality protection. At the same time various countries launched research programs of which outstanding results have been published, for example, in four IAHR/IAWPRC conferences [5, 24, 50,22].

The first projects to demonstrate the feasibility of RTC were also initiated in the late 1960s [10, 32, 47]. Although research provided a wide variety of pollution abatement techniques their actual application beyond the demonstration scale has fallen behind. A major reason is the fact that runoff pollution abatement programs are usually financed with grants up to 95 % coming from sources other than the polluter (e.g. a city). For example, the United States Environmental Protection Agency (USEPA) used to support eligible projects with up to 75 % of the investment cost but not support any operation and maintenance (O&M) cost. Naturally, projects were designed to reach a minimum O&M cost level, almost regardless of the investment cost (an extreme example is separation). However, RTC of CSS is a typical example for a low investment, high O&M cost runoff pollution abatement technique.

1.2.2 **Sanitary Districts and Public Works.** The implementation of RTC is also strongly influenced by features of local administration. Typically, various cities in a metropolitan area form a sanitary district which is governed by a "Municipality of Metropolitan..." This configuration is perfect to manage a "clean" SSS in that storm and sanitary sewers are operated by the public works departments of the cities but interceptors and treatment plants by the sanitary district. However, difficulties arise in the operation of "dirty" SSS (i.e. with false connections, infiltration/inflow, high stormwater pollution) or CSS. Typically, the sanitary district is interested in reducing the treatment plant throughput to reduce O&M cost whereas the concern of the city is basement flooding. Even if a sanitary district is willing to accept additional combined sewer flows in times of storms, the usable storage in the interceptor might be very small. The cities operating large trunk sewers, on the contrary, are not eager to activate this storage since this might increase the flooding hazard. Different drainage authorities operating in a metropolitan area usually assure each other that, whatever measures are applied, backwatering will not occur. Obviously, these conditions are not in favour of the implementation of RTC where preferably in–line storage is activated by intended backwatering.

1.2.3 **Design and Operation.** The management of urban drainage systems is imbedded in public service legislation and regulations. Their main difference to the free market economy is that productivity increases have no benefit to those who achieve them. "Economic" solutions therefore are not identical to those which would originate in the framework of a competitive market.

On the operations level an interesting discrepancy between the planning/design stages of a project versus its operation/ maintenance can be observed. Whereas the former is executed by engineers and technicians, operations are usually carried out by personnel who are sometimes hardly educated in "what it's all about". This is often used as an argument that RTC cannot be successful because the appropriate personnel is lacking. Indeed, performance limits of existing RTC system (RTCS) are sometimes due to the fact that operators do not understand the purpose of the system. Operation of a RTCS requires intensive communication between all divisions and levels of a drainage agency, a management task that is extremely difficult to reach especially in large organizations.

2. REAL TIME CONTROL SYSTEMS

2.1 Introduction to Real Time Control

An urban drainage system is operated in real time if process data which is currently monitored in the system is used to operate flow regulators during the actual runoff process. For the operation of complex systems (e.g. with multiple detention tanks) the notion of systems control becomes important. Real time systems control is applied if the measurements are allowed to determine the state of the entire system. The data is transferred to some central intelligence where decisions are made on how to operate regulators with respect to the current and future states of the entire system. Ultimate performance might be reached with optimal control where control decisions are made to achieve a maximum benefit of a pre–specified performance criterion. Automatic control means that operational decisions are made by machines rather than humans. In a computer control system a pre–programmed computer decides how to activate regulators. In a supervisory RTCS operators make strategic decisions on set points (i.e. desired flow rates) whereas the local control actions (i.e. operating a gate) are automatically actuated by a controller. In a manual RTCS all actions are initiated by operators.

In a RTCS at least one set of the following elements apply:

- a **sensor** (e.g. water level gauge) that monitors the ongoing process,
- a **regulator** (e.g. pump or gate) that manipulates the process,
- a **controller** that activates the regulator to bring the process back to its desired value (set point, and
- a **telemetry** device that carries the measurement data from the sensor to the controller and the signals of the controller back to the regulator.

These four elements form a **control loop** which is a common feature of every RTCS, (Fig. 11.1). The control loop is influenced by external factors such as disturbances of the process and the adjustment (manual or automatic) of the control action.

2.2 Operational concepts

In this section the concepts of how a given system of RTC hardware can be operated are discussed. Operational concepts are concerned with the way process information is used.

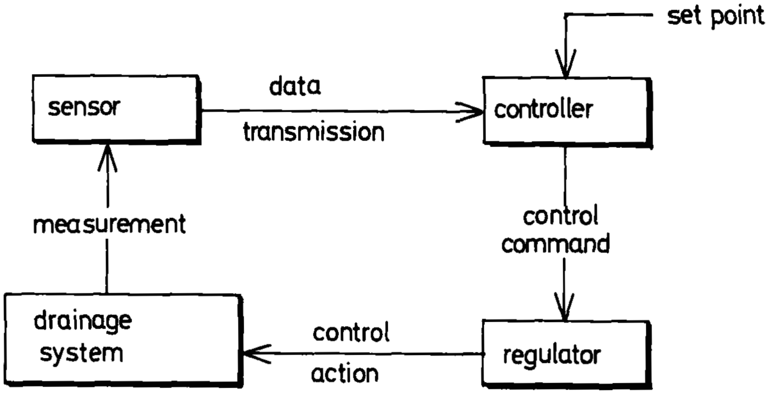


FIGURE 11.1 *Elements of a real time control*

2.2.1 **Local Control.** Typically, the task in operating a drainage system is to activate a number of pumps, sluice gates and weirs to allow adverse effects only (e.g. sewage diversions to receiving waters) if the system is at capacity. In static systems, this can only be achieved in some rare cases namely if the system is receiving its design load. If, for example, the outflow of a detention pond is controlled by an orifice, the optimal outflow rate is reached when the pond is full. During other periods the outflow is less and, consequently, the emptying time is longer.

Since this deficiency of static systems is well known, regulators are implemented in drainage system that have been designed to maintain a pre-set flow or water level. Common to all these regulators is that process measurements are taken at the regulator site. Therefore, a system of such regulators is termed a local control system.

The performance of local controllers (e.g. avoid instability such as "gate hunting", avoid transient generation, increase control speed) cannot be remotely improved from a control centre. Individual specification of set points during each event is not possible. In a local control system, set points can only be modified at the regulator site. Sometimes, operational data is centrally acquired but under local control to ease supervision of the system. If problems arise, maintenance crews can be dispatched immediately.

2.2.2 **Regional Control.** Local control is a good solution if the system has only one regulator (e.g. inflow equalization tank at a treatment plant). However, if more than one regulator is controlled independently from each other it might happen that an upstream pond is emptying into a downstream pond which is already overflowing. In those cases, better operation is possible if the set points are modified in an appropriate way and at the right time. This has to be done in view of process measurements further up or down of the regulator site (regional control).

2.2.3 **Systems Control.** If a RTCS is more complex or if all regulators are operated in coordination, systems control is applied. Set points are specified in view of process measurements throughout the system. The time sequence of all set points is called control strategy. Systems control in drainage systems is required if

- many regulators exist that affect each other,
- loading patterns are temporally and spatially variable, and
- strict performance criteria are to be kept.

Consider a cascade of two tanks. Each is actively controlled to not release more than the maximum capacity of the downstream conduit. Fig. 11.2 shows that this local (fixed) control strategy results in a downstream diversion. A better strategy with time–varying pond releases avoids this. Note that for the better solution no additional capacities are needed. This example shows that the operational flexibility is substantially enhanced if set points of local control loops are modified according to the actual system loading. This can be achieved in a number of ways.

In older control systems, measurements are displayed on meters located in a control centre. Regulators are actuated manually by operators. Since only limited information is available, operators need full understanding of the dynamics of both the control and the drainage system.

In an supervisory control system, regulators are actuated by automatic controllers but set points are specified by the operational personnel. As with manual control, supervisory control requires experience with the flow dynamics in the drainage system. It can be interactively supported by a flow simulator that allows modelling of the control effects before they are executed. It can also be supported by a data bank of previous control decisions. The simplest way of automatic control is frequently termed matrix control. There, each entry of a matrix represents a control action, given a set of actual measurements. An expert system might also be used for automatic control, possibly in a self–learning mode. Finally, the control problem might be formulated as an optimization problem. This allows an evaluation of control performance on an absolute ('the best') rather than a relative ('a better') scale.

2.3 Physical concepts

The basic elements of a RTCS have been introduced above as sensor, regulator, telemetry, controller, and central intelligence. The arrangement and interconnection of multiple control loops determines the physical concept of the RTCS. Generally, RTCS can be divided into the two classes of decentralized and centralized systems. The groups are distinguished as to where the intelligence is located that provides the decisions on appropriate control strategies. A supervisory or manual control system is necessarily a centralized system.

2.3.1 **Decentralized Systems.** If the sensors are each connected to a regulator and a controller, the control loops are isolated from each other (local control system). Here, the

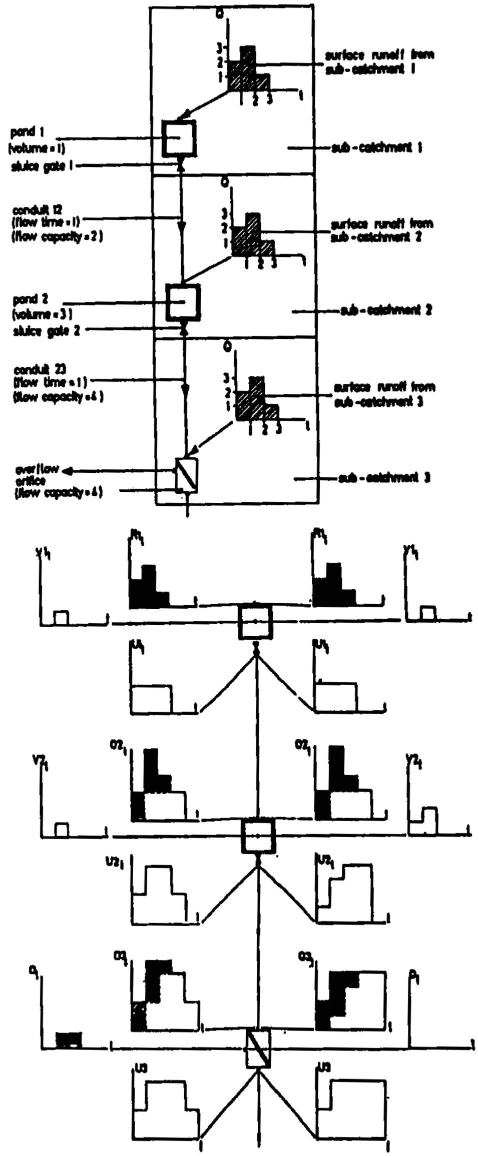


FIGURE 11.2 *Operation of two storage tanks with maximum outflow (left) and time variable outflow (right).*

set points are fixed and cannot be changed during the process. Usually, attempts are made to keep the devices as simple as possible (no external power, no electronic sensors) to minimize investment and maintenance costs. Examples are, among others, vortex valves, float regulated gates, and air regulated siphons. In an analog RTCS, all data is transmitted in analog form (e.g. pneumatic, hydraulic, electric current, voltage). Any kind of computer control, however, requires a digital RTCS.

Long time delays (i.e. flow time between the regulator and the sensor) can require that local control loops are augmented with additional sensors or interconnected between each other. An example is a feedforward/feedback loop where the controller uses the measured signals from both the controlled variable (e.g. storage outflow) and the disturbance (e.g. storage inflow) to determine the control action (e.g. gate opening). Another way to interconnect control loops is cascaded control where the output of a downstream controller is taken as the set point for an upstream controller. These advanced control concepts obviously require a larger and more expensive data transmission network.

2.3.2 **Centralized Systems.** Any kind of centralized control system requires motorized regulators, electronic data transmission and a control centre. In a direct digital control (DDC), a central computer replaces all controllers of the system. A DDC system requires wiring between all sensors to the computer and back to all regulators. It relies heavily on the availability of the central intelligence. Therefore, a DDC system is usually backed by hardwired local control loops (relay controls) to maintain simplified operation when communications or the central computer fail.

The disadvantage common to RTC concepts described above is that set points are fixed. However, their optimum values change and depend on the actual loading of the system. If a centralized system already exists (e.g. a DDC system) additional hardware requirements are relatively small. For automatic operation, extra computer capacity is needed for the strategic software. The state–of–the–art of RTCS design is a combination of decentralized and centralized systems. Microprocessors work as local controllers whereas set points are updated by a central intelligence (i.e. computer and/or operator). This distributed control concept combines the flexibility of a DDC system with the reliability of independently working multiple control loops. With the recent development of powerful microcomputers it is foreseeable that these will be installed as local controllers and, at the same time, communicate with each other to determine the optimal control policy. In such an integrated decentralized RTCS the central computer would eventually become obsolete.

2.4 Real Time Control Hardware

2.4.1 **Sensors.** Although a large variety of sensors do exist only very few fulfill the requirements for urban drainage systems. These include [40]:

- a wide measurement range,
- reasonable accuracy at routine operation on–site,
- suitable for on–site calibration,
- rugged, foolproof, maintenance friendly,
- sensing free surface, pressurized, as well as reverse flow,
- unaffected by humidity, solids, foam, aggressive chemicals,
- suitable for manhole installation,
- inexpensive,
- automatic indication of malfunction,
- suitable for remote data transmission.

Because of the difficulties and high costs of in–sewer flow measurements, rainfall–runoff modelling is applied in the analysis of urban drainage systems. Based on the physical understanding of the runoff generating processes, as reviewed in Chapter 4, rainfall data is computationally transformed into runoff and conduit flows. Therefore, raingauges are an important part in a drainage system monitoring network. Furthermore, advantage can be drawn by the time delay between rainfall and runoff to provide short term runoff forecasts. Point measurements of rainfall intensities are state of the art with errors less than 10% for moderate to high intensity storms. A major source of uncertainty, however, is the spatial variability of rainfall. A large number of raingauges is required to keep the errors of computed runoff resulting from undetected rainfall below 10 % [37]. Most types of raingauges feature a collector funnel which leads the precipitating water into a calibrated container. Either the rising velocity of a float, the increasing weight of a bucket, or the time interval between the "tips" of a tipping bucket indicate the actual rain intensity. A different principle involves the formation of standard size drops through a small outlet in the collector funnel.

Particularly suited for supervisory or manual RTCS are rainfall measurements by radar [11, 33]. In some countries, radar data is available for user subscription. A number of cities either experiment or already apply radar to aide operations, e.g. Detroit, Chicago, Milwaukee, Seine St–Denis/ France, Emschergenossenschaft/Germany.

The following principles are used to measure the water level. With a bubbler a constant rate of air, or other gas, is injected into the wastewater close to the bottom of the conduit. The measured backpressure is proportional to the water depth above the air outlet. An air pressure sensor measures the water depth through the compression of a static air column in a pipe. The pipe is hanging in the wastewater and is slightly inclined into the flow direction to prevent clogging. In a water pressure sensor a diaphragm/spring system is used to measure the water pressure acting on the sensor, thereby indicating the water depth. With a sonic level sensor the travel time of ultrasound from the probe to the water level and back is converted into the water depth. Other level sensors such as conductivity probes, capacitance probes, and floats are less suitable for drainage systems.

Few flow sensors are available to measure sewer flow directly. Either their high cost, inaccuracy, or limited range have prevented routine application so far. The most commonly applied flow measurement technique is the conversion of a level measurement into a flow rate using flow formulas such as the Manning equation. Here, errors result from the facts that

- the flow is proportional to the 8/3 power of the level, hence any level measurement error is amplified,
- the actual water level gradient is unknown,
- roughness and gradients of old conduits are difficult to assess [28],
- sediments might reduce the cross sectional area,
- pressure flow cannot be handled at all.

More accurate techniques combine depth and velocity measurements. Methods applied for velocity measurements either use the Doppler effect where the frequency shift of ultrasound passing through the wastewater is proportional to the flow velocity, or electromagnetic induction where the conductor wastewater induces a voltage by passing a magnetic field that is generated by coils above the conduit. This technique is very accurate for pressurized flow (e.g. through an inverted siphon).

In RTCS, regulators can be used for flow measurements. Structures such as perpendicular sharp crested weirs (bascule gates), sluice gates, constrictions, and vortex valves can be calibrated and, in conjunction with level sensors, used for flow monitoring.

Water quality sensors play a very minor role in RTC of urban drainage systems. Except for some explosives monitors and quality sensors in receiving waters (SS, pH, DO, temperature, redox potential, conductivity), quality sensors are not routinely used.

Frequently applied are limit switches such as the mercury float switch, diaphragm switch, thermal dispersion sensor, or proximity sensor. These sensors do not have a continuous measurement range but only indicate whether a pre–set value is met. They are widely used in hardwired backup control loops.

2.4.2 **Regulators.** Flow or water level regulators in urban drainage systems are often very large and custom designed. However, some basic principles are common to all successful devices.

1. Regulators are fail–safe designed in that malfunction of vital parts result in an acceptable functional decline of the system. For example, sluice gates would have by–passes, weirs would move into a safe position in case of a power failure, etc.
2. All parts exposed to sewage and the sewer atmosphere are drastically simplified and corrosion resistant. The preferable material is stainless steel.
3. Sensitive parts are located in an appropriate environment, i.e. dehumidified vault for hydraulic and electric machinery, dehumidified and heated vault for transducers, telemetry equipment, etc.
4. All parts of a regulator station (including gates, sensors, motors, etc.) are accessible, maintainable, and exchangeable.
5. Vital functions of regulators can be remotely supervised from the control centre.

The best known regulators for sewer flows are pumps. Pumping stations in urban drainage systems are usually equipped with a number of constant or variable speed radial pumps to handle the wide range of flow rates. If deep interceptors are used for in–line storage the sump levels can vary considerably. In these cases the proper selection of the optimal operating points of the various pumps is important. Otherwise cavitation and excessive wear and tear can occur. Measures for transient suppression have to be carefully designed to avoid upstream damage due to emergency shut off and subsequent surges and water hammer especially if large flows have to be pumped.

Weirs are used to store water in ponds or sewers. Depending on their design concept, there exist either perpendicular, side–spill, or leaping weirs. Self–operating weirs use a counter–weight or buoyancy of an air tank to adjust the height of the crest. A special design is an air regulated siphon–weir which funtions as a weir or a syphon depending on the air supply in its crest part. The air supply is controlled by a float–regulated valve. Inflatable dams are broad crested weirs, to be used to activate storage volume in large trunk sewers.

Gates (e.g. sluice, radial, sliding) are movable plates to constrict the flow in a sewer or in the outlet structure of a tank. The require a fail–safe by–pass to ensure safe operation in times of blockage.

Valves are devices which rotate inside a pipe to throttle flows (i.e. plug, knife, butterfly). They allow for a relatively adjustment of the flow rate. In a vortex valve the fluid is rotating. This motion builds an increasing resistance with increasing flow rate. It features neither external power supply nor moving parts. Other regulators used in RTCS include air regulated inverted siphons, movable tide gates, and flow separators which allow to the separation of incoming flow in two parts.

2.4.3 **Telemetry and Data Transmission Systems.** Any regulator featuring external power needs a special data transmission system. If very short distances apply between the sensor/transducer, the controller and the regulator/motor, data transmission can be analog (pneumatic, hydraulic, or electric current). If the signals are frequency modulated transmission distances can be increased. Here, the voltage or current transducer output is transformed into a frequency.

For catchment wide RTC, digital data transmission is increasingly applied. Advantages compared to analog transmission are that

- some transducers have already digital output (e.g. tipping bucket raingauge),
- digital data is suitable input for digital computers,
- transmission reliability (against noise) can be as high as desired, and
- high information transmission rates (bits per second, bps) can be obtained.

If analog data is to be transmitted in digital form it first has to be decoded with an analog/digital (A/D) converter. There, the analog signal is sampled with a frequency higher than twice the maximum process frequency to avoid loss of information. The sampled signal is then approximated by the closest binary number given a selected word length. For example, 1% conversion accuracy requires a 7 bit word length. The words are arranged and transmitted in blocks. Each block consists of words indicating the beginning of a transmission, source, number, type (request/ transmit), block length, the information, the termination of the transmission, and control bytes for data processing.

Transmissions can be facilitated by wire or wireless. However, in most European countries wireless transmission is restricted to mobile transceiving stations. Transmission by wire uses either privately owned, leased, or dialled public telephone lines. Leased lines are mostly used to take advantage of the services offered by the telephone companies. Dialled lines are preferred if data transmission over long distances is not required at all times, e.g. only for security checks, transfer of stored data, or raindata transmission. If public telephone lines are to be used, data has to be fed into an officially approved modem (modulator–demodulator) which connects the RTCS with the telephone network. The modem provides the appropriate frequency modulation for phone transmission.

Several logical channels can be transmitted in one physical channel if multiplexing is applied. With frequency multiplexing a frequency band, and with time multiplexing a time slot, is assigned to each logical channel.

Required capacities of transmission channels depend on the number of data points, the scanning frequency, and the amount of information per scan and data point. A standard for data transmission in public telephone lines is 1200 bits per second. Typically a sluice gate regulator station has in the order of 10, a pumping station in the order of 100 data points.

2.4.4 **Controllers and Process Computers.** Mechanical or pneumatic controllers are widely used in industry. However, these types of analog controllers cannot be remotely modified. Remote set point adjustment is not feasible with these devices. With the development of digital computers, many analog controllers could be replaced by one central computer. This allows a more flexible controller calibration, interconnection of control loops, and set point adjustment.

With the recent advent of inexpensive microprocessors, the vulnerability of such a system could be overcome by implementing a central minicomputer and several local microcomputers. Although they have separated tasks they can back up each other. Typically, the on–site microprocessor (programmable field panel, programmable logic controller, remote terminal unit) controls and coordinates all functions of an outstation, including

- acquisition of measurement data,
- processing (smoothing, filtering, etc.) of data,
- status, functional, and limit checks,
- temporary data storage,
- control computations and output, and
- receive and report data from and to the central station.

To perform these tasks, the microcomputers are equipped with input/output interfaces for local/manual operation, A/D converter, modem, energy supply and backup, real time clock, and storage for software and temporary data recording.

Differences between microcomputers and central process computers are gradual. In distributed RTCS their tasks are somewhat interchangeable. However, a number of major tasks usually remain for the central computer.

1. System wide data acquisition, long term storage, and data management is performed through the central computer. This data base provides the performance record of the RTCS and is used for all kinds of further planning.
2. Usually the operator interfaces are controlled by the central computer. It might also be used for interactive simulation/optimization software (decision support software).
3. In case of automatic systems control (optimized control, expert system) set points are adjusted by the central computer.
4. Since the central computer acquires system–wide rainfall and flow data it can be used to generate automatic forecasts of system inflows using rainfall forecasting and rainfall–runoff models.

Central process computers are equipped with hardware similar to microcomputers. Generally, they feature more user friendly operator interfaces, higher computing capability, disk and magnetic tape storage, and more input/output devices such as teleprint, hardcopy, plotter, etc. For reliability and/or background computations often two central computers are employed (front end/host).

3. PROCESS CONTROL

3.1 Introduction to Process Control

The control loop as the basic element of a RTCS has been introduced above. In a feedback loop, control commands are actuated depending on the measured deviation of the controlled process from the set point. Unless there is a deviation a feedback controller is not actuated. A feedforward controller anticipates the immediate future values of these deviations using a model of the process. Then it activates controls ahead of time to avoid the deviations. A feedback/feedforward controller is a combination of the two.

3.2 Continuous Control

Consider the regulation of the gate opening of a detention pond outlet to maintain constant outflow. A flow level sensor might be located downstream of the gate. Whenever the gate opening is changed the flow shows a typical reaction. Based on the physical behavior of the system (i.e. its differential equation) these reactions can be of a proportional (P), integral (I), or derivative (D) type, Fig.11.3. They can also be a combination thereof including a time delay between regulator action and sensor reaction.

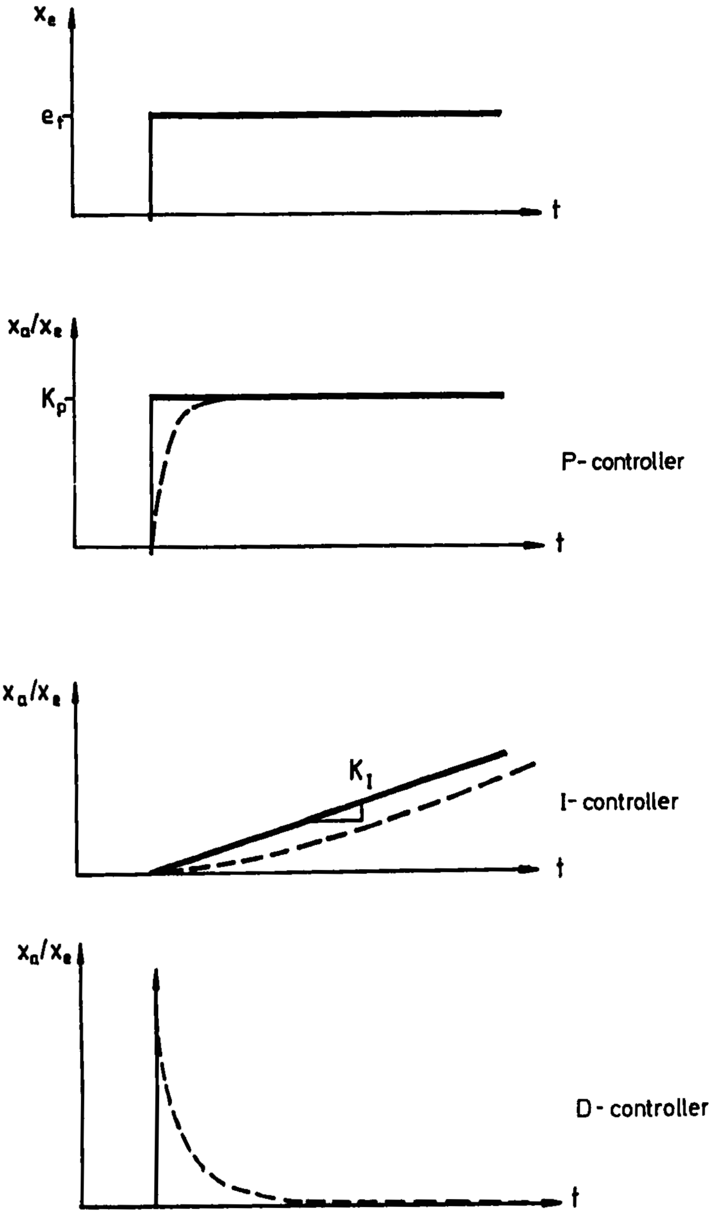


FIGURE 11.3 *Operating diagrams of a P–, I–, and D– controllers*

3.2.1 **Proportional Control.** The simplest controller behaviour is that of a P–controller. Here the signal u to actuate the regulator is proportional to the difference e between the measured variable and the set point. At time t it is

$$u(t) = K_P \cdot e(t) \tag{1}$$

The intensity of the controller reaction is given by the proportional gain K_P. Too low a K_P makes the controller slow, a value too high causes "over–reaction" of the regulator known as instability. A simple example for a P–controller is a float actuated gate, where the gate displacement is proportional to the vertical float motion, Fig.11.4. The disadvantage of a P–controller in a loop with pure P–behaviour is that constant deviations from the set point (offset) cannot be avoided. In Fig.11.4, for example, levels and flow might represent an equilibrium state with the set point level of 2.0 m. If a disturbance occurs (say, the outflow is increased from 2.0 to 3.0 m^3/s) the P–controller will open the inflow gate to 0.75 m and a new pond level of 1.25 m will be obtained. Thus, the offset would be –0.75 m. Other disturbances or proportional gain will yield different offsets.

3.2.2 **Integrated Control.** The output of an I–controller depends on the past history of set point deviations, namely

$$u(t) = K_I \sum_{i=0}^{t} e(i) \tag{2}$$

An I–controller reacts slower than a P–controller but always forces the process back to the set point. Fig.11.5 shows the behaviour of the water level h in a pond outlet, regulated by a sharp–crested weir. A higher K_I accelerates the controller ($K_I = 1.0$). Too high a K_I, however, causes instability ($K_I = 1.5$).

The disadvantages of P– and I– controllers can be avoided by combining them. The output of a PI–controller is

$$u(t) = K_P\, e(t) + K_P \frac{T}{T_N} \sum_{i=0}^{t} e(i) \tag{3}$$

with T_N called the reset time and T being the time step between regulator actions. Fig.11.6 displays level reactions of the inflow/level–loop for the pond shown in Fig.11.4. Only the PI–controllers allow fast and precise control to the set point level of 2.0 m. In this case the

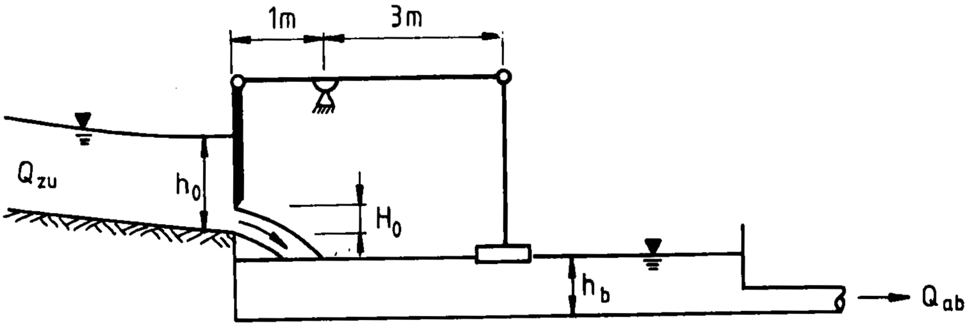


FIGURE 11.4 *Float controlled gate*

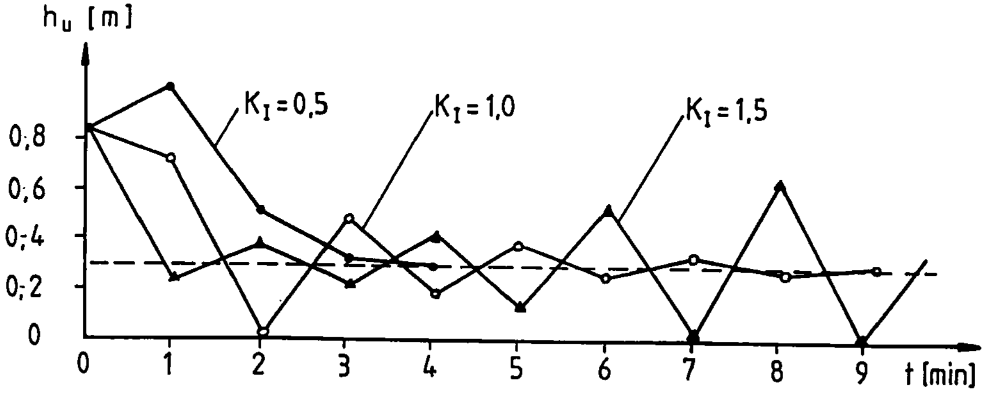


FIGURE 11.5 *Water level as function of I–controllers*

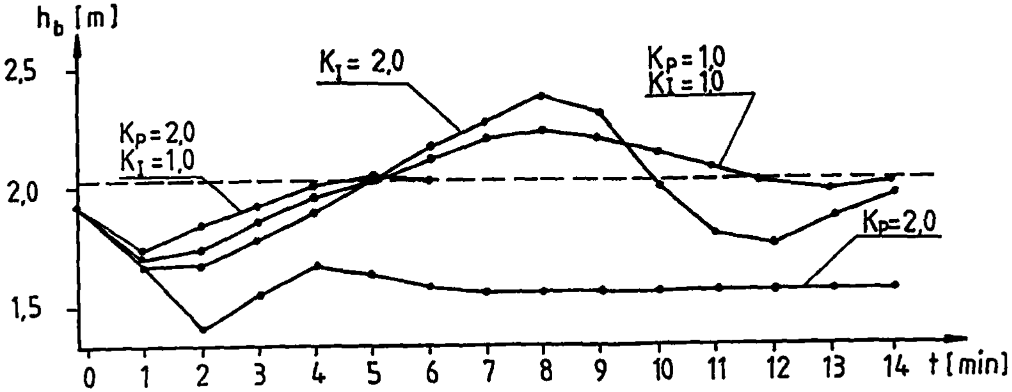


FIGURE 11.6 *Water level as function of various controllers*

combination of $K_P = 2.0$ and $K_I = 1.0$ seems to be a good set of parameters.

3.2.3 **PID – Control.** If a loop involves long time delays it might be worthwhile to make the controller more "nervous". If derivative action is added to the PI–controller the output of the resulting PID–controller is

$$u(t) = K_p\, e(t) + K_p \frac{T}{T_N} \sum_{i=0}^{t} e(i) + K_p \cdot \frac{T_V}{T} \left\{ e(t) - e(t-1) \right\} \tag{4}$$

The rate time $T_V > 0$ results in a short over–reaction of the controller.

Controller calibration, i.e. selection of type and parameters, can be done through analysis of the underlying differential equations, or real or simulated experiments.

1. Given a regulator displacement the reaction of the controlled variable can be measured or computed.
2. Depending on the desired controller type (P, PI, PD, PID) and the reaction above the parameters KP, KI, and KD can be estimated using calibration formulae.
3. If the controlled process is nonlinear (e.g. gate flow as a function of gate opening, channel flow as a function of water level) the controller parameters are only valid in the vicinity of a reference point (e.g. a particular flow rate or water level). For other reference points, other sets of parameters have to be found.
4. Once implemented, the behaviour of a controller has to be tested. If possible, full scale experiments over the whole range of control variables have to be carried out to ensure that instability cannot occur. Otherwise, regulators would permanently be activated and wear out very quickly. During routine operation, the initially selected control parameters can be fine–tuned to eventually reach optimum controller behaviour.

3.3 Discrete Controllers

3.3.1 **Two Point Controller.** Two point or on/off control is the simplest and most frequently applied way of control. An advantage of this type of control is the simple regulator needed. It has only two positions: on/off or open/closed. An example is the two point control of a pump filling a reservoir. When the measured value (the water level) reaches a limit (low level) the regulator (pump) will switch on and the pump discharge obtains the maximum value. Now the water level rises. When the high level limit is reached, the pump will be switched off. Then the level will decrease again, etc. The difference between the two switching levels is called dead band.

3.3.2 **Three Point Controller.** Three point controllers are typically used for regulators such as sluice gates, weirs, etc. In the middle position of the controller, the output signal is indifferent (0) and in the other positions: maximum (+) and minimum (–), respectively. In Fig.11.7 the operating curve of a three point controller can be compared to a two point controller. If the dead band (a) and the range of insensitivity a' are chosen properly the effect of a three point controller might be similar to a continuous controller.

4. CONTROL STRATEGIES

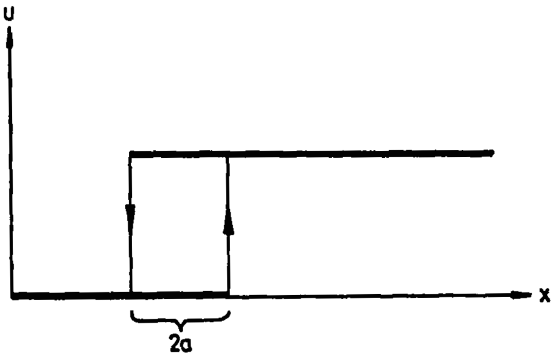
4.1 Introduction

In the preceding section it has been shown how a controller adjusts a regulator to achieve minimum deviation of the regulated flow from the set point. This section discusses how to find these set points. A control strategy is defined as the time sequence of all regulator set points in a RTCS.

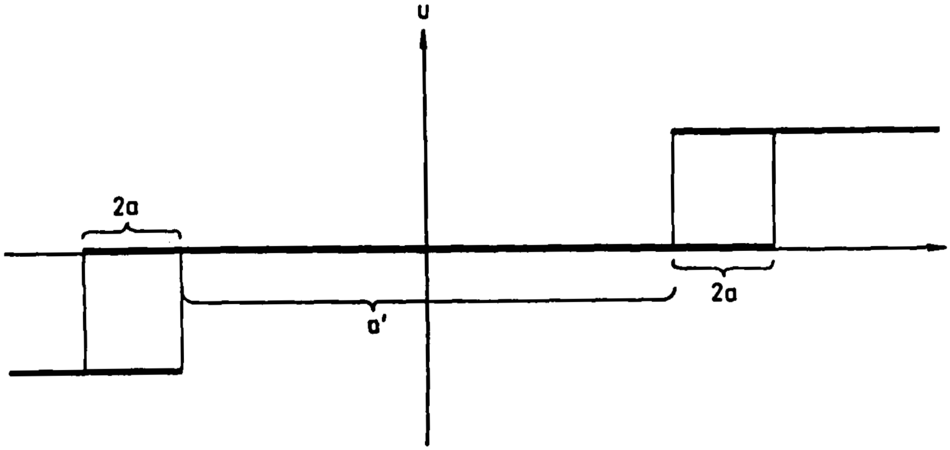
In Fig.11.2 it was demonstrated that the control strategy is a crucial feature of a RTCS. A cascade of two ponds is operated to minimize overflows. On the left hand side a fixed set point strategy is applied: release as much as possible but no more than the downstream conduit capacity. It was shown that the fixed set point strategy yields overflows whereas a better strategy with time varying set points of pond outflows allows to avoid overflows completely (right hand side).

Obviously, the simplest control strategy is to keep set points constant, Fig.11.8. For example, this option might be advisable for a regulated pond at the inflow of a treatment plant. One might also design a cascade of controllers, the output of a master controller being the set point of a slave controller. In Fig.11.9, this arrangement is used to modify the set point of the lower pond outflow to avoid overflows.

In most cases optimum set points vary with each flow pattern. If these patterns would display periodical variability, set point selection could simply follow a time schedule. A typical example is a water supply system where water demand shows a well known regularity. RTC of drainage system, however, is control of transient stormflows and pollutant loads which show no regular pattern. Here, the optimum set points differ for every storm and even change within a storm. Hence, a flexible method has to be used to react to whatever transients do occur.



two - point - controller



three - point - controller

FIGURE 11.7 *Operating diagrams of two–point and three–point controllers*

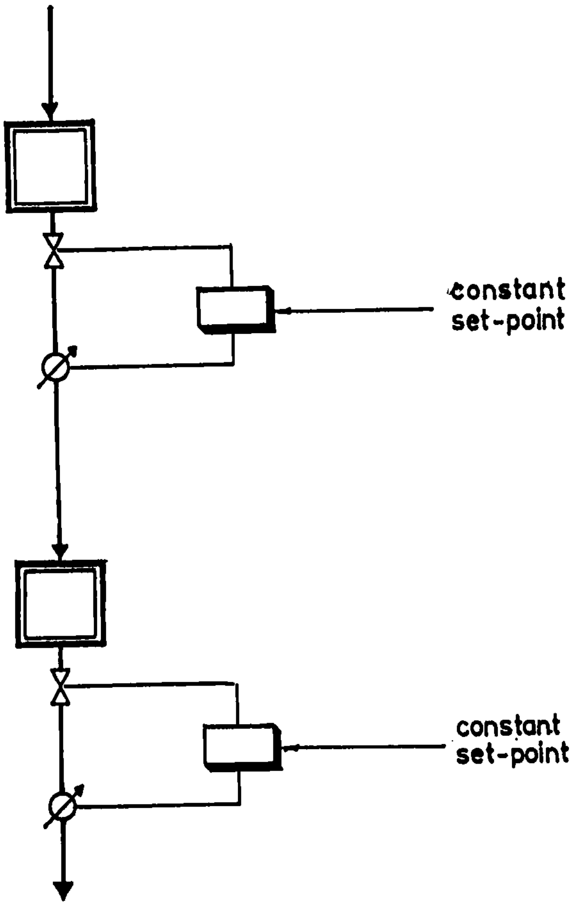


FIGURE 11.8 *Local control*

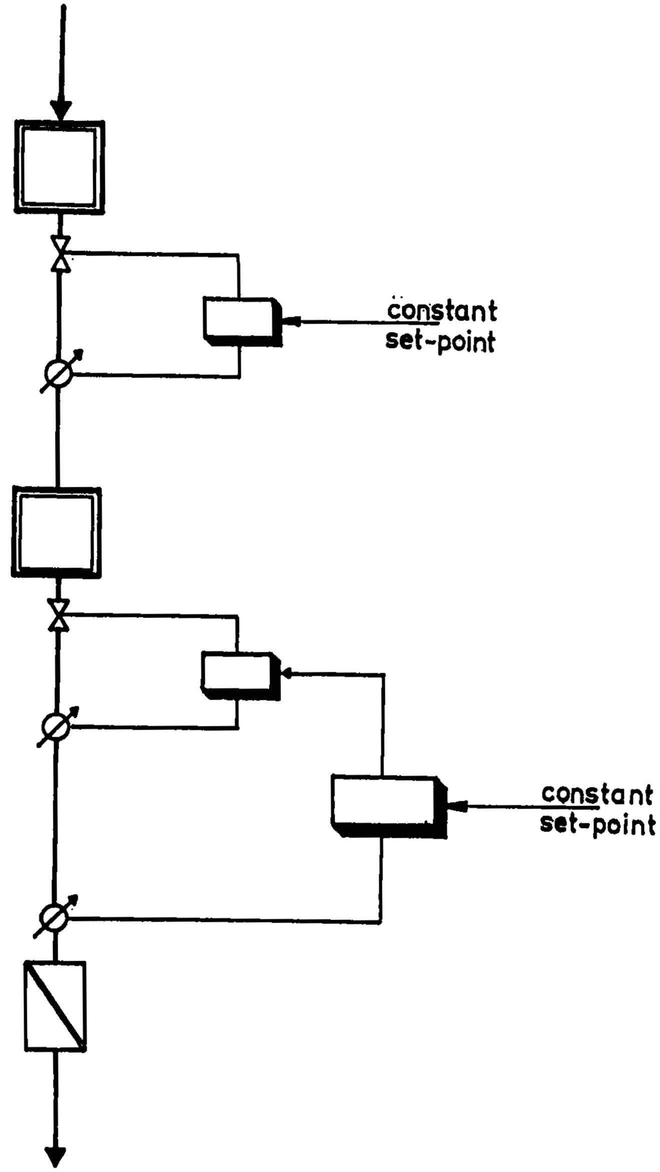


FIGURE 11.9 *Matrix control*

4.1.1 **Operational Objectives.** Before the problem to determine the control strategy can be addressed, the objectives of the operation of the drainage system have to be specified. Mostly these are to avoid accidents, flooding, pollution through combined sewer overflows, and excessive O&M costs. It is not only useful to rank these objectives due to their priority but also to specify some "ideal" operation and "costs" if this optimum is not reached. The costs allow to evaluate the performance of the control strategy with respect to every single objective.

Some objectives cannot be fulfilled at the same time because they are conflicting. For example, during a storm combined sewer overflows can be reduced by storing storm sewage in the conduits (in–line), at the higher risk of flooding, though. In these cases, a trade–off between the conflicting objectives has to be defined and a best compromise strategy has to be found. Usually a much higher priority is given to avoid flooding than to avoid pollution. This again is much more important than to avoid O & M costs. Most important, of course, is safety for maintenance crews and town–dwellers. Hence, a trade–off between the various objectives is usually not applicable because the operational priorities are very distinct.

4.1.2 **Physical Constraints.** A control strategy has to be physically executable. In a drainage system, this requires that pumping, flow through conduits etc. cannot be greater than the physically possible rates (static constraints). Other static constraints which are less obvious but nevertheless important are the present (initial) and desired (final) state of the drainage system (e.g. ponds empty, dry weather flow, etc.).

Additionally, the control strategy has to obey the physical laws of water motion in a drainage system, i.e. continuity and energy balances (dynamic constraints). For example, the dynamic constraint of a detention pond is

(storage at time t+1) – (storage at time t) =
(inflow during interval t) – (outflow during interval t)

A dynamic constraint of a pipe describes the flow transport, e.g.

(upstream flow at time t–1) = (downstream flow at time t)

It should be noted that the latter example is a simplification of the underlying physics of the flow process. Because of nonlinearities of friction this flow routing equation is only a rough approximation. It is these constraints that make the problem of finding a control strategy complicated, especially if they change accidentally during a storm (e.g. blockage of conduit, defect of pump, etc.).

4.1.3 **System Loading.** The physical capacities of a drainage system are either known to the engineer or can be determined with sufficient degree of accuracy. The loading of the system, though, be it storm inflows or pollutant loads are more difficult to determine in real time.

It is obvious that some knowledge of "what will enter the drainage system next" would be very precious information for the decision on how to control flows. It is also clear that the longer these forecasts reach into the near future the better a control strategy can be. Options to determine the input of a drainage system are

- flow and water level measurements in upstream sewers which allow to react within the travel time of the sewage,
- rain measurements and application of rainfall/runoff models which extend the available reaction time by the flow concentration time on the surface of the catchment, and
- rain forecasts which would gain additional time depending on the forecast time horizon.

If none of this information is available the control strategy can only be of the fixed set point type.

The monitoring of drainage processes was described earlier. It is important to recognize that measurements include measurement errors. It is therefore important to check control strategies with respect to measurement errors. Practically speaking, control strategies have to be "cautious" to avoid "surprises". These could be unexpected storm development, inflows from non–monitored tributary sewers, etc. Control strategies are usually based on measurements alone; however, it is useful to develop them using off–line simulation of the drainage processes as well.

4.2 Tools for Strategy Development

4.2.1 **Optimization.** The most rigorous approach to finding a control strategy automatically is to use mathematical optimization techniques, Fig.11.10. Here, the problem is reduced to the minimization of an objective function subject to initial state constraints, state capacity constraints, control constraints, final state constraints, dynamic constraints, and non–negative constraints.

One of the better known techniques is linear programming where all decision variables, i.e. state and control variables, are linear. Once a control problem is formulated as a linear

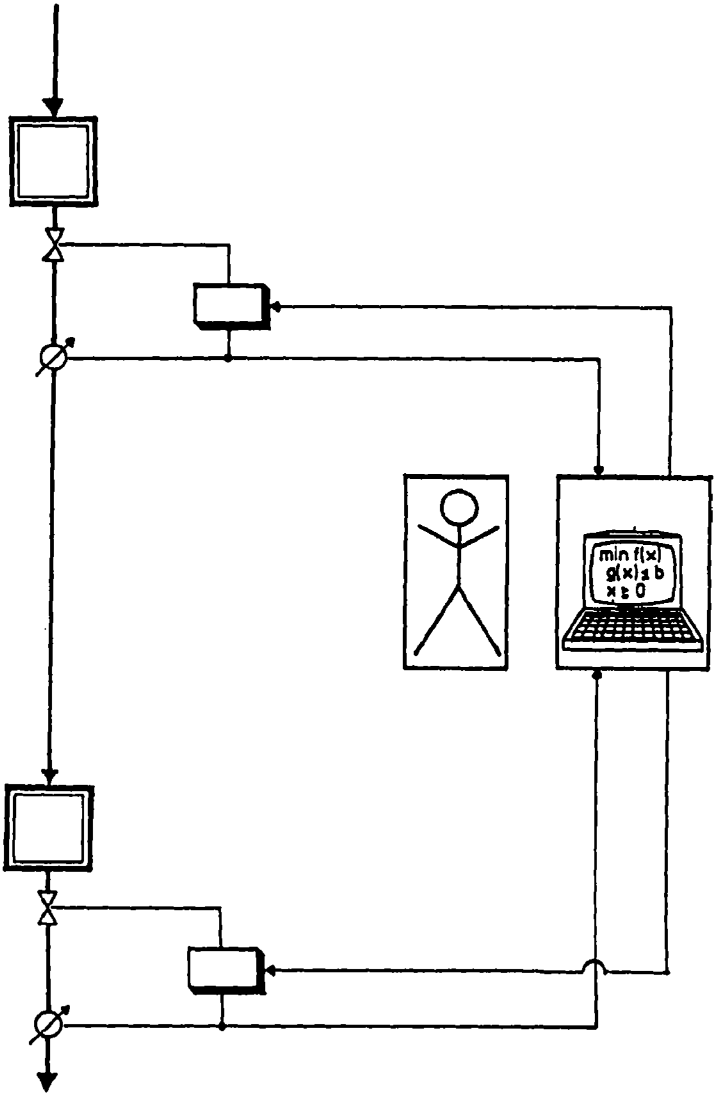


FIGURE 11.10 *Optimal control*

programming problem (LPP) it can be easily solved with commercially available software packages.

For the example of Fig. 11.2 the LPP can be formulated as to minimize the objective function (here: minimize total overflows)

$$\min \quad \sum_{t=1}^{n} O(t)$$

where n is the time horizon for which inflows can be specified and during which the desired final state shall be reached. The constraints which are not allowed to be violated at any time step t=1,2,..,n are sub–divided into:

(1) initial state constraints, (here: storage V, flows Q and overflow O are assumed to be zero) at time step t = 1

$V1_1 = 0$

$V2_1 = 0$

$O_1 = 0$

$Q2_1 = 0$

$Q3_1 = 0$

(2) state capacity constraints, (here: maximum storage V and maximum flows Q)

$V1t \leq V1max = 1$

$V2t \leq V2max = 3$

$Q2t \leq Q2max = 2$

$Q3t \leq Q3max = 4$

(3) control constraints, (here: maximum pond releases U)

$U1t \leq U1max = 2$

$U2t \leq U2max = 4$

$U3t \leq U3max = 4$

(4) final state constraints at time step t = n+1

$V1n+1 = 0$

$V2n+1 = 0$

$On+1 = 0$

$Q2n+1 = 0$

$Q3n+1 = 0$

$U1n+1 = 0$

$U2n+1 = 0$

$U3n+1 = 0$

(5) non–negative constraints

$V1t \geq 0$

$V2t \geq 0$

$Ot \geq 0$

$Q2t \geq 0$

$Q3t \geq 0$

$U1t \geq 0$

$U2t \geq 0$

$U3t \geq 0$

(6) The dynamic constraints require some simplifications of the flow process that is governed by the nonlinear St.Venant equations. Under the assumptions of

- no backwatering,
- constant flow velocities,
- no attenuation of flow hydrographs within a conduit, and
- ideal performance of the downstream orifice with $U3 = Q3$ if $Q3 < U3max$ and $U3 = U3max$ if $Q3 \geq U3max$

the dynamic constraints are for

- the mass balance in the upstream pond: $V1t{+}1 = V1t + R1t - U1t$
- the flow routing in the upstream conduit: $Q2t{+}1 = U1t$
- the mass balance in the downstream pond:
  $$V2t{+}1 = V2t + R2t - U2t + Q2t$$
- the flow routing in the downstream conduit: $Q3t{+}1 = U2t$
- the diversion of overflows: $Ot = Q3t + R3t - U3t$

It should be noted that the dynamic constraints (6) vary with every time step, because inflows R are never exactly known ahead of time. Hence, on–line optimization might have to be re–executed whenever actual inflows deviate too much from their forecasts. For given storm inflows and a time horizon of, say, $n = 5$ a solution of this problem was displayed in Fig. 11.2. In this case the control strategy are the hydrographs of the decision variables U (pond releases). These are the set points against which sluice gate controllers for the two pond outlets are compared and actual outflows are corrected. It can be seen that the optimum control strategy avoids overflows completely. The local control with fixed set points (release maximum allowable pond outflows), on the contrary, yields overflows during two time intervals.

Other optimization techniques are discussed in detail in the literature. Applicable techniques include dynamic programming and its derivatives, derivatives of linear programming, and calculus of variation [6, 15, 34, 38, 42, 48].

Common to any optimization technique are a number of difficulties:

(1) Operational objectives of an agency might be monetary, intangible, and/or conflicting. It is very difficult to aggregate all objectives into one single equation. Multiple objective optimization techniques, however, cannot be fully automatic. They require interaction with a decision maker to find best compromise ("Pareto optimum") solutions.

(2) If a single objective function has been specified it is usually of a mixed integer/continuous, nonlinear, and non–monotone type. Since powerful optimization techniques are not available for this kind of objective the function has to be further simplified.

(3) Flow routing has to be simplified to allow application of standard optimization techniques. This involves spatial and temporal aggregation, and linearization. The effects of these simplifications on final control performance, although probably not very important have not been fully investigated yet.

4.2.2 **Search.** Search is a technique that can intuitively be carried out (like a chess player does) or, similarly to optimization, be formulated as a mathematical problem. Here, one specifies an initial control strategy (e.g. the default fixed set point strategy). Based on additional information about the current state of the system and future input one would then try to change an appropriate control variable to gain some additional benefit (e.g. start pump earlier). If done automatically by a computer the strategy is changed for variables in a way that a pre–specified performance index (i.e. objective function) gains maximum improvement. If no improvement is possible anymore it is assumed that an optimum strategy has been found.

Automatic search techniques require some insight into the search algorithm (e.g. steepest descent search, Newton–Raphson search, etc.) to reduce the number of required iterations. In practical applications such a large number of iterations might be required that the necessary computer time would not allow on–line applications. However, search techniques allow for a more flexible formulation of the objective function and the constraints (e.g. non–linear).

4.2.3 **Decision Matrices.** Decision matrices are a tool that, compared to optimization techniques require extensive development work but allow for very fast on–line execution of control strategies. Essentially, control variables are specified in advance for all possible combinations of input and current state variables, Fig.11.11. Therefore, if n state variables (e.g. pond storage, conduit flow) and p inflow variables (e.g. current inflow, next inflow, next but one inflow, etc.) are assumed, an (n+p)–dimensional matrix has to be specified. Each entry of the matrix represents the control decision which has to be executed for a given combination of state and input.

For the two ponds in Fig.11.2 and given inflows and state data a decision matrix such as Table 11.1 might be applied. From this matrix it is obvious that for high input flooding cannot be avoided. However, if one–step forecasts for the input R2t+1 would be available one can try to avoid upstream releases U1 if at all possible. Thereby, at least flooding at the downstream pond could be circumvented.

4.2.4 **Heuristics.** Heuristic methods to find a control strategy are based on experience of the operating personnel. Here, an operator would either directly actuate the regulators (Fig.11.12), specify only the set points of each regulator (Fig.11.13) or even use an interactive flow simulator as a decision aid to find an appropriate control strategy (Fig.11.14). Heuristic control introduces the potential to use sources of information that are not accessible to a computer (e.g. intuition, view out of the window, phone calls, etc.). In case of emergency an operator can immediately seek assistance, advice maintenance crews, etc. An experienced operator will probably carry out near–optimum control. He disregards (like a good chess player) all control options that are possible but not advisable.

However, heuristic control also has serious drawbacks. Once an operator leaves his job his experience will also be gone. His successor will make mistakes all over again. Experience with stormwater RTC can only be gained slowly. It is raining approximately 5 % of the time. An operator spends approximately 20 % of the time for his job. Hence, only during 1 % of the time is he really exposed to a stormwater RTC problem. Unless his job is integrated in treatment plant operation, 3 to 4 operators working in shifts have to be educated, trained, and paid just for stormwater operation. In conclusion, there are arguments in favour of automatic systems control.

4.2.5 **Expert Systems.** Recently, attempts have been made to apply artificial intelligence to decision problems with so–called expert systems (Fig. 11.15). With an expert system it is programmed how experience is gained and how this experience is applied for decisions such as the specification of RTC strategies.

TABLE 11.1

Decision matrix for the control of two stormwater ponds (V storage, R inflow, U outflow)

V1t/V2t R1t/R2t+U1t−1	0/0	0/1	0/2	0/3	1/0	1/1	1/2	1/3
0/0	0/0	0/1	0/2	0/3	1/0	1/1	1/2	1/3
0/1	0/1	0/2	0/3	0/4	1/1	1/2	1/3	1/4
0/2	0/2	0/3	0/4	0/4	1/2	1/3	1/4	1/4
0/3	0/3	0/4	0/4	0/4	1/3	1/4	1/4	1/4
0/4	0/4	0/4	0/4	0/4	1/4	1/4	1/4	1/4
0/5	0/4	0/4	0/4	*0/4	1/4	1/4	1/4	1/4
1/0	1/0	1/1	1/2	1/3	2/0	2/1	2/2	2/3
1/1	1/1	1/2	1/3	1/4	2/1	2/2	2/3	2/4
1/2	1/2	1/3	1/4	1/4	2/2	2/3	2/4	2/4
1/3	1/3	1/4	1/4	1/4	2/3	2/4	2/4	2/4
1/4	1/4	1/4	1/4	1/4	2/4	2/4	2/4	2/4
1/5	1/4	1/4	1/4	*1/4	2/4	2/4	2/4	2/4
2/0	2/0	2/1	2/2	2/3	2/0	2/1	2/2	2/3
2/1	2/1	2/2	2/3	2/4	2/1	2/2	2/3	2/4
2/2	2/2	2/3	2/4	2/4	2/2	2/3	2/4	2/4
2/3	2/3	2/4	2/4	2/4	2/3	2/4	2/4	2/4
2/4	2/4	2/4	2/4	2/4	2/4	2/4	2/4	2/4
2/5	2/4	2/4	2/4	*2/4	2/4	2/4	2/4	*2/4
3/0	2/0	2/1	2/2	2/3	*2/0	2/1	*2/2	*2/3
3/1	2/1	2/2	2/3	2/4	*2/1	*2/2	*2/3	*2/4
3/2	2/2	2/3	2/4	2/4	*2/2	*2/3	*2/4	*2/4
3/3	2/3	2/4	2/4	2/4	*2/3	*2/4	*2/4	*2/4
3/4	2/4	2/4	2/4	2/4	*2/4	*2/4	*2/4	*2/4
3/5	2/4	2/4	2/4	*2/4	*2/4	*2/4	*2/4	*2/4

* flooding cannot be prevented

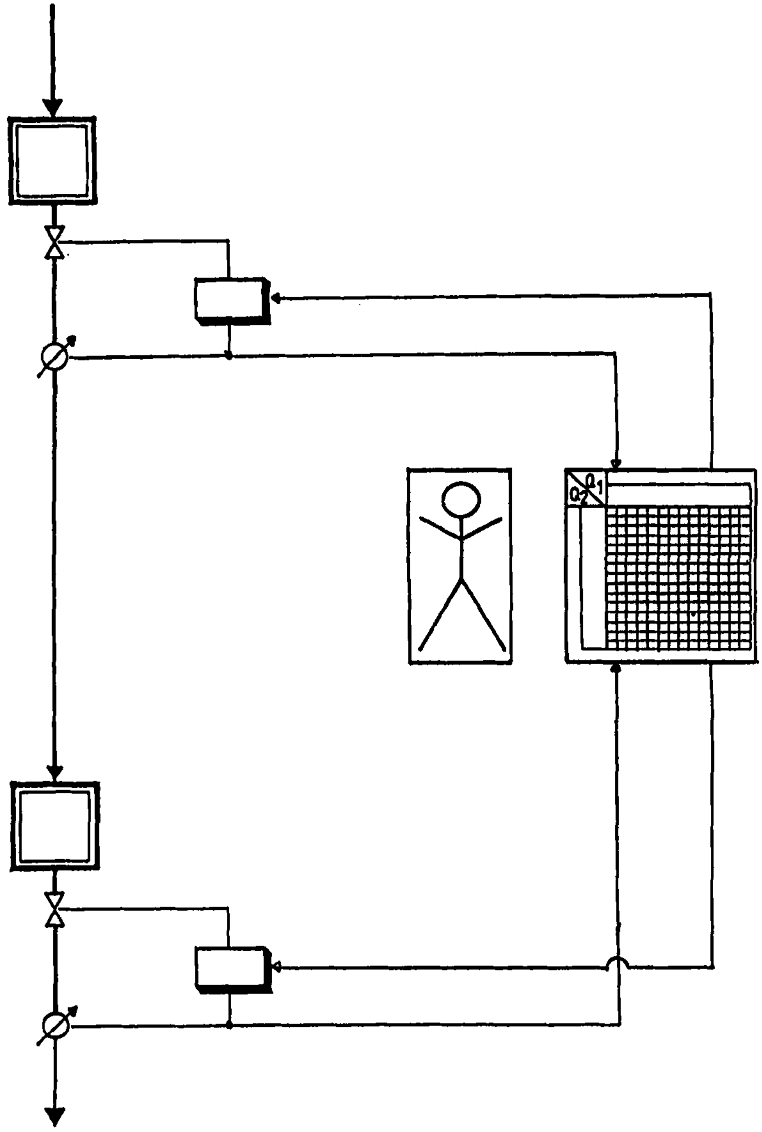


FIGURE 11.11 ***Matrix control***

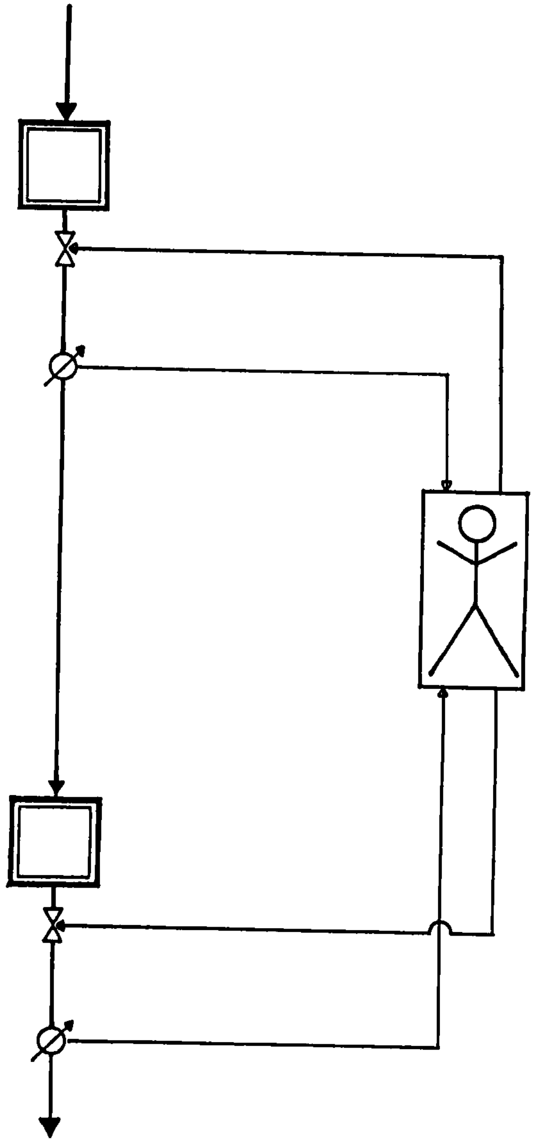


FIGURE 11.12 *Manual control*

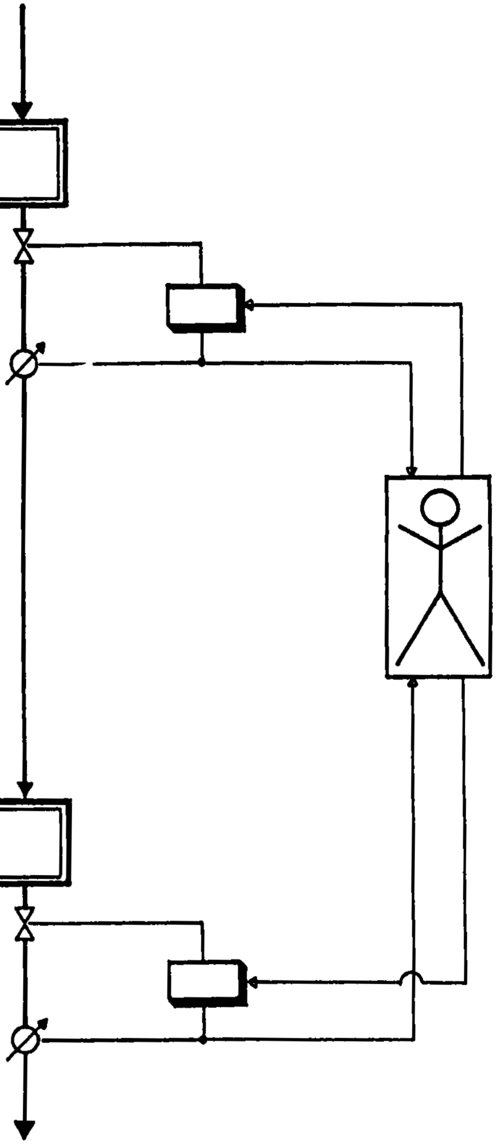


FIGURE 11.13 *Supervisory control*

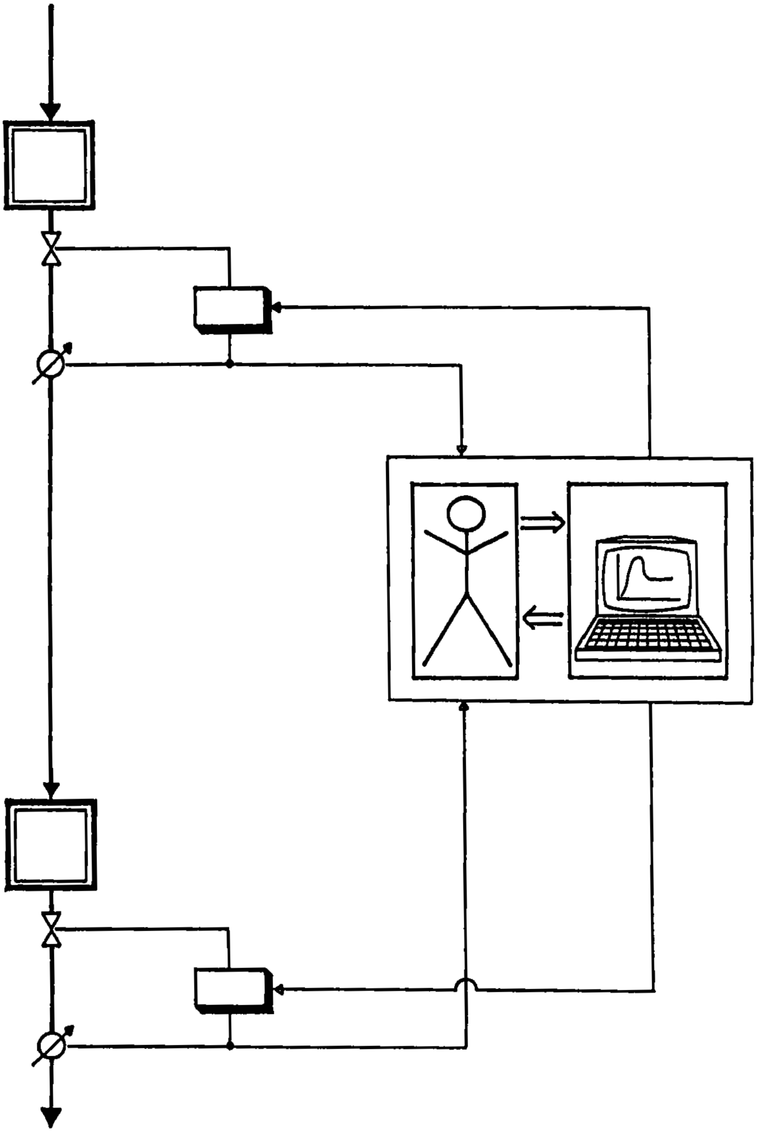


FIGURE 11.14 *Global control with decision aid system*

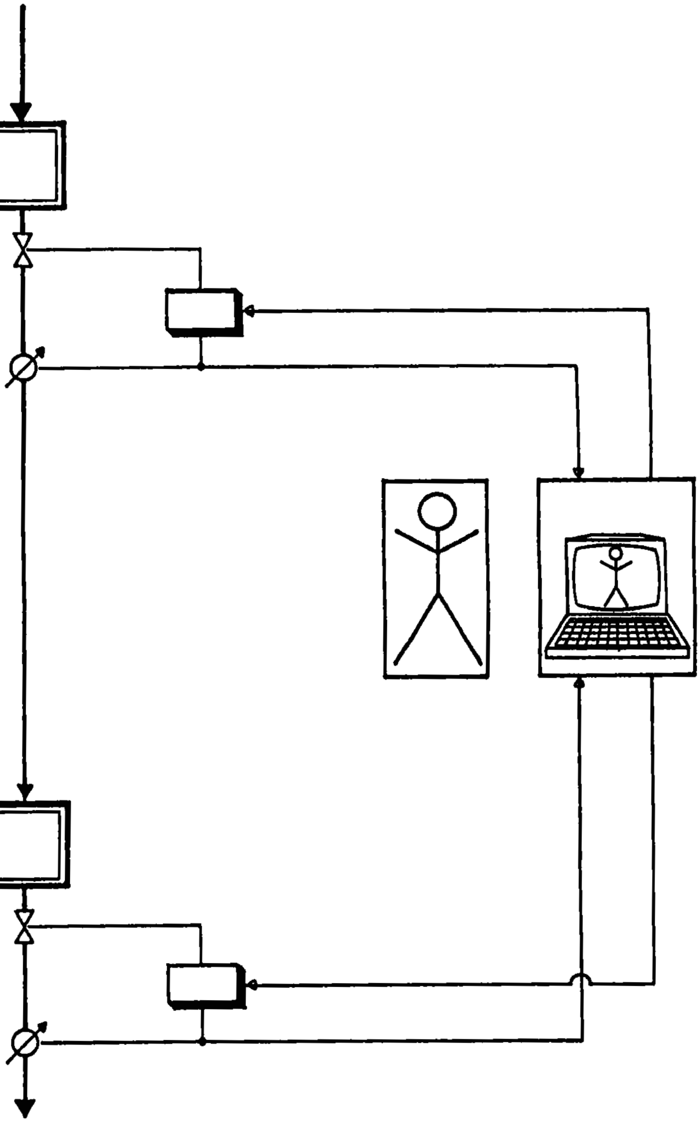


FIGURE 11.15 *Global control with expert system*

An expert system uses known information about the system, its operational state and precisely described reasoning processes to retrieve (or produce) new and previously unknown information about the system. For the development of a control strategy in real time one needs to specify the state of the flow process (past, present, and possibly future) and the goals to be reached.

In an expert system three knowledge levels have to be distinguished. Level 1 concerns knowledge about the state of the drainage system to be dealt with (basis of facts). Level 2 is knowledge about how the desired information is to be derived from the actual state. These are the production rules. Level 3 is the interpreter which decodes these rules, applies them to each particular state of the system to allow consequences to be drawn.

For the development of a RTC strategy, level 1 knowledge is derived from the results of simulation models and measurements.

Level 2 knowledge corresponds to the operator who, in real cases proposes a solution and applies it. This is the expert's knowledge which gives its name to the methodology.
In opposition to optimization techniques, the purpose of finding the best solution is replaced by the one of finding a good solution or an acceptable solution.

Level 3 knowledge deals with the actual mechanisms involved in the reasoning process and how it is to be translated into computer programs.

Ideally, a self-learning expert system is obtained that should be able to accumulate more experience than any human being in his life. In fact, the basis of rules can not only be used to determine the strategy, but also to evaluate the rules that lead to the strategy, analyse them in relation to the operational situation and eventually come up with more appropriate rules (meta rules).

Expert systems have great flexibility in formulating different scenarios in that quantitative (e.g. m^3/s flow) and qualitative (e.g. heavy rain), deterministic (e.g. storage full) and probabilistic information (rain likely) can be combined. If subjective statements are to be included, obviously the expert system can only be used interactively with an operator. Expert systems are extremely application oriented and hardly transferable to other RTCS. In fact, their name includes the idea that the control decisions are not found through a model of the physical system but through a model of an expert (here: the operator).

5. APPLICATIONS

The first comprehensive approaches to RTC in drainage system had been initiated in the United States in the end of the 1960s. In the 1970s a number of demonstration projects were implemented and are mostly still under operation. Some of these are described below.

The county of Seine–St–Denis, close to Paris, has operated a large RTCS for more than a decade. Other existing or planned RTCS in France include the counties of Hauts–de–Seine, Val–de–Marne, and the cities of Nancy and Bordeaux.

Large parts of The Netherlands are below sea level. There, virtually every drop of water in drainage system has to be pumped, mostly from CSS. Hence, in a strict sense, all drainage systems in The Netherlands are under RTC. Integrated systems, of course, are not so frequent, e.g. Rotterdam, Wervershoof, Utrecht, and Eindhoven.

After the re–organization of water resources management and administration in England the number of integrated RTCS is steadily increasing. Solely dedicated to wastewater control, for example, are the systems in Newcastle–upon–Tyne, Birmingham/ Wolverhampton, and Grimsby/ Cleethorpe.

German states enforce specific combined sewer overflow regulations. These resulted in the construction of thousands of small detention tanks to catch the first flush of pollutants in combined sewer systems. Some 20 % are equipped with controllable regulators. Quite a few employ some form of regional control, mainly to avoid downstream CSO caused by upstream tank releases. As for large supervisory RTCS examples can be found in cities such as Bremen, Hamburg, and Munich.

5.1 United States

The Detroit Metro Water Department (DMWD) serves 11 communities with 35 % of the Michigan population, and large industrial inflows [47]. From rather low gradient sewers CSO is diverted into the Detroit, Rouge and Huron rivers through 110 outfalls. The controllable in–line storage, related to the entire catchment area is only 8.3 m^3/ha. DMWD operates an extensive flow level monitoring system at some 250 sites using very simple and apparently reliable air pressure cells. Operation routinely relies on National Weather Service radar data which allows the water level in the system to be pumped down up to three hours before an approaching storm starts to rain over the catchment. The conditions of 70 tide gates are monitored to detect blockage and other operational problems. The RTCS is operated in 24 hour supervisory control, together with the water supply system. A reduction in CSO duration from 5 % to 1.3 % has been achieved. Central automatic control is not implemented.

The Municipality of Metropolitan Seattle has operated a RTCS for approximately 15 years to reduce CSO into Puget Sound (Pacific Ocean), Duwamish River and Lake Washington [32]. The system can be run in local automatic, central supervisory, or central automatic control modes. Original plans to adjust the decision matrix as a function of local rainfall have not been implemented yet. Average CSO volumes for different modes of control are as follows: 7.0 million m^3 for "no RTC", 4.4 for local control, 1.9 for central automatic control, and 1.5 for central supervisory control. Planning is currently underway to upgrade the system, and replace the old computer hardware by state–of–the–art technology. Because of the success of the RTCS, plans for separation have been deleted. Additional benefits of the system, according to the operational personnel, have been reduced flooding and monitoring and source detection of gasoline spills.

The Northeast Ohio Regional Sewer District operates major trunk sewers, interceptors, and treatment plants for the City of Cleveland, Ohio and 33 suburban communities [9]. Before remedial measures, almost every rain event created CSO at some 600 overflow points into the Cuyahoga River and Lake Erie. The RTCS was initiated in 1975 with 3 regulators, and a greatly expanded system became operational in 1983. The system now controls some 50% of the original CSO volume. Operation up to now has been local automatic with central supervision. Centrally adjustable control actions are actuated by on–site microprocessors. The control centre has a back–up computer which is intended for later simulation and optimization of the control strategy. Operator interfaces include colour CRTs with graphic displays, "trending" software, alphanumeric and dedicated keyboards. In terms of hardware, this system is one of the most advanced in North America.

The City of Lima, Ohio operates a RTCS for its combined sewer system that covers some 15 km^2 [8]. It and Seattle are the only two RTCS with central automatic operation. However, neither system employs real time modelling of the rainfall/runoff process. In Lima, 8 sluice gate regulators are operated from the central control room. The system has been operating for some 5 years without major problems. The treatment plant inflows are not only regulated during wet but also during dry weather by the control gates. After storms the system is automatically flushed by opening interceptor gates beginning downstream. After hours the system is running automatically without supervision.

5.2 Japan

A typical Japanese RTCS is the supervisory control system of the Neya River Basin. Similar systems are located in Tokyo, Okayama, etc. This system consists of two treatment plants and 17 pumping stations in a catchment of approximately 270 km^2. The data, which includes water levels in the sedimentation tank, the number of operating pumps and the valve operation in the pumping stations is gathered by the telemetry network using public telephone lines. The most important goal is to prevent flooding which occurs in urban areas once in a few years. The operation of the drainage system, therefore, is linked to the systems of other organizations such as the river department or the rescue department.

Communication among several departments allows control of the overall system by also using the regulator units of other departments. Examples of such control units are the detention basins and the gate at the junction of two rivers. By coordinating these units, the operator may find additional control capacity compared to what would be available in the local system only.

The global control of the drainage system is expected to benefit from a connection with the meteorological information system. This system is equipped with radar and AMeDAS (Automated Meteorological Data Acquisition System). 17 radar sites are presently operating in Japan covering all the country and the coastal waters. AMeDAS is also equipped with remote monitoring stations over all Japan (one station per 289 km^2). There, rainfall precipitation, wind velocity and direction, temperature and atmospheric pressure are measured. These data indicate the spatial distribution of rainfall and allow the paths of cloud masses and precipitation to be followed. By using these data, the Japanese Meteorological Agency developed rainfall forecasting models and, in the near future, will commence with releasing the information of forecasted rainfall for a time horizon of several hours. These forecasts are very useful for RTC of drainage system.

In the City of Osaka control units were equipped with a computer to execute control of a neighbouring relay pump station remotely and automatically. However, application was limited to small scale units which consist of a single pump located near to other manned control units. Thereby, in cases of failure of the automatic system the staff is able to take over manual operation.

In the Kanagawa District a demonstration system was installed featuring water level and velocity sensors and a glassfibre data transmission network. The system is to monitor sewage flows within a large urban drainage area. Operators can use this data to override local control functions if need be. In–line storage can also be activated if this kind of flow information is available. Since such a data transmission system is rather expensive it is used for other purposes as well.

5.3 France

The RTCS in the Departement Seine–Saint–Denis covers an area of 256 km^2. Both flooding and pollution problems led to an RTCS which seemed to be the only way to handle the phenomenon of localized storms in such a large drainage system.

At present, with approximately half of the projected hardware installed, the Seine–Seine–Denis RTCS features

- a telemetred monitoring network consisting of 12 rain gauges, 20 flow gauges, 30 overflow gauges,
- local monitoring stations, i.e. 8 rain gauges, 80 flow gauges, 20 overflow gauges),
- 20 locally controlled pumping stations with central supervision,
- a real time information retrieval and alarm system at the homes of operators on–call.

This system is still under expansion. One of the six main drainage districts is now experimentally under global control mode. Its decision aid system is designed to cope with the large amount of available information, with short term rainfall forecasting and flow propagation, with the evaluation of the best control strategy and its presentation to the operator. Additionally 10 units such as diversion structures and retention basins are controlled separately.

Heavy flooding has been experienced in Bordeaux while upstream retention basins were not filled. Off–line simulation of automatic control have been carried out. They showed the efficiency of automatic control for a retention basin. It was decided to install an automatic RTCS for this basin. The other basins are still manually operated, i.e. crews are dispatched to the basins whenever heavy storms are forecast. No on–line decision aid system is planned. However, use of radar information on storm activity is going to be implemented.

In the Departement Hauts–des–Seine, overflow gates to the Seine River required automatic control. This is to reduce the frequency of combined sewer overflows and to avoid river water entering the drainage system at high flow levels. The stations are controlled locally.

As yet, the performance of this local RTCS has not been simulated off–line. No evaluation of the routine operation has been carried out. However, the management is aware of a need for a new design of the existing RTCS.

The City of Marseille experiences frequent beach and harbour pollution due to combined sewer overflows and extensive floodings during heavy Mediterranean storms. Off–line modelling of these phenomena has been started. A monitoring network including 5 remotely transmitting raingauges was installed. Two regulator stations (one overflow gate, one retention basin) started operation in 1988. A decision aid system, based on the concept of expert systems is going to be developed.

The City of Nancy experienced heavy flooding in the downtown area, after which two large detention basins have been constructed within that area. Moreover, the District of Nancy is aware of the potential benefits RTC can offer to other public services such as public transport and drinking water distribution. Therefore, a comprehensive RTCS has been installed that allows to operate the drainage system and the water distribution using the same control computer. The two basins are locally controlled with central supervision. Global control has not yet proven useful. The district management is presently concentrating on rain and flow measurements including laboratory and field testing. Also, a radar for storm surveillance is under evaluation. Up to now no decision aid system is envisioned.

The Departement Val–de–Marne, located South of the Departement Seine–Saint–Denis is planning a RTCS to overcome basically the same problems as are occurring in its neighbouring departement. As an additional problem, some major water intake structures of the City of Paris water supply system are located just downstream the Val–de–Marne catchment. According to the Clean Seine River Program operations of the drainage system have to be drastically improved. One of the first actions is a RTCS for a new interceptor serving the new Valenton treatment plant. The existing system features a monitoring system of 12 digitally recording raingauges, 40 digitally recording flow gauges and a radar image processing system. Two pumping stations and 3 overflow structures are under local control. There is also an on–line communication link to the planning and operation divisions including a data base improve planning and operation. Off–line simulation of the controlled drainage system is underway.

5.4 The Netherlands

One of the first RTCS in the Netherlands was installed by the water authority of the Dommel at the treatment plant of Eindhoven. The system controls a large gravity flow sewer main to which many small villages are connected. The storage as well as the capacity of the main is controlled by 3 regulating stations equipped with regulating gates. The global control is carried out in the central control room manually by means of set point adjustments. Also fully automatic overall control is implemented, with varying results. The system has been in operation since 1975.

In the region of West–Friesland in the province of North–Holland a fully automatic global control system has been functioning with very good results since 1983. The system controls 28 sewage pumping stations, using spatial distribution of rainfall in the area of the regional treatment plant situated at Wervershoof.

In the City of Amsterdam a large management information system for more than 200 sewage–pumping stations is in operation. The pump data is monitored in a central control room and used for maintenance or operational purposes. The pumps can be controlled manually from the control room. The pumping stations are provided with telemetry equipment, which can be adapted with software for local pump control.

In the City of Rotterdam the sewer system is also used for drainage of the groundwater. In former days the water was pumped to the receiving waters. Nowadays the water is pumped to the treatment plants with a hydraulic capacity far below the total pumping capacity of the sewage–pumping stations. The municipality of Rotterdam installed an advanced remote–control system for several dozens of pumping stations. The pumping stations are equipped with PLC–controlled and speed regulated pumps, maintaining a certain receiving water level under local control mode. This water level can be manually adjusted from the central control room. Automatic integrated control of the storage capacity is planned for the future.

Many cities in the Netherlands have planned to install systems with central monitoring and remote control of pumping stations. The City of Utrecht will realize a fully automatic system by the end of 1987 also optimizing the use of the storage in the sewer system.

The City of Schiedam is about to start the implementation of a RTCS for optimal water management under extreme rainfall conditions. The city is situated almost completely below sea level, so there is no possibility of gravity overflow out of the sewer system.

5.5 United Kingdom

Along the Tyneside over 200 outfalls discharge crude sewage into the river. Because of the flow patterns and tidal effects it was found that raw sewage was taking up to 10 days to clear the estuary. The river water quality suffered and the neighbouring beaches were affected by sewage deposits. A scheme was initiated to clean up 32 km of estuary and 14 km of beaches by constructing sewers on either bank of the river Tyne to intercept the old outfalls. Sewage is now conveyed by gravity and pumping mains to the new treatment work at Jarrow and Howden. The majority of the old outfall pipes have been retained as overflow pipes. Sluice gates control the flow of sewage into the interceptor sewer and at times of heavy rainfall the interceptor is used for storage. All the gates, siphons and pumping stations are controlled from the telemetry centre at Howden.

Similarly in Liverpool where 24 outfalls, discharging crude sewage into the Mersey estuary, are intercepted by a deep sewer. At each interceptor site, an on–line storage tank has a calibrated vortex outflow device discharging sewage into the interceptor sewer. Controllable sluice gates allow the storage tanks to be activated and isolated. The overflow pipes are below high tide level and estuary water has to be prevented from entering the tanks.

In the Black Country to the West of Birmingham the poor quality of effluent from a number of old sewage treatment works has led to an unacceptable water quality in the river Tame. To address this situation, the Severn Trent Water Authority is constructing a trunk sewer system to enable the works to be closed and the flow transferred to Minworth Water Reclamation Works, where treatment capacity is available. The catchment served is some 1500 km^2 and, as such, can give rise to storm flows which cannot be coped with by the trunk sewer system. There may be value in using a RTCS to optimize trunk sewer capacity and minimize impact on the river from sewer overflows by using balancing and inlet control. Flow gauges and level sensors have been installed in the sewer, together with water quality monitors in the river to provide the information necessary to model the system, predict the value of options and design whichever systems of control and operation is proven to be necessary.

RTCS has been employed at a number of sea outfalls, i.e. Weymouth and Grimsby where sewage following preliminary or partial treatment is pumped up to the headworks. Discharge through the sea outfall is by gravity. The optimisation of the pumping regime to prevent flooding is the main objective but in addition other objectives are sought, such as initiating flushing velocities periodically to scour the pipe of deposits or saline water, control the treatment processes and at the same time minimize the energy costs.

5.6 Germany

One of the first RTCS in Germany was installed in the City of Bremen. Two large storage tanks, additional in–line storage, 5 pumping stations and overflow gates are under supervisory RTC. The system has been in operation since the early 1980s and is presently under review for improvement and extension.

The City of Hamburg operates one of the oldest "modern" sewer systems in the world (construction began in the 1850s). Serious sedimentation, corrosion and overflow problems require a spectrum of actions of which the installation of a RTCS is only one. Ultimately 5 regional control centres will control a number of detention ponds, gates, pumping stations, and inverted syphons in the system.

The City of Munich is planning to expand the storage capacity in its combined sewer system drastically. Before the turn of the century some 500 000 m^3 of additional pond storage will be available, mostly as underground concrete tanks. For the development of the control strategy of these ponds the existing remote monitoring system is going to be expanded to 20 raingauges, 66 water level gauges and 4 continuous water quality monitoring stations along the receiving Isar River.

The City of Nürnberg presently constructs an oversized interceptor sewer. The storage volume of 50 000 m^3 will be controlled by 7 large sluice gates with by–passes. It is planned to allow for both simple local control as well as advanced systems control strategies.

The Ruhrverband, a large sanitary district in the heavily industrialized Ruhr area, is in charge of 150 detention ponds (another 350 planned) many of which are controlled by fixed set point regulators. In the networks of Brilon, Neheim, and Ense, RTCS are either already installed or planned.

A number of small town systems (Herrenberg, Münsingen, etc.) have some semi–automatic RTCS for their stormwater ponds. Whereas the activation of the ponds is automated the emptying commands are manually given from the treatment plant personnel.

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Chapter 12

LEGISLATION RELATING TO HIGHWAY POLLUTION

D. Hughes

Contents Page

1. MOTOR VEHICLES AS POLLUTANTS : LEGAL PRINCIPLES

Legal regulation of the motor vehicle is no new phenomenon [1]. Such regulation is part of the laws relating to the environment, but there is no one world–wide code for controlling motor vehicles as pollutants. There are many reasons for this. International Environmental Law is still in a formative state. Within nations there may be no concensus as to the degree of control that should exist over motor vehicles. However, many legal commentators would not disagree that an effective system of control must be based on three essential principles with regard to pollutants : prevention, mitigation and compensation [2]. Few legal systems deny the notion that a person injured by an activity should receive compensation, either from the author of the activity or from the State, or an insurance fund. Likewise legal systems commonly lay down provisions for mitigating effects of deleterious activities, for example by allowing courts to make orders stopping activities, or by making provision for grant aided schemes to lessen an activity's impact. Prevention, however, of polluting activities is not always well represented in law. The deleterious nature of an activity must be clearly observable, and its domestic consequences will often have to outweigh the national advantages it brings before a state takes preventive action. Furthermore, adequate preventive technology may be a pre–condition to action. Action once taken may assume a number of legal forms. For example, control may take the form of specification standards which seek to prevent harm from a process by laying down design and construction, etc., requirements, or they may be emission standards which lay down upper limits for emissions from specified activities [3].

To eradicate an environmentally deleterious activity by law the following conditions are required: generally agreed perception of a clear hazard, whose nature and consequences are understood, and which has been authoritatively scientifically analysed and described; technological or other means to deal with the problem, and a convergence of interests between those responsible for the hazard and the rest of society so it becomes socially and economically worthwhile to take action, and to bear any increased costs, inconveniences or loss of individual rights and freedoms flowing therefrom. Where these conditions are not met legal action can probably be no more than a 'control', that is a set of measures that accept the continuing existence of a problem and seek to set mitigating curbs on it, while not seeking eradication. This is 'the best practicable means' approach. In overall terms many, if not most, legal measures in respect of vehicle pollution currently fall into this latter category.

There are some aspects of the vehicle pollution issue that are, or may be, subject to eradicating or preventive legal measures. The Experts Group on Environment and Development have urged that States should establish specific environmental standards, and in particular emission standards prescribing maximum permissible releases of specified pollutants from, inter alia, means of transport [4]. These commentators are not alone in stressing that mitigating the effects of pollution, and paying compensation to those damaged by it are not substitutes for preventing it in the first place.

The difficulties of establishing standards for limiting emissions from motor vehicles have been examined by Georgiades, Chiron and Joumard in their paper "Establishment of Atmospheric Pollution Standards for Motor Vehicles" [5]. These authors point out that emission standards are both controversial and complicated, and take account of a diverse range of considerations, which differ from nation to nation. Thus emission standards may be based on constant speeds for engines, or on the basis of acceleration; they may specify particular gear ratios; they may be based on idling engines; they may take into account the opacity of smoke or its ability to colour a filter; they may take account of volumes of exhaust gases; they may be based on tests carried out on a rig designed to reproduce certain kinematic cycles that indicate emissions at particular speeds. The European Communities ECE 15 cycle is based on a maximum speed of 50.0 km per hour with an average speed of 18.7 km p.h. starting with a 'cold' engine. Such a set of test values is good for determining emissions of carbon monoxide and hydrocarbons, but is less satisfactory with regard to nitrogen oxide emissions which increase at high speeds and it was for this reason that the Extra Urban Driving Cycle was introduced, as described in Chapter 2. In the United States, save for California, the test parameters for the kinematic cycle are based on slow or fluidly moving urban traffic, and also on main road speeds of up to 91.2 km p.h. with an average speed of 34.3 km p.h. California uses parameters that take into account even faster speeds of 96.4 km p.h. and average speeds of 77.4 km p.h. In addition emission standards may include endurance requirements and guarantees that after a certain mileage, or length of use the emission limits will still be met by the vehicle in question – The United States, Canada and Japan all have such requirements. Standards may also be flexible in that production line vehicles may be allowed to produce more emissions than the individual vehicle selected for testing for approval of the type as a whole with regard to emissions. This is the case in the European Community where type approval standards are utilised. (See further below).

It is against the foregoing that the material below must be examined. The principal area of study will be United Kingdom law, but the law of other jurisdictions will be examined on a comparative basis, and special attention will be paid to the legal regimes adopted by the member states of the European Communities, and the United States of America.

2. LEGISLATION RELATING TO EMISSION CONTROL

2.1 The problem and methods of dealing with it

The quantities and composition of pollutants produced by motor vehicles were described in Chapter 1. The following summary reviews the properties of those pollutants which can be controlled by legislation applied at source. Petrol engined vehicles are the main source of carbon monoxide (CO) emissions [6]. In addition they also produce nitrogen oxides (NO_x), hydrocarbons and some particulate matter, which, in the case of vehicles using leaded petrol will include lead compounds. Vehicles utilising unleaded fuel, however, emit few particulates [7]. Diesel engined vehicles produce 'smoke' which is made up of carbon particles which have absorbed exhaust gases such as hydrocarbons. Indeed, comparatively, diesel powered vehicles, in terms of unit weights of fuel used, emit some four times as much particulate matter as vehicles powered by leaded petrol [8, also 1.3 of Chapter 1].

Each of these pollutants can have deleterious effects. NO_x is implicated in the formation of acid precipitation which may affect plant and animal life. NO_2, in high concentrations, can damage health, as can CO.

Vehicle emissions in the United Kingdom of NO_x have substantially increased over a period of ten years according to a written answer to a question in the House of Commons (H.C. Written Answers, 30th November 1988 col. 278). In 1977, 849 thousand tonnes of NO_x were emitted, and this rose to 1031 thousand tonnes in 1987, with a 7.4% increase in that year on previous figures. 40% of NO_x in the United Kingdom comes from vehicles. Vehicle carbon dioxide emissions have also risen over the same period from 20.0 to 26.6m tonnes, and carbon monoxide from 3.70 to 4.47m tonnes. Concentrations, of NO_2 exceeding World Health Organisation guidelines have been detected in certain urban areas in the United Kingdom, in particular in London [9]. Also in London there are records of severe concentrations of smoke, principally resulting from particulate matter released by motor vehicles [10].

An average family car running 13,000 miles p.a. produces some 78 lbs (hydrocarbons), 50 lbs (NO_x) and 1,056 lbs (CO) – about half a ton of polluting material. With a three way catalytic converter, such as have been standard since the early 1970s in the U.S.A., pollutant emissions can be reduced to 7 lbs (hydrocarbons), 8 lbs (NO_x) and 30 lbs (CO).

Additionally hydrocarbons emitted from vehicles, or as evaporated from fuel spillages, can be implicated in photochemical smog formation (Section 2 of Chapter 1). Smog damages human and plant life, corrodes materials and is unpleasant to smell [11]. Less volatile hydrocarbons are encountered in particulates. Some hydrocarbons, the polycyclic aromatic group, are known to be carcinogenic, though a precise epidemiological link between exposure to vehicle exhausts and increased risk of cancer was not known by the Royal Commission on Environmental Pollution in their 1984 report [12], also Chapter 7. Lead in petrol as a pollutant was examined by the Royal Commission on Environmental Pollution in their 1983 report [13]. Lead is generally toxic to vertebrates, and much of this pollutant is now irretrievably released into the environment, (Chapter 8). Though lead is taken into the body primarily through food and drink, the Royal Commission concluded that lead from petrol can enter the human body through a number of ways, and that most adults could derive up to 20% of their level of blood lead from this source [14]. The Commission's 1983 recommendation was that lead in petrol should be reduced and finally phased out by 1990 [15].

There are a number of methods by which atmospheric pollution caused by vehicles can be controlled, or, in some cases, eradicated : regulation of the way in which vehicles, in particular their engines and exhaust systems, are constructed; regulation of fuels used, and regulation of vehicle use, by fiscal means for example. Vehicle emissions can also be controlled under general "across–the–board" air quality standards, for which see Section 5. Many of these methods of regulation are to some extent represented in United Kingdom law.

2.2 Regulation of Construction

United Kingdom law in relation to construction controls is based on obligations deriving from membership of the European Communities. European Community Law has as its primary source the various treaties made by the member states, and especially the Treaty of Rome 1957 and The Single European Act 1986 [16]. The treaties confer many powers on the various organs of the Communities, but in the present context we are concerned with Article 189 of the Treaty of Rome, under which the Council and Commission of the E.E.C. may issue binding directives to member states. Such measures only bind as to the end to be achieved, not as to the means to achieve that end: those are left to the choice of member states. Directives therefore are not issued to bring about total uniformity of law and practice, but to advance approximations between the laws of member states. So far as Community Directives relating to vehicle engine emissions are concerned, their development is further guided by non–binding regulations issued by the United Nations Economic Commission for Europe (ECE) [17].

With regard to the implementation *within* the United Kingdom of European obligations, it is the general rule that "secondary" or "delegated" legislation, as opposed to Acts of Parliament, is used to embody legal requirements. A wide power to make such delegated legislation exists under Section 2 of the European Communities Act 1972,[18] and, in relation to road vehicles, the more specific powers used are those under Sections 41 and 64 of the Road Traffic Act 1988. Regulations are made under those powers by the Secretary of State for Transport after consultation with representative organisations under Section 195 of the Road Traffic Act 1988. Currently we have the Road Vehicles (Construction and Use) Regulations, SI 1986 No. 1078, which came into force on 11th August 1986. These have been subsequently amended by, inter alia, The Road Vehicles (Construction and Use) (Amendment) Regulations, SI 1987 No. 676 and the Road Vehicles (Construction and Use) (Amendment) (No. 2) Regulations, SI 1987 No. 1133 and The Road Vehicles (Construction and Use) (Amendment) (No. 6) Regulations, S.I. 1988 No. 1524. Hereafter these will be collectively referred to as the Construction and Use Regulations.

Regulation 61 of these Regulations contains detailed rules relating to the emission of smoke, vapour, gases and oily substances, etc. The general rule is that every vehicle must be constructed so that it does not emit any avoidable smoke or visible vapour, and, more particularly, must comply with specific requirements contained in the table to Reg. 61(3). Motor vehicles are defined by the regulations as those that are mechanically propelled and intended or adapted for use on roads.

Thus in the case of vehicles propelled by compression ignition engines, and equipped with devices to facilitate engine starting by the supply of excess fuel to the engine, it is a requirement that such devices cannot be readily operated by persons inside the vehicle, *unless* (a) the vehicle is a works truck (i.e. it is essentially designed for in premises use) or (b) the vehicle having the device is so designed and maintained that *either* use of the device after the engine is started cannot cause an excess of fuel to be supplied to the engine, *or* the device does not cause any increase in the smoke or visible vapour emissions from the vehicle. In the case of vehicles first used on or after 1st April 1973 and propelled by compression ignition engines, it is a further requirement that the engine shall be of a type test certificated in accordance with *British Standard Specification for the Performance of Diesel Engines for Road Vehicles*, BS AU 141 a; 1971, published on 19th May 1971. Where a vehicle was first used on or after 1st January 1972 and is propelled by a spark ignition engine (other than a two stroke engine) it is a requirement that the engine must be equipped with means sufficient to ensure that, while the engine is running, any vapours or gases in the engine crank case, or in any part of the engine to which vapours etc., may pass from the crank case, are, so far as is reasonable practicable, prevented from escaping into the atmosphere other than through the engine's combustion chamber. However, this requirement does not apply to two wheeled motor cycles, nor to vehicles falling within the next classification. This applies to vehicles first used on or after 1st October 1982 and propelled by spark ignition engines. Here the requirement is that the vehicle must comply at the time of first use with European Community Directive 78/665 or 83/351 (or ECE Regulation 15.03 or 15.04). There are exempted from this classification: (a) vehicles with a maximum gross weight exceeding 3500 kg; (b) vehicles having only two wheels; (c) vehicles with an unladen weight of less than 400 kg, and (d) vehicles with less than four wheels or having maximum speeds not exceeding 25 km/h [19].

There is a general exemption from the basic Regulation 61(1) requirement that vehicles should not emit avoidable smoke or vapour for vehicles complying at the first time of their use with, in the case of compression ignition propelled vehicles, Community Directives 72/306 or 77/537 (the latter applying to agricultural vehicles) (or ECE Regulations 24.01, 24.02 or 24.03), or, in the case of spark ignition engine vehicles, Community Directives 78/665 or 83/351 (or ECE Regulations 15.03 or 15.04) [20]. Rather there is a scheme of regulatory emission control based on European Community Directives under the Construction and Use Regulations which is mandatory for spark ignition vehicles first used on or after 1st October 1982. Further under Regulation 61(5) no person may use, or cause or permit the use on a road of any motor vehicle from which any smoke, visible vapour grit, sparks, ashes, cinders or oily substances are emitted, if such emissions cause, or are likely to cause, damage to any property or injury or danger to any person who is, or who can reasonably be expected to be, on the road. Where, in the case of a compression ignition engine, there is a start–facilitating excess fuel delivery device, the vehicle so equipped is not to be used on a road if the device is used while the vehicle is in motion. Likewise in the case of compression ignition engined vehicles first used on or after 1st April 1973 (whether or not deemed to comply with the requirements of the law by virtue of compliance with Community Directives 73/306 or 77/537) if the fuel injection equipment, the engine speed governor, or any other propelling parts of the engine have been altered or adjusted to increase smoke emissions, the vehicle may not be used on a road.

Under Regulation 61(6) where a vehicle was first used on or after 1st January 1972 and is propelled by a spark ignition engine, other than a two stroke engine, it may not be used on a road unless the means adopted to ensure that vapours and gases escape, so far as is reasonably practicable, only through the engine's combustion chamber are in good working order. This requirement does not apply to vehicles first used on or after 1st October 1982 and which comply with Community Directive 78/665 or 83/351 (or ECE Regulation 15.03 or 15.04). Under Regulation 61(2) where motor vehicles use solid fuel (e.g. a traction engine) they must be fitted with trays or shields to prevent ashes and cinders from falling onto roads, and must have efficient appliances to prevent emissions of sparks or grit. The 'first use' of a vehicle for the purpose of the Regulations is generally the date of its registration [21].

Though the Construction and Use Regulations provide the legal emission standards which must be met, they did not in 1988 completely translate into United Kingdom law the most stringent requirements of the European Community Directives on vehicle emissions (see Directive 83/351/EEC and 88/76/EEC). The Regulations did, however, enforce the standards of Directives 72/306/EEC (O.J. L190 20.8.72) (Com (71) 148/4) on diesel engine exhaust opacity, and 78/665/EEC (O.J. L223 14.8.78) on Petrol engine emissions. The emission standards of 70/220/EEC (O.J. L76 6.4.70), the first community directive on petrol engine emissions, amended and added to by the subsequent Directives required that, in tests (the manner of performing which was [and is] laid down in the Directives) the mass of carbon monoxide, the mass of hydrocarbons, and the mass of nitrogen oxides had to be less for vehicles of a given reference weight than certain specified figures. Reference weight was [and is] expressed in terms of the 'Reference Mass' in kilograms of the vehicle, i.e. the mass of the vehicle in running order less the uniform mass of the driver of 75 kg and increased by a uniform mass of 100 kg [22]. As examples of permitted emissions, where a vehicle is of a Reference Mass of less than, or no more than, 750 kg, the mass of Carbon Monoxide in grams per test must not exceed 65, hydrocarbon mass must not exceed 6.0, and nitrogen oxides must not exceed 8.5. For cars of greater masses the limits are higher, rising to 143 grams (carbon monoxide), 9.6 grams (hydrocarbons) and 13.6 grams (nitrogen oxides) for vehicles of 2,150 kg or more reference mass [23].

It should be notied that in the context of the regulations 'vehicle' means any vehicle with a positive ignition engine intended for road use and having at least four wheels, a permissible maximum weight of at least 400 kg, and a maximum design speed equal to or exceeding 50 km/h (agricultural tractors and public works vehicles being excluded) [24]. It is also a requirement of the Directive that components in vehicles likely to affect the emission of gaseous pollutants must be so designed and constructed as to enable the vehicle, when normally used, and despite vibration, to comply with the requirements of the Directives.

It should further be noted that the Construction and Use Regulations were amended in 1988 so that the more stringent requirements of recent Community Directives will be incorporated shortly into United Kingdom law, see further below.

72/306/EEC lays down standards with regard to the opacity of emissions of pollutants from road vehicles equipped with diesel (i.e. compression ignition) engines. As with 70/220/EEC tests to check emissions are specified in the Directive, and the measure of opacity as tested by the prescribed method must not exceed limits prescribed in the Directive. It is also a general requirement of the Directive that components liable to effect the emission of pollutants from diesel engines must be so designed, constructed and assembled as to enable the vehicle, when used normally, and despite any vibration to which it may be subjected, to comply with the emission limits. The basic requirement is that the light absorbant co–efficient of the gases emitted by the engine at steady speeds as measured by tests prescribed by the Directive must not exceed prescribed limits.

The emission controls for petrol engined cars contained in 83/351/EEC have not, as stated above, been mandatory in the United Kingdom, but vehicles may have complied with them. This Directive introduces lower limit values for emissions to prevent adverse environmental consequences of vehicle emissions, and to reflect advances in vehicle design. The basic rule is that for a vehicle of a given reference mass the mass of CO and the combined mass of bydrocarbons and NO_x obtained in tests must be less than values tabulated in the Directive. So, for example during the period covered by the 83/351/EEC, for a vehicle of a mass of up to and including 1,020 kg, CO in grams/test could not exceed 58, and the combined hydrocarbons and NO_x could not exceed 19.0 g/test. These figures rose, for vehicles of 2,150 kg or more mass, to 110 and 28.0 respectively [25]. With effect from 1st April 1991 the Construction and Use Regulations are amended so that emissions from vehicles propelled by spark ignition engines first used before that date may comply with the requirements of Community Directives 78/665, 83/351 or 88/76 (ECE Regulations 15.03 or 15.04) or in any other case with Directives 83/351 or 88/76 (ECE Regulation 15.04). These requirements will bring United Kingdom law into line with European Community requirements with respect to vehicle emission limits for hydrocarbons, nitrogen oxides and carbon monoxide, and take away the option for relevant vehicles first used on or after 1st April 1991 to comply with the exhaust emission limits laid down in Directive 70/220/EEC as amended by 78/665, instead, however, they may comply with 70/220 as amended by 88/76.

As is described later in this chapter, in Section 4, the amendments made by Directive 88/76 (O.J. 1988 No. L36/1) move from emission limits based on reference mass to those based on engine cubic capacity, so that for vehicles whose capacity is greater than 2000 c.c. the mass of CO must be equal to or less than 25 g/test, the combined mass of hydrocarbons and NO_x must be equal to or less than 6.5 g/test and the mass of NO_x must be equal to or less than 3.5 g/test. For cars between 1400 and 2000 c.c. the respective figures are 30 g/test, and 8 g/test and for those below 1400 c.c. the figures are 45 g/test, 15 g/test and 6 g/test. However, for production series vehicles a little more latitude in emission limits is allowed; for cars over 2000 c.c. the respective figures are CO, 30 g/test, hydrocarbons and NO_x 8.1 g/test; and the mass of NO_x 4.4 g/test, for those between 1400 and 2000 c.c. the respective figures are 36 g/test and 10 g/testm and for those under 1400 c.c. the figures are 54 g/test, 19 g/test and 7.5 g/test. It will be noted that for "middle range" cars there is only a combined emission limit value for hydrocarbons and NO_x together.

The emission limits for gaseous pollutants from diesel engines used in vehicles were further revised in Directive 88/77 EEC, O.J. 1988 No. L.36/33, so that the mass of CO in grams per kWh must not exceed 11.2, hydrocarbons must not exceed 2.4 grams per kWh, and NO_x must not exceed 14.4 grams per kWh. Again a degree of tolerance is allowed to production line engines. Particulate emissions from diesel engines are further restricted by Directive 88/436 EEC, O.J. No. L214/1 so that the basic requirement is that the mass of particulates must be less than 1.1 g/test, with a declared objective of a further reduction to 0.8 g/test in the future.

It should further be noted that the Directives provide requirements for 'type approval' of those vehicles that conform with specified design and construction standards. 'Type approval' is a system whereby competent authorities may issue regulations to approve types of vehicles referred to them by manufacturers, on the basis that they meet, for example, exhaust emission standards. Where such type approval is given, manufacturers may then proceed to produce series of vehicles conforming to the type specification as approved. In the United Kingdom the competent authority in the Secretary of State for Transport, and a comprehensive type approval system exists under Sections 54 – 63 of the Road Traffic Act 1988. Provided the standards of the Directives are met, it is not competent for a member state of the EEC to refuse 'type approval' (either national or EEC) to a vehicle. In other words, vehicles fulfilling Directive requirements must be approved as satisfying legal emission requirements of member states. Furthermore no member state may refuse or prohibit the sale, registration, entry into service or use of a vehicle on grounds of air pollution arising from gases from that vehicle's positive ignition engine if the vehicle satisfies the requirements of 70/220/EEC Annexes I, II, III, IV and VI [26]. For type approval regulations issued in the United Kingdom see, inter alia, S.I. 1984 No. 981 (motor vehicles), S.I. 1982 No. 1271 (goods vehicles) [27]. Currently Type Approval with regard to emissions from motor vehicles (including diesel powered vehicles) exists for those complying with Council Directives 70/220, 74/290, 85/351, 88/76 and 88/436 as a general rule. However, see also further below for fuel requirements. The type approval requirements in general mirror the emission limits of the Construction and Use Regulations and follow the reductions brought about by EEC Directives, See S.I. 1988 Nos. 1522, 1523 and 1669.

It is generally an offence under Section 91 of the Road Traffic Offenders Act 1988 to contravene or fail to comply with Construction and Use regulations, or to use, or cause or permit to be used, on a road a motor vehicle which does not so comply. Enforcement of the regulations is thus a matter for the criminal law, and it would not appear that an action by an individual in the civil courts is appropriate in such circumstances, see Phillips v Britannia Hygenic Laundry Co. Ltd. (1923) 2 K.B. 832 and Tan Chye Choo v Chong Kew Moi (1970) 1 W.L.R. 147 [28]. Provision is made for tests of the condition of vehicles by Sections 45 to 48 and 66 of the Road Traffic Act 1988, (vehicles other than goods vehicles), and by Section 49 of that Act (goods vehicles). Tests for non–goods vehicles are carried out by persons authorised by the Secretary of State for Transport, or by inspectors appointed by him or by district or London Borough councils designated by him. Motor vehicles other than goods vehicles registered after 1st April 1963 now require test certification three years after their first registration, see, generally, Section 47(2)(b). It is an offence, under Section 47 of the 1988 Act, to use, or cause or permit to be used, a vehicle requiring a test certificate in respect of which no certificate has been issued. Goods vehicles are subject to comprehensive testing under Section 49 of the 1988 Act and S.I. 1988 No. 1478. Where a goods vehicle is refused certification as not complying with requirements an appeal may be made to an area mechanical engineer appointed by the Secretary of State, and thence to the Secretary of State himself. Notice of prohibition of use is thus, in the United Kingdom, the ultimate sanction for failure to comply with regulations. Enforcement of such controls lies with examiners appointed by the Secretary of State. Even so, the Royal Commission on Environmental Pollution in their 10th Report drew attention to concern over the problem of ensuring effective supervision of goods vehicles, and good standards of maintenance, needed to control emissions, particularly of smoke [29]. A 'goods vehicle' is defined by Section 192 of the 1988 Act as a motor vehicle constructed or adapted for the carriage of goods.

2.3 Fuel content regulations

Vehicles' emissions can also be controlled by the specification of their fuels. Specific regulation of the levels of sulphur and lead in vehicle fuels is provided for by United Kingdom Law which, as with vehicle emission standards, is influenced by Directives issued by the EEC, see for example on Sulphur 75/716 EEC which was overtaken by the effect of 87/219 EEC as from 1st January 1989 [30]. The formal law in the United Kingdom is contained in Sections 75 and 77 of the Control of Pollution Act 1974. These provisions empower the Secretary of State to make regulations to limit or reduce air pollution by imposing composition requirements on motor vehicle fuels, and by restricting the production, treatment, distribution, import, sale or use of fuels failing to comply with the requirements. There is a consultation process before regulations are made, with representatives of motor vehicle manufacturers and users, fuel producers and users and others conversant with air pollution issues. Enforcing agencies for the regulations are local Weights and Measures Authorities (i.e. generally County Councils, Metropolitan District Councils and London Borough Councils).

With regard to the sulphur content of gas oil, that is a liquid petroleum refined product used to propel diesel engined vehicles, it is the general rule that no person may produce, distribute, import, sell, possess for sale, use, or cause or permit to be used, any gas oil for a diesel engined motor vehicle which contains more than 0.3% by weight (in grams) of sulphur. To implement Directive 87/219 EEC further regulations have been made, the permissible sulphur content is 0.3 grammes per hundreds S.I. 1990 No. 1096. In respect of lead in petrol, a variety of circumstances and pressures have led to gradual reductions of content since 1970 [31]. United Kingdom law once again here mirrors EEC requirements (see Directives 78/611 EEC and 85/210 EEC), and the objective of vehicles running on lead–free petrol is in view, with that commodity to be available widely by 1989. Even so at the start of 1989 only some 5% of fuel used by cars in the United Kingdom was unleaded. Detailed rules relating to lead in petrol are to be found in S.I. 1981 No. 1523, S.I. 1985 No. 1728 and S.I. 1989 No. 547. The maximum permitted amount of lead in petrol from 31st December 1985 has been 0.15 grams, and the production and sale, etc., of unleaded petrol is also permitted. 'Unleaded petrol' is defined as containing no more than 0.020 grams of lead per litre up till 1st April 1990, and 0.013 grams thereafter. The amendment of the Construction and Use regulations in 1988 (1988 S.I. 1988 No. 1524) also inserts relevant new regulations. The new regulation 39A requires petrol engined vehicles first used on or after 1st April 1991 to be designed and constructed to run on unleaded petrol, and further prohibits the modification of such a vehicle so that for road use it is incapable of running on unleaded petrol. Supplementary requirements relate to the means of filling such vehicles with petrol and to symbols indicating that they can run on unleaded petrol. Certain vehicles are, however, excluded from these requirements, that is those complying with EEC type approval certificates issued before 1st October 1989, or where the manufacturer of the vehicle has supplied to the authority which issued the relevant EEC type approval certificate his own certificate stating that adapting relevant vehicles to the unleaded fuel requirements of EC Directive 88/76 would involve a change in material specification of inlet exhaust valves, or a reduction in compression ratio or an increase in engine capacity to compensate for loss of power, [in other words, major engineering work] and that certificate has been accepted. These exemptions will apply for a limited time; for vehicles with an engine capacity of less than 1400 c.c. the vehicle must be first used before 1st April 1992 to comply, for vehicles between 1400 and 2000 c.c. the date is 1st April 1994, and for those over 2000 c.c., 1st April 1993. It is thought, however, that under 2% of new registrations are likely to have to take advantage of these exemptions. The new type approval requirements which will apply generally from 1st October 1989 will provide that type approved vehicles with petrol engines will have to be able to run on unleaded petrol. However, exemptions may be gained in respect of type approved vehicles needing major engineering modifications for a period of time down to 1st October 1993; see S.I. 1988 No. 1522 which also generally modifies type approval requirements relating to exhaust emissions so that compliance with Council Directive 88/76 is acceptable for type approval. Compliance with older less stringent emission standards for type approval purposes is to be phased out over a period of time from 30th September 1989.

Turning briefly from regulation to compensation, in Budden v B.P. Oil Ltd (1980) 124 Sol. Jo. 376 the Court of Appeal held that it is not negligent for a supplier of petroleum products to comply with regulations concerning fuel composition, and that a civil action could not be brought in respect of lead levels in petrol that were in line with what had been sanctioned otherwise by the law.

2.4 Fiscal Regulation

Unleaded petrol is more expensive to produce than leaded, and takes rather more feedstock. To encourage production a tax reduction on duty for unleaded petrol was introduced by Section 1 of the Finance Act 1987 to ensure that the 'pump price' of leadless should be equivalent to that of leaded petrol. It has, however, been arguable that further tax reductions are needed to boost sales of lead–free petrol, as in West Germany where tax concessions of up to 4.8 pence per gallon helped to increase lead–free petrol sales to 22% of the market by the end of 1987. By March 1988 only some 700 out of 20,000 filling stations sold unleaded petrol, and only one motorist in 2,000 was buying the product. In the 1988 Budget on March 15th the Chancellor of the Exchequer increased petrol duties generally, adding some 5p to 6p to the pump price of a gallon of fuel. However, the increase was not imposed on 'unleaded', or 'low–lead' petrol, making this product some 5p per gallon cheaper than ordinary 4–star petrol. Even so, other European nations still had more favourable tax regimes for unleaded petrol, resulting in a price advantage for the product of 18p in Denmark, 15p in Luxembourg and 11p in Switzerland. The 1989 Budget proposals of 14th March introduced, however, a 10 pence differential between leaded and unleaded fuel. The Budget changes were expected to boost demand for unleaded fuel, but the United Kingdom still lags behind many other European nations in this respect. In the Netherlands every petrol station retails unleaded fuel and it accounts for 36.3% of sales. In Denmark 90% of stations retail unleaded fuel and this accounts for 32.5% of sales, while in West Germany where 75% of stations retail unleaded fuel it accounts for 41.8% of sales. On the other hand in the United Kingdom before 1989 unleaded petrol accounted for only 0.4% of sales.

At the start of 1989 the majority (four out of every five) of new cars coming onto the roads can run on lead–free petrol, and 25% of cars currently in use can run on the fuel, though engine adjustments are required in some cases to enable the change over to be made. Though leaded petrol will still be available for sale for some time, reductions in lead content are already reflected in lower lead pollution levels. Lead in London's air fell by 50% in 1986, though the effects of reductions in the lead content of petrol appear to take a long time to work themselves through into measurements. Lead contamination of plants and dust alongside busy motorways, such as the M25, continues to give cause for concern. Lead levels in petrol have gone down, but there has been a substantial increase in traffic levels and this has cancelled out gains otherwise made [32]. Indeed increased numbers of vehicles generally may make it difficult to achieve the EEC's objective that, by the 1990s, total vehicle emissions of CO and hydrocarbons will be no more than 1965's levels, and that NO_x emissions should be down to 60% of the 1965 level. Furthermore there was in 1988 and 1989, despite the price advantage, a degree of sales resistance to unleaded petrol, and a feeling that if a major vehicle user such as the Crown were to announce a change over to unleaded petrol for its vehicles that might give a lead to companies and private motorists. The Queen's personal fleet of cars was to be converted to unleaded petrol it was announced on 14th February 1989 and the Post Office Fleet on 9th March 1989, though 28,000 of its 35,000 vehicles already ran on lead free diesel fuel. That tax incentives can induce a change in the fuel buying practices of motorists in supported by reports on March

16th (see "The Times") that major fleet users of cars are moving towards the use of lead free petrol. Thus British Airways announced on March 15th that it would convert 4,500 vehicles at Heathrow to unleaded petrol, thus saving £23,000 on its fuel bill. GUS was similarly to convert 3,500 company cars. Further publicity for the increased availability of the product may, however, be required – only 15% of garages stocked unleaded petrol by the end of 1988, i.e. 3,000+ outlets, rising to 4,300 by the end of January 1989; though 60 outlets a week were taking on board the sale of unleaded petrol, and Texaco expects unleaded petrol at all its 1,400 stations by the end of 1989. There are, however, 21,000 filling stations in the country. (See Parliamentary written answers, 23rd January 1989, reported in "The Times" of that date. See also page 494 for recent developments).

Some fiscal laws act counter to pollution control measures. In the United Kingdom it is thought that, normally in each year, some 58% of new cars are company owned (the 1987 figure being 50% of new vehicle registrations). Overall some 3.4 million cars are so owned, with eight out of ten motorists who enter central London receiving some sort of company car subsidy. 42% of all cars commuting into London are company owned, while some 81% are in some way subsidized by firms and companies. The average benefit of a company car per driver per annum (1987–88) was thought to be £2,450 (the benefit of a 1.3 litre car being worth £2,700 p.a. to an employee), yet the tax on this benefit was only between £114 and £200 per annum. The pre 1988 effective tax loss to the Treasury was some £2.35bn, £500 m. more than public transport subsidies. Company cars tend to be 20% larger than average privately purchased family cars, and it has been argued they have raised petrol consumption by 20% (the equivalent of 907m gallons of petrol in 1988), and, in 1980, accounted for 20% of vehicle emissions. They certainly contribute to pollution problems in urban areas, London especially, where increased subsidies to public transport might actually reduce company car use. However, it is government policy to eliminate London Regional Transport's subsidy within six years. The effective subsidisation, and hence promotion, of company car use undermines enviromental and resource conservation measures. Taxation of company cars was, however, substantially increased by the 1988 Budget, not because of environmental pressures, but, it would seem, because the discrepancy between the value of a company car as a 'tax break' and the amount of tax paid on it was too great to be ignored. Tax scales on such cars were doubled for 1988–89. Even so on a 1.3 litre car worth £2,700 to an employee tax is still only paid as if it were worth £1,050, even though to buy such a car an employee would have to earn a further £3,250 gross, at 25% tax. The tax savings are even higher for larger cars and those with higher salaries: a Ford Sierra 2.0 litre car was worth £3,900 p.a. to an employee and a BMW 735 was worth £13,700, equivalent to a salary rise of £19,800. Even after the 1988

Budget changes the cost of tax foregone on company cars was some £1.75bn, for a far higher proportion of cars in Britain are company owned than on the main–land of Europe. Tax concessions were still currently twice British Rail's entire subsidy and greater than the total subsidy to all public transport in the United Kingdom, while the cost of congestion in London to industry is also some £1.5bn p.a. Indeed 8% of the working time of commercial vehicles nationally is lost through traffic congestion, and the removal of congestion could lead to the removal of up to 8,000 lorries from the nation's roads [33].

The 1989 budget again increased tax levels on company cars, however, once more not because of environmental arguments but because of a feeling that the 'perk' was undertaxed. The Chancellor of the Exchequer increased scales of taxation on company cars by a third for the year 1989–90, though he did not increase taxation on fuel benefits given to employees. The result is that a car of up to 1400 c.c. capacity taxed in 1987–1988 at £525 p.a. will in 1989–90 be taxed at £1,400, and the tax increases will be commensurately higher for more expensive and larger cars driven by high paid employees. There is, of course, a delicate balancing exercise behind the increases in taxation : too great a 'hike' in tax rates could lead to employees demanding pay increases simply to meet the increase in tax.

The benefit of free car parking provided for employees is not taxed however. Some 50% of those who commute by driving into London each day park in a firm's reserved space, yet the annual commercial fee for such a space in central London is some £8,500 – £15,000 p.a. Tax paid on such a benefit could amount to £262m p.a. It can be argued that the existence of guaranteed parking places is a major incentive encouraging car borne commuting, though there are some who argue that such is the convenience to individuals of car travel that any restrictions placed on company provided cars and parking would only lead to a greater number of private vehicles on the roads, with two thirds of drivers prepared to make such a switch. Certainly few cars entering London are used for business in the city during the day (only some 10–13% of entrant vehicles) thus indicating that most cars are simply used for the convenience of their drivers in getting to work. Overall it seems only 20% of company cars travel more than 15,000 miles p.a. on business.

3. LEGISLATION RELATING TO NOISE AND VIBRATION CONTROL

Traffic noise has been a subject for attempted control in the United Kingdom throughout the century [34] and Chapter 9. The problem now disturbs more people than all other noise nuisances [35]. Controls over traffic noise, as with emissions, take a variety of forms, eradicatory, preventive, regulatory (or mitigatory) and compensatory.

3.1 Control under Planning, Highway Construction and Traffic Regulation Legislation

Noise, and the associated problem of vibration, from vehicles can be mitigated by planning powers, existing in England and Wales under the Town and Country Planning Act 1990. Department of the Environment Circular 10/73 [36] gives guidance on using these powers, counselling against permitting the building of dwellings in areas where particular road traffic noise levels are encountered [37]. Indeed as noise control legislation in the United Kingdom does not specifically deal with traffic vibration which can be a cause of damage to buildings, control under planning, highway or traffic regulation legislation is the legal means of countering the phenomenon. Local Planning Authorities may incorporate guidelines to assist in achieving the policy suggested in Circular 10/73 when drawing up strategic and tactical development plans for areas, the Structure and Local Plans. These will then assist in deciding applications for permission to develop land [38]. Planning control does not, of course, eradicate traffic noise, it is merely an attempt to keep other developments away from noise.

It is also possible to regulate noise levels under the Highways Act 1980 by building roads of particular types, either to re–route traffic away from other structures, or to control the speed of traffic flow, or the gradients that it faces, both of which contribute to noise levels. The design, surfacing and layout of roads built using Highways Act powers can help reduce the impact of traffic noise [39].

It is possible to mitigate the effect of traffic noise by sound insulating buildings. Section 20 of the Land Compensation Act 1973 empowers the Secretary of State to make regulations in respect of giving traffic noise insulation grants. Under the Noise Insulation Regulations, S.I. 1975 No. 1763, there is a duty imposed on the appropriate highway authority to carry out noise insulation works, or make a grant in respect of such works in or to an eligible building, where the use of a new highway, or additional carriageway, opened to the public after 16th October 1972 causes noise in excess of certain levels. These levels are as follows. The specified level is a noise level of L_{10} (18 hour) of 68 dB(A) where L_{10} is the sound level in dB(A) which is exceeded for one tenth of a period of an hour, and L_{10} (18 hour) is the arithmetic mean of all levels of L_{10} during a normal working day measured from 06.00 to 24.00 hours. The 'relevant noise level' of traffic is measured 1 metre in front of the most exposed part of a building's facade and, first, must be greater by at least 1 dB(A) than the noise level before road construction, and, secondly, must not be less than 68 dB(A) i.e. the specified level, and thirdly, the traffic noise must make an effective contribution to the relevant noise level of at least 1 dB(A). Most residential buildings within 300 metres of relevant roads will qualify for grant aid.

Under Sections 1 and 2 of the Land Compensation Act 1973 compensation is payable to freeholders, or tenants with at least three years of their leases to run, of dwellings occupied as residences by such persons, and to such owner occupiers of other land which is an agricultural holding or another unit of land whose annual value does not exceed an amount prescribed under Section 149(3)(a) of the Town and Country Planning Act 1990. Compensation is payable in respect of depreciation of the value of land in consequence of noise, fumes, etc., caused by the use (as opposed to the construction) of a new highway opened after 17th October 1969. The claimant must have acquired his land before the new highway came into use, and must, under Section 3(2) of the Act, generally make his claim within the period from one to three years of the first use of the highway. Compensation is payable by the appropriate highway authority, and is computed under Section 4 of the 1973 Act.

No compensation is payable under these provisions for depreciation caused by intensified use of existing highways, or following the making of traffic management schemes or the specification of lorry routes, for which see below.

Traffic flow may be regulated outside London by traffic regulation orders made under Section 1 of the Road Traffic Regulation Act 1984, where expedient, inter alia, to prevent danger to persons or other traffic on roads, to prevent damage to the road or any building on or near it, to prevent the use of a road by unsuitable traffic, or for preserving or improving the amenities of an area through which the road in question runs. Such orders are made by the Secretary of State for Transport in respect of trunk roads (i.e. certain statutorily specified through–traffic routes), and otherwise in England and Wales by the relevant county council. The prescribed procedures for making orders are contained in Sched. 9, Part III of the 1984 Act and S.I. 1986 No. 180 and S.I. 1989 No. 1120. An order may, under Section 2 of the 1984 Act, prohibit, restrict or regulate the use of a road by vehicular traffic, generally or with specified exceptions, and either at all or specified times. Through traffic may be prohibited. Heavy commercial vehicles, i.e. those defined under Section 138 of the Act as having operating weights exceeding 7.5 tonnes, may be required to use specified routes, or their use may be prohibited or restricted within specified zones if this is expedient for preserving or improving the amenities of the area or some parts of the area of a county authority. The authority may consider the interest of the whole of their area, not merely that part containing the regulated road, see Freight– Transport Association Ltd v Royal—Berkshire County Council (1981) R.T.R. 95. Similar powers to regulate traffic in London are conferred on The Secretary of State and the London Borough Councils by Section 6 of the 1984 Act.

In certain areas in England and Wales of high landscape or other natural value, such as National Parks, areas of outstanding natural beauty and nature reserves, a traffic regulation order may be made, under Section 22 of the 1984 Act, to conserve or enhance natural beauty, or to afford better public opportunities for the enjoyment of the amenities of the area, or for recreation or the study of nature. The Secretary of State has an extended jurisdiction in this connection over non–trunk roads alongside the powers of the county authorities, and the Countryside Commission may make submissions to the Secretary of State as to the desirability of his making an order [40].

Prevention of noise and vibration pollution by the use of the 1984 Act powers is not easily achieved, however. Section 122 of that Act makes it clear that the prime purpose of the powers of local authorities is to secure the expeditious, safe and convenient movement of traffic, though the effect of traffic on the amenities of an area must be had in regard when deciding whether and how to exercise the powers. It may not be possible to use road traffic regulation powers where there is no supply of adequate, alternative, easily accessible routes for heavy vehicles to use. Restrictions on road use may in any case be ignored and are difficult to enforce. Furthermore it can be argued that neither planning, nor transport, nor highway or road traffic regulation powers with regard to traffic can work effectively unless they are possessed by strategic authorities that can integrate them into a properly co–ordinated transport and traffic scheme. This is true of all large cities, and especially of London where, it was reported in "The Times" for 16th January 1989, traffic congestion costs the nation £1.5bn p.a. The demise of the former Greater London Council under the Local Government Act 1985 has robbed the capital of such a strategic authority, so that responsibility for traffic and transport are effectively fragmented between thirty two London Boroughs, The City of London Corporation, five central Departments of State, several 'quangos' (special statutory agencies) together with a network of committees and advisory bodies (see below, page 494).

Fiscal means of control can, however, be used in addition to, and in conjunction with, other traffic powers, as 'The Times" reported on 7th and 25th January 1989. 'Road Pricing' can be introduced as a more efficient way of rationing road space than the congestion which does the same job in a chaotic fashion at the moment. A 20% reduction in the number of private cars entering London each day would improve traffic flow considerably, and could reduce the need for new roads, cut accidents, many of which stem from a condition identified as 'congestion frustration', and diminish noise and atmospheric pollution. Road pricing would add to the time and petrol costs of motorists an element in respect of wear on roads and a 'congestion element': the fractional cost increase in the petrol and time costs of others. Payment is related directly to use and so the old adage 'is your journey really necessary?' could begin to have currency, and wasteful car use would be discouraged. Such schemes, however, probably need enhanced public transport services to cater for the needs of those who are discouraged from using their own cars. The technical means for introducing road pricing exist, for example electronic number plates which 'trigger' a recording device in the road surface so a bill can be sent for a journey. What is lacking in Britain is the political will to introduce road pricing, and it may be that congestion may have to increase even more significantly before there is sufficient public pressure to force action on this issue.

The problem is, of course, not just confined to London. Other cities have adopted differing policies : Paris relies on massive investment in public transport, particularly rail and underground routes, while Lagos has a system whereby odd numbered vehicles may enter the central area one day in every two, alternating with even numbered vehicles. The result here, however, has been for the wealthy to purchase two cars, one with an odd and one with an even number. Singapore, however, has introduced supplementary licence requirements so that those entering the central area between 7.30 a.m. and 10.15 a.m. must pay for the privilege, with company cars being licensed at a double rate. Cars carrying four or more passengers are exempted from the fee. Traffic levels have now been reduced by 76%, following a 44% reduction after 1974 when the scheme was first introduced, and much more pooling of car use is common. The Netherlands is considering raising the price of private motoring in an effort to reduce the congestion which cost the Dutch economy some £216m in 1988. Road tax for private cars will increase, and road pricing, using electronic tagging devices, on some routes is proposed to apply from 1996. Sweden is considering the introduction of road pricing in Stockholm with a view to reducing the flow of private car trips by 35% and also to reduce air pollution. As from August 1988 Milan barred its city centre to cars during working hours; the ban extended to all locally registered cars, though some were given permits to enter the centre, and further daily permits were made available from September 1988 to allow drivers to purchase a limited amount of time to enter the centre. Financial reasons seem to have inspired the Milanese decision, but similar bans in Rome and Florence have been motivated by fears over vehicle created pollutants attacking historic buildings. Athens has restrictions on private vehicles which ban them from the inner part of the city between 7.00 a.m. and 8.00 p.m. on alternate days. This alone has not prevented Athens from suffering smog. In August 1988 NO_x levels exceeded the warning level of 500 mg p.c.m. A particular problem for Athens is the fact that one fifth of its car population, about 200,000 vehicles, is some fifteen or more years old, and emissions increase as vehicles age.

3.2 Restrictions on vehicle weight

Though not exactly 'highway pollution' there is a disbenefit to society in the form of excessive wear on highways, bridges, etc., from heavy road vehicles. Though the numbers of lorries in Great Britain fell from 600,000 in 1967 to 435,000 in 1986, modern lorries are heavier than their predecessors, and larger numbers of heavier lorries are expected in the future. 75% of total road freight is carried by the heaviest lorries (those over 25 tonnes): 89% of the nation's freight is carried by road, the third highest percentage in the world. Road wear costs £500m p.a., and impact damage to surfaces, bridges, footpaths, etc., a further £110m p.a. There are also 'hidden' costs in the form of damage to sub–surface installations and vibration damage to buildings [41]. Under the Road Vehicles (Construction and Use) Regulations, S.I. 1986 No. 1078 Part IV the maximum permitted lorry vehicle weight is 38 tonnes, and by 1986 there were 34,000 lorries weighing over 33 tonnes. The weight problem is, however, complex: some vehicles are 'heavy' because of their construction; other vehicles cause problems primarily because of the way in which their weight is distributed and the number of their axles – a six axle 38 tonne lorry generally has less "damaging power" than a five axle lorry – some lorries cause damage because they are overloaded.

Maximum weight limits are specified by the regulations for individual axles of lorries, but there are no requirements as to the design of suspension systems with a view to reducing road wear. Some lorry types, as has been said, cause more wear than others, e.g. the two axle tractor type, which comprises 70% of all 38 tonne lorries. Attempts to wean operators away from such vehicles by incentives under the Vehicle Excise Duty Scheme do not seem successful. Though powers exist under the Road Traffic Act 1988 to prevent overloading of heavy vehicles and to enforce weight limits, enforcement of the law is currently piecemeal and uncoordinated, though the Department of Transport was seeking to introduce a national enforcement plan in 1987–88, and a power for local authority inspectors to stop and check vehicles (currently the exclusive preserve of the police) is under consideration. Better use could be made of modern technology, e.g. sensors in roads, to facilitate law enforcement. Though it is government policy that lorries should pay, according to class of weight and size, their 'share' of road track costs in the form of vehicle taxation, when such costs are being calculated by the Department of Transport 'costs' of issues such as accidents, damage to underground services, vibration damage to buildings, traffic delays, and social and environmental nuisances, are not taken into account. If the disbenefits of heavy lorries are a form of pollution it would seem in the United Kingdom that 'the polluter pays' principle is not being fully applied in relation to this issue. Lighter vehicles certainly seem to bear a disproportionate burden of track costs, though it is difficult to arrive at an objective method of allocating costs to vehicles.

The United Kingdom has been under some pressure from its European partners to raise the upper limit on lorry weights to 40 tonnes (allowed under EEC directives) but has refrained from allowing these limits in this country before roads and bridges are strengthened to withstand loads. However, it appears the 1986 regulations may one day be amended to allow an increase in the permitted weight of articulated lorries. On 9th March 1989 EEC proposals for yet heavier lorries were rejected by the House of Commons without a vote, and Her Majesty's Government undertook to oppose the proposals within the EEC. The official position is that there are 1,200 bridges on trunk roads, and 11,250 on local authority maintained roads, too weak to support heavier lorries, and strengthening them will not be completed before the end of the century and would cost £700m. There is, however, disagreement between the British government and the Commission of the EEC on the point, with the latter maintaining there are only 230 trunk roads, and 4000 local authority road, bridges unable to carry the EEC permitted 40 tonne vehicles. There is a programme to upgrade trunk road bridges but this will take 15 years to implement, and in the meantime the United Kingdom has used its powers under the treaties setting up the EEC to obtain "derogations" (i.e. permissions to depart from the objective of directives) in respect of weight limits for five and six axle lorries (from 1984) and also in respect of two, three and four axle lorries. The current EEC proposal would raise weight limits for these latter types from 32.5 tonnes to 38 tonnes, and there is a further proposal to raise weight limits for five and six axle lorries to 44 tonnes by 1996. The United Kingdom may attempt to use powers under the EEC treaties to veto such proposals. Officials of the Commission of the EEC, however, believe the United Kingdom will have to accept 40 tonne lorries by 1996, on the basis that the weight limit is part of the harmonisation programme for the internal market and is thus subject to majority voting under the terms of the Single European Act. Currently, as stated above, the United Kingdom enjoys a derogation from EEC requirements enabling it to put a ceiling on lorry weights overall of 38 tonnes, with a 10.5 tonne limit on drive axles. It is this that the Commissions wishes to phase out by the end of 1996. (See also Section 5 of this Chapter).

Public concern continues over the lack of enforcement of the current law with regard to overloaded lorries, and the complexity of our enforcement procedures which do not contrast well with those in mainland Europe where drivers of over– weight vehicles can be fined on the spot and vehicles detained until fines are paid [42]. In Britain fines for overloading offences average only £161, yet 22%–33% of lorries may be overloaded. and 25% of lorries coming from the continent in 1987 were overloaded [43]. Not only does current United Kingdom law need to be enforced more rigorously, and have more effective surveillance systems provided, but thought needs to be given to future lorry design standards, to fiscal measures appropriate to encourage use of less harmful designs, and to find methods of reflecting in vehicle tax rates at least the higher capital element in allocated road track costs consequent on use of heavier lorries, even if it is much harder to include elements in respect of social and environmental costs [44].

3.3 Noise control by vehicle construction standards

Noise standards for motor cars, lorries, buses and motorcycles are to be found in the Construction and Use regulations for vehicles; type approval schemes also exist for cars and lorries. United Kingdom regulations reflect EEC requirements which, inter alia, have sought to reduce vehicle noise, though member states may not restrict sale or use of vehicles conforming to EEC standards on grounds that they have more stringent national requirements. Basic EEC requirements are laid down in 70/157/EEC, and have been progressively modified by Directives down to 84/424/EEC. As from 1st October 1988 it will be permissible for member states of the EEC to refuse national type approval where vehicles do not conform to Directive requirements, and, from 1st October 1989, to refuse to allow such non–conforming vehicles to enter service [45]. The most recent EEC limits for vehicle noise in dB(A) are: for cars 77; for buses over 3.5 tonnes but with less than 200 HP 80; for similar vehicles with more than 200 HP, 83; for buses and goods vehicles between 2 and 3.5 tonnes 79; for similar vehicles less than 2 tonnes 78; for goods vehicles over 3.5 tonnes with less than 100 HP, 81; for similar vehicles between 100 and 200 HP, 83, and for similar vehicles over 200 HP, 84. These figures relate to petrol engined vehicles, for diesel engined vehicles of less than 3.5 tonnes the limits are increased by 1 dB(A).

British law is to be found in the Road Vehicles (Construction and Use) Regulations S.I. 1986 No. 1078, Parts K and E. Under Regulation 54 every vehicle propelled by an internal combustion engine must be fitted with an exhaust system including a silencer, and exhaust gases may not escape without passing through the silencer, which must be maintained in good order, and not modified so as to increase exhaust gas noise. However, alternatively a vehicle may comply with the requirements of EEC Directives 77/212, 81/334, 84/372 or 84/242, or 78/1015 in the case of motorcycles. Further under Regulation 55 the general rule is that wheeled motor vehicles first used on or after 1st October 1983 must be so constructed as to comply with specified sound limit requirements, measured in specified tests. Currently the British legal limits are based on the requirements of Directive 77/212/EEC which sets decibel limits for cars of 80 dB(A), rising to 88 dB(A) for goods vehicles over 3.5 tonnes and having over 200 HP. As from 1990/91 further reduction in noise limits for vehicles first used after 1st April 1990 (or 1st April 1991 with regard to certain buses and lorries) will apply. These will reflect the requirements of Directive

84/424/EEC. Thereafter a further revision of EEC noise limits is expected. Noise limits for type approved cars are also to be found in regulations made under the Road Traffic Acts, see, for example, S.I. 1982 No. 1271, S.I. 1984 No. 981 and S.I. 1987 No. 1509 which reflect requirements of EEC Directives down to 1984. Limits for type approved lorries are to be found, see, for example S.I. 1987 No. 1508.

So far as motorcycles are concerned, current EEC requirements are set out in Directive 78/1015/EEC which sets dB(A) limits according to engine cubic capacity, rising from 78 dB(A) where capacity is less than 80 cc to 86 d B(A) where it is over 500 cc. Under the Construction and Use regulations (Regulation 57) every moped or two wheeled motorcycle, whether or not equipped with a sidecar, first used on or after 1st April 1983 must be so constructed that its sound does not exceed specified limits. For mopeds the limit is 73 dB(A) as they are not subject to EEC requirements. Mopeds are defined by para. 5 of Schedule 9 to the 1986 Regulations, and have engine cylinder capacities of not more than 50 cc, and maximum speeds of not more than 30 m.p.h. For other motorcycles the limits are those required by the EEC Directive. For the future Directive 87/56 EEC will further reduce motorcycle noise limits in two stages from 1988 and 1991. Limits will range from 77 dB(A) to 82 dB(A) at first, reducing to 75 dB(A) to 80 dB(A). Under Regulation 57(3) of The Construction and Use Regulations the silencer which forms part of the exhaust system of a motorcycle first used after 1st January 1985 must either be the original fitted on manufacture, or clearly marked that it either accords with British Standards, or complies with the cycle manufacturer's specifications. (For certification of motorcycle sound levels for signifying that a type of motorcycle complies with the requirements of Directive 78/1015 EEC see S.I. 1980/765 and S.I. 1988 No. 1640.)

Under Section 1(1) of the Motor Cycle Noise Act 1987 it is an offence for any person in the course of carrying on a business to supply or offer, or to agree to supply or offer, or to possess for the purposes of supply, a motor cycle exhaust system, or a silencer or other component for such a system, unless the item complies with regulations made by the Secretary of State. The power to make regulations under Section 1(2) of the Act extends to preventing the supply of exhaust systems, silencers and other components likely to result in excessive motor cycle noise. Regulations may prescribe requirements by reference to British Standard Specifications (made under Section 41 of the Road Traffic Act 1988) or to EEC instruments, or other international requirements. Regulations are to be made after consultation with such representative bodies as the Secretary of State thinks fit. The Act applies to mopeds as well as other motorcycles, and in the context of the Act "supply" includes gifts.

The 1986 Regulations Part E further provide that no motor vehicle shall be used on a road in such manner as to cause excessive noise which the driver could have avoided by reasonable care, see Regulation 97. Regulation 98 generally requires vehicle engines to be stopped when stationary to prevent noise, and Regulation 99 generally restricts use of audible warning instruments by any person on vehicle. There are exceptions in respect of emergencies and other urgent uses, reversing alarms fitted to heavy goods vehicles, crime prevention, and certain sounds made to indicate that the vehicle is conveying goods for sale as allowed under Section 62 of the Control of Pollution Act 1974. (In this connection see also Section 71 of the Control of Pollution Act 1974 authorising the Secretary of State to prepare and approve codes of practice on minimising noise. A Code of Practice has been issued with respect to Ice Cream Van Chimes, see S.I. 1981 No. 1828). Section 62 of the 1974 Act also generally prohibits the use of loudspeakers in streets, but under Section 62(2)(d) this prohibition does not apply where the loudspeaker is fixed to a vehicle and is operated solely to entertain, or to communicate with, drivers or passengers of the vehicle, or where it forms part of the vehicle's warning system and, in any case, is so operated as not to give reasonable cause for annoyance to persons in the vicinity.

Under Section 7 of the Road Traffic (Consequential Provisions) Act 1988 nothing in the Act authorises a person to use on a road a vehicle so constructed as to cause a public or private nuisance, and so civil liability for nuisance caused by vehicles is preserved.

4. THE EUROPEAN DIMENSION OF LEGISLATIVE CONTROL : THE FUTURE

British environmental law, policy and practice in relation to highway pollution is increasingly guided by obligations imposed under European Community Law. Though the legal basis of community action in respect of environmental issues was somewhat suspect, [46] the Treaty of Rome was amended in July 1987 by The Single European Act and a firm legal basis for Community action on the environment provided [47]. However, the European Community is still a long way from being a nation state in its own right; the member nations are still independent nation states [48] and the approaches of the member states to the development and implementation of pollution controls remain disparate [49]. With regard to vehicle emission controls the initial object of the EEC Directives was, first, to promote freedom of trade by introducing standards compliance with which ensured a vehicle could not be refused type approval by member states, and, secondly, to ensure the progressive harmonisation of national laws of member states on an optional basis.

However, with the approval by national ministers of the EEC's fourth Environment Action Programme in December 1987 a review of the implementation of EEC legislation may be expected [50].

The EEC has, as stated, a programme, under Articles 100–102 of the EEC Treaty for the harmonisation of the laws of member states, and the basis for national action is Directives issued under Article 189 of the Treaty, with enforcement proceedings being available against those states who fail to translate Directives into their own law, though the legal process involved is cumbersome and time consuming. Some further freedom is allowed to member states, as stated earlier, under Art. 189 because, though 'binding as to the result to be achieved', choice of method of implementation remains with member states. This freedom is limited, much depending on the degree of detail in Directives. Certainly choice of method does not allow non–implementation of a directive as fully as possible. The vehicle emission Directives are extremely detailed, and, as has been seen, have been generally incorporated into United Kingdom law. However, it is possible under Article 36 of the EEC Treaty to derogate from community obligations on a number of stated grounds, including the protection of health, though such a derogation may not act as a disguised restriction on trade. How far the power under Art. 36 extends in relation to Directives on vehicle emissions, etc., is uncertain. However, in Commission of the European Communities v U.K. [1988] "The Times" 6th September the Court of Justice of the European Community had to consider whether a British Statutory Instrument on vehicle lighting (S.I. 1984/821) complied with European requirements under Directive 76/756 dealing with the harmonisation and approximation of the laws of member states concerning type approval of vehicles, in this case particularly with regard to the lighting and light signalling of vehicles. It was pointed out by the Court that the Directive had the purpose of reducing, and indeed eliminating, hindrances to internal community trade resulting from different mandatory technical lighting requirements in Member States. The Directive contained an exhaustive list of lighting and light signalling devices, and there was an explicit obligation on member states to adopt these "either in addition to or in place of ... existing rules". It was held, therefore, that a member state could not unilaterally require manufacturers of vehicles which complied with the Directive's requirements to comply with other requirements not imposed by the Directive. The UK was accordingly in breach of its community obligations (see below, page 494).

The EEC does not, however, constitute the only European body whose deliberations have relevance for future highway pollution controls. The UN Economic Commission for Europe (ECE) is active in setting limits for the emission of NO_x, though there is international disagreement as to the date by which such emissions should be stabilized [51]. The World

Health Organisation (WHO) has also issued air quality guidelines in relation to many toxic substances discharged into air, and it appears that in Britain these limits are transgressed on busy roads in respect of nitrogen dioxide [52]. Nevertheless vehicle emission controls are still largely a matter for each nation to develop and implement, even though each may be able to utilise considerable amounts of material internationally agreed and adopted by other states. This is particularly well illustrated by the problem of the future direction of vehicle emission control.

Technologies here conflict with no consensus between North America, Japan and the EEC (nor internally in the latter case) as to the best way to proceed. Japanese manufacturers lead the world in advanced engine technology, particularly in relation to 'multi–valve' cylinder heads generally considered necessary to meet future EEC emission standards for vehicles above 1.4 litres [53], see also Chapter 10. Laws for the protection of the Environment date from 1967 in Japan, and economic considerations have not been paramount over the demands of environmental protection. Stringent United States emission controls (currently dating from 1983) require the fitting of three–way catalysts, requiring lead–free petrol. European car exports to the USA have to be so equipped. There is evidence that without the fitting of three–way catalysts there will continue to be a high level of nitrogen pollutant fall–out from vehicle emissions [54]. The EEC, however, has a compromise policy for future exhaust standards.

The EEC's 1984 proposal [55] was for further reductions of vehicle emissions. This was not acceptable to all member states [56]. It took till December 1987 to find a compromise fomula, enabling member states to introduce new emission standards between 1988 and 1991. The compromise was voted in by a majority, under the provisions of the Single European Act, supplementing Article 100 of the EEC treaty, which allow decisions to be taken in this way, against the wishes of Denmark and Greece, the former intending under its Article 36 derogation rights to introduce stricter emission standards for small cars [57]. The compromise moved away from the notion of uniform emission requirements for all cars irrespective of engine size, and 'banded' cars into three groups, 2 litres and above, 1.4 to 2 litres and below 1.4 litres, with emission requirements becoming more lenient as engine capacity decreases. The compromise means that large cars (over 2 litres capacity) should be fitted with three way catalysts if they are to meet EEC requirements, the medium category will require 'lean burn' engines with oxidation catalysts only, while small cars will be fitted with only lean burn engines. However, part of the compromise was that emission standards for the smallest cars would be made more stringent in 1992/93 [58].

Under the proposal (now enshrined in Directive 88/76) for cars of 2 litres or more capacity, as from 1988/89 CO emissions in grams per test should be 25, hydrocarbons and NO_x should be 6.5 and NO_x should be 3.5. For medium sized cars the limits would be, as from 1991/93, 30 and 8.0 with no specific limit for NO_x separate from hydrocarbons. For small cars the interim limits from 1990/91 would be 45, 15 and 6, with these becoming the same as for medium cars as from 1992/93. A car with a lean burn engine only (i.e. one in which the proportion of air to petrol burnt within the combustion chamber is increased from 15.1 up to 22.1) can achieve the EEC limits as they currently stand, but not if they are further restricted to approach current USA standards. If the EEC were to apply the principle that vehicle emissions should be governed by the "as low as technically achievable" principle (ALATA) then three way catalytic converters would have to be introduced for all vehicles. The irony so far as the United Kingdom is concerned is that while the major British car manufacturers are opposed to fitting catalytic converters on grounds of cost, it is a British engineering company which is the world leader in their manufacture. (For currrent limits see below, page 482).

In favour of "lean burn" engines is the argument that they reduce fuel consumption *significantly, thus also reducing the amount of carbon dioxide emitted, and therefore* helping to reduce the global warming effect of this gas. However, in view of the increased numbers of vehicles likely to come onto the roads, it might well be that "lean burn" technology might only stabilise the level of CO_2 emissions.

Georgiades, Chiron and Joumard [5] have pointed out that the EEC approach to setting emission standards has relied on adapting the law to suit existing technology, and thus stricter emission limits have emerged generally only as technologies have improved, (as opposed to following the ALATA principle mentioned above.) Moves away from this stance have, however, come as a result of political pressure within the Community from the Germans who, since the early 1980s, have been increasingly concerned about the effects of acid deposition on their forests, and who have a major Green political party. France, however, found herself in a difficult position in relation to calls for reduced vehicle emissions. The French car industry and oil companies urged caution, and favoured limiting motorway speeds only, and France's two largest producers favoured a staged reduction approach, a little at a time over five year periods. The French ecological movement was largely preoccupied at this time with France's nuclear industry, while within the government itself there was disagreement between the Ministries of Industry and the Environment. In Germany, on the other hand, emission limits became a major public issue because of the activities of the various political parties, and ministers there perforce had to press for stricter EEC regulations. Georgiades, Chiron and Joumard argue that the progress made over EEC emission standards indicates the complex nature of the standard setting process, involving economic, industrial and political considerations in a process of debate and compromise. They go on to argue "In such a climate, scientifically–based decisions can be no more than a compromise and the political establishment of the standard no more than the expression and transformation of the different considerations involved. The theoretical limiting value is never discussed and is clearly not the subject of disagreement. Other considerations predominate and the respective weights of the different parties concerned have a lot of influence on the choices that are made."[59].

The object of the EEC proposal is, inter alia, to reduce NO_x emissions from cars in the EEC from 3.17m. tonnes p.a. to 1.67m. tonnes, with 1.33m. tonnes of this coming from petrol engined cars. Thus emission limits for European cars should be 'equivalent' to those required under 1983 U.S. standards. However, there is doubt as to which technology is the most appropriate means to use to achieve reductions. In particular many argue that NO_x emissions can only be substantially reduced by 2010 by three way catalysts fitted to all cars. These might involve substantial cost and fuel consumption penalties on vehicles. There is further doubt as to whether the EEC proposals are strict enough, bearing in mind likely growth in car ownership well into the next century, particularly in the small car range where most NO_x will continue to be emitted.

For the longer term future of exhaust standards in the EEC the prospect is one of uncertainty [60]. A review is scheduled for the early 1990s, but the nations of the community are divided, with the United Kingdom, Spain, France and Italy remaining opposed to very strict emission controls. Much of the United Kingdom's opposition stems from its attachment to "lean burn" technology as described above. However, West Germany and the Netherlands aim to reduce emissions, particularly of NO_x at a much faster rate, and other nations not in the EEC (Switzerland, Austria, Norway, Sweden) have emission limits equivalent to those in the USA. Those who argue for stricter limits point out that the defect in the present "emissions package" under EEC law is that it fails to take account of increases in the number of vehicles and of journeys made. The growth in the number of small cars is predicted to be greatest and their NO_x emissions, where only 'lean burn' technology is used, increase rapidly as speed increases; emissions of carcinogenic polyaromatic hydrocarbons can also not be curbed by 'lean burn' technology. On 12th April 1989 the European Parliament voted for much stricter emission limits for small cars (i.e. those under 1400 cc). The Commission of the EEC had proposed early in April 1989 emission limits to come into force in 1993 to further reduce CO emissions by 33% and those of NO_x by about 50%. The European Parliament, however, voted (309 to 5) for stricter standards parallelling those of the USA and Japan, which will involve the fitting of catalytic converters which are capable of reducing emissions by some 90% for hydrocarbons, 88% (CO) and 50% (NO_x). The British car industry claims such converters would add some £500 to the price of a car because though converters cost only, on average, some £50, there are extra costs involving in some re–engineering of vehicles. This claim is, however, disputed, and costs may only be some £100 – £300 per vehicle. In addition British car makers already fit catalysts to cars for sale in the USA and Japan which have strict emission limits, and some Japanese and German makers offer catalysts as standard on some models.

The European Parliament's vote forced a rethinking of policy by the Commission for, though the Parliament has no power under the Single European Act to act as a legislative body for the whole community, its consultative position enables it to oblige the Commission to re–submit its proposals for approval by the Council of Ministers. The Council of Ministers can, of course, reject such a re–submitted and amended proposal, but, effectively, only by a unanimous vote (which is unlikely to occur) or by certain states (for example, the United Kingdom, France and Italy) using their voting power under the Treaty of Rome to prevent the proposal being accepted by means of the "qualified majority" procedure which is applicable to the matter of emission standards. In the result Directive 89/458 was agreed, reducing small car emission limits at 19 (g/test) for carbon monoxide and 5 (g/test) for the combined mass of hydrocarbons and NO_x as from 1990.

The Dutch government, as part of a programme to reduce NO_x emissions from vehicles by 75% by the year 2010 has therefore introduced financial incentives to persuade purchasers of new cars to buy those cars which meet the emission standards of the USA. Such incentives may be contrary to EEC law, but the Commission of the EEC appears unwilling to pursue rapid action against the Netherlands, who would rely on the judgment in Commission of the European Communities v Kingdom of Denmark Case 302/86, Notice 88/C269 05 (a case concerning the legality of Danish legislation requiring that certain drinks should be in returnable containers) that there are occasions when protection of the environment takes precedence over eliminating trade barriers [61] (see below, page 494).

In December 1987 the EEC also agreed a further directive on gaseous emissions that is CO, NO_x and hydrocarbons from heavy vehicles [63]. The gaseous pollutant emission limits are fixed as from 1st July 1988 at 11.2 grams (CO), 2.4 grams (hydrocarbons) and 14.4 grams NO_x, per test. Engine component parts must be so designed, constructed and assembled as to enable the engine to comply with the limits despite vibration. Further controls over gaseous emissions and particulate emissions were mooted in 1987/88 [64]. In particular the House of Lords Select Committee on the European Communities has called for mass emission limits for particulates discharged by lorries and buses, improved diesel engine designs control particulate emissions, and improved diesel fuel quality [65]. Recent developments are reported in the final section of this chapter.

4.1 A Comparative view of other European Nations' Legislative Systems

The legislative patterns of highway pollution law in the other member states of the EEC are generally similar to that of the United Kingdom – a mixture of the preventive, the prohibitive and the mitigatory. Planning and land use controls, for example, can be used in Belgium under a Law of 28th July 1973 to protect health against, inter alia, mobile sources of noise, and to lay down noise requirements with regard to new road construction. Legislation in Denmark enables the issue of guidelines for protection of the environment, including road noise quality objectives, and noise is a relevant planning consideration in planning issues, with areas above specified dB(A) levels being considered unsuitable for residential development unless screening can reduce noise. Noise is also a relevant consideration in French planning law, and since 1983 there has been a programme to reduce road noise, both as regards new and existing roads, either by reducing noise at source, as for example by the erection of noise barriers, or by sound insulation of buildings. Mayors have road traffic regulation powers to restrict or prohibit vehicles use on some roads, and may fix routes for some vehicle types. The Federal Republic of Germany has also issued guidelines since 1983 for traffic noise control on federal roads, and these also influence provincial road noise control practices. Highway planning in Germany also takes account of noise emissions. Where unavoidable effects of traffic noise occur compensation schemes are available to enable landowners to instal sound proofing. Ireland utilises land use planning and development controls to site developments sensitive to, or creative of, noise in appropriate places. Noise is also a relevant factor in plan making in Italy. In the Netherlands planning law can be used to prevent noise nuisances from arising; ministerial circulars have also been issued giving guidance on reduction of traffic noise. In 1984 the Minister of Housing, Physical Planning and Environmental Management gave guidance under noise nuisance legislation on noise zoning along new roads.

With regard to the suppression of pollution by vehicle construction and fuel composition standards, the past decade has seen considerable increases in legislative activity, with member states of the EEC generally taking steps to harmonize their laws in line with community directives. Control of air pollution from vehicles in Belgium is a central government responsibility; type approval systems for vehicle construction and design follow EEC models. Noise levels for classes of vehicles are fixed by law, and there are controls over the import, distribution, sale or use of petrol with a lead content above specified levels. Denmark requires observance of vehicle design and construction standards, and since 1977 has had stricter standards over petrol and diesel engine exhaust emissions than previously. Vehicle engines must be designed so as not to produce unecessary smoke. Though Denmark operates a type approval system for vehicle inspection at first registration, there is a continuing obligation to comply with air pollution controls, and the police have power to inspect vehicles at any time. Denmark has also progressively reduced the maximum permitted level of lead in petrol. France has controls over both air and noise pollution from motor vehicles, though type approval systems exist for classes of vehicles. Emission of polluting fumes is controlled by design standards, as are noise levels, and vehicle engines must be maintained, fuelled and used by vehicle users so that harmful or annoying fumes are not emitted. The sulphur and lead content of fuels is also fixed by law.

In the Federal Republic of Germany use of vehicles is controlled by operating permit, and vehicles' emissions must not transgress limit thresholds. Regular examination of vehicles to ensure compliance with permits takes place, and vehicles may not be used so as to cause nuisances or environmental damage. There are also noise standards for vehicle construction, and a special regime for use of heavy vehicles during certain times of day, during holiday periods and on Sundays. Germany has introduced strict standards to reduce the lead content of petrol and the sulphur content of diesel fuel. Tax advantages are given in respect of sales of low–lead petrol. Greece has a type approval scheme for all vehicles used in that country, and generally also requires that vehicle construction methods should prevent, so far as possible, emissions of gases, smoke, fumes and noise, it being the duty of vehicle owners and drivers to so maintain and drive vehicles that emission of pollutants is avoided. Vehicles in Greece are subject to periodic testing. It is a further requirement that vehicles should be driven in such a way as to avoid disturbance, and using a vehicle to produce noise disturbance is prohibited. Greece has established noise emission standards, and local regulations may provide for restrictions on times and places of vehicle use. There are further regulations as to the CO content of vehicle exhaust fumes, the soot content of diesel exhausts, and the sulphur content of motor fuel. Ireland likewise operates vehicle type approval schemes which incorporate EEC requirements. Emission of smoke, visible vapour, noxious gases and offensive odour is not permitted under construction requirements. There is a further requirement that vehicle maintenance should prevent emissions avoidable by reasonable steps; this is enforced by criminal sanctions. The Irish type approval scheme extends to noise suppression requirements; Irish law requires the fitting of efficient silencers, and their subsequent efficient maintenance. At the same time modification of silencers so as to increase noise is prohibited, as is the fitting of vehicle warning systems that emit excessive noise. Audible warning devices are prohibited in certain places at particular times. There is a general requirement that vehicles should be so used in public that excessive noise avoidable by reasonable care is not generated. Irish law sets limits for the sulphur and lead content of petrol, and lead content has been progressively reduced in line with EEC standards.

Turning aside for the moment from the national standards to their implementation, it will be found that the practical results of the various controls differ widely from nation to nation. This is well illustrated by the figures for the sale of unleaded petrol as a proposition of total petrol sales. West Germany heads the league here with 41.8% of all petrol sold being unleaded. West Germany is followed by the Netherlands (36.3%), Denmark (32.5%), Luxenbourg (4%), Italy (1.3%), The United Kingdom (0.4%), Belgium (0.2%), and Spain (0.1%). In Greece hardly any lead free petrol is sold, though 100 outlets are equipped to sell it. (1989 figures – see above, page 466.)

Italy also operates vehicle type approval systems. Since 1966 there have been limits on polluting emissions, for example CO, and as to opacity of diesel exhausts. Regular testing of vehicles is required by Italian law, which also regulates use of fuels. Noise emission standards are fixed for classes of vehicles; it is a general requirement that vehicles must be equipped with efficient silencers maintained in good order. Use of audible vehicle warning devices is restricted generally, and particularly in built–up areas. Local authorities have traffic regulation powers to forbid vehicular access to particular localities. Use of electrically powered vehicles by municipalities is encouraged by grant aid. Luxembourg's system of type approval is based on EEC models, and the law also generally requires that vehicles should not emit fumes causing inconvenience to others. The type approval scheme extends to noise emissions. There is a general requirement for the fitting of efficient silencers, with prohibitions on the use of excessively noisy vehicles. Use of audible vehicle warning devices is restricted generally, and particularly in built–up areas. Luxembourg has requirements restricting lead and sulphur content of fuels, with lead content being reduced progressively over a period of time. Environmental protection is a specific constitutional responsibility in Dutch law. There are general vehicle construction requirements that nuisance arising from the emission of smoke, vapour or fumes should be avoided, and as to fitting of efficient silencer systems. The Netherlands operates a type approval scheme based on EEC models. There are specific limits on vehicle exhaust emissions for both petrol and diesel engined vehicles, and there has also been progressive reduction in the lead content of petrol, benzene content of which has also been reduced [66]. (Switzerland, which is not a member of the EEC, requires all new cars to be fitted with three way catalytic converters to filter out the majority of CO and NO_x emissions. These cars can only run on lead free petrol which now accounts for 34.5% petrol sales.)

5. CONTROL UNDER GENERAL AIR QUALITY STANDARDS

"Air quality standards ... are legal upper limits imposed on levels of pollutants in ambient air during a given period of time." [67]. Georgiades, Chiron and Joumard [5] amplify this by stating: "Environmental standards apply to public places and are aimed at safeguarding public health and protecting the material, biological and plant life environment. A distinction needs to be made between values having the force of law, where any infringement leads as a matter of course to the application of measures to ensure limits are respected, and those serving for guidance or that are regarded as no more than desirable or recommended limits" [68]. They go on to point out that such standards may be based on absolute concentration limits integrated over a chosen period of time, or on average values or percentiles. In setting a standard the object is to fix an acceptable level of a pollutant in the ecosystem consistent with the "good health" (itself a notoriously indeterminate concept) of those exposed to it. This is achieved by a mixed scientific and nonscientific process. The physical and chemical properties of the pollutant can be determined, and its toxicity, likely ability to be absorbed or accumulated, etc. This will enable an estimate to be made of the amount of the pollutant in question likely to be, for example, lethal. Studies of the toxicity of the substance can be carried out on cell cultures and animals, and experimental models evolved to predict a substance's effects. Experimentation enables the scientist to evaluate toxic effects, and to predict any serious effects, such as the incidence of cancer likely to result from exposure to the substance. Epidemiological studies can, in addition, evaluate the effect of exposure on the human population, and these will consider the varieties of environmental conditions experienced by the exposed population and their varying susceptibilities to affectation. These approaches were reviewed in Chapter 7. However, the information thus derived will not be utterly objective and conclusive since there will have been extrapolations from data and a number of observed uncertainties. It is thus necessary to modify the experimental figures by dividing them by a safety factor which brings the experimental level at which no adverse effects are observed to a lower level which then becomes the limit value for exposure. This safety factor is determined evaluatively, not on a scientific basis, by considering factors such as the varieties of sensitivity to exposure, the range of pathological conditions found in humans, the possible consequences of combinations occurring between pollutants, the quality of the available data on the effects of exposure. The result is that limit values are empirically rather than totally scientifically determined: mathematical and biological considerations are balanced by ethical and philosophical ones, and technical, economic and political factors also play a major part in the limit value setting process.

Within the European Community Directives have been made with regard to: smoke (sometimes referred to as TSP or 'total suspended particulate matter') and sulphur dioxide, giving guide values for limiting the amounts of SO_2 and particulate matter (see 80/779/EC, originally proposed in 1976); [69]. Nitrogen Dioxide (see 85/203/EEC proposed in 1983) [70] and Lead (see 82/884/EEC, proposed in 1975) [71]. These are, of course, 'across the board' controls applying to levels of pollutants wheresoever derived, being general provisions to protect human and environmental health and are additional to and separate from, the emission controls for vehicles considered earlier. With regard to sulphur dioxide and airborne particulates, uniform emission limits for their presence at ground level were set by Directive 80/779, with limits being met, where possible, by 1st April 1983, and a mandatory date for compliance of 1st April 1993, together with a duty generally to implement more stringent limits contained in Annex II of Directive 80/779. Furthermore there is a requirement that air quality is not to be allowed to deteriorate to a significant degree, even where it is currently better than the standard envisaged by the Directive. Subject to mandatory compliance by 1993 member states may seek temporary exemption for clearly defined zones provided they can show they are taking genuine steps to improve the quality of air in these areas. A complex system of values, which is open to quite serious scientific criticism, [72] sets limits of 80 $\mu g\ m^{-3}$ for smoke, over a year (taken from the median of daily values) with a winter limit (i.e. between 1st October and 31st March) of 130, and a year (peak) limit of 250. More controversially SO_2 limits are allowed to vary according to the measure of smoke, so that where smoke levels are low more SO_2 is allowed to be present, i.e. there is a 'trade off' principle. For the year as a whole where there are less than 40 $\mu g\ m^{-3}$ of smoke the SO_2 limit is 120 $\mu g\ m^{-3}$; where smoke is present in quantities of more than 40 $\mu g\ m^{-3}$ the SO_2 limit is 80. The relevant proportional limits for the winter period are: smoke less than 60, SO_2 up to 180, smoke more than 60, SO_2 up to 130. The year (peak) limits are: smoke less than 150, SO_2 up to 350, smoke more than 50, SO_2 up to 250.

So far as the United Kingdom is concerned it is the official view that compliance with the Directive can be achieved under legislation which is concerned with stationary sources of pollution, for example the Clean Air Acts 1956 and 1968, The Alkali etc. Works Regulation Act 1906 and The Control of Pollution Act 1974. Planning legislation may also be relevant, and one English district has an air quality management system with a core component of 40 $\mu g\ m^{-3}$ for both smoke and SO_2. [73]. However, it must be admitted that the setting of mandatory air quality limits is alien to traditional British air pollution policy which relies on a 'best practicable means' approach to control emissions; the United Kingdom seems to lack the necessary coordinating legal measures needed to bring together the relevant authorities and bodies so that all appropriate means of control are used. Control of smoke and SO_2 for the present purpose under Road Traffic regulation powers does not seem to have been considered by the British Government when it made its formal response to the EC Commission on the Directive in 1982. In any case the vehicle emission limits set by other Directives would prevent much freedom of manoeuvre in this connection. [74]. Further United Kingdom legislation is promised to deal with the issue of air quality standards consequent on the comprehensive review of air pollution legislation initiated in 1982. A consultation paper issued in 1986 invited comments by 31st March 1987 on this issue, but Parliamentary time is needed for major legislation to be introduced. The Consultation paper proposed that the EC standards for air quality for smoke and SO2 (and also for lead and NO_2): "should be formally recognised in law by empowering the Secretary of State to set air quality standards (whether nationally or by area) by Regulations. These standards would be taken into account in the setting of b.p.m. (best practicable means) for particular processes, while recognising the effect of emissions from other sources, e.g. vehicles ... In this context, and against the background that EC Directive 84/360 makes provision for the setting of emission limits on a community–wide basis, based on the best available technology not entailing excessive costs, and taking into account the nature, quantities and harmfulness of the emissions concerned, it is proposed that the Secretary of State should be empowered to set emission limits (whether nationally or by area) by Regulations for specified emissions or in relation to specified processes." [75]. However, it should be noted that the Consultation Paper was overwhelmingly concerned with stationary sources of pollution.

So far as, at least, the United Kingdom is concerned it is arguable that air pollution sources should be treated together with a co–ordinated programme of reduction. However, it is doubtful whether this can be achieved given the existing structure of national and EEC laws, and the absence in the United Kingdom of a single overall environmental protection agency such as an 'Environment Protection Commission', though the Department of the Environment is making moves towards the introduction of integrated pollution control [76].

With specific regard to NO_2 air quality standards for the ambient atmosphere were set for the EC by Directive 85/203. The date for compliance with the limit was set at 1st July 1987, and the limit itself is 200 μgm^{-3} of NO_2 (calculated on the basis of the 98th percentile of mean values per hour recorded throughout the year) with more stringent guide values to be applied to specifically protected zones. Exemptions may be given for states unable to meet the requirements, but subject to a requirement that they must plan for improvements in air quality, and a final compliance date of 1st January 1994 is set. Member States also remain free to set stricter limits on NO_2 than those in the Directive. In the United Kingdom the EEC limit is generally met in respect of NO_2, and only very heavily trafficked areas are likely to exceed it. This being so it is likely that NO_2 control within the United Kingdom would be seen primarily as a task for requirements relating to vehicle emissions to achieve, but, of course, those requirements cannot conflict with the general EC vehicle emission standards already discussed [77].

The general air quality requirements as to lead are contained in Directive 82/884, which sets limit values to be adhered to from 9th December 1987, though derogations may be obtained down to 9th December 1989. The limit is that the lead concentration in air must not exceed two μgm^{-3} as an annual average mean concentration, though member states may set stricter limits. Various powers exist in the United Kingdom to comply with the Directive, such as: Section 79 of the Control of Pollution Act 1974 empowering local authorities to monitor lead levels in air; powers under Public Health legislation to enable local authorities to take steps for securing the abatement of statutory nuisances, and powers under the Health and Safety at Work Act 1974 to control emissions from scheduled processes. However, it is generally believed that the power to bring about a reduction of the lead content of petrol contained in Section 75 of the Control of Pollution Act 1974, already considered above, will be sufficient to reduce lead levels in general to the standard required by the Directive [78].

In the United States of America air quality standards exist for both stationary and moving sources of atmospheric pollutions. Particularly stringent standards for emissions from motor vehicles and fuel standards are laid down, and it is to these that attention must now be turned.

6. HIGHWAY POLLUTION STANDARDS IN THE UNITED STATES OF AMERICA

The place of the motor vehicle, both as an object of desire and loathing, in the United States is almost too well known to require further documentation, but the following, from 1967, serves as an example of the literature:

> "One more word about our shiny master, the motor car. As motors are stepped up for higher compression, year by year, nitrogen oxides are stepped up also. And as gasoline manufacturers vie for more 'pick–up' by adding new substances like tetraethyl lead and nickel to the gasoline, these extremely toxic substances are added to our atmosphere. The insane competition for speed and power bows neither

> to safety nor to health. It is unreasonable to blame the manufacturers. In the end they put out what the public demands. ... Detroit is convinced the customer prefers the fast jump to a long and happy life." [79].

Certainly from the 1960's onwards there has been a vocal lobby in the United States for controls over motor vehicles and the pollution they produce. Georgiades, Chiron and Joumard comment that action in the United States was forced because of the urgent problems of California where high emission levels, the particular local micro–climate and hydrocarbon evaporation had produced severe pollution problems, such as the photochemical 'smog' of Los angeles. Despite disagreements between politicians, who wanted a 90% reduction of polluting emissions, and technical experts who envisaged a 60% reduction, the legal limits ultimately set were what the politicians desired, being based on the most advanced technical possibilities, and, indeed, on possible future developments. In consequence vehicle manufacturers had to adopt complex and expensive measures to meet the new legal requirements [80].

In 1970 the US Congress passed the Clean Air Act (amended in 1977) laying down emission standards for vehicles, more stringent than those previously applying, to become effective from 1975. This led to the introduction of catalytic converters to deal with CO and hydrocarbon emissions. Furthermore the Environmental Protection Agency, a federal body, established in 1970, decided to use its powers under the clean air legislation to prohibit the use of lead as a fuel additive, partly because that was necessary to make catalytic converters work. However, because of general health fears the agency also instituted a more general programme of phasing out lead from petrol. The legality of this decision was challenged, but the EPA's action was upheld in the US Court of Appeals [81]. Regulations became effective requiring unleaded fuel for catalytic converter equipped vehicles in 1974, and in 1978 the amount of lead to be added for conventionally fueled cars was reduced from 2.5 grammes per gallon to 1.1 grammes per gallon, with effect from October 1982. This has generally reduced population exposure to ambient lead, and by the mid 1980's over 50% of vehicles on the road in the United States used unleaded fuels. In 1984 EPA proposed that lead levels in petrol should be reduced to 0.1 gramme per gallon, and this was reached in stages between July 1985 and January 1986. EPA further proposed totally lead free petrol from 1st January 1988. (The position in Japan is broadly similar[82]). EPA has also set national air quality standards in respect of lead, particularly with a view to protect the health of young children aged under five, and the limit is 1.5 μg Pb m^{-3}. Though challenged in Court this was upheld [83]. One of the chief features of interest of Lead Industries Association v EPA was that the court examined EPA's processes for arriving at the ambient air standard for lead. EPA's decision was not based on unanimous scientific opinion, but there was a sufficient weight of authority to justify the line taken, and – a most important issue – EPA was also justified in providing a margin of safety in its standard. (See also further below and above for Georgiades, Chiron and Joumard's comments on the limit setting process.)

In addition to the controls over lead outlined above various other controls exist over highway pollution in the United States. Though the 1970 and 1977 Clean Air Acts are federal legislation, the individual States are also required to implement the legislation via State Implementation Plans (SIPs). EPA has also drawn up the National Ambient Air Quality Standards (NAAQS), for example the lead standard referred to above. These standards are subject to periodic review under the terms of the 1977 Act, and standards exist for, inter alia, total suspended particulate matter (smoke), carbon monoxide, SO_2 and NO_2 [84].

Emission controls for vehicles are strict in the United States. For example in 49 States (California is an exception for which see further below) petrol engined vehicles of up to 6000 lb gross vehicle weight may emit only up 0.41 g/mi of hydrocarbons, 3.4 g/mi of CO, 1.0 g/mi of NO_x and 0.6 g/mi of particulate matter. In California the limits are 0.39 g/mi (hydrocarbons), 7.0 g/mi (CO), 0.4 g/mi (NO_x) and 0.4 g/mi (particulate matter). For light–duty vehicles up to 8,500 lb gross vehicle weight, the general emission limits are: 0.8 g/mi (hydrocarbons), 10.0 g/mi (CO), 2.3 g/mi (NO_x) 0.26 g/mi (particulate matter), and for heavy–duty vehicles over 8,500 lb gross vehicle weight the figures are 1.3 g/bhp–hr (hydrocarbons), 15.5 g/bhp–hr (CO), 10.7 g/bhp–hr, (NO_x), 0.25 g/bhp–hr (particulate matter). Again California has separate limits for both heavy and light duty vehicles [85]. In this respect California is in a special position vis–a–vis the other States of the Union. The US emission standards are national requirements and the States may not depart from them save for California which may do so under waiver from the EPA [86]. California's standards overall since 1977 have had to be as strict as those elsewhere, but there have been variations from the federal standards from time to time, as for example in 1978 when California's requirements on CO emissions were relaxed and the NO_x requirements were tightened up by comparison with general federal requirements [87] (and see also the beginning of this Chapter for further comment on how California's limit values are determined.)

The EPA emission standards have not gone without legal challenge, which, as we have seen was also the case with its requirements over lead in fuel. EPA has been generally upheld by the US Courts in a series of actions; see for example Automotive Parts Rebuilders Association v EPA where the Court upheld a requirement of the emission controls that manufacturers should warrant that their emission control equipment would comply with federal standards for a period of some five years,[88] and Chrysler Corporation v EPA where an order from the EPA to Chrysler to recall a number of vehicles not conforming to emission requirements was upheld [89]. Under the amendments to the Clean Air Act of 1977 vehicle manufacturers must warrant that a vehicle is free of defects that could lead to it not complying with emission standards, and checks are carried out by EPA at vehicle assembly lines to check that standards are being met. It would also seem to be established that, under the relevant U.S. legislation, where vehicles do not conform with emission standards the only remedy is recall and repair by the manufacturer, see Center for Auto Safety v Ruckelshaus [90]. EPA has also taken successful action against individual states which have failed to implement satisfactory inspection and maintenance programmes for vehicle emission systems. The requirement for such a programme arises after a state has asked for more time to meet Air Quality Standards (see above) in respect of CO and NO_x, see for example United States v Ohio Department of Highway Safety [91]. Indeed there has been a general increase since 1977 in the number of jurisdictions in the United States where annual gaseous emission inspections for used vehicles are required [92]. Inspection and maintenance (I/M) programmes for motor vehicles were introduced on a mandatory basis by New Jersey, Oregon, Arizona and Rhode Island, and these conformed with EPA standards. By 1986 30 States had I/M programmes in operation, and these related to 55 urban areas (including Washington DC), five further urban areas were due to come under the aegis of I/M programmes by 1988. I/M programmes tend, however, to affect mostly those driving older cars as it is generally found that emission problems increase with a vehicle's age. The drivers of such vehicles in the United States tend to be among the less well off, and so some States, e.g. Pennsylvania, have enacted laws to set maximum scales of charges for the repair of vehicles failing I/M tests. If costs over the maximum have to be imposed for some reason the state subsidizes the bill [93].

In the United States transportation planning and management powers are also used to curb highway pollution, especially in view of the much higher level of dependence in that country on private transport than is the case in the nations of the EEC. Examples of this method of dealing with the problem include the establishment of 'Park and Ride' schemes at the edges of urban areas, and the setting up of 'bus and cycle lanes'. Fiscal measures have likewise been used, such as the reduction of bridge tolls for those using pooled transport arrangements, and their increase for vehicles singly occupied. However, EPA's proposed parking surcharges for vehicles were not pursued [94].

CONCLUSIONS AND RECENT DEVELOPMENTS

Highway pollution is in some ways a discrete topic. The motor vehicle is a major producer of pollutants. This is recognized by Georgiades, Chiron and Joumard who identify three "rationalities" in the attempt to fix emission standards: the scientific which aims to protect populations and the environment agianst pollutants and which involves both scientific and empirical techniques, and which may postulate low limit values hard to achieve in practice; the economic which is concerned to protect industries giving rise to polluting emissions, and the political which expresses the sensitivity of society towards the issue. A standard is a compromise reached between these "rationalities". Georgiades, Chiron and Joumard conclude: "scientific evaluations ... provide an indication of the size of the necessary reductions and hence a basis for discussion. Greater understanding of the subject, increased sensitivity and the adaptation of the industries concerned lead to periodic reductions in the limiting values which accordingly approach those considered to be acceptable from a medical point of view." [95]. The noise and congestion caused by motor vehicles, and the demand for land to be devoted to road building are examples of how pollution arises from the use of the highway. Yet this pollution is truly only part of a wider air pollution issue, and cannot be studied or dealt with effectively in isolation, though, of course, reductions in the polluting emissions from vehicles would go a very long way to reducing overall atmospheric pollution. The motor car produces, it is true, the common atmospheric pollutants. but these then combine with other similar products from stationary sources of pollution, e.g. coal fired power stations. With regard to such combined sources of pollution, air quality controls which seek to limit the levels of pollutants generally in air are clearly more far reaching measures than emission controls for vehicles which serve only to control one source of pollutants. Obviously the latter can serve to advance the goals of the former: whether they do is another matter. The United States is a nation where attempts are made to bring 'can' and 'do' together. In the nations of the EC on the other hand, and particularly within the United Kingdom, it is harder to see such an integrated approach towards air pollution. On the contrary a fragmented, piecemeal approach, perhaps with conflict between the various standards and methods of regulations applicable to the problem, would seem to characterise the position.

At the start of this chapter it was stated that eradication of environmentally deleterious activities by law requires agreed perception of perceived hazards of a known variety, technology or other means appropriate to deal with the problems, and a convergence of interests amongst those who create hazards and those who suffer from them so that eradication becomes acceptable. In conclusion, to those factors can be added a need to see a problem 'in the round' and not just as a discrete issue, and also the necessity for strong co–ordinating agencies able to direct, harmonise and integrate all means of approach to the task of eradication. Highway pollution in all its forms does, of course, raise particular and discrete issues, but it is also part of a much wider issue of atmospheric pollution generally whose solution requires a much greater breadth of vision and action than is current in many states.

The following developments in the law should also be noted.

A/ Unleaded Petrol Sales in the EC

By October 1990 sales of unleaded petrol accounted for 38% of UK petrol sales, but that was still behind nations such as the former West Germany and Denmark, though in advance of, for example, Ireland, France, Italy, Spain, Portugal and Greece. See ENDS *Report* 191, p.26.

B/ Further changes in EC vehicle emission standards

It is proposed to consolidate the EC Directives on vehicle emissions, reducing permitted emissions for large and medium cars to the levels for small cars as from July 1992. Germany, The Netherlands, Denmark and Greece have also argued for further emission reductions to take effect from 1995, though the nations of the EC may compromise on this issue by agreeing that a sliding scale of tax incentives may be introduced by Member States to encourage the purchase of "cleaner" cars as opposed to the imposition of stricter mandatory emission standards. See ENDS Reports 184 page 27, and 191, page 32.

C/ The use of Road Traffic Regulation Powers to control vehicle noise, and the impact of EC legal requirements

In R v London Borough Transport committee, ex parte Freight Transport Association Ltd (1989) "Times" November 17th the Queens bench divisional Court was called upon to determine the legality of a scheme introduced by a number of London Boroughs, purportedly made under the Road Traffic Regulation Act 1984, Section 6, whereunder lorries over 16.5 tonnes maximum gross weight were banned from certain roads at certain times of the day, though an exemption from the ban could be obtained by fitting specified airbrake silencers. The Divisional Court found the scheme illegal in that it trespassed on the powers of the Secretary of State to regulate vehicle construction under Section 41 of the Road Traffic Act 1988, and also because it went beyond the technical requirements of relevant EC Directives (70/156, 70/157, 71/320). The London Boroughs appealed against this decision, and the Court of Appeal decided the matter in October 1990 ("Times" October 4th). In the Court of Appeal the matter was disposed of on the basis that the exemption requirements transgressed EC Directive requirements. The Court applied Commission v United Kingdom (re dim-dip car lights [1988] 3 CMLR 437 arguing that the relevant Directives are exhaustive as to the technical requirements that may be required by Member States in respect of vehicle braking systems, and that they may not be supplemented. Indeed any supplemental requirements would contravene the principle of the free movement of goods which is central to the functioning of the EC. Thus vehicle construction and use, though nationally regulated, must be in line with EC requirements on a community wide basis, and cannot be allowed to vary according to where vehicles ply their trade.

This decision should be read alongside Commission of the EC v Federal Republic of Germany (1990) "Times" 24th October. Here Germany had introduced new road tax systems effectively penalising certain types of goods haulage by road. The issue arose as to whether Germany was in derogation of EC obligations not to take any unilateral measure which has the effect, directly or indirectly, of changing, to the detriment of other Member States, the conditions under which international transport across or between Member States takes place. Germany argued the measure was designed to protect the environment by encouraging freight to move by rail or canal. The Court of Justice of the EC found a prima facie case had been made against Germany. Though the Court accepted that environmental protection is a Community objective, see Commission v Denmark (1988) ECR 4607, that does not of itself allow a Member State to introduce a transport regime which disadvantages the carriers from other Member States.

In consequence of these decisions it may be argued:

- Environmental considerations are important but not paramount within the scheme of values of the EC;
- EC law may act as a force to retard environmmental protection in that the need for Member States to move in harmony can inhibit individual national initiatives on the environment;
- Where a measure has the effect of damaging free trade and transport within the EC that measure in general needs to be collectively supported;
- Just as national environmental measures must be consonant with EC obligations so too must any action taken by a subordinate agency such as a local authority.

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APPENDIX

ENVIRONMENTAL QUALITY STANDARDS

1. AIR QUALITY STANDARDS

(i) EEC Limit Values, all in $\mu g\ m^{-3}$

smoke and sulphur dioxide (Directive 80/779)

reference period	smoke	sulphur dioxide
Year, median of daily values	68	if smoke < 34 : 120 if smoke ≥ 34 : 80
Winter, median of daily values, October to March	111	if smoke < 51 : 180 if smoke ≥ 51 : 130
Year (Peak), 98th percentile of daily values	213	if smoke < 128 : 350 if smoke ≥ 128 : 250

lead (Directive 82/882
the limit value is 2 $\mu g\ m^{-3}$ (annual mean)

nitrogen dioxide (Directive 85/203)
the limit value is 200 $\mu g\ m^{-3}$, expressed as the 98th percentile of hourly average concentrations measured throughout a calendar year.

(ii) USA National Ambient Air Quality Standards (NAAQS) :

Pollutant	**Standard ($\mu g\ m^{-3}$)**		**Comments**
	Primary*	**Secondary‡**	
***total suspended particulate matter* (PM$_{10}$ standard)**	65	60	annual geometric mean,
	150	90	*maximum* 24–hour concentration,
sulphur dioxide	80	60	annual arithmetic mean
	365	260	maximum 24–hour concentration
	–	1,300	maximum 3–hour concentration
carbon monoxide	10,000	↑	maximum 8–hour concentration
	40,000		maximum 1–hour concentration
ozone	235	↑ same as primary	maximum 1–hour concentration
lead	1.5	↓	3–months average
nitrogen dioxide	100	↓	annual arithmetic mean

* to protect human health
‡ to protect welfare

(iii) WHO Guideline Values

a) Guideline values for combined exposure to sulphur dioxide and particulate matter

			Reflectance assessment	Gravimetric assessment	
	Averaging time	Sulphur dioxide ($\mu g\ m^{-3}$)	Black smoke ($\mu g\ m^{-3}$)	Total suspended particulates (TSP) ($\mu g\ m^{-3}$)	Thoracic* particles ($\mu g\ m^{-3}$)
Short term	24 hours	125	125	120	70
Long term	1 year	50	50	–	–

* estimated equivalent values for a sampler having a 50% cut–off point at 10μm

b) Guideline values for individual substances based on effects other than cancer, odour or annoyance.

Substance	Time–weighted average	Averaging time
Cadmium	1 – 5 ng m^{-3}	1 year (rural areas)
	10–20 ng m^{-3}	1 year (urban areas)
Carbon disulphide	100 $\mu g\ m^{-3}$	24 hours
Carbon monoxide	100 mg m^{-3}	15 minutes
	60 mg m^{-3}	30 minutes
	30 mg m^{-3}	1 hour
	10 mg m^{-3}	8 hours
1,2–Dichloroethane	0.7 mg m^{-3}	24 hours
Dichloromethane (Methylene chloride)	3 mg m^{-3}	24 hours
Formaldehyde	100 $\mu g\ m^{-3}$	30 minutes
Hydrogen sulphide	150 $\mu g\ m^{-3}$	24 hours
Lead	0.5–1.0 $\mu g\ m^{-3}$	1 year
Manganese	1 $\mu g\ m^{-3}$	1 year
Mercury	1 $\mu g\ m^{-3}$ (indoor air)	1 year
Nitrogen dioxide	400 $\mu g\ m^{-3}$	1 hour
	150 $\mu g\ m^{-3}$	24 hours
Ozone	150–200 $\mu g\ m^{-3}$	1 hour
	100–120 $\mu g\ m^{-3}$	8 hours
Styrene	800 $\mu g\ m^{-3}$	24 hours
Sulphur dioxide	600 $\mu g\ m^{-3}$	10 minutes
	300 $\mu g\ m^{-3}$	1 hour
Tetrachloroethylene	5 mg m^{-3}	24 hours
Toluene	8 mg m^{-3}	24 hours
Trichlorethylene	1 mg m^{-3}	24 hours
Vanadium	1 $\mu g\ m^{-3}$	24 hours

(c) Guideline values for individual substances based on effects on terrestrial vegetation

Substance	Guideline value	Averaging time	Remarks
Nitrogen dioxide	$95\mu g\ m^{-3}$ $30\ \mu g\ m^{-3}$	4 hours 1 year	In the presence of SO_2 and O_3 levels which are not higher than $30\mu g/m^3$(arithmetic annual average) and $60\mu g/m^3${average during growing season} respectively.
Total nitrogen deposition	$3\ g\ m^{-2}$	1 year	Sensitive ecosystems are endangered above this level.
Sulphur dioxide	$30\mu g\ m^{-3}$ $100\mu g\ m^{-3}$	1 year 24 hours	Insufficient protection in the case of extreme climatic and topographic conditions.
Ozone	$200\mu g\ m^{-3}$ $65\mu g\ m^{-3}$ $60\mu g\ m^{-3}$	1 hour 24 hours averaged over growing season	
Peroxyacetylnitrate	$300\mu g\ m^{-3}$ $80\mu g\ m^{-3}$	1 hour 8 hours	

2. WATER QUALITY STANDARDS

Water quality legal standards have not been routinely applied to highwaty runoff. Instead, most countries operate on the basis that

i) standards apply to the receiving water, and
ii) the quality of the runoff discharged should not result in serious deterioration of the quality of the receiving water.

Rivers and streams are classified according to intended use (see Tables which follow) and the impact of discharged highway runoff should not result in a change of classification. More advanced legislation exists in the United States of America. Following the introduction of the 1987 Water qualtiy Act, discharges from a municipal separate storm sewer system serving a population of more than 100,000 will require a permit by October 1992 (ref: J.D. Gallup and K. Weiss, in : L.A. Roesner, B. Urbonas and M.B. Sonnen, editors, Design of Urban runoff Quality Control, ASCE, 1988).

EEC Water Quality Guide Values
(μg/*l* unless states otherwise)

	Drinking Water	Protection of Salmonid Freshwater Fish
BOD		3 mg/*l*
total ammonia (as N)	0.038	0.031
nitrite (as N)		3
nitrate (as N)	5.65	
aluminium	50	
antimony	10 (MAC)	
arsenic	50 (MAC)	50 (*)
barium	100	
cadmium	5 (MAC)	5 (*)
chromium	50 (MAC)	5 (*)
copper	100	1 (*)
iron	50	
lead	50 (MAC)	4 (*)
manganese	20	
mercury	1 (MAC)	1 (*)
nickel	50 (MAC)	50 (*)
silver	10 (MAC)	
zinc	100	10 (*)

MAC maximum allowable concentration

* hardness < 50 mg/*l* $CaCO_3$

RIVER CLASSIFICATION, ENGLAND AND WALES

River Class	Quality	Remarks	Current potential uses
1A	i) Dissolved oxygen saturation greater than 80%.	i) Average BOD probably not greater than 1.5 mg/ℓ	i) Water of high quality suitable for potable supply abstractions and for all other abstractions.
	ii) Biochemical oxygen demand not greater than 3mg/ℓ	ii) Visible evidence of pollution should be absent	ii) Game or other high class fisheries
	iii) Ammonia not greater than 0.4mg/ℓ		iii) High amenity value
1B	i) DO greater than 60% saturation	i) Average BOD probably not greater than 2mg/ℓ	i) ii) Water of less high quality than Class 1A but usable for substantially the same purpose.
	ii) BOD not greater than 5mg/ℓ	ii) Average ammonia probably not greater than 0.5mg/ℓ.	
	iii) Ammonia not greater than 0.9mg/ℓ	iii) Visible evidence of pollution should be absent.	
		iv) Waters of high quality which cannot be placed in Class 1A because of high quality effluent present or because of the effect of physical factors such as canalisation, low gradient or eutrophication.	
2	i) DO greater than 40% saturation.	i) Average BOD probably not greater than 5mg/ℓ	i) Waters suitable for potable supply after advanced treatment.
	ii) BOD not greater than 9mg/ℓ.	ii) Water not showing physical signs of pollution other than humic coloration and a little foaming below weirs.	ii) Supporting reasonably good coarse fisheries
			iii) Moderate amenity value.

River Class	Quality	Remarks	Current potential uses
3	i) DO greater than 10% saturation. ii) Not likely to be anaerobic. iii) BOD not greater than 17mg/ℓ		Waters which are polluted to an extent that fish are absent or only sporadically present. May be used for low grade industrial abstraction purposes. Considerable potential for further use if cleaned up.
4	Waters which are inferior to Class 3 in terms of dissolved oxygen and likely to be anaerobic at times.		Waters which are grossly polluted and are likely to cause nuisance.
X	DO greater than 10% saturation.		Insignificant watercourses and ditches not usable, where objective is simply to prevent nuisance developing.

SUBJECT INDEX