The Handbook of Environmental Chemistry 64
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Peter Pluschke
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Indoor Air Pollution

Second Edition

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The Handbook of Environmental Chemistry

Founded by Otto Hutzinger

Editors-in-Chief: Damià Barceló • Andrey G. Kostianoy

Volume 64

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Indoor Air Pollution

Volume Editors: Peter Pluschke · Hans Schleibinger

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Aims and Scope

Since 1980, *The Handbook of Environmental Chemistry* has provided sound and solid knowledge about environmental topics from a chemical perspective. Presenting a wide spectrum of viewpoints and approaches, the series now covers topics such as local and global changes of natural environment and climate; anthropogenic impact on the environment; water, air and soil pollution; remediation and waste characterization; environmental contaminants; biogeochemistry; geoecology; chemical reactions and processes; chemical and biological transformations as well as physical transport of chemicals in the environment; or environmental modeling. A particular focus of the series lies on methodological advances in environmental analytical chemistry.

Series Preface

With remarkable vision, Prof. Otto Hutzinger initiated *The Handbook of Environmental Chemistry* in 1980 and became the founding Editor-in-Chief. At that time, environmental chemistry was an emerging field, aiming at a complete description of the Earth's environment, encompassing the physical, chemical, biological, and geological transformations of chemical substances occurring on a local as well as a global scale. Environmental chemistry was intended to provide an account of the impact of man's activities on the natural environment by describing observed changes.

While a considerable amount of knowledge has been accumulated over the last three decades, as reflected in the more than 70 volumes of *The Handbook of Environmental Chemistry*, there are still many scientific and policy challenges ahead due to the complexity and interdisciplinary nature of the field. The series will therefore continue to provide compilations of current knowledge. Contributions are written by leading experts with practical experience in their fields. *The Handbook of Environmental Chemistry* grows with the increases in our scientific understanding, and provides a valuable source not only for scientists but also for environmental managers and decision-makers. Today, the series covers a broad range of environmental topics from a chemical perspective, including methodological advances in environmental analytical chemistry.

In recent years, there has been a growing tendency to include subject matter of societal relevance in the broad view of environmental chemistry. Topics include life cycle analysis, environmental management, sustainable development, and socio-economic, legal and even political problems, among others. While these topics are of great importance for the development and acceptance of *The Handbook of Environmental Chemistry*, the publisher and Editors-in-Chief have decided to keep the handbook essentially a source of information on "hard sciences" with a particular emphasis on chemistry, but also covering biology, geology, hydrology and engineering as applied to environmental sciences.

The volumes of the series are written at an advanced level, addressing the needs of both researchers and graduate students, as well as of people outside the field of "pure" chemistry, including those in industry, business, government, research establishments, and public interest groups. It would be very satisfying to see these volumes used as a basis for graduate courses in environmental chemistry. With its high standards of scientific quality and clarity, *The Handbook of Environmental Chemistry* provides a solid basis from which scientists can share their knowledge on the different aspects of environmental problems, presenting a wide spectrum of viewpoints and approaches.

The Handbook of Environmental Chemistry is available both in print and online via www.springerlink.com/content/110354/. Articles are published online as soon as they have been approved for publication. Authors, Volume Editors and Editorsin-Chief are rewarded by the broad acceptance of *The Handbook of Environmental Chemistry* by the scientific community, from whom suggestions for new topics to the Editors-in-Chief are always very welcome.

Damià Barceló
Andrey G. Kostianoy
Editors-in-Chief

Preface

In the year 1751 John Arbuthnot published “An Essay concerning the Effects of Air on Human Bodies” [1] and gave a comprehensive review of the scientific state of the art of his time. He summarized his knowledge and his findings in a number of “Practical Aphorisms” such as “Private Houses ought to be perflated once a Day, by opening Doors and Windows, to blow off the Animal Steams” and a second example “Houses for the sake of Warmth fenc’d from Wind, and where the Carpenters Work is so nice as to exclude all outward Air, are not the most wholsom.”

His thoughts are still valid and his treaty is still valuable – not in all details. Building technologies and practices have changed much and Indoor Science became a very broad, inter-disciplinary field of knowledge.

Going through the agenda of international conferences on indoor air and recent publications, there are still a number of topics, which have been discussed already in the eighteenth and nineteenth centuries, such as moisture and dampness [2], mould and bacteria [3], and bad odours and carbon dioxide [4].

Relatively early research on hygienic conditions in urban dwellings and establishments by Pettenkofer and others in the eighteenth and nineteenth centuries covered some of still today relevant topics in indoor air quality. Pettenkofer developed some basic principles. From a hygienic point of view he analysed the indoor living conditions and came to the conclusion that science could contribute significantly in improving everyday life by defining rules for indoor environmental quality and health.

He revealed [5–8] that human emissions and activities must be taken into consideration and

– He developed a concept for chamber studies of indoor emission sources [6]. He summarized: “substances which pass off from the skin and lungs require as close an investigation as those that are eliminated by the urine. Hitherto no method has been devised, or apparatus constructed, adequate to the conditions of such an inquiry. . . . The present state of physiology requires conditions of inquiry in which a man can breathe and move freely without any apparatus attached to his body… This can only be done by placing the man in an accurately measurable current of air, which can be tested for certain constituents before it comes in contact with him, and can be investigated again after the air has taken up the gaseous matters thrown off by the skin and lungs."

- He underlined the importance of source control to ensure good indoor air quality [7]. Still today – despite all the technical means, which are at hand nowadays to enhance ventilation and to clean air – it is a valid principle; in Pettenkofer's words: "If I had a nuisance in my room, I should be a fool if I kept it there and trusted to stronger ventilation. The rational way is to do away with the pollutions, not to keep them and to fight them by ventilation."
- And through his thorough studies on human bio-effluents [7] he could deduce general rules for ventilation and for the acceptable limit of carbon dioxide in indoor environments. The carbon dioxide abundance of 1,000 parts per million, which he proposed, is consistent with current understanding of the value that marks the threshold of significantly degraded air by human bio-effluents. According to Pettenkofer, this concentration "is, in my mind, the measure only for all the other alterations which take place in the air simultaneously and proportionately, in consequence of respiration and perspiration; its increase shows to what degree the existing air has been already in the lungs of the persons present. On an average, in spaces in which the air kept good, there existed a ventilation of more than 2100 cubic feet per head and hour." – a rate of about 16.5 l/s per person, not far from the value prescribed in today's ventilation standards.

Still today, human occupants play a significant role in indoor chemistry – may be an even more significant role than at the times of Pettenkofer, because we are still emitting bio-effluents but we add nowadays a variety of personal care products which introduce a large number of chemicals into indoor environments and may be reactive with other components in air, as recently reviewed by Weschler [9].

Even though England, France and Germany may have been the homes of the first scientific treatment of indoor air quality, meanwhile Indoor Air Science has become a worldwide subject and a matter of concern and we try to reflect this in this volume of the *Handbook of Environmental Chemistry*.

The range of indoor spaces is broad: from very basic households with open fire and simple cook-stoves to sophisticated urban architecture and to mobile indoor environments like aircraft cabins, cars and public transport systems.

The first edition of this volume of the *Handbook of Environmental Chemistry* on Indoor Air Pollution was published in the year 2004 [10]. It focused on the analysis of basic factors determining indoor air quality. Still a lot of research is dealing with emission scenarios [11], pathways of contaminants, toxicological effects of individual compounds as well as of their mixtures and epidemiological evaluation of the effects of indoor air pollution. Human well-being and performance are becoming often the focus of indoor air research [12]. The second edition follows the same concept by updating most of the reviews. Tunga Salthammer and Godwin Ayoko give broad reviews in this volume on emission processes and on the large variety of volatile organic pollutants. Furthermore, new topics are introduced, such as the reflections of Carl-Gustaf Bornehag on the relation of chronic illnesses and indoor chemical exposure.

Beyond these basics of Indoor Sciences, it must be indicated that a number of recent developments will open new horizons for indoor air research:

(a) During the last few years we became aware that climate change may result in new challenges in building research and indoor air quality studies [13, 14]. Challenges due to climate change bring more severe periods of heat, heavy rainfalls and high humidity or – on the contrary – extreme regional dryness. Changing meteorological conditions will alter architecture and building equipment as well as living habits of the people. Smart technologies will serve people under various circumstances but will require new ways of management of the buildings and may lead to challenges in the management of indoor air quality: more sensors will be installed in houses, control devices must be followed and maintained and a risk of introducing un-voluntarily new sources of odours and chemicals may follow.

(b) As a consequence of increasing energy prices and of the limits of fossil energy resources, more emphasis is now put on energy efficiency of buildings. This changes definitely the concepts for ventilation because buildings are becoming tight structures. Effects are manifold: less exposure to ambient air pollutants, change to mechanical ventilation, more significant influence of indoor pollution sources, new factors influencing function, comfort and cosiness of a building. Traditional building considered ventilation as a design characteristic to be guaranteed by adequate orientation of a building, the good choice of building materials and a concept considering dimension and function of windows and doors [15]. It is a certain risk that we forget Pettenkofer's statement: "If I had a nuisance in my room, I should be a fool if I kept it there and trusted to stronger ventilation." Modern technical equipment makes it easy to fight nuisances by increasing ventilation but source control will always be the most favourable way to guarantee a safe and cosy indoor environment.

(c) Special emphasis must be put on those households using traditional cookstoves. They are worldwide still large in number as the following diagram (published by Kirk Smith et al.) shows [16, Fig. 1]:

Over the last two decades many intervention strategies and modernization processes have been launched to improve living conditions and health in rural areas as well as in the rapidly developing urban agglomerations in Asia, Africa and Latin America by implementing new designs of stoves which help to create better indoor air quality. Lidia Morawska has – coming from an Australian background – updated her review on indoor particles, combustion products and fibres and covered a wide range of non-gaseous pollutants resulting from a wide variety of human activities, i.e. from tobacco smoke.

Whereas in the Northern hemisphere cooking is mostly a minor factor in indoor air quality, it determines largely indoor air quality in households and certain establishments in the Southern hemisphere. Kalpana Balakrishnan provides an updated report on the situation focusing on India in this volume. More reports

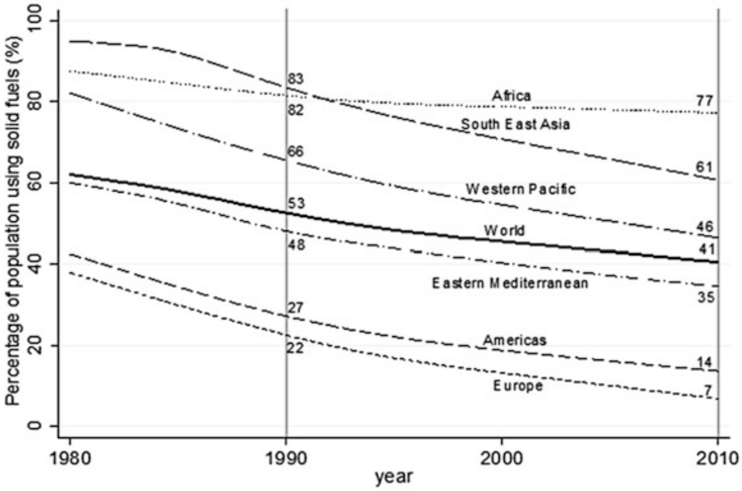


Fig. 1 Percentage of population using solid fuels for cooking 1980–2010

Fig. 2 Open fire in a Nicaraguan household – before intervention

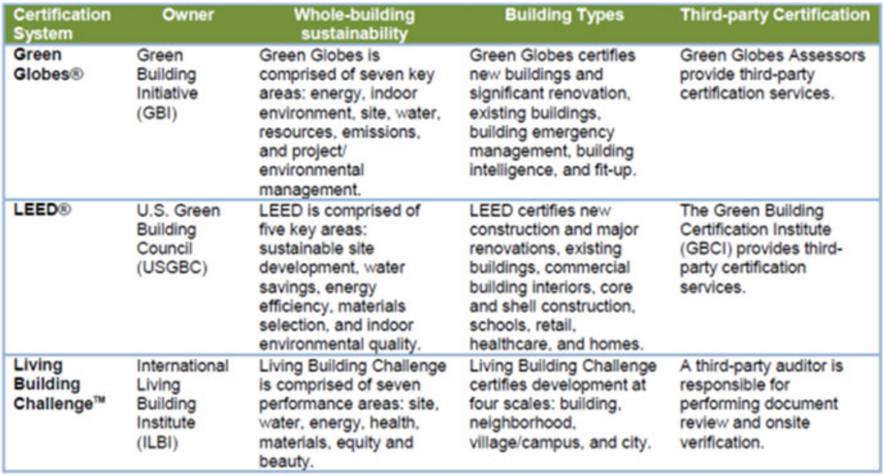
from Africa, Latin America [17, 18] and Asia [19] have brought forward similar findings and led to a wide range of activities and changes, such as demonstrated in the pictures from a kitchen in San Carlos, Nicaragua (Figs. 2 and 3).

Fig. 3 Improved kitchen and new stove in a rural kitchen in Nicaragua – after intervention

(d) In the industrialized world intervention strategies in order to improve building quality focus on building certification as a market-oriented tool to strengthen energy efficiency and indoor air quality in the building sector.

More and more applied research is dealing with concepts to prevent indoor air pollution: Green Buildings should be energy efficient and free from pollutants – clean, proper and comfortable. Building certification may be one concept to deal with these requirements but specific points have to be made also in architecture, building design, technical equipment and control mechanisms during building and renovation activities. Economic aspects will play a crucial role in defining the target lines for a Green Building [20].

Internationally accepted standards for Green Buildings tend to evaluate the environmental impact of a building throughout its entire life cycle. This takes account of not only factors such as energy efficiency, use of natural light and use of environmentally friendly building materials that are available at a regional level, but also transport routes or the integration of the building into the public transport network. According to these concepts indoor air quality is



Certification System	Owner	Whole-building sustainability	Building Types	Third-party Certification
Green Globes®	Green Building Initiative (GBI)	Green Globes is comprised of seven key areas: energy, indoor environment, site, water, resources, emissions, and project/ environmental management.	Green Globes certifies new buildings and significant renovation, existing buildings, building emergency management, building intelligence, and fit-up.	Green Globes Assessors provide third-party certification services.
LEED®	U.S. Green Building Council (USGBC)	LEED is comprised of five key areas: sustainable site development, water savings, energy efficiency, materials selection, and indoor environmental quality.	LEED certifies new construction and major renovations, existing buildings, commercial building interiors, core and shell construction, schools, retail, healthcare, and homes.	The Green Building Certification Institute (GBCI) provides third-party certification services.
Living Building Challenge™	International Living Building Institute (ILBI)	Living Building Challenge is comprised of seven performance areas: site, water, energy, health, materials, equity and beauty.	Living Building Challenge certifies development at four scales: building, neighborhood, village/campus, and city.	A third-party auditor is responsible for performing document review and onsite verification.

Fig. 4 Summary of Green Building Certification Systems [21, 22]

> just one aspect to be dealt with, yet a crucial one. The following table shows a summary of the Green Building Certification Systems in the USA (Fig. 4).

In more countries national certification schemes have evolved and have been put into practice. In Germany the market for the certification of Green Buildings is mainly shared between three systems:

DGNB (Deutsche Gesellschaft für Nachhaltiges Bauen): a system which is based on European norms and standards but adaptable to a large range of local conditions. To that extent the DGNB system may be applied in a wide range of geographical, social and technical settings.

Secondly LEED plays a major role in Germany and thirdly the BREE system (Building Research Establishment Environmental Assessment Method) is well known and often used [23]. Additionally CASBEE (Comprehensive Assessment System for Building Environmental Efficiency) from Japan may be mentioned – a system widely spread in Asia.

Finally, Jiming Hao and Tianle Zhu give a comprehensive view in this volume on Chinese standards and strategies to overcome existing problems in indoor air quality: a combination of traditional cooking behaviour and home equipment with modern home decoration and refurbishment products contributes to the deterioration of indoor air quality – not to talk about ambient air quality and its contribution to indoor air pollution. Wherever you are, indoor air quality is a work in progress – much has to be done to improve and guarantee indoor air quality – 266 years after John Arbuthnot's statement "Private houses ought to be perflated once a day . . . ."

Nürnberg, Germany Peter Pluschke

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Contents

Release of Organic Compounds and Particulate Matter from Products, Materials, and Electrical Devices in the Indoor Environment

Tunga Salthammer

Abstract In the indoor environment, the need for the reliable assessment of human exposure to indoor pollutants is still attracting increasing attention. This, however, requires a detailed understanding of the relevant compounds; their sources, physical and chemical properties, dynamics, and reactions; and their distribution among the gas phase, airborne particles, and settled dust as well as the availability of modern measurement techniques. With respect to a healthy indoor environment, only low-emitting products, which do not influence the indoor air quality in a negative way, should be used in a building. Therefore, materials and products for indoor use need to be evaluated for their chemical emissions. The demand for standardized test methods under laboratory conditions has resulted in several guidelines for determination of emission rates by use of test chambers and cells. Many studies have shown that the types of sources in occupational and residential indoor environments, the spectrum of emitting compounds, and duration of emission cover a wide range. Furthermore, it has now been recognized that both primary and secondary emissions may affect indoor air quality. The problem might become dominant when components of different materials can react with each other or when catalytic materials are applied. As a consequence, modern product development should also consider secondary products, which seem to be of importance for long-term emissions. In order to characterize the release of compounds and particles from materials under realistic conditions, it is important to study the influence of processing, substrate, and climatic parameters on emitting species and emission rates.

Keywords Building products, Cell, Emission rate, Emission testing, Particles, SVOC, Test chamber, VOC

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Contents

Abbreviations

CEN	European Committee for Standardization
D	Diffusion coefficient
D_a	Aerodynamic particle diameter
DEHP	Di(2-ethylhexyl) phthalate
DIBP	Diisobutyl phthalate
DIN	Deutsches Institut für Normung
DINCH	Diisononyl cyclohexane-1,2-dicarbo
DINP	Diisononyl phthalate
DMF	Dimethylformamide
DnBP	Di(n-butyl) phthalate
DNPH	Diphenylhydrazine
ECMA	Previously: European Computer Manufacturers Association
FID	Flame ionization detector
FLEC	Field and Laboratory Emission Cell
FMPS	Fast Mobility Particle Sizing
GC/MS	Gas chromatography mass spectrometry
ISO	International Organization for Standardization
K_OA	Octanol–air partition coefficient
L	Loading (m^2/m^3)
n	Air exchange rate (h^{-1})
NDIR	Nondispersive infrared spectroscopy
OSB	Oriented strand board

PAS	Photoacoustic spectroscopy
PCA	Principal component analysis
PID	Photoionization detector
PM	Particulate matter
PTR-MS	Proton-transfer-reaction mass spectrometry
SBR	Styrene–butadiene rubber
SER_A	Area specific emission rate
SMPS	Scanning Mobility Particle Sizing
SVOC	Semi-volatile organic compounds
TCB	Trichlorobenzene
TDI	Toluene diisocyanate
TEA	Triethylamine
TSP	Total suspended particles
TVOC	Total volatile organic compounds
VOC	Volatile organic compounds
VVOC	Very volatile organic compounds
ZDEC	Zn-diethyldithiocarbamate
μ-CTE	Micro Chamber Thermal Extractor

1 Introduction

Research into the air quality in the interiors of noncommercial buildings has been of great importance for years [1–3]. There are many reasons for this, but according to Salthammer and Bahadir [4], the following facts have special significance:

- In countries in the cold and temperate climatic zones, people spend the vast majority of their time not in the open air but in many types of interior rooms closed off to outdoors, generally spending more than half of this time in their own homes [5]. In certain sectors of the population (e.g., small children and older persons), this percentage can even reach 90% and more.
- Indoor air contains a wide spectrum of pollutants like organic and inorganic components, particulate matter, and microbes. Particularly in the private sphere, many activities of the room occupants make a contribution to contamination of the air – especially smoking but also the use of a large number of household and hobby products.
- One consequence of the energy crises of the 1970s and the rising energy costs resulting from this was that many rooms were much better sealed off against the outside air than previously had been the case. This trend towards energy saving is still unbroken today due to the push for a global reduction in CO_2 emissions. More effective sealing – when unaccompanied by adequate mechanical ventilation – will result in a sometimes considerable reduction in the air exchange rates in these rooms, which in turn means the concentrations of substances emitted into the indoor air will rise.

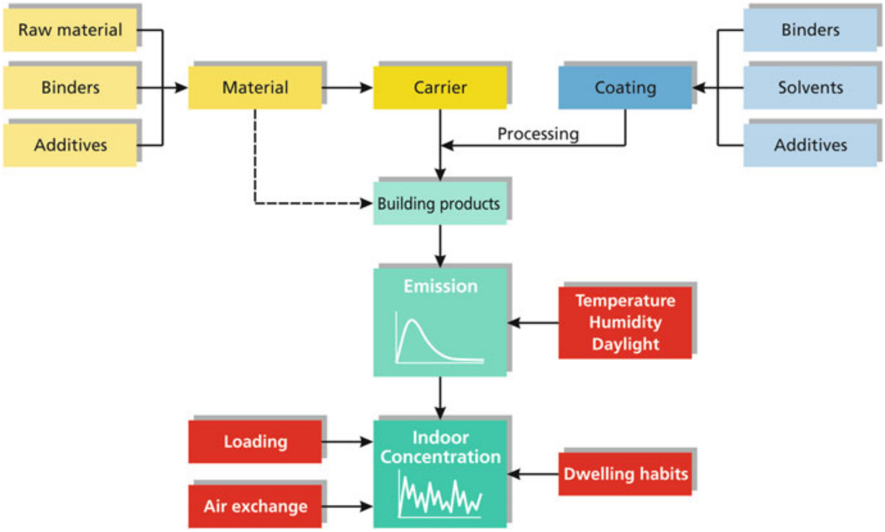


Fig. 1 Influence of applied materials, processing, climatic parameters, and living behavior on emissions from a building product and indoor concentrations

- Today office buildings are often equipped with air-conditioning systems. This can not only bring about the release of organic and microbial contaminations but also even produce climatic conditions in the rooms which are perceived as unpleasant.

Pollutants and their analysis and evaluation of their sources have a long tradition in indoor-related sciences. The interrelationships which lead from emission from a source to a particular room air concentration are very complex (see Fig. 1). Not only the emissions from the primary source itself must be taken into consideration but in all cases also the boundary conditions governing the situation in question. In addition to the surface area of the product in the room, these will in particular include building-related physical parameters, the building and dwelling characteristics, and the utilization conditions and also possibly social factors as well. Over the years, in industrialized countries, the release of volatile organic compounds (VOCs) from building products has been decreasing [6]. For this reason, VOCs temporarily slipped out of the primary focus of indoor research. Soon, they experienced a renaissance when being implicated with the formation of oxidation products and ultrafine particles from chemical reactions under indoor conditions. In parallel, indoor scientists became interested in the interaction of semi-volatile organic compounds (SVOCs) between the gas phase, airborne particles, and settled dust [7–9], because it was recognized that human exposure to SVOCs can occur via ingestion, inhalation, and dermal absorption [10, 11]. As far as the very volatile organic compounds (VVOCs) are concerned, the discussion still focuses on formaldehyde [12, 13], since its classification as a human carcinogen by the International Agency for Research on Cancer [14].

The evaluation of the emission potential of individual products and materials under realistic conditions and over defined timescales therefore requires the use of climate-controlled emission testing systems, so-called emission test chambers and cells. As shown in Fig. 2, the size can vary between a few cm^3 and several m^3, depending on the application [15]. Such emission measurements are performed for a number of purposes [16] as, for example:

- Compiling of substance-specific emission data from various sources to back up field investigations into indoor air quality
- Determination of the influence of environmentally relevant factors such as temperature, humidity, and air exchange on the emission characteristics of the products
- Processing of characteristic emission data to estimate product emissions
- Processing of characteristic emission data to develop models that can be used to predict indoor concentrations
- Ranking of various products and product types on the basis of the characteristic emission data

On the basis of characteristic emission data, there is an increasing tendency to award a quality label to particularly "low-emission" products [17].

2 Measuring Organic Indoor Pollutants

2.1 Environmental Test Chambers and Cells

The selection of the testing system, the sampling preparation, and the test performance all depend on the task to be performed. According to ISO, chambers and cells are defined as follows:

Emission Test Chamber: Enclosure with controlled operational parameters for the determination of volatile organic compounds emitted from building products (ISO 16000-9) [18].

Emission Test Cell: Portable device for the determination of volatile organic compounds emitted from building products. The emission cell is placed against the surface of the test specimen, which thus becomes part of the emission cell (ISO 16000-10) [19].

During an emission investigation, the product/material to be investigated is tested with regard to temperature (T), relative humidity (r.h.), air exchange rate (n), air velocity, and product loading factor ($L =$ ratio of surface of product to be investigated to the volume of the emission test chamber) under standardized testing

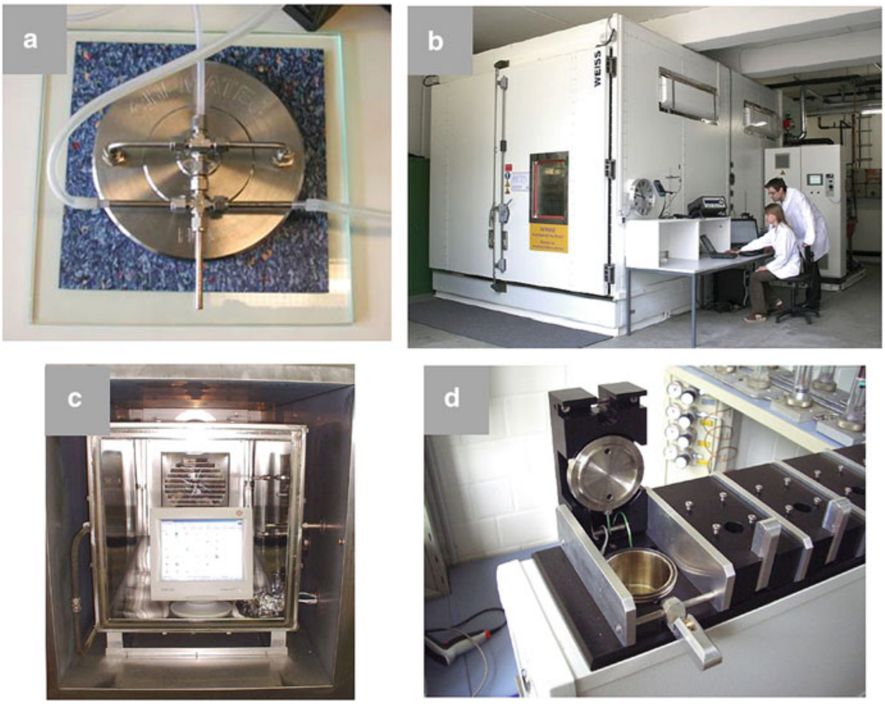


Fig. 2 Different devices for emission testing of building products. (**a**) FLEC; (**b**) 48 m^3 stainless-steel chamber; (**c**) 1 m^3 stainless-steel chamber; (**d**) micro chamber (μ-CTE)

conditions in an emission test chamber that can be sealed gastight against the outside atmosphere. The test procedure is suitable for emission investigations of both surfaces and of volume samples. This is a convention process where the boundary conditions are selected in such a way that they reflect those to be found in realistic indoor rooms. In interpreting the results of test chamber investigations, it must, under certain circumstances, be taken into account as a limiting factor that not all realistic conditions to be found in an indoor room can be simulated.

Today, emission test chambers typically range in size from 0.020 to 50 m^3. They are broadly classified into small scale (<1 m^3) and large scale. A common interior volume for an emission test chamber is 1 m^3 and the interior is usually made of glass or stainless steel. The specific emission rates determined in test chambers are associated with application of the product in a reference room under specified climate and ventilation conditions. A reference room is needed since it is not possible to evaluate emissions by testing in all possible use scenarios. The European Reference Room dimensions, as defined by CEN TC 351 WG2, are provided in Table 1. The resulting product loading factors, climate, and ventilation conditions are selected to represent the general indoor environment.

Table 1 Dimensions of the European reference room (CEN TC 352)

Parameter	Dimension
Chamber volume	30 m^3
Chamber dimension (L × W × H)	4 m × 3 m × 2.5 m
Surface walls	31.4 m^2
Surface floor or ceiling	12 m^2
Surface window	2 m^2
Surface door	1.6 m^2

It is often useful to have an instrument or an indicator for quality control of products with regard to emissions arising during the utilization phase. To enable action to be taken during the production process as quickly as possible, fast emission investigations accompanying the production process would be a worthwhile objective. It is therefore desirable to have a measuring system that can be used to carry out emission testing and quality assurance on the spot. The relevant principle of a transportable emission testing cell for mobile application was implemented in Scandinavia for the first time in 1991 with the so-called Field and Laboratory Emission Cell (FLEC) [20, 21]. The FLEC opens up the opportunity of carrying out nondestructive emission testing on surfaces within the framework of field investigations. In this way it is possible to identify emissions from building products when they are already installed and also the sources for air-polluting substances.

The micro chamber (μ-CTE) is a new device comprising small cylindrical chambers each with an interior volume of approximately 45 mL [22]. The μ-CTE consists of six individual stainless-steel cylindrical chambers ($d = 4.5$ cm), located in one unit. All six micro chambers are supplied simultaneously with the same, controlled flow of synthetic air. The μ-CTE is a complementary tool for standard emission test facilities and intended to produce fast information about the composition and level of VOC emissions for development of new, low-emission products/materials. Micro chamber measurements have already been shown to provide a useful basis for fast emission screening prior to formal product certification. Moreover, since the device has only a small chamber volume and offers a relatively high loading factor, it has also been shown to offer enhanced sensitivity relative to conventional chambers for compounds of low volatility (SVOCs = semi-volatile organic compounds) [23] which is due to reduced sink effects.

ISO standards defining emission test chambers and cells as well as the preparation of test specimen and analytical issue are summarized in Table 2.

2.2 *Sink Effects*

A sink effect is the fact that the released components partially adsorb within the test chamber, e.g., at the chamber walls. This can result in an incorrect reading of the

Table 2 ISO standards for the emission testing of building products using test chambers and cells

ISO standard	Description	Reference
ISO 16000-6	Determination of VOCs in test chamber air	[24]
ISO 16000-9	Emission test chamber method	[18]
ISO 16000-10	Emission test cell method	[19]
ISO 16000-11	Preparation of test specimen	[25]
ISO 16000-25	Micro chamber method	[26]
ISO 16000-28	Odor emissions	[27]

concentration determined at the chamber outlet, which can lead to the wrong emission rate being computed. In principle every test chamber demonstrates a low sink effect. The degree of adsorption on chamber walls and in materials as well as the extent of the recovery can vary extremely for different VOCs [28]. This depends in part on the volatility of the relevant substance. It was recently demonstrated that absorption within the test chamber is favored as the boiling point increases [23]. A comparatively higher polarity of a compound can also be favorable to the adsorption at the chamber walls. Sollinger et al. [29] showed that the sink effect is clearly increased by introducing an adsorbing sample surface (carpet) into the test chamber. The fact that the sample to be investigated in a chamber investigation also acts as a sink has been reported by several authors [30–32]. An important factor influencing sinks is the air distribution in the device, which can be determined by use of anemometers [33, 34] or can be calculated by computational fluid dynamics [22].

Whereas the sink effects caused by the chamber itself can be reduced to a minimum by using appropriate construction materials, it is not possible to influence the sink effects attributable to the actual sample. Many materials subjected to emission testing are also good sinks for the substances emitted by them. Especially with porous building materials with a large surface area or with foams, it must be expected that the intrinsic material sink effect leads to a clear delay of the substance emission. Figure 3 shows an experiment where the test substance 1,2,3-trichlorobenzene (1,2,3-TCB) was charged into the test chamber continuously over a period of 3 days (sorption phase). The increase in the chamber concentration and the decay after the charging was completed (desorption phase) at an air exchange rate of 1 h^{-1}. The experiment was then repeated using a gypsum board in the chamber. This revealed that both the increase and the decay of the concentration of the test substance were considerably delayed. Whereas with the empty chamber the 1,2,3-TCB concentration had dropped to almost zero already after 300 min after the charging was completed, this process lasted more than 3 days with the sink in the chamber. The sink effect of the sample material therefore means that the measured emission rate is smaller than the actual rate, as emitted substances are initially absorbed in the sink. At the same time the measured duration of the emission process is longer, as even after the completion of the actual emission, substances are still released from the sink and lead to the chamber concentration dropping more slowly. This sink effect caused by the sample to be investigated

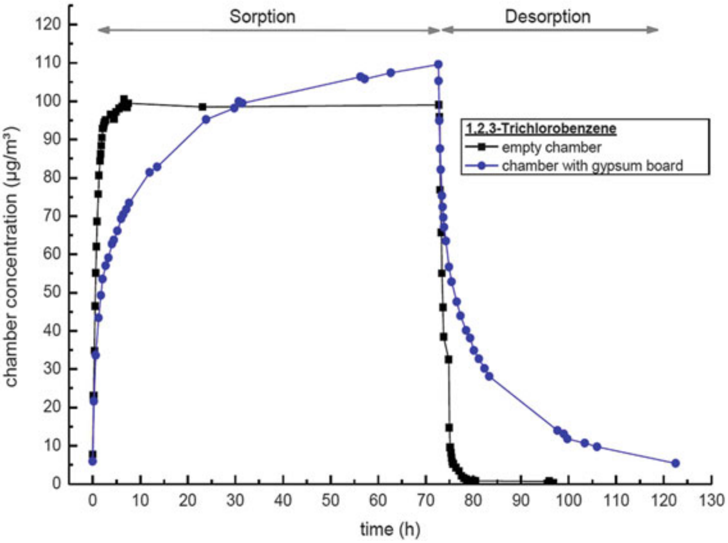


Fig. 3 Concentration vs. time profile for 1,2,3-TCB in the empty chamber (*black*) and with gypsum board (*blue*) (Picture by courtesy of E. Uhde)

plays a role in each chamber investigation and cannot be avoided. For this reason it is of fundamental importance to guarantee that the chamber has as good a recovery as possible. Uhde and Salthammer [35] have evaluated different test chambers by cluster analysis. Principal component analysis (PCA) revealed that the sink effect essentially depends on a compound's boiling point. In connection with the sink effect, it should also be taken into consideration that a so-called "memory effect" can result for subsequent investigations, which must be avoided by appropriate cleaning of the chamber. It is advantageous here if the test chamber can be thoroughly heated at higher temperatures for cleaning purposes.

2.3 Calculation of Emission Rates

Air measurement in a chamber or cell initially produces the concentration $C(t)$ at the time t of the measurement. To enable better comparability of the measured data, the specific emission rate (SER) independent of air exchange and loading is to be preferred. The SER describes the product-specific emission behavior, e.g., as surface-specific emission rate (SER_A) with the unit µg/(m^2 h) or as unit-based specific emission rate (SER_U) with the unit µg/(unit h).

The time-dependent determination of the emission potential is carried out according to the balance equation (1), whereby $C(t)$ = chamber concentration in µg/m^3, n = air exchange rate (in h^{-1}), and L = loading in (m^2/m^3):

$$\mathrm{d}C/\mathrm{d}t = L\,\mathrm{SER}(t) - nC(t) \tag{1}$$

For a decaying concentration-time function, SER(t) is obtained by transition to the difference quotient according to Eq. (2):

$$\mathrm{SER}(t) = [(\Delta C/\Delta t) + nC(t)]/L \tag{2}$$

with

$$\Delta C_i/\Delta t_i = [(C_i - C_{i-1})/(t_i - t_{i-1}) + (C_{i+1} - C_i)/(t_{i+1} - t_i)]/2 \tag{3}$$

Thus, if there are $n + 1$ experimental data available for concentration, $n-1$ emission rate values can be obtained by this method. In the steady state ($\mathrm{d}C/\mathrm{d}t = 0$), Eq. (2) progresses to Eq. (4):

$$\mathrm{SER} = (nC)/L \tag{4}$$

More sophisticated emission source models that also include sinks can be found in the literature [36]. However, the calculation of emission rates using physically based models requires nonlinear curve fitting and a sufficient number of data points. Large errors in parameter estimates can result from rough chamber data and/or wrong models [37].

2.4 Kinetics and Mass Transfer

The dynamics of emission processes from building product surfaces has been studied in detail, and a number of physical and empirical models of different complexity have been described [31]. The emission can be characterized by two fundamental physical processes (see Fig. 4) [38]:

(a) Gas-phase mass transfer (i.e., external diffusion)
(b) Source-phase mass transfer (i.e., internal diffusion)

The gas-phase mass transfer model (a) is based on molecular diffusion across a laminar boundary layer as described in Eq. (5):

$$\mathrm{SER} = \frac{D}{\delta}\cdot(C_\mathrm{S} - C_\mathrm{i}) = k_\mathrm{g}\cdot(C_\mathrm{S} - C_\mathrm{i}) \tag{5}$$

SER is the specific emission rate, D is the diffusion coefficient, δ is the thickness of the boundary layer, C_S is the concentration of the target VOC at the source surface, C_i is the concentration of the target VOC in the air, and k_g is the gas-phase mass transfer coefficient.

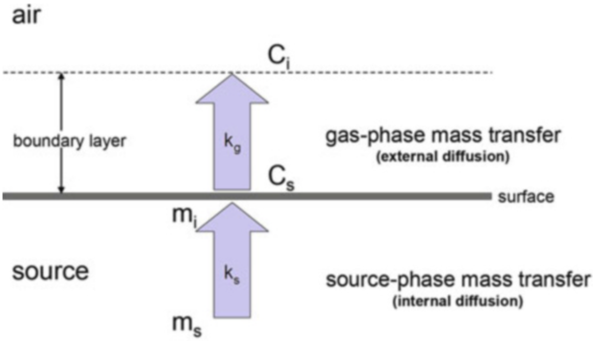


Fig. 4 Diagram of the kinetic processes involved in the mass transfer model of VOCs from material surfaces; adopted from Sparks et al. [38]

Source phase mass transfer is limited by diffusion from the interior of the source to the surface and can be described by Eq. (6):

$$\mathrm{SER} = k_{\mathrm{s}} \cdot (m_{\mathrm{S}} - m_{\mathrm{i}}) \tag{6}$$

Here, m_{s} is the mass of the target VOC in the source, m_{i} is the mass of the target VOC at the surface, and k_{s} is the source-phase mass transfer coefficient.

In terms of the comparability of emission test results obtained from different chambers and cells, three different scenarios have to be considered. For $k_{\mathrm{g}} \gg k_{\mathrm{s}}$ the emission is controlled by the external diffusion process, and the thickness of the boundary layer δ is directly related to the air velocity above the surface. This applies to most wet-applied or liquid products during the drying/curing phase. In this case, the airflow conditions in the test facility (i.e., air velocity and turbulence) are important. This means that precise control of the air velocity may be critical in the short term (i.e., 1–14 days) if it is not to influence the emission test result. For $k_{\mathrm{g}} \ll k_{\mathrm{s}}$ the emission is controlled by the internal diffusion process and the influence of the airflow condition in the test facility should be negligible. This applies to most materials manufactured in the solid phase and to wet-applied or liquid products after they have dried or cured. A more difficult situation arises for $k_{\mathrm{g}} \approx k_{\mathrm{s}}$ or if the ratio k_s/k_g changes over time. This situation arises in case of curing two-component diisocyanate adhesives, where a two-step process can be observed. In the first step, the emission is dominated by surface evaporation, and the decay of emission is mainly caused by the decrease in monomer content due to reaction. In the second step, the release is limited by internal diffusion [39].

2.5 Online Monitoring

Time-fluctuating concentration levels are found in indoor rooms since in addition to the type and strength of the emission sources, the air exchange situation and room-climate conditions are also changing. If a temporarily high-resolution concentration profile or peak values are to be determined, continuously recording instruments which record concentrations in time segments of just a few minutes or in individual cases of even a few seconds will be indispensable [40].

Nondispersive infrared spectroscopy (NDIR) uses the radiation absorption of molecular vibration bands in the infrared region and is of importance in the measurement of carbon monoxide (CO), carbon dioxide (CO_2), and dinitrogen oxide (N_2O). The last two mentioned can be used as indicator gases in determining the air exchange rate. Also counted among the infrared methods is photoacoustic spectroscopy (PAS). This is a technique which converts the excitation energy adsorbed in the infrared region during radiationless relaxation into an intensity-modulated acoustic signal. Commercial instruments allow up to five components to be measured in parallel. In addition to CO, CO_2, and sulfur hexafluoride – which is also an indicator gas for air exchange measurement – these instruments are primarily important in measuring hydrocarbons, these being determined as total volatile organic compounds ($TVOC_{PAS}$) calibrated vs. propane.

Ultraviolet absorption serves as the measuring principle for ozone measurement. The excitation source is a low-pressure mercury lamp. At the measurement wavelength of 254 nm, ozone exhibits a high absorption coefficient. The measuring principle is less susceptible to problems, and equipment of this type has been used for many years in measurement networks.

One of the oldest and most important continuous methods is to register the concentration by means of a flame ionization detector (FID). Hydrocarbon compounds are oxidized in a hydrogen flame. This creates ions which are registered electrically. The intensity of the signal depends on the number of C–H groups in the molecule. A total value is obtained which is given as the "total hydrocarbons" or $TVOC_{FID}$.

In the photoionization detector (PID), ionization is effected using ultraviolet light and is dependent on the ionization potential of the individual compound. Typical excitation energies fall in the 8–14 eV range. Operation often takes place at 10.2 eV, and with this excitation energy, aromatic hydrocarbons can be picked up with particular sensitivity.

Some work has been carried out to evaluate TVOC values obtained from different methods. The comparison of the FID and PAS response for the same chamber concentration showed remarkable deviations between the two detector types. Therefore, it will be almost impossible to compare $TVOC_{PAS}$ and $TVOC_{FID}$ for complex substance mixtures. Although direct reading instruments offer a very useful and convenient means to assess general concentration trends of VOC during emission testing or chamber experiments, their application for the determination of reproducible, comparable TVOC values is quite limited [41, 42].

Proton-transfer-reaction mass spectrometry (PTR-MS) allows the monitoring of selected $[M + 1]^+$ ions; the high time resolution makes it a powerful tool for indoor-related research. The PTR-MS technique was successfully applied in test chambers for the determination of material diffusion coefficients [43] and the time-resolved measurement of material emission signatures [44] and for investigating the effect of catalytic air purifiers on indoor air quality [45].

2.6 Discontinuous Sampling and Analysis of VOCs

Three general types of solid sorbents are mainly used for trapping VOCs in air: inorganic sorbents, carbon-based porous materials, and porous organic polymers. The main types of inorganic sorbents are silica gels, molecular sieves/zeolites, aluminum oxides, and magnesium silicates. Carbon-based sorbents include activated charcoals, carbon blacks, graphitized carbon blacks, and graphitized molecular sieves. Phenylene oxide polymers and polyurethane foams represent the most widely used polymer sorbents. The surface area of a sorbent influences the amount of a given substance that can be adsorbed by the medium, whereas the surface polarity determines the general type of compounds a sorbent can be used for. The sorbents offer different suitability for VOC analysis depending on the type and amount of substance to be sampled. The inorganic sorbents are often used to trap hydrocarbons and polychlorinated biphenyls (PCBs). Carbon-based sorbents with large surface area and molecular sieves are useful to trap very low-boiling compounds but are unsuitable for higher boiling substances; labile or reactive compounds may even decompose on active sites on the sorbent surface. Porous polymers with a comparatively small surface area allow the adsorption and desorption of high-boiling compounds like glycols and phthalates. On the other hand, it is difficult to sample low-boiling compounds like C2–C5 alkanes. The different characteristics of the presented sorbents show the need to carefully choose the right adsorption medium for a given VOC mixture [46].

The state of the art in emission investigations is sampling on Tenax TA or combinations of Tenax/Carbotrap. This allows accumulation of polar and nonpolar VOCs [47, 48]. Following thermal desorption (TD), GC separation on a capillary column, and MS analysis using quadrupoles [49], identification takes place on the basis of spectrum libraries [50] and quantification by means of internal and external standards [51]. This method is suitable for concentration ranges from $<1\ \mu g/m^3$ to several mg/m^3. With regard to sampling, it should also be noted that Tenax TA is degraded by nitrogen oxides [52]. A total of 25 Tenax TA degradation products from the sampling of NO, NO_2, O_3, H_2O_2, OH, and limonene-O_3 mixtures were identified by Klenø et al. [53].

The dinitrophenyl hydrazine (DNPH) process has become established for aldehydes and ketones. Here the substances are derivated to hydrazones and quantified using liquid chromatography with UV detection (HPLC/UV). Alternatively, formaldehyde can be determined by using acetyl acetone (acac) method [54]. There is an excellent correlation between the acac and the DNPH method for formaldehyde. However, the DNPH method underestimates higher aldehydes (>C5) in comparison to TD-GC/MS [55].

2.7 Air and Fogging Sampling of SVOCs

Quantitative analysis of SVOCs in room air takes place after sampling with a combination of particle filters and adsorbent (typically polyurethane foam or XAD-2). This method takes into account the fact that only one part of the SVOCs is present in the gas phase while another part is bound to suspended dust particles. Conventional methods are used for analysis, such as GC/MS. The sampling of SVOCs on Tenax followed by thermal desorption GC/MS is limited to compounds with a boiling point <370°C (C22, docosane). In test chambers, the sampling on glass wool is convenient for compounds of lower volatility [8].

To assess the condensable amount of SVOCs, a fogging apparatus can be mounted in a chamber. The device was firstly developed by Bauhof and Wensing [56] for SVOC measurement in automobile interiors. It is made of a cooled aluminum body (usually 15°C at 23°C chamber temperature) with high-polished stainless-steel collection plates mounted on each side. After the exposure to the chamber atmosphere, the plates are dismounted and put into a stainless-steel extraction apparatus, where the condensed substances are eluted in an ultrasonic bath. The extracts are then analyzed. The fogging value (in μg) determined over a period of 14 days is a characteristic value for the amount of SVOC that can be expected to condense on cooler surfaces in the indoor environment. The fogging method is based on a convention. The comparison of results requires identical experimental performance.

2.8 Analysis of Airborne Particles

Particles in the atmospheric environment constitute a major class of pollutants, in addition to those occurring in gaseous or vapor form. The actual shape and chemical composition of airborne particles can only be ascertained with a great deal of effort. For this reason the "aerodynamic diameter" D_a as defined in Eq. (7) is used to characterize the particles. D_P is the physical particle diameter and ρ_P is the particle density. For a particle of any shape or size, this is equal to the diameter of a sphere of density 1 g/cm^3 which in still or laminar flowing air has the same gravitational settling velocity as the particle [57]:

$$D_a = D_P \cdot \sqrt{\rho_P} \tag{7}$$

While previously it was mostly only the TSP (total suspended particles) which were measured and only in special cases was an attempt made to break the suspended dust down by particle size, today the focus is on specific particle size fractions. At the forefront of interest, here are the fractions with defined particle diameter, in particular PM_{10} and $PM_{2.5}$; recently also PM_1 has become a matter of scientific research. PM_{10} refers to the sum of inhalable suspended dust particles with an aerodynamic diameter <10 µm while $PM_{2.5}$ is the sum of respirable particles < 2.5 µm. The so-called ultrafine particles (UFPs) with a diameter < 0.1 µm have recently been attracting increasing interest as well. For measuring the particle number concentration vs. size distribution of UFPs, Scanning Mobility Particle Sizing (SMPS) and Fast Mobility Particle Sizing (FMPS) are the methods of choice. The SMPS enables the analysis of particles in the size range from 10 to 1,000 nm with a size resolution of 64 channels per decade. The FMPS (5.6–560 nm) provides a much lower resolution in size with 16 channels per decade. On the other hand, the low scan time of 1 s offers the recording of fast dynamic processes.

2.9 Sample Preparation for Emission Testing

In selecting parameters for emission testing and interpreting the results, it must be taken into consideration that this is a convention process and that not all real conditions in indoor rooms can be simulated. The results of such investigations can be increasingly used in the future to develop “low-emission” products and, by selecting such products in the form of source checking, make a considerable contribution to achieving good indoor air quality.

The so-called sample preparation is of particular importance with regard to emission test results. This involves, for example, the question whether liquid samples (paints, adhesives, etc.) are to be applied on absorbent or inert substrates. Investigations of paints have found that clearly different emissions occur depending on the substrate [58]. A sample preparation as close to the realistic situation as possible is always appropriate where the results are to be used to derive health-related statements. In investigating floor coverings, it can be useful to investigate only the product surface that is exposed to the indoor room and to seal off the backing and any side edges during the emission test with as “low-emission” material as possible. To achieve as realistic an examination as possible, emission investigations of complete sandwich systems, e.g., lime flooring, leveling compound, primer, adhesive, and top layer, are of particular interest. Such test scenarios can provide valuable information on compatibility and possible chemical reactions between the individual building products. If only the individual products are subjected to separate emission testing procedures, then follow-up reactions [59] cannot be found.

3 Nonreactive and Reactive VOCs

Most materials and products for indoor use contain organic compounds, which can be released in the indoor environment. The majority of the pollutants described are typical industrial chemicals like aliphatic and aromatic hydrocarbons that are not reactive under indoor room conditions.

Table 3 Influencing factors in an emission investigation

Product history	Age of product
	Transport to the test laboratory and storage time
	Sample preparation
	Climatic conditioning prior to testing
Chamber measurement	Supply air quality
	Background concentration
	Sink effects/recovery
	Air exchange ratio
	Air tightness of the environmental test chamber
	Internal air mixing
	Air velocity
	Accuracy of temperature, relative humidity, air exchange ratio, product loading factor
Sampling of test chamber air and chemical analysis	Choice of sorbent tube
	Recovery of specific VOC and SVOC
	Detection limit
	Precision
	Accuracy

However, the results of research work have shown that “indoor chemistry” is also of great importance in the evaluation of indoor air quality. Various products contain reactive components, which decompose under the influence of oxidants, heat, moisture, or light in the material [43]. In many cases volatile compounds result in this way with sensory effect [60] and are released into the room air as secondary emissions. In addition new VOCs can be formed even during the manufacturing process. Wolkoff et al. [61] pointed out that especially reactive organic compounds and their reaction products can cause irritation in persons and therefore make a significant contribution to the sick building syndrome (SBS). Many building materials, furnishings, and household products have nowadays been documented as sources of reactive VOCs.

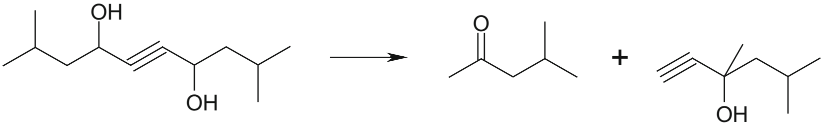
Tucker [62] has summarized the major factors that are now thought to influence emission of vapor-phase organic compounds from surface materials (see also Table 3):

- Total amount and volatility of constituents in the material
- Distribution of these constituents between the surface and the interior of the material
- Time (i.e., age of the material)
- Surface area of the material per volume of the space it is in (“loading”)
- Environmental factors such as temperature, air exchange rate, and relative humidity
- Chemical reactions in the source (e.g., conversion in varnishes and some adhesives)

In his publication, Tucker [62] has also compiled emission factors (SER_A) for various compounds from a variety of sources as reported in the literature. Note that SER_A values vary by a factor of many thousands from one material to another.

3.1 Solvents

The wide variety of common solvents and film formers (aromatic hydrocarbons, aliphatic hydrocarbons, ketones, esters, glycols, alcohols) still represents a very important group of emitting compounds, as freshly manufactured products may cause high SER_A values. On the other hand, most solvents do not have any chemical reactions under normal conditions, unless so-called reactive solvents (e.g., styrene) are involved. An important exception is the substance 2,4,7,9-tetramethyl-5-decyne-4,7-diol (T4MDD), which is frequently used as a film former and foam inhibitor in water-based coating systems. T4MDD degrades to 4-methyl-2-pentanon (MIBK) and 3,5-dimethyl-1-hexyne-3-ol. As MIBK is a common solvent, the degradation of T4MDD can be mistaken for the use of MIBK in the analysis [63].



(8)

In test chamber investigations, hemiacetals and acetals such as 1-hydroxy-1-ethoxy-cyclohexane and 1,1-diethoxy-cyclohexane can sometimes be demonstrated qualitatively in the chamber air. These are typical reaction products of cyclohexanone with ethanol. A known artifact in gas-chromatograph analysis is the formation of 1,1-dimethoxy-cyclohexane from cyclohexanone and methanol in standard solutions. The solvent *n*-butanol plays an ambivalent role in the indoor environment. It is frequently used in formulations, but it is also a reaction product of *n*-butyl acetate, *n*-butylacrylate, etc. Moreover, *n*-butanol is released into indoor air from condensation particle counters (CPCs) for aerosol measurement.

Usually, the emission of VOCs from building products is relatively fast, as shown in Fig. 5 for a chamber experiment with a water-based paint surface and the target compound triethylamine (TEA) (boiling point 89°C). The PTR-MS method provides a sufficiently high density of data for the numerical solution of Eq. (2) and for kinetic analysis of the time vs. concentration curve using nonlinear regression [37]. In this case the release of TEA could be described with a double-exponential function as expected for a decaying source in the absence of sinks. More experimental details are provided in the captions of Fig. 5 (see also [43]).

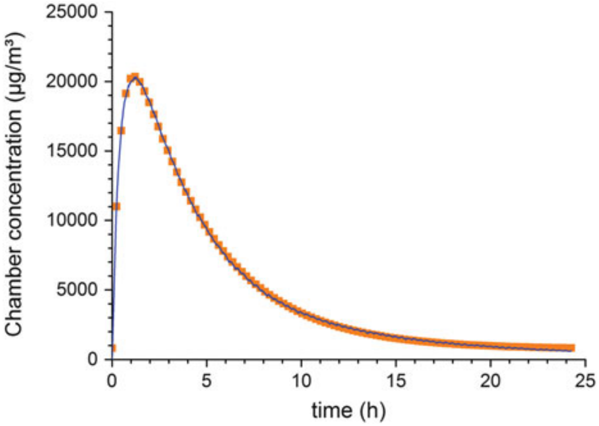


Fig. 5 Emission of TEA from a freshly applied water-based paint (2 g paint on 0.035 m^2 glass plate) in a 0.5 m^3 stainless-steel chamber at $T = 23°C$, r.h. = 50%, and $n = 1\ h^{-1}$. The *solid curve* was obtained from a no-sink double-exponential function $f(t) = A_1 \cdot (\exp(-k_1 \cdot t) + A_2 \cdot \exp(-k_2 \cdot t))$. A total release of 29 mg TEA was calculated from Eq. (2) and numerical integration [43]

3.2 Degradation of Unsaturated Fatty Acids

Saturated and unsaturated aldehydes with chain lengths C5-C11 are some of the most problematic and undesirable substances in indoor rooms. Aliphatic aldehydes are very odor intensive, and the odor is generally described as unpleasantly "rancid" or "greasy." With sensitive persons or in high concentrations, the perception of aliphatic aldehydes can cause nausea. Emission sources in indoor rooms are essentially unsaturated fatty acids such as oleic acid, linoleic acid, and linolenic acid as components of linoleum [64, 65], coating systems containing alkyd resin as well as paints, oils, and adhesives on a natural basis. The aldehydes generated during the oxidation processes of the unsaturated fatty acids can continue to react to the corresponding low-chain acids. Typical degradation products of oleic acid are saturated aldehydes from heptanal (C7) to decanal (C10), whereas linoleic acid is mainly degraded to hexanal (C6). Unsaturated aldehydes such as 2,4-heptadienal result from oxidizing the double unsaturated linolenic acid. The degradation of fatty acids was investigated particularly with regard to food chemistry aspects as aldehydes are strong aroma components [66]. This property is also used in so-called air freshening sprays, which also contain saturated aldehydes [67]. The surface of a piece of furniture can under certain circumstances demonstrate high emission rates for aldehydes for months and even years. The reason for this lies in the constant slow degradation of unsaturated fatty acids under real-life conditions. It was observed with various furniture surfaces that odor problems can occur even after several years of use. In these cases a cracking of the paint film allows oxidative processes after the penetration of atmospheric oxygen.

Chamber tests carried out on wood and wood-based products showed that in addition to the wood species pine and the drying process [68], the pressing process is particularly responsible for the later release of VOCs [69, 70]. Freshly manufactured OSBs made of pine strands show besides the emission of monoterpenes also a considerable emission of pentanal, hexanal, heptanal, octanal, and nonanal. These compounds are generated by thermal and oxidative degradation processes of wood chips during the drying process and then during the pressing process in the surface layers of the OSB [59].

Cork products can also emit considerable amounts of the aldehyde furfural (2-furancarboxaldehyde) [71], whereby the release of the furfural generally corresponds with that of acetic acid. The cause for the formation of furfural is the thermal decomposition of the hemicelluloses contained in the cork at temperatures above 150°C, while acetic acid is formed by the separation of acetyl groups. Further identified by-products in the thermal treatment of natural cork were formic acid and hydroxymethylfurfural [72].

3.3 Terpenes

Terpenes can be emitted in large amounts both by plants and by building products and items of furnishing. The terpenes involve mostly odor-intensive natural materials that are made up of units of 2-methyl-butane and 2-methyl-1,3-butadiene (isoprene) [73]. Of relevance for the indoor area are the monoterpenes (C_{10}) such as α-pinene, β-pinene, limonene, and 3-carene as well as the sesquiterpenes (C_{15}) such as β-caryophyllene and longifolene. Especially resin-rich softwoods such as pine and spruce are strong emission sources for monoterpenes. However, wood, wood products, and coatings are by far not the only emission sources for terpenes in indoor rooms. The substances are also to be found in cleaning agents, cosmetics, air fresheners, paints, lacquers, and oils. A very interesting compound is carvone, which on the one hand is a component in natural terpene balsam. On the other hand, it is also a possible oxidation product of limonene. 3-Carene is generally not an emission product of coatings, as it is removed from the raw products due to its known effect as a contact allergen.

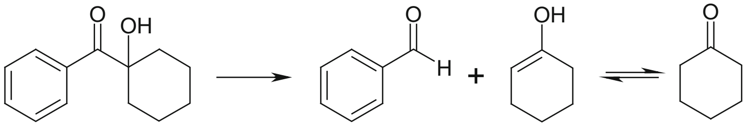
The formation of ultrafine particles due to terpene/ozone reactions is a phenomenon in atmospheric chemistry which has been known for decades [74]. In the indoor environment, terpenes can reach high concentrations due to the diversity of emission sources, while ozone usually makes less than 50% of the outdoor values [75]. Today, there are numerous publications available dealing with product-related terpene/ozone reaction and the formation of ultrafine particles (see Table 4).

Table 4 Compilation of some emission studies on building products and indoor equipment

Products	Description	Ref.
Latex paint	Influence of substrate on VOC emission	[76]
Latex paint	Study of VOC emission	[77, 78]
Latex paint	Influence of ozone on VOC emission	[79]
Photocatalytic wall paint	Influence of light on VOC emission	[80, 81]
Photocatalytic ceilings	Influence of light on VOC emission	[82]
Water-based paint	Influence of film thickness on VOC and SVOC emission	[83]
UV-curing lacquer	VOC emission in dependency of climatic parameters	[84]
Wood-based panels	VOC and formaldehyde emission	[85]
Oriented strand board	VOC emission	[69, 70]
Biocides	Long-term emission	[86]
PVC flooring	VOC emission	[87–89]
Consumer products	Flame retardants	[90]
Polyurethane (PUR) products	Emission of 2,4- and 2,6-TDI	[91]
Polyurethane (PUR) adhesives	Emission of MDI and HDI, influence of temperature, monomer content, curing mechanism	[39, 92]
Cork	VOC emission (chamber)	[71, 72]
Building products (miscellaneous)	Compilation of VOC emission studies	[93]
Building products (miscellaneous)	Degradation of ozone in dependence of building products	[94]
Building products (miscellaneous)	VOC emission in dependence of climatic parameters	[95]
Linoleum	VOC emission, sensory testing	[64, 65]
Furniture surface coating	VOC emission	[63]
Hardcopy devices	Different conditions and parameters	[27, 96–101]
Textile floor covering, PVC flooring, water-based paint	Round-robin test	[102]
Wall coverings	SVOC emission	[103]
Textile floor coverings	VOC emission	[29, 104]
Building products (miscellaneous)	Sensory emission, impact of climatic parameters	[60, 105–107]
Building products (miscellaneous)	Sorptive interactions between VOC and materials	[28, 108, 109]
Essential oils	Particle formation	[110]
Household appliances	Particle formation	[111, 112]
Wood and wood products	Ozone	[113]
Wood and wood products	Processing parameters	[69, 70]
Natural paint	Ozone, particle formation	[114]
Air fresheners and cleaning products	Ozone	[115–117]
Nanosprays	Particle formation	[118, 119]
Electronic cigarettes	Particle formation, nicotine, aroma compounds	[120]
Carpet	Ozone	[121–123]

3.4 Light as a Cause for Secondary Emissions

In many countries legal requirements regarding the consumption and release of volatile solvents have encouraged manufacturers of coating systems to introduce more and more UV-curable formulations. The technique reduces the amount of solvents required for the coating of metal, wood, or other materials significantly and can even be utilized for powder-based systems. Photoinitiators start the curing process and are essential ingredients of UV-curable coating systems. However, photoinitiators are usually overdosed to avoid undercuring of the lacquer film, and a certain amount of residual photoinitiator is left in the finished product after curing. The photochemistry of most common photoinitiators is a chemistry of the carbonyl group. The three important fragmentation processes forming radical species are Norrish I reaction (α-cleavage), Norrish II reaction, and electron transfer. Fragmentation processes of photoinitiators form a number of volatile products, which may contribute to indoor air pollution [84]. Benzaldehyde and alkyl-substituted benzaldehydes are usual components, because Norrish I is the most important reaction for cleavage. A well-known example is 1-phenyl-2-hydroxy-2-methyl-propane-1-one (PHMP). α-Cleavage generates two radicals in the first step. The benzoyl radical may recombine to benzil, reduction of PHMP leads to 1-phenyl-2-methyl-1,2-propane and acetone, and recombination of the 2-hydroxy-propyl radical gives 2,3-dimethyl-2,3-butanediol (pinacol). Cyclohexanone is formed from the hydroxy-cyclohexyl radical upon α-cleavage of 1-hydroxy-cyclohexyl-phenone (HCPK), followed by hydrogen abstraction and keto-enol tautomerization as shown in Eq. (9). Photoinitiator fragments may reach considerable concentrations in indoor air. It should, however, be mentioned that some photoinitiator fragments like benzaldehyde and acetophenone are typical degradation products of the polymeric Tenax TA.



(9)

Nowadays, the reduction of indoor air pollutants by photocatalysis has received considerable interest. Typical building products like wall paints and ceilings are equipped with modified TiO_2 to produce ·OH radicals and the superoxide radical ion $\cdot O_2^-$ and act as a catalyst under indoor daylight conditions (>400 nm). In chamber experiments it was shown that the method works well for nitrogen dioxide (NO_2) under dynamic conditions and for formaldehyde under static conditions. When the chamber was spiked with VOCs under dynamic conditions, no significant effect could be observed [80]. The reliability of photocatalysis for purification of indoor air has to be studied in more detail, because oxidation products with negative impact on human health might be formed by typical indoor chemistry [80–82]. Salthammer and Fuhrmann found a variety of undesired reaction products when exposing a wall paint, which was designed for the catalytic removal of indoor air pollutants, with artificial daylight [80] (see Fig. 6).

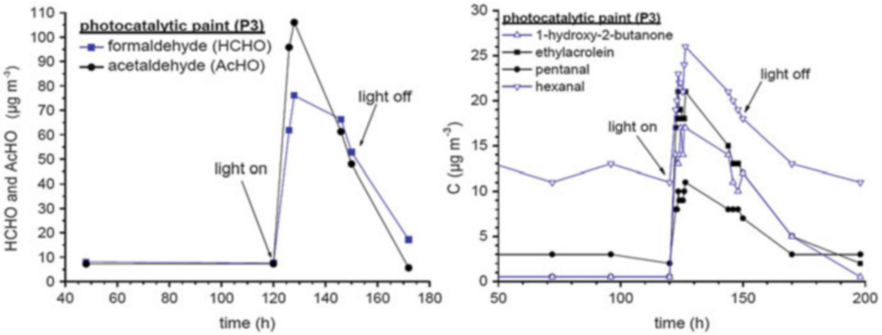
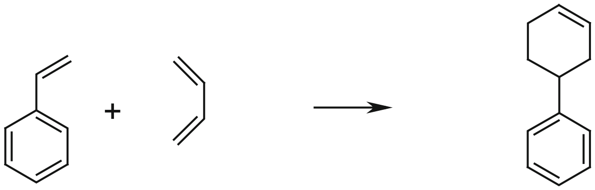


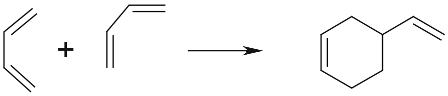
Fig. 6 Emission of secondary compounds from a photocatalytic wall paint in a 1 m^3 test chamber in dependence of irradiation [80]

3.5 Production Conditions as Source of Emissions

If the production of a material utilizes extreme conditions (heat, moisture, chemicals), degradation processes can be started in the material. An example already mentioned is the thermal treatment of softwood strands during production of oriented strand boards (OSB). In the following, some more case studies will be presented.

Very remarkable substances can be formed during the production of styrene–butadiene copolymers (SBR), which are used as foam backs for carpets. The reaction is a radical-induced polymerization. The residual monomers styrene and butadiene are removed by distillation, but the odor-intensive compounds 4-phenylcyclohexene (4-PCH) from styrene and *cis*-butadiene (see Eq. (10)) and 4-vinylcyclohexene (4-VCH) from *cis*- and *trans*-butadiene (see Eq. (11)) can be formed from the remaining monomers under the conditions of a thermally permitted $_{\pi}2_s + {}_{\pi}4_s$ Diels–Alder cyclic addition. 4-PCH was mainly responsible for the "new carpet smell" [29] and can be detected by the human nose in concentrations of only few µg/m^3.

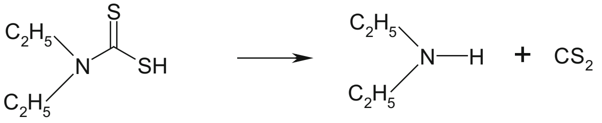

(10)


(11)

Another type of a $_{\pi}2_s + {}_{\pi}4_s$ Diels–Alder reaction in a building product is the dimerization of 2-chloro-1,3-butadiene, which is used for the production of synthetic rubber. Due to the substituent in 2-position, the two different isomers 1-chloro-4-

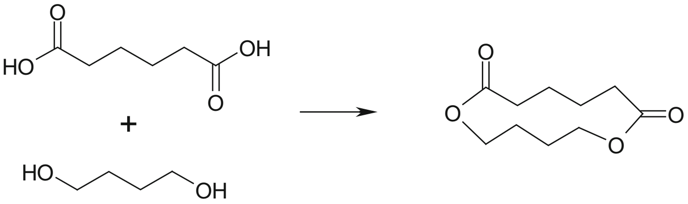
(1-chlorovinyl)-cyclohexene and 1-chloro-5-(1-chlorovinyl)-cyclohexene can be identified [59].

Zn-diethyldithiocarbamate (ZDEC) is used as a vulcanization accelerator for the production of styrene–butadiene rubber (SBR). During polymerization ZDEC decomposes to carbon disulfide (CS_2) and diethylamine (see Eq. (12)). From diethylamine the carcinogenic compound *N*-nitrosodiethylamine can be formed from the reaction with NO_x.



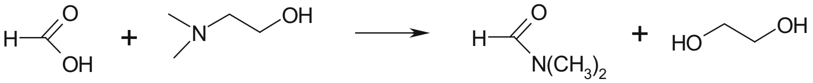
(12)

When two or more different kinds of material are fixed together, a variety of in-material reactions leading to VOC emissions are possible. The hydrolysis of esters fixed on wet concrete has already been discussed. In some cases such interspecies reaction may produce "exotic" compounds. A good example is the appearance of 1,6-dioxa-cyclododecane-7,12-dione, which emitted from a mouse pad. Adipinic acid was a component of the decorative layer, 1,4-butanediol was an ingredient of the adhesive, and the carrier was made of PU foam. The reaction is a simple cyclization under elimination of water (see Eq. (13)).



(13)

During the chamber test of a "green" paint, the formation of dimethylformamide (DMF) from dimethylaminoethanol and formic acid was observed (for suggested reaction, see Eq. 14). While dimethylaminoethanol (and probably traces of dimethylamine) were intentionally present in the paint, the formic acid (together with other carboxylic acids) was generated during the curing process of the lacquer film. Several micrograms of DMF per cubic meter air could be detected after 24 h of drying/curing in the chamber. The appearance of DMF may cause problems in case of product ranking or labeling because of its classification as a toxic compound.



(14)

4 Semi-volatile Organic Compounds and Their Degradation Products

The physical properties of polymer materials are commonly set by the use of additives. In this context, semi-volatile organic compounds (SVOCs), being widely used as plasticizers, film formers, stabilizers, and flame retardants, are of special relevance. Usually, these substances are not chemically bound to the polymer matrix and thus migrate within the material. Due to the slowness of the migration process, SVOC emission into indoor air from polymeric products may take place over months to years [87, 124]. Within the plasticizer group, esters of phthalic acid (phthalates) are still dominating the market. Di(2-ethylhexyl) phthalate (DEHP) is usually put at the focus of environmental consideration due to its large production volume in the past. However, some phthalates are reputed to have negative health impacts, such as on children's development [125] and children's respiratory diseases [126]. Although there is still some controversy about the health effects associated with DEHP [127, 128], a shift from low to high molecular phthalates [3] or even to other compound classes (adipates, succinates, glutarates, terephthalates, trimelliates, citrates) has taken place over the last decade. Some of these so-called "alternative" plasticizers are used on account of the lower risk they present to human health. One example is diisononyl cyclohexane-1,2-dicarboxylate (DINCH), which is derived from diisononyl phthalate (DINP) via catalytic hydration. Since 2002 this plasticizer has been widely used in sensitive application fields such as medical articles, toys, and food packaging [129].

As Weschler and Nazaroff [130] pointed out, there is a continual growth of interest in the indoor occurrence of SVOCs, but the severity of the analytical challenges involved has impeded progress in studying them. The use of models to estimate indoor concentration development is thus becoming more relevant. These models need compound-specific parameters such as vapor pressure (p_0) and the octanol–air distribution coefficient (K_{OA}) to be able to predict emission rates [131, 132] and their interaction with indoor sinks [9]. The availability of reliable measurements of these parameters is limited for many SVOCs. Even for well-researched phthalates, vapor pressure ranges of several magnitudes are found in the literature [8].

In recent years there have been few emission studies with low-volatile plasticizers as target compounds [133]. The experiments by Uhde et al. [103] were performed with PVC wall coverings in 1 m^3 test chambers of 14 days. Maximum concentrations of 5.10 $\mu g/m^3$ (DnBP), 0.50 $\mu g/m^3$ (DIBP), 2.08 $\mu g/m^3$ (DPP), and 0.94 $\mu g/m^3$ (DEHP) were reported. Similar results were obtained by Afshari et al. [134], Katsumata et al. [135], and Schripp et al. [136] for DnBP and DEHP emission from different polymeric materials or from the pure liquid. A FLEC study carried out by Clausen et al. [87, 88] showed that the emission rate of DEHP from PVC flooring material remains constant for more than 1 year.

Previous investigations with PVC have shown that more than 60 days were needed for the DEHP equilibrium concentration to be reached in the FLEC.

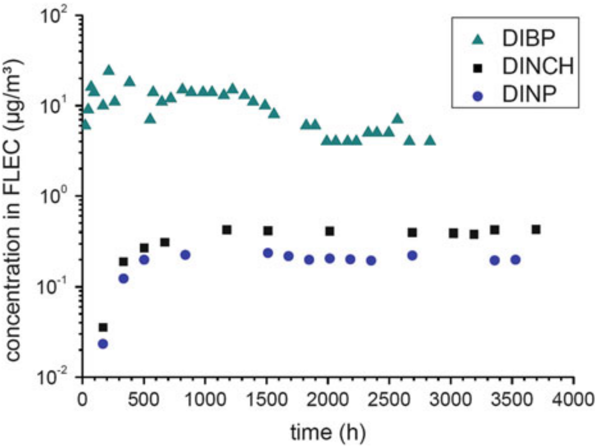


Fig. 7 Time vs. concentration curves for DINP (source PVC), DINCH (source PVC), and DIBP (source paint) in the FLEC at $T = 23°C$, 50% r. h., $n = 514\ h^{-1}$, and A/V $FLEC = 505\ m^2/m^3$ [8]

The setup of a FLEC study by Schossler et al. [8] with PVC was therefore similar to the experimental conditions as described by Clausen et al. [87]. The time it took for DINP and DINCH to reach the steady-state concentration is in the same order of magnitude or even faster (see Fig. 7), although both compounds are less volatile compared to DEHP. However, the steady-state concentrations were considerably lower with 0.22 µg/m^3 for DINP and 0.42 µg/m^3 for DINCH.

Results for such low-volatile plasticizers are helpful in estimating indoor air concentrations and the gas/particle interaction of DINP, DINCH, and other chemicals with similar properties. Taking into account the fact that for DEHP all studies come more or less to the same conclusion that the concentration in indoor air is below 1 µg/m^3 and comparing the physical properties of DEHP with DINP and DINCH, it can be stated that gas-phase concentrations higher than 0.5 µg/m^3 are not expected for these two compounds. The concentration of DIBP in the FLEC over 3,000 h was in the order of 4–24 µg/m^3 [8]. This result is in accordance with previous investigations for DnBP, an isomer of similar volatility [136].

5 Indoor Particles

With the growing importance of nanotechnology and its possible effects on the environment, the public interest in possible exposure to airborne ultrafine particles in the indoor environment has also increased [111, 137, 138]. It has become clear, due to the simultaneous trend towards using semi-volatile organic compounds (SVOCs) in products [3, 139], that it is not only the gas phase which has to be considered in the indoor environment but also the fact that complex processes of formation, adsorption, desorption, and reaction are taking place between particles, molecules, and surfaces [9]. The relevance of the particle-creating potential of terpene/ozone to the quality of indoor air has been documented [115, 140]. Studies have also been carried out into the effects of human activities on particle formation [120, 141–143] and into the use of nanotechnological products [118, 119]. The work of He et al. [144] revealed that many electrically operated devices in the indoor environment, such as printers [96–98, 145, 146] or household appliances [99, 112], can be significant emission sources for ultrafine particles.

For the investigation of hardcopy devices in test chambers, the international standard ECMA-328 has been developed. In a recent study, which considered 27 different laser printers, it was found that the investigated devices represented to very different extents a source of particles for the indoor environment. The calculated total number *N* [96] of emitted particles (5.6–560 nm) measured by an FMPS during a 10-minute period of printing ranged from $3{\cdot}10^{10}$ particles per print to $4{\cdot}10^{12}$ particles per print [99]. For all devices investigated, the vast majority of particles were smaller than 100 nm, with an individual time-resolved size-class distribution being found for each printer. The upper size limit of the particles detected with the FMPS was approx. 250 nm.

An assessment of the relevance to health required information not only about size distribution but also about the chemical nature of the particles released during printing. In the case of the thermodenuder investigations with a group of 19 desktop printers, it was noted that the particles detected by the FMPS completely vaporized above 400°C. Morawska et al. [98] observed that printer particles have a hydrophobic character. These findings clearly suggest that the particles originate from organic chemical components.

A variety of SVOCs could be detected in the test chamber air during printing when the 27 devices were investigated. Basically, during qualitative analysis, polysiloxanes and homologous series of long-chain alkanes in the range from C21 to C45 (with waxlike properties) were detected. The polysiloxanes were identified as homologous series of open-chain and cyclic compounds, composed of $(Me)_2SiO$ units which extended over a broad volatility range. Probably, the polysiloxanes result from technical mixtures of silicone oils or silicon grease which are used as heat-resistant lubricants. With some printers bis(2-ethylhexyl) adipate and bis(2-ethylhexyl) decanedioate as well as some other not precisely identified long-chain carboxylic acid esters were found.

Household appliances also exhibit a high degree of variability in particle emission intensity. A recent study reports on the measurement of 12 household appliances such as toasters, grills, and hair dryers in an emission test chamber regarding the release of particles between 5.6 and 560 nm [112]. The devices were new at the time experiments started and had never been used for their original purpose. For instance, toasters and sandwich makers were tested without the presence of food or residues from prior usage. During the experiments the devices released aerosols with count median diameters mainly below 100 nm (see Fig. 8). Within the operating phase, high quantities of 10 nm particles are released which form larger particles by agglomeration. The origin of the particles can be attributed to the heated surfaces, but cleaning these surfaces only had a minor influence on the emission strength. The released particles are evaporated between 150°C and 200°C.

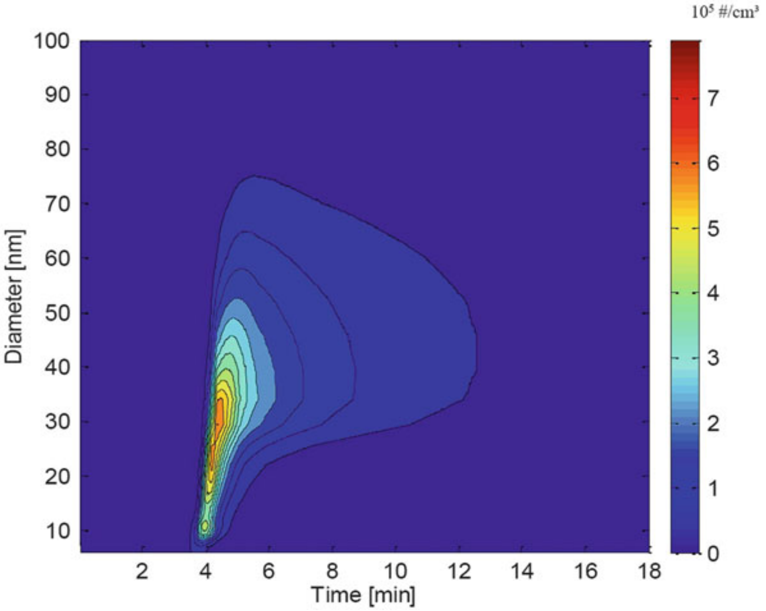


Fig. 8 Particle concentration and size distribution of a flat iron in a 1 m^3 stainless-steel chamber at $T = 23°C$, 50% r.h., $n = 3\ h^{-1}$, and an operating time of 1 min [112]

These findings indicate the particles to be formed from semi-volatile organic compounds. However, the compounds are not located on the heated surfaces and are not released as a supersaturated vapor because emission is continuous over the operating phase of the device. The contribution of oxygen to the formation process could be neglected because the emission was also detected in a nitrogen atmosphere.

6 Conclusion

Scientific research and field studies in the field of indoor air contamination were initially to a great extent concerned with solvent components and those volatile organic compounds which were also to be found in outdoor air. It was however realized at an early stage that even those reactive substances which had not yet been registered make a contribution to contamination of indoor air, as do their reaction products and further organic compounds. Alongside VOCs and their chemical reactions, current research interests focus on semi-volatile organic compounds, on chemical reactions indoors, and on the generation of ultrafine particulate matter from the reaction of unsaturated hydrocarbons with ozone.

With new substance groups and new findings, the procedures used in registering and evaluating indoor pollutants have also experienced major changes over the years. Today, materials can be tested beforehand in test chambers to determine their emission potential as regards air-polluting substances. Before the measured data can be evaluated, experts will need to work out recommendations and also guideline and reference values.

Through the results of a large number of interdisciplinary research activities, it could be demonstrated that in making a reasonable assessment of the indoor situation, it will not in most cases suffice to consider the concentrations of indoor contaminants in isolation. Instead it is now generally recognized that the quality of the indoor air and also above all its perception by room users is actually a complex interplay of many influencing factors.

With the creation of reliable measurement and testing methods and the further development of guideline values for the indoor air and quality criteria for products used indoors, conditions are now satisfying not only for the health-related evaluations of a particular indoor situation. The development of products with lower emissions and of modern living-space concepts also includes preventive aspects. Today many problems relating to indoor rooms have still not been solved. However, numerous activities in the fields of science, industry, and politics are contributing to a continuous improvement in indoor air quality.

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Indoor Particles, Combustion Products and Fibres

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Abstract Pollutants in the indoor environment are a complex mixture of gases, vapours and particles in either liquid or solid phase, suspended in the air, settled or adsorbed on or attached to indoor surfaces. The pollutants originate from a multiplicity of indoor and outdoor sources. The pollutant mixture is dynamic, involved in numerous physical and chemical processes and changing its characteristics with time. Its composition and concentration depend on the strengths of indoor sources, pollutants' concentration outside and the properties of heating ventilation and air conditioning systems. The spatial distribution of pollutant concentration within the indoor environment is often inhomogeneous.

Particulate matter in the indoor environment includes particles, which are airborne as well as those which are settled on indoor surfaces, dust. The particles vary in chemical properties, which depend on the origin of the particles and differ for particles in different size ranges. The particles can, for example, be combustion or nucleation products, dust or bioaerosols and can act as carriers of adsorbed chemicals, bio-contaminants or condensed gases. Particles are a key component of emissions from all the combustion sources. In particular, a significant indoor combustion product, environmental tobacco smoke (ETS) is a mixture of particle and gaseous products of smoke exhaled to the air by smokers and mixed with the smoke resulting from smouldering of a cigarette between the puffs.

This chapter is focused on particulate matter, its origin, characteristics and behaviour in the indoor environment. In addition, several important classes of indoor pollutants are discussed, those which are entirely or partially composed of particulate matter. These include ETS and combustion products from other sources, such as wood smoke or vehicle emissions, and also fibres, in particular, asbestos.

Keywords Dust, ETS, Fibres, Indoor air pollution, Indoor particles

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Contents

1 Origins of Indoor Airborne Particles

A large number of sources contribute to the ambient airborne particulate matter and include motor vehicles, power plants, windblown dust, photochemical processes, cigarette smoking, nearby quarry operation, etc. Particles encountered in indoor air can be generated from either indoor or outdoor sources. The indoor sources of particles include:

- Occupants (which are humans and pets contributing to air pollution through functions natural to life processes or through activities conducted)
- Soil and water
- Cooking
- Tobacco combustion
- Combustion appliances
- Building materials
- Furnishings
- Consumer products
- Maintenance products
- Cleaning activities
- Photocopiers and laser printers

There is a significant variation between particles generated not only by different sources but even by the same type of source. The most significant of indoor residential sources are smoking, cooking, cleaning and occupant movement.

Some of the particles present in the air are primary and some secondary in nature. A primary particle is a particle present in the air in the form in which it was generated by a source, such as smoking, while a secondary particle is formed in the air by gas-to-particle conversion, such as secondary organic aerosols. An important characteristic of airborne particles is their size distribution. The size of particles strongly affects particle behaviour and fate in atmospheric systems as well as deposition in the human respiratory tract. Size distribution is also the main factor in choosing the instrumentation to be used for particle detection. Particles in supermicrometre size ranges (larger than 1 μm) are usually primary in nature and result mainly from mechanical processes such as cleaning and physical activity of the occupants' indoors. Particles in submicrometre ranges are generated mainly from combustion processes as well as from secondary processes such as gas-to-particle conversion and nucleation or photochemical processes. In the indoor domestic environment, the main sources of submicrometre particles include smoking, cooking (particularly frying and broiling) and operation of gas burners, gas ovens and electric toasters [1], while in an office, printing and cleaning [2, 3].

A source signature or fingerprint of a source consists of the physical and/or chemical characteristics of the emissions, which are specific and at best unique for that source. Availability of such signatures is very important for the determination of absolute and fractional contributions from specific sources (source apportionment) and, in turn, for developing of emission inventories. Quantitative source apportionment is a very complex undertaking as on one hand, ambient air contains a dynamic mixture of pollutants emitted from various sources. This mixture undergoes continuous change in time as the interactions between pollutants take place and as the components of the mixture are removed from the air due to the presence of various sinks. On the other hand, it is only rarely that specific emission characteristics are unique to a particular source. More often, emissions from other sources display some of these characteristics as well. The following are used as source signatures:

- Physical aspects of particles (number or mass size distribution, density and shape, electric charge and optical scattering)
- Chemical aspects of signatures: elemental composition, elemental ratios, characterisation of chemical form, isotope ratios and organic compounds

In addition to the source signatures, certain elements or compounds have been used as markers of emissions from specific sources [4]. A suitable marker should be (1) unique or nearly unique to the emissions from the source under consideration, (2) similar in emission rates for a variety of the same type of fuels, (3) easily detected in air at low concentrations and (4) present in consistent proportion to compounds that have effects on human health. Additionally, an ideal marker should also be easily (in real time), accurately and cost-effectively measurable.

Quantitative measures of emissions are emission factors or emission rates. A source emission factor is typically defined as the amount of a chemical species, mass, particle number, etc., emitted per unit mass of fuel burned or per a defined task performed [5]. The former is often referred to as mass-based emission factor and has a unit such as g kg^{-1}. The latter can be called task-based emission factor. The unit of task-based emission factor depends on the definition of tasks. For example, a task can be the number of cigarettes smoked or a certain distance driven by a motor vehicle and thus the units may be g $\mathrm{cigarette}^{-1}$ or g km^{-1}, respectively. Emission rate on the other hand is the amount of a chemical species, mass, particle number, etc., emitted by the source per unit time. For example, emissions from stoves are usually characterised in terms of emission factors. Similarly, re-entrainment of settled dust to the air is represented by resuspension rates. Emission factors and emission rates vary significantly not only between different types of sources but also between sources of the same type.

Certain sources such as, for example, cooking or smoking, or activities such as movement are always associated with relatively high emission factors or rates and thus always contribute in a measurable way to indoor concentration levels of particles [6]. For other sources or activities, however, such as, for example, cleaning or vacuuming, conflicting results have been reported in the literature. While a number of studies, for example, found cleaning to contribute to increase of particle concentration, the PTEAM study determined the contribution from cleaning to be not statistically significant [7]. The reasons for the conflicting results relate on one hand to varying emission rates (of up to four orders of magnitudes) [8] from these sources, which are sometimes very low, and also to the degree of dilution of particles introduced from these sources to indoor environments, which depend on air exchange rate. The combination of low emission factors or rates and high exchange rates can result in an insignificant contribution from certain sources in certain environments. It is important to keep in mind that the choice of measuring method is also a factor of significance: if an inappropriate method were chosen for detection of the particles emitted, the conclusions about emissions from the sources would be erroneous.

The concentration of particles as well as of other pollutants in the indoor environment depends on a number of factors:

- Type, nature and number of sources
- Source used characteristics
- Building characteristics
- Outdoor concentration of pollutants
- Infiltration or ventilation rates
- Air mixing
- Removal rates by surfaces, chemical transformation or radioactive decay
- Existence and effectiveness of air contaminant removal systems
- Meteorological conditions

The role of the outdoor air on the indoor particle characteristics cannot be overestimated. In the absence of active indoor sources, particles generated by outdoor sources, which penetrated indoors, are the main constituent of indoor particles. In a typical outdoor urban environment, motor vehicle emissions constitute the most important source of all pollutants including particles. The emissions from motor vehicles penetrate indoors and their concentration for natural ventilation buildings in indoor air is often comparable to the concentration outdoors.

The relative importance of indoor and outdoor sources depends on the environment, building characteristics and lifestyle of the occupants. For example, there will be little contribution from indoor combustion sources in an indoor environment, which does not require heating, where cooking is conducted using electric stoves and where there are no smokers. On the other hand, the environments with operating open fires or where cigarettes are smoked inside could have concentrations, which are orders of magnitude higher than in the outdoor environment. Open-fire burning presents a particularly severe problem in developing countries, where, in many places, it is the most affordable or the only available way of cooking or heating [9].

2 Physical, Chemical and Biological Properties of Particles

Airborne particles can be classified and characterised in a number of ways and, for example, according to their physical, chemical or biological properties. Also, many different terms are used in relation to airborne particles. Some of them identify particles by their sizes; others by the processes, which led to their generation; and some by the particles' ability to enter the human respiratory tract [10, 11]. In particular, an *aerosol* is an assembly of liquid or solid particles suspended in a gaseous medium long enough to enable observation or measurement, and a *particle* or *particulate* is a small, discrete object.

The most important physical properties of aerosol particles include number and number size distribution, mass and mass size distribution, surface area, shape and electric charge. To a larger extent, these are the physical properties of particles, which underlie particle behaviour in the air and ultimately removal from the atmospheric systems. Of particular importance is the size of the particles. Efficiency of various forces acting on particles and processes to which they are subjected in the air depends strongly on particle physical properties, of which size is one of the most important. Health and environmental effects of particles are strongly linked to particle size, as it is the size, which is a predictor of the region in the lung where the particles would deposit or the outdoor and indoor locations, to which the particles can penetrate or be transported. Also, sampling of particles and choice of an appropriate instrumentation and methodology are primarily based on particle physical properties.

2.1 Particle Size and Its Relation to Formation Mechanisms

Various classifications and terminologies have been used to define particle size ranges. The division most commonly used is between *fine* and *coarse particles*, with the boundary between these two fractions widely accepted as 2.5 μm. However, this division has been defined differently by different authors, in relation to different aerosols and for different applications, and ranges from 1 to 2.5 μm. The division line often used in aerosol science and technology is somewhere between 1 and 2 μm [10–12]. The rationale behind this is that this is the range of a natural division between smaller particles, which are generated mainly from combustion as well as process leading to gas-to-particle conversion, and larger particles, which are generated from mechanical processes.

Ultrafine particles have been defined as those, which are smaller than 0.1 μm. Another classification is into *submicrometre* particles, which are smaller than 1 μm, and *supermicrometre* particles, which are larger than 1 μm. The terminology that has been used in the wording of the ambient air quality standards and also for characterisation of indoor and outdoor particle mass concentrations includes $PM_{2.5}$ and PM_{10} fractions and the *total suspended particulate* (*TSP*). $PM_{2.5}$ (fine particles) and PM_{10} are the mass concentrations of particles with aerodynamic diameters smaller than 2.5 and 10 μm, respectively (more precisely, the definitions specify the inlet cut-offs for which 50% efficiency is obtained for these sizes). TSP is mass concentration of all particles suspended in the air. There have been references made in the literature to PM_1 or $PM_{0.1}$ fractions, which imply mass concentrations of particles smaller than 1 and 0.1 μm, respectively. These terms should be used with caution, as particles below 1 μm, and even more those below 0.1 μm, are almost always measured in terms of their number rather than their mass concentrations, and therefore, these terms could be misleading.

It should be kept in mind that the divisions between the different particle-size classes are somewhat arbitrary. On the one hand, there are no natural boundaries between these size classes as nature itself does not provide a perfect division. On the other hand, all natural sources (versus laboratory sources) produce particles within a certain range of diameters (polydisperse particles). Therefore, there is no sharp boundary delineating the contribution of particles from a given particle source. This argument also extends to effects produced by the particles. For example, it cannot be expected that there will be much difference between 0.09 and 0.11 μm particles in terms of their composition and behaviour in atmospheric systems, nor in their penetration into the lung or the health effects they cause, despite the second particle being outside the defined ultrafine range.

While the above classification of particles considers only their sizes, as already discussed, particle size is a consequence of the process, which led to its generation, and thus is also dependant on the source. Submicrometre particles are generated mainly from combustion, gas-to-particle conversion, nucleation or photochemical processes, while larger particles result mainly from mechanical processes such as cutting, grinding, breaking and wear of material or dust resuspension. Particles in the submicrometre size range typically contain a mixture of components including soot, acid condensates, sulphates and nitrates, as well as trace metals and other toxins. Coarse particles contain largely earth crustal elements and compounds.

Particle-size distribution can also be described in terms of modes, which correspond to peaks within the distribution. This classification relates to particle formation mechanisms; however, it also implies particle-size ranges. The location of the modes is variable, depending on the specific sources and other local atmospheric conditions. Particles can be classified into the following modes:

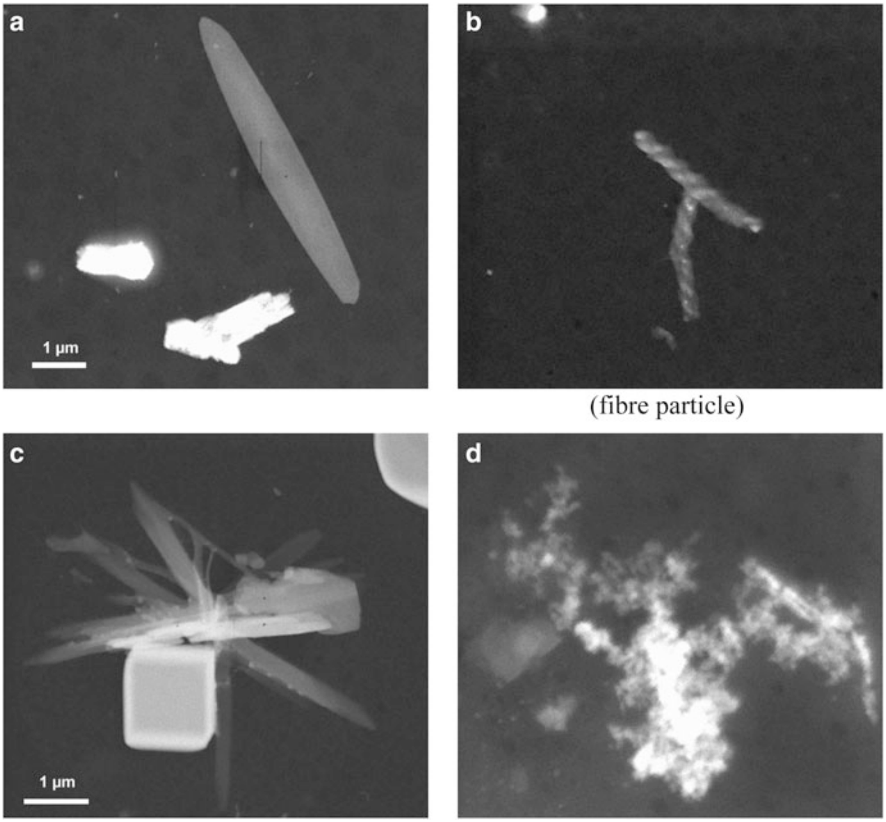
- *Nuclei mode*: particles in this mode are formed by nucleation of atmospheric gases in a supersaturated atmosphere and their size is of the order of nanometres.
- *Accumulation mode*: particles in this mode originate from primary emissions as well as through gas-to-particle conversion, chemical reactions, condensation and coagulation.
- *Coarse mode*: particles generated by mechanical processes.

2.2 Particle Shape and Equivalent Diameter

Particles vary significantly in shape, which in general relates to the particle formation process or their origin. Some particle shapes are fairly simple and regular; however, the majority is of a varying degree of irregularity or complexity. Particles resulting from coagulation and agglomeration of smaller solid particles are usually highly irregular and display fractal properties. Examples of these are particles resulting from combustion process such as vehicle emissions or tobacco smoking, which are agglomerates of carbonaceous particles. Similarly, dust particles and particles resulting from mechanical breaking, grinding, etc., have generally irregular shapes. By contrast, liquid aerosol particles are usually spherical, while simple fibres are rod shaped. Biological particles are of complex shapes and differ significantly between each other. Figure 1 presents microscopic images of particles of various shapes collected inside and in the vicinity of residential houses in Brisbane, Australia.

For practical applications, particles of complex or irregular shapes are usually characterised by only one or two parameters, those which can be measured. These are most commonly particle diameter and for fibres, their lengths and width. Diameter is a characteristic of spherical objects; however, as explained above, only a small fraction of airborne particles are spherical. A means of representation of particle irregular shapes is by particle equivalent diameter, which is the diameter of a sphere having the same value of a physical property as the irregularly or complex-shaped particle being measured. Equivalent diameter relates to particle behaviour (such as inertia, electric or magnetic mobility, light scattering, radioactivity or Brownian motion) or to particle properties (such as chemical or elemental composition, cross-sectional area, volume-to-surface ratio). Therefore, particle diameter determined experimentally depends on choice of particle properties or behaviour investigated, and thus, application of different methods for measurements of particle diameter usually results in somewhat different values of the diameter reported.

The most commonly used equivalent diameters are aerodynamic (mainly for particles larger than 0.5 μm), diffusion (for particles smaller than 0.5 μm), light scattering (for various ranges from about 0.1 μm and larger) and mass equivalent (mainly for larger particles). The aerodynamic (equivalent) diameter D_a is the



(fibre particle)

Fig. 1 Particles collected in and outside residential houses in Brisbane, Australia, and examined with an energy-dispersive X-ray analyser attached to a transmission electron microscope. (**a**) There are three big particles in this picture, two lighter particles with dominant elements: S, Ca, O and Mg and one darker particle with dominant elements, Ca, S, Na, O and Mg. (**b**) There are two big particles in this picture with dominant elements: C, Cl, Na, Mg, K, O. The particles are likely fine pieces of insect body or plant material. (**c**) There are two types of particles in this picture, a square particle (NaCl crystal) and many big fibrous particles with dominant elements: Ca, S, Na, O and Mg. D: there are many particles jointed together in this picture with dominant elements: Fe, Pb, Si. The particles are likely from vehicle emissions

diameter of a unit-density sphere having the same gravitational settling velocity as the particle being measured. D_a is given by Eq. (1),

$$D_\mathrm{a} = D_\mathrm{P} k \sqrt{\frac{\rho_\mathrm{P}}{\rho_0}} \tag{1}$$

where D_P is the physical particle diameter, ρ_P is the density of the particle, ρ_0 is the reference density (1 g cm^{-3}) and k is a correction factor, which can be approximated by $k \approx 1$ for many applications.

An important feature of many types of particles, particularly those of complex shapes resulting from combustion processes, is their fractal structure. For example, Schmidt-Ott [13] measured fractal dimensions of ultrafine particles resulting from gas-to-particle conversion and showed that such particles have very large surface areas. Thus, large quantities of chemicals can be adsorbed by such particles, and in such cases knowledge of the aerodynamic diameter alone provides no information on the possible amount of adsorbed compounds. Understanding of fractal objects and their links and relationship to various processes in nature can help in understanding the structure and growth of particles [14, 15].

2.3 Particle Sizes and Size Distribution

The smallest and the largest airborne particles can differ in size by up to five orders of magnitude and range from about 1 nm to about 100 μm. The former is molecular size and the later is the size above which particles sediment rapidly due to gravitational force. Almost all of the sources generate particles with some distribution of sizes, so-called polydisperse aerosol, rather than particles of a single-size, monodisperse aerosol. The spread of particle-size distribution is characterised by an arithmetic or geometric (logarithmic) standard deviation. The most common ways of characterisation of a particle distribution are in terms of its mean size, which is average of all sizes; median size, which means equal number of particles above; and below this size or mode size, which is the size with the maximum number of particles. The terms used include count, number or mass median diameter, which are abbreviated as CMD, NMD or MMD, respectively. MMAD is mass median aerodynamic diameter.

The size distribution of particles generated by most sources is lognormal, which means that the particle concentration versus particle-size curve is “normal” (bell shaped) when the particles are plotted on a logarithmic scale. The width of the peak in the distribution is characterised by geometric standard deviation. Different emission sources are characterised by different size distributions. These distributions are not unique to these particle sources alone; however, the information on the size distribution can help to identify source contribution to particle concentrations in ambient air and also serve as a source signature. In general, submicrometre and supermicrometre particles result from different generation processes and only occasionally the same source generates particles with broad-size distributions, covering both fine and coarse ranges.

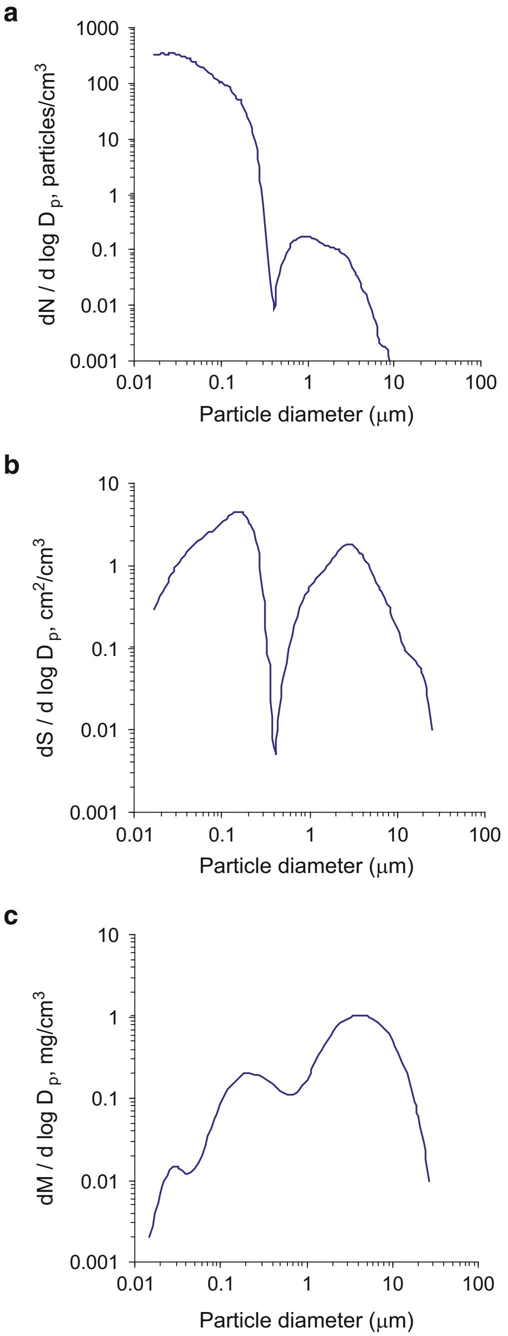
Numerous researchers have presented information about size distribution of particles originated from various sources. Wallace [16] reported that at the peak of number concentrations, the diameters of particles produced by several sources are in the range of gas burners, gas oven and toaster in range of 0.01–0.02 μm; incense 0.1 μm; frying and broiling from 0.05 to 0.1 μm; and walking, moving one’s arms and even sitting in front of a computer produce particles of 5–10 μm. Abt et al. [17] showed that cooking (including broiling and baking, toasting and barbecuing) produced particles with volume diameter in a size range between 0.13 and 0.25 μm and cleaning and smoking particles smaller than 1 μm, while moving of people and sautéing resulted in particles in the range between 3 and 4.3 μm. Frying was associated with both fine and coarse particles. Measurements conducted in an apartment in Taipei by Li et al. [18, 19] showed that a number median diameters of particles originating from the background, smouldering cigarettes, burning mosquito coils and joss sticks were found to be 0.07, 0.085, 0.08 and 0.07 μm, respectively. In a study conducted by Kleeman et al. [20], natural gas, propane and candle flames generated particles between 0.01 and 0.1 μm and meat charbroiling showed a major peak in the particle mass distribution at 0.1–0.2 μm with some material present at larger particle size but not above 1.0 μm. Recent studies (He et al. [2], Knibbs et al. [8]) show laser printer-generated particles with the CMD between 0.04 and 0.076 μm and vacuum cleaner generated particles with the CMD between 0.015 and 0.152 μm, respectively. Biological particles are another class, which include many different species, varying not only in their biological composition but also size. In general, viruses range from 0.02 to 0.3 μm, bacteria from 0.5 to 10 μm, fungi from 0.5 to 30 μm, pollen from 10 to 100 μm and house dust mites about 10 μm.

Particles in ambient air constitute a mixture originating from different sources, and thus, the individual components of the mixture are characterised by different size distributions. The measured distribution of this mixture may or may not display individual peaks from the contributing sources and thus may or may not be used for source identification. In many cases, however, the characteristics of size distribution can be a useful tool in source characterisation.

Concentration of particles in the air as well as particle-size distributions can be considered in terms of either particle number or mass. In terms of number, the vast majority of airborne particles are in the ultrafine range. For example, in urban outdoor air where motor vehicle emissions are a dominant pollution source, over 80% or more of particulate matter in terms of number is in the ultrafine range [21]. Since outdoor particles significantly contribute to indoor particle concentrations, also in the indoor air, particle number concentration is usually dominated by the smallest particles. However, most of the mass of airborne particles is associated with large particles since the mass of ultrafine particles is often very small in comparison with the mass of the larger particles. Particle surface area in turn is largest for particles somewhat above the ultrafine size range.

The relationship between particle number, surface area and volume is presented in Fig. 2a–c, respectively, on an example of a typical urban air particle-size distribution measured in Brisbane, Australia [22]. This relationship was derived using measured particle number size distribution and calculating particle surface and volume distribution (assuming their sphericity) and by plotting $dN/d\log D_\mathrm{P}$, $dA/d\log D_\mathrm{P}$ and $dV/d\log D_\mathrm{P}$, which represent particle number, surface are and volume, respectively, per logarithmic interval of size. Particle mass can be calculated from the volume when particle density is known or can be assumed. It can be seen from Fig. 2 that the peak in the number distribution spectrum appears in the area where

Fig. 2 Typical urban ambient air particle number size distribution measured in Brisbane, Australia, and calculated from the number distribution (**a**), surface (**b**) and volume size distributions (**c**), respectively



there is almost no volume in the volume distribution spectrum and vice versa; the peak in the volume distribution spectrum is where the particle number is very low.

Since different sources contribute to generation of particles in the submicrometre range, which is predominant in particle number, and different sources to larger particles, which predominate in mass, it is only occasionally that there is a correlation between particle number and mass. In general, however, only limited information or no information at all can be obtained about particle number from the measurements of particle mass and vice versa. The degree of correlation depends on specific local conditions, and in particular, better correlations are achieved for conditions when the majority of particles in the fine and coarse size ranges are related to the same generation process.

Comparison between different particle mass fractions, for example, $PM_{2.5}$ and PM_{10}, reveals that there is often a relatively high degree of correlation between the fractions. One of the main reasons for this is that $PM_{2.5}$ is a fraction of PM_{10}, often quite a significant, and for sources operating in a stable manner, the relation between $PM_{2.5}$ and PM_{10} emissions remains more or less constant. Outdoor ratios of particle mass fractions could be indicators of these ratios in the indoor environment where natural ventilation is used and in the absence of indoor sources. In the presence of operating indoor sources, however, the indoor and outdoor ratios could differ substantially. For example, Chao et al. [23] reported that the outdoor PM_{10} in Hong Kong was around 50–70% of the TSP level; however, indoor ratios of PM_{10} to TSP measured in eight residential premises varied from 81.9 to 97.7%.

2.4 Chemical Composition of Airborne Particles

The chemical composition of indoor particles is very complex and depends on particle origin as well as any post-formation processes, in which the particle is involved. Some types of particles like asbestos and glass fibres consist of inorganic materials, while other types like cellulose fibres are purely organic. The most important chemical properties of particles include:

- Elemental composition
- Inorganic ions
- Carbonaceous compounds (organic and elemental carbon)

Interest in different aspects of particle chemical compositions is driven either by the risk associated with certain components or with a potential for application of the knowledge about the components towards some practical purposes. For example, interest in elemental composition derives from the potential health effects of heavy elements like lead, arsenic, mercury and cadmium and the possibility of using the elements as source tracers [6]. Water-soluble ions such as potassium, sodium, calcium, phosphates, sulphates, ammonium and nitrate associate themselves with liquid water in the indoor environments and can also be used for source apportionment. Carbonaceous compounds are composed of organic and elemental carbon.

The former can contain a wide range of compounds such as polycyclic aromatic hydrocarbons, pesticides, phthalates, flame retardants and carboxylic acids, some of which are tracers for certain sources, while the latter is sometimes termed “soot”, “black carbon” and “graphitic carbon”.

Due to their irregular shapes, a majority of particles present in the indoor environment have large surface areas, which provide an opportunity for particles to serve as sinks for a variety of organic species. Semi-volatile substances are found both in the particulate and vapour phases, volatile compounds occur mostly in the gas phase and substances with very low vapour pressure are adsorbed almost exclusively. In addition to the surface area of the particles, the vapour pressure of organic compounds also plays an important role in determining whether they will be found in the gas phase or adsorbed on particle matter. More information on particle chemistry is provided in the Sect. 4.3.

2.5 Bioaerosols

A certain fraction of particles in indoor and outdoor air is of biological origin. In addition to particles, some volatile organic compounds are also of microbial origin (MVOC). According to the definition formulated at the IGAP workshop in Geneva in June 1993: “Biological Aerosol Particles (BAP) describe airborne solid particles (dead or alive) that are or were derived from living organism, including micro-organisms and fragments of varieties of living things” [24]. The sources of biological particles can be classified as (a) animal or human, (b) terrestrial including rural (plants) and urban, (c) aquatic and (d) atmospheric. Biological particles in the indoor environment include viruses; bacteria, which, at cell destruction, release endotoxins; animal dander (cats, dogs, rabbits, rodents, birds), which contains allergens (e.g. cats – Fel d 1); cockroaches and other insects, which contain allergens; mites, which contain allergens (e.g. Der p1) and release faeces; and moulds (filamentous fungi), which release mould spores (which, in turn when damaged, break into spore fragments which include allergens and contain beta-glucans) and release primary metabolites (microbial volatile organic compounds, MVOC) and secondary metabolites (mycotoxins) and pollens [6].

Various units are used in relation to the concentration of biological particles in the air and in particular (1) for viable microorganisms, colony-forming units (CFU m^{-3}) for bacteria and fungi and plaque-forming units (PFU m^{-3}), for viruses, and (2) for nonviable microorganisms: number of individual microorganisms per m^{-3}, number of microorganism-containing particles per m^{-3} or μg m^{-3}.

The presence and distribution of biological particles vary significantly between different indoor environments. The particles can be suspended in the air, attached to indoor surfaces and to the dust accumulated in the building and also present in any internal parts of the building structure or its operating systems (inside walls, air conditioning units, ducts, etc.). The concentration levels of the particles can vary by orders of magnitude and depend on a range of local conditions and factors affecting their growth, survival transport and removal from the air. Most commonly, the concentration of biological particles is significantly lower than the concentration of nonbiological particles, with concentrations of the former being in the range of 10^1–10^4 m^{-3} and the latter on the range of 10^9–10^{11} m^{-3} and 10^6–10^7 m^{-3} for particles in submicrometre and supermicrometre ranges, respectively [6].

3 Transport and Behaviour

Airborne particles are subjected to a multiplicity of processes, interactions and reactions, which change the characteristics the particles initially had when introduced into the air. The most important processes include sedimentation, deposition on surfaces, coagulation, changes by evaporation or condensation. Some of the emission products undergo rapid changes, such as, for example, combustion related, which are reactive mixtures of hot gases and particles, while others, like mechanical dust, are less so. Particles generated indoors and measured some time after emission often have different characteristics to those measured immediately after formation. The residence time of particles in the air depends on the nature of the processes they are involved in and varies in indoor air from seconds to minutes or hours. Larger particles (of a micrometre size range in aerodynamic diameter and more) are removed from the air mainly through gravitational settling (with particles above 100 μm settling almost immediately after becoming airborne), while smaller particles are removed by precipitation or diffusional deposition.

The processes of the highest significance in affecting indoor particle concentration levels and other characteristics include:

- *Penetration of outdoor particles indoors through open doors and windows as well as through the building envelope*: A measure of the ability of the particles to penetrate the building envelope is defined as *penetration factor* [25] (sometimes also called *penetration coefficient*). In cases when windows or doors of a building are open, they provide the main penetration route for the particles, and the relative importance of penetration through the building envelope becomes insignificant. Under such circumstances, the penetration factor for particles in all size ranges is very close to unity, which means that particles enter buildings very easily with the air which carries them. Such situations are characterised by large air exchange rates. Studies conducted by Mosley et al. [26] and Long et al. [27] showed that penetration factor is generally lower than unity for situations when the particles travel a torturous path through a building envelope and also that it is strongly size dependant.
- *Deposition of particles on indoor surfaces*: Fundamentals of the theory of particle deposition are presented in [11] and describe the process of particle flux towards the surface due to the gradient in particle concentration established in the region of the surface. At the surface, the concentration is close to zero due to the deposition and increases at a distance away from the surface (e.g. wall)

to achieve equilibrium usually a few centimetres from the surface. *Deposition rate* is defined as the number of particles depositing per unit surface area per unit time ($m^{-2}\ s^{-1}$). Particle deposition on indoor surfaces strongly depends on particle size and is governed by the processes of particle diffusion towards the surfaces, which is of particular significance for very small particles, and of gravitational sedimentation, which is significant for larger particles. In addition, the presence of airflows induced by convection currents or action of fans, as well as air turbulence, can increase particle transport towards the surface and thus the deposition. Deposition is also dependant on the size of the surface area and on surface characteristics, with sticky surfaces resulting in higher and smoother in lower deposition. The larger the surface area, the higher the probability of particle deposition, and therefore, furnished rooms with lots of surface area will have a higher total deposition rate than bare rooms. Additional factors affecting particle deposition are the presence of surface charge, which increases the deposition rate; temperature gradient, which results in convective currents and thermophoretic deposition; and room volume. This multiplicity of factors affecting particle deposition results in deposition being a highly variable process, site-specific and difficult to quantify either through experimental studies or modelling. Many studies have shown that there is a considerable difficulty associated with decoupling and quantifying separately particle deposition and other parameters describing particle dynamics, such as penetration factor [27]. Studies conducted by He et al. [28] in 14 residential houses showed that deposition rates were particle-size dependant and they varied from house to house. The lowest deposition rates were found for particles in the size range from 0.2 to 0.3 μm for both minimum (air exchange rate: 0.61 ± 0.45 h) and normal (air exchange rate: 3.00 ± 1.23 h) ventilation conditions. The results of statistical analysis indicated that ventilation condition (measured in terms of air exchange rate) was an important factor affecting deposition rates for particles in the size range from 0.08 to 1.0 mm, but not for particles smaller than 0.08 mm or larger than 1.0 μm.

- *Resuspension of particles deposited on surfaces*: Particles that have deposited on indoor surfaces, settled dust, may be resuspended from the surfaces and re-entrained into the air. To resuspend the particles, certain force must be applied and energy used to detach the particles from the surface. According to the theory of aerosol interactions [10], while most adhesion forces are linearly dependant on particle diameter, most detachment forces are proportional to particle diameter to the second or third power. The differences between adhesion and detachment forces in their dependence on particle diameter result in large particles being more readily detached than small ones. The theory was confirmed in a number of field studies and, for example, Thatcher and Layton [29] concluded that submicrometre particles are essentially non-resuspendable under circumstances encountered in residences. The resuspension rate was shown to increase as particle size increases and also similarly to the several other studies showed that all the normal activities of the occupants, like walking (even walking in and out of the room), moving around, children playing or cleaning, result in an increase of supermicrometre particles.

- *Removal of particles from indoor environment by ventilation and filtration*: As a result of operation of natural or mechanical ventilation system, particles generated indoors are removed from indoor environments, but at the same time, particles from outdoors penetrate indoors. Filters remove a certain fraction of particles from the air supplied indoors. Ventilation and air filtration are the main remedial actions available to reduce concentration to airborne contaminants indoors. Of critical importance are the design and operation of the filtration and ventilation systems and the type of filters used.

Generally, ventilation represents a dilution control of indoor pollution, which means that contaminated indoor air is diluted or displaced with "clean" outdoor air. Ventilation performance depends on room geometry, ventilation method applied and on its operating conditions, as well as on the location and strength of the sources and the types of contaminants generated [30]. Ventilation rate or air exchange rate is defined as the ratio between the outdoor air flow rate and the effective volume of ventilated space. Air exchange rate varies and depends on climate, type of building and its operation as well as the lifestyle of the occupants. For example, as summarised by Biggs et al. [31] in residential houses in Australia, with mild to warm climate, air exchange rate at 50 Pa (ACR(50)) was reported to be relatively high reaching on average 19.2 h^{-1}, while in Canada and Sweden of much cooler climates, the ACR (50) value was reported to be 4.4 and 3.7 h^{-1}, respectively. The ventilation rate in office buildings is usually much lower than that in residential houses, and in the USA was found to be 0.9 h^{-1} and in Brisbane, Australia, about 0.8 h^{-1} [32].

- Mechanical filtration systems are intended to limit the introduction of particular pollutants from outdoors to indoors. The efficiency of such systems generally depends on the filter properties and aerodynamic properties of filtered particles [32]. The efficiency of filters varies from 5 to 40% for low-efficiency filters, such as dry media filters and panel and bag filters, from 60 to 90% for electrostatic precipitators to over 99% for HEPA filters. Further, filtration efficiency is also particle-size dependant. For example, He et al. [33] reported that the lowest filtration efficiency was observed for particles approximately 60–100 nm in size. The overall average filtration efficiency of the HVAC systems in an office building was $59.5 \pm 12.6\%$ and $45.7 \pm 11.0\%$ for particles in the size range 8–300 nm and $51.0 \pm 8.3\%$ and $44.4 \pm 8.0\%$ for particles in the size range 6–3,000 nm, respectively. Not only the filters but the whole HVAC system contributes to particle reduction, due to particle losses on the cooling/heating coil and other parts of the system. The selection of a system depends on the type of indoor environment, outdoor and indoor sources, demand on the level of reduction of pollutant concentrations and the cost associated with purchase, operation and maintenance of the system.
- *Chemical reactions involving vapours and gases leading to particle generation*: Examples of such processes are reactions between ozone and various terpenes in indoor environments, which have been shown to result in a significant increase in the number and mass concentrations of submicrometre particles [34].

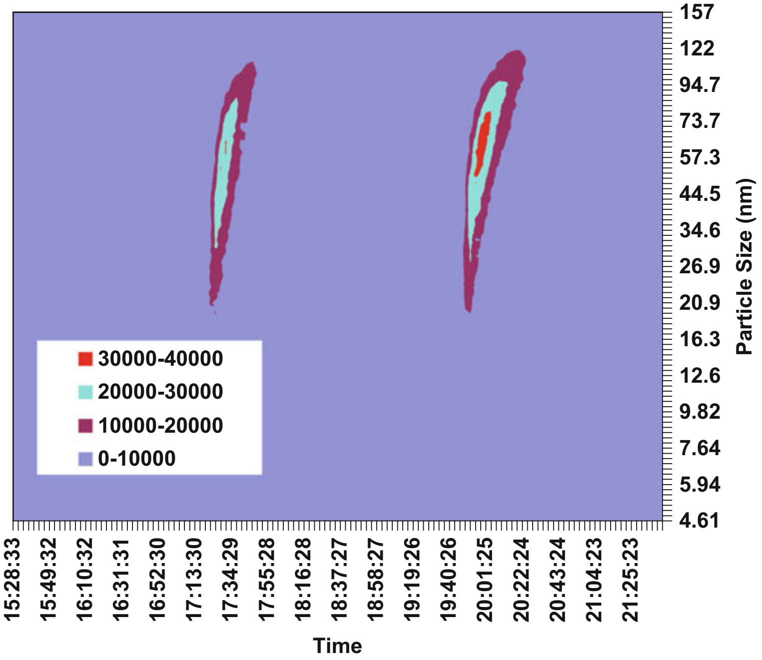


Fig. 3 An example of particle-size distribution variation during the particle formation process. The first set of data (*left*) is for four trays while the second (*right*) is for five trays of the detergent

For example, 20–40 ppb of O_3 and several hundred ppb of terpenes can generate an additional 5–40 μg m^{-3} of fine particles [35]. Initially, more than half of the mass increase is in the ultrafine fraction; however, particle number and mass size distribution evolve over hours, shifting the peak of the distribution towards larger sizes. Controlled experiments were conducted by Morawska et al. [36] in a lecture theatre, in which indoor O_3 and monoterpene concentration levels were adjusted by an O_3 generator and different number of tries of detergent, respectively. Their results showed that when indoor O_3 concentration levels were lower than 0.005 ppm, there was no clear increase in particle number concentration; however, rapid particle formation was found to occur above this O_3 concentration. An example of particle-size distribution during the experiments is given in Fig. 3. It can be seen from Fig. 3 that the particle-size distribution for five tries was larger than for four trays and the difference was found to be statistically significant ($p < 0.01$) for particles in the size range 0.005–160 nm. This indicates that a large number of nanosized secondary particles formed, which subsequently grew in size. They also found that $PM_{2.5}$ concentration in the lecture theatre did not increase above the background level during the particle formation process. The products of ozone/terpene reactions include hydroxyl radicals that can, in turn, react with other indoor organic compounds such as toluene to produce low volatility products, which further contribute to particle formation. The reaction of ozone with isoprene produces much less particulate matter than the reactions involving terpenes. This is related to lower molecular weight of isoprene (half of the weight of terpene) and also to the differences in volatility of the major products. In general, more secondary particles are produced under more humid conditions.

There are also processes other than those listed above, which affect particle physical properties, most importantly size. The most significant of them are coagulation, which results from Brownian motion and collision of particles, mainly of similar sizes; deposition of smaller particles on the surface of bigger particles; changes to particle size due to changes in its moisture content including hygroscopic growth or shrinking by evaporation [10, 11]; as well as to changes in its organic composition by ageing growth or volatility [37]. However, typically the significance of these processes in affecting indoor particle characteristics is lower than of processes such as deposition or removal by ventilation. This is because of the relatively long-time scale of these processes and also of relatively low particle concentration levels. For example, the process of coagulation is strongly dependant on particle concentration and while the time needed for number concentration to halve is 0.2 s for particle concentrations of 10^{10} cm^{-3}, which could be encountered in concentred exhaust emissions, it is as much as 55 h for concentrations of 10^4 cm^{-3}, which could be encountered in indoor environments [11].

It is a complex task to untangle the role of individual processes on particle characteristics because many of the processes take place simultaneously, affect differently particles of different size ranges and are dependant on a large number of factors and characteristics of the indoor environments. Therefore, rather than investigating the role of individual processes, many studies have considered the combined impact of several such processes on particle characteristics. An example of this is particle loss rate, which includes surface deposition of smaller particles, due to diffusion, gravitational settling, and convective transport as well as removal of particles by ventilation.

A model developed by Koutrakis et al. [38] describes particle behaviour in the indoor environment and incorporates the role of various processes. The model assumes that perfect, instantaneous mixing takes place and also steady-state conditions in terms of outdoor concentrations and indoor source emission rates. The main model equation is

$$C_{\mathrm{in}} = \frac{PaC_{\mathrm{out}} + V^{-1}\sum_{i=1}^{n} Q_{\mathrm{is}}^{i}}{a+k} \tag{2}$$

where C_{in} is the indoor concentration (number of particles or mass per m^3 of air); P is the penetration factor (coefficient); a is the air exchange rate (h^{-1}); C_{out} is the outdoor concentration (number of particles or mass per m^3 of air); ${Q_{\mathrm{is}}}^{i}$ is the mass flux generated by the source i; n is the number of investigated indoor sources; V is the volume of room or house (m^3); k is the decay rate (h^{-1}) due to diffusion and sedimentation.

A situation often investigated is when no particles are generated from indoor sources for suitably long periods and the only contribution is from outdoor air. The indoor-to-outdoor concentration ratio of the particles is then equivalent to infiltration factor (F_{INF}), which is defined as the equilibrium fraction of ambient particles that penetrate indoors and remain suspended [39]:

$$F_{\mathrm{INF}} = \frac{C_{\mathrm{in}}}{C_{\mathrm{out}}} = \frac{Pa}{a + k} \tag{3}$$

4 Selected Types of Indoor Pollutants

4.1 Dust

According to the terminology from aerosol science, *dust* is defined as solid particles formed by crushing or other mechanical breakage of a parent material, larger than about 0.5 μm [10]. According to USEPA [40], *house dust* is defined as "a complex mixture of biologically derived material (animal dander, fungal spores, etc.), particulate matter deposited from the indoor aerosol and soil particles brought in by foot traffic". German Guideline 4300-8 [VDI, 41] states that "there is currently no generally binding definition of the term settled house dust. To delimit the term from suspended particulate matter, it is intended to mean all types of particles, which are encountered indoors in deposited form. The dust may be solids of the most varied inorganic or organic materials which can be of natural or synthetic origin. The term includes not only fractions which originate indoors themselves, but also those which are introduced from the outside". The same document makes a distinction between *old dust*, which is dust of unknown age found on indoor surfaces, and *fresh dust*, whose age is known and which is usually of the order of 1–2 weeks.

As described above, as a result of various human activities, settled dust can be resuspended into the air. Physical and chemical properties of dust vary and depend of the source from which the dust originated.

Size distribution of house dust particles ranges over several orders of magnitude and includes particles of the order of micrometres in aerodynamic diameter as well as those of the order of several millimetres. The majority of house dust particles are of the order of tens of micrometres. For example, Quee et al. [42] showed that 58% of dust was in the size range from 44 to 149 μm; Seifert [43] reported that 6–35% of the dust was in the range from 30 to 63 μm and Morawska and Salthammer [44] showed that 12.8–76.4% of dust was in the range from 63 μm to 2 mm and 9.5–35.5% was smaller than 63 μm. Molhave et al. [45] reported that 41% of office dust was in the range from 50 to 125 μm and 40% was larger than 125 μm.

Dust particles of different size ranges also differ in composition and content of organic and inorganic material. The differences are related to the origin of the particles, and in particular, the smaller dust particles include skin flakes, fragments of hair, microorganisms such as fungal spores and pollen, food crumbs, abrasion of textiles and fittings, sand, loam, clay and soot. Larger dust components can include parts of a plant (e.g. leaves and needles), hair or gravel.

As could be seen from the above listing, house dust contains many compounds of biological origin. The presence and concentration of certain compounds can be used as an indicator or marker of certain biological contaminants or indoor practices. For example, concentrations of β (1 → 3)-glucans and extracellular polysaccharides (EPS) are good markers for the overall levels of fungal concentrations in floor dust according to Chew et al. [46], while endotoxin concentrations in settled dust can be used as an indicator for residential hygiene according to Bischof et al. [47].

Due to the different origins of dust, samples collected from different indoor environments vary significantly in composition. For example, the dust from kindergartens most commonly consists almost completely of inorganic materials such as sand loam and clay from sand pits, while house dust from the residences of animal owners having at the same time heavy abrasion of carpets can consist virtually solely of organic material [6]. By contrast, dust collected in offices contained the following components: microorganisms, endotoxins, allergens, minerals and adsorbed organic compounds [45]. Residential and office cleaning removes a certain fraction of dust; however, excessive cleaning may increase concentration of hazardous components of cleaning agents in house dust [3, 48].

4.2 Fibres

In general, fibres are particles of an elongated shape, with one dimension significantly larger than the other two. The dimension of the fibres can cover a wide range with diameters as small as 0.025 μm while with lengths reaching several 100 μm [49]. Fibre dust particles have been defined as those which [50]:

- Exceed a lengths-to-diameter ratio of 3:1
- Have a length of more than 5 μm
- Have a diameter of less than 3 μm

Mineral fibres are of natural and synthetic (man-made mineral fibres, MMMF) origin. These two classifications, in turn, can be subdivided into inorganic and organic fibres. From the wide range of available fibres, only those which are significant from both technological and health aspects are asbestos, mineral wool and ceramic fibres.

Asbestos is a collective term for silicate minerals of serpentine and amphibole groups that occur naturally as fibres. The serpentine group includes chrysotile (white asbestos) and the amphibole group includes crocidolite (blue asbestos), amosite, anthophyllite, tremolite and actinolite. *Mineral wool* is a term used collectively for products that consist of synthetically manufactured inorganic fibres. These vitreous (amorphic) fibres are produced from a melt and depending on the starting material are referred to as glass, rock or slug fibres. Mineral wool generally serves as heat and sound insulation in construction engineering. *Ceramic fibres* can have both vitreous and crystalline structures. The former are aluminium silicate fibres (refractory ceramic fibres), which can be imparted with increased temperature by the addition of certain substances (e.g. zirconium oxide) and are used, for example, in refractory linings in industrial furnaces. Crystalline ceramic fibres include aluminium fibres and single crystalline whisker fibres, which are manufactured for special applications.

The sources of asbestos and other mineral fibres in the indoor environment are fire retardant, acoustic, thermal or electric insulation and structural material. Some of the mineral fibres such as asbestos were used commonly in the past as a part of building construction or, for insulation purposes, however, have been banned from such applications. Exposure to fibres has been associated with serious health effects including asbestosis, lung cancer in connection with asbestosis and mesothelioma (form of cancer disease).

4.3 Combustion Products in the Indoor Environment

Indoor combustion sources are related mainly to cooking, heating and tobacco smoking. In addition, outdoor combustion products, which in urban environments originate most commonly from vehicle emissions, penetrate inside and contribute to indoor pollution. Under ideal conditions, complete combustion of carbon results only in generation of carbon dioxide (CO_2) and water vapour. Any products other than CO_2 are often called products of incomplete combustion and include particulate matter and gases.

The majority of particles resulting from combustion processes are in terms of number in the ultrafine size range and in terms of mass in the submicrometre range. For example, natural gas, propane and candle flames generate particles in the size range between 0.01 and 0.1 μm, and meat charbroiling shows a major peak in the particle mass distribution at 0.1–0.2 μm with some material present at larger particle size but not larger than 1.0 μm [20]. Aerosol particles generated through combustion can grow hygroscopically 10–120%, depending on initial particle size and its origin [51].

Combustion particles are of complex chemistry, carrying most of the trace elements, toxins or carcinogens generated from the combustion process. Combustion of different types of fuels results in emissions of various trace elements, which are present in the fuel material. In most cases, there is not just one specific element that is related to the combustion of a particular fuel, but a source profile of elements [4]. For example, motor vehicle emissions contain Br, Ba, Zn, Fe and Pb (in countries where leaded petrol is used); biomass (wood) burning contain Cl, K, Na, Fe and Br; and coal combustions contain Se, As, Cr, Co, Cu and Al. For comparison, the crustal elements include Mg, Ca, Al, K, Sc, Fe and

Mn. Since most of the trace elements are nonvolatile, associated with ultrafine particles and less prone to chemical transformations, they often remain in the air for prolonged periods of time in the form in which they were emitted.

All of the combustion sources generate large amounts of volatile and semi-volatile organic compounds. Polynuclear aromatic hydrocarbons (PAH), some of which are strongly carcinogenic, are an important class of compounds contained in the organic fraction of the fine particulate matter. PAH compounds are synthesised from carbon fragments into large molecular structures in low-oxygen environments, such as occurs inside the flame envelope in the fuel-rich region of the flame structure [4]. If the temperature is not adequate to decompose compounds upon exiting from the flame zone, then they are released into the free atmosphere and condense or are adsorbed onto the surface of particles. Many different combustion systems are known to produce PAH compounds. The most studied PAH is benzo[*a*]pyrene (B[*a*]P), which is a physiologically active substance that can contribute to the development of cancer in human cells.

Semi-volatile organic compounds can be present in the air either in the vapour or in particle form (solid or liquid). From the point of view of the effect on human health, it could be of significance in what physical form the semi-volatile compounds are when they are inhaled. There is very little information available on this aspect, and this is due to the interest but mainly to the difficulties in investigating organic composition of small amounts of mass. The mass of particles in the submicrometre and ultrafine range is very small, and in order to collect sufficient mass for standard organic chemistry analyses, long sampling times are required, which is prohibitive for many exposure or health effect studies.

4.3.1 Environmental Tobacco Smoke

Almost all of the major studies have found that a significant source of fine particles is cigarette smoking, resulting in an increase in average fine particle levels indoors in the range from 10 to 45 $\mu g\ m^{-3}$ [52]. Thus, environmental tobacco smoke (ETS) is one of the most significant indoor pollutants. ETS is a mixture of two components: mainstream smoke drawn through the tobacco, taken in and exhaled by the smoker, and sidestream smoke, which is emitted by the smouldering cigarette between puffs. Exposure by nonsmokers – passive smoking – is of great concern as it is an involuntary risk, often incurred by the most susceptible members of society, unborn children, infants and young children. Over 4,000 compounds have been identified in ETS, the most important of them being CO, NO_x, nicotine, acetone, benzene, phenol, toluene, formaldehyde and B[*a*]P, e.g. [20]. Other organic compounds emitted present in ETS include iso-alkanes, anteiso-alkanes (anteiso-trioacontane, anteiso-hentriacontane, anteiso-dotriacontane, iso-tritracontane) [53]. Once generated and introduced to the air, both gas and particle phases interact with each other

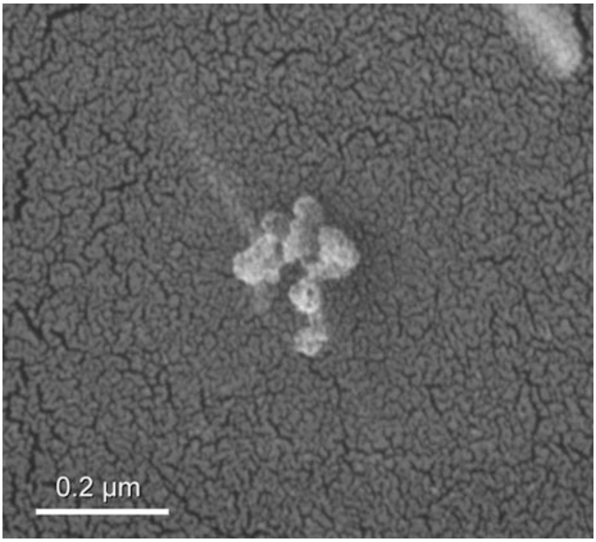


Fig. 4 Environmental tobacco smoke particle examined with an energy-dispersive X-ray analyser attached to a transmission electron microscope. The dominant element of the particle is carbon

(gas-to-particle conversion and particle-particle interactions), with atmospheric aerosols and with the environment.

Physical Characterisation of the Particulate Phase of Cigarette Smoke

Particles generated from combustion of a cigarette, range in size quite substantially, with the vast majority of them, however, being very small, below 1 μm, and a significant fraction below 0.1 μm. Figure 4 presents a microscopic image of an ETS particle. A literature review conducted by Nazaroff and Klepeis [52] revealed that most studies reported ETS particles as occurring in a single-size mode, with a large majority of the particle mass distributed among particles with diameters in the range 0.02–2 μm. The median of mass median diameters reported in the reviewed studies was found to be 0.37 μm, with the central 90% ranging between 0.2 and 0.5 μm. The median reported GSD was 1.4, with the central 90% ranging between 1.2 and 2.1. Figure 5 presents an example of a number size distribution of ETS measured in chamber testing [54].

The analysis conducted by Nazaroff and Klepeis [52] also showed that the median emission factor for cigarette smoking is 12.7 mg cigarette^{-1}, with the distribution exhibiting positive skew. The lognormal parameters of the distribution were a geometric mean of 12.0 mg cigarette^{-1}, with a geometric standard deviation of 2.1. The central 90% of the distribution lay between approximately 5 and 40 mg cigarette^{-1}, with ten of the seventeen results being in the range 7.8–13.8 mg cigarette^{-1}. The total mass of particulate matter emitted from cigarettes in a particular indoor environment can be estimated as the product of such emission factors and the number of cigarettes smoked indoors.

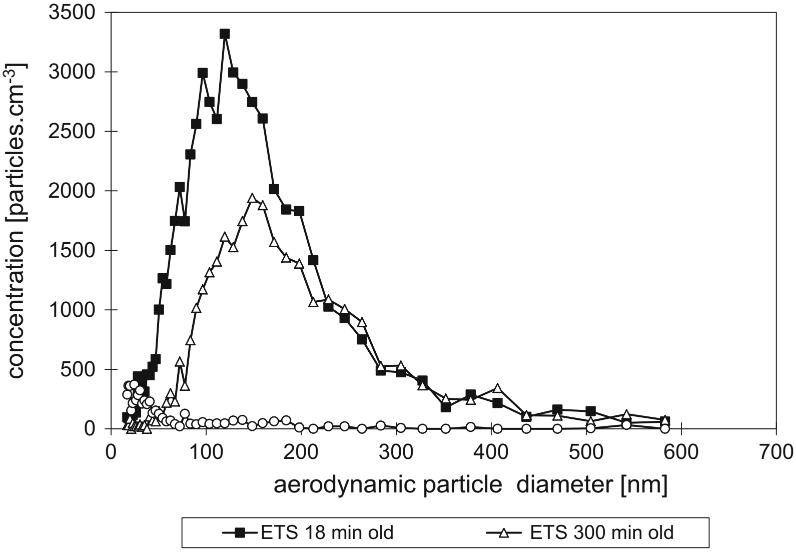


Fig. 5 Size distribution of environmental tobacco smoke measured 18 and 300 min after generation [54]

Chemical Characterisation of Cigarette Smoke

In addition to inorganic gases such as carbon monoxide (CO) and nitrogen oxides (NO_x), a large number of elements and compounds are generated from cigarette combustion. The particulate phase of the smoke is of special importance because it contains a significant amount of cigarette combustions products, for example, all of the tar and most of the nicotine [55]. Some of the compounds are present in cigarette smoke in very minute quantities, often difficult to measure; some of them, however, are emitted at much higher concentrations. Those emitted at higher concentrations and which are specific to cigarette smoke are called markers of the smoke. Nicotine, carbon monoxide (CO), 3-ethenylpyridine, nitrogen oxides, pyridine, aldehydes, acrolein, benzene, toluene and several other compounds have been used or suggested for use as markers for vapour phase constituents of cigarette smoke. Repairable suspended particulate matter (mass fraction of inhaled particles, which penetrates to the unciliated airways), solanesol, *N*-nitrosamines, cotinine, chromium and potassium are among the air contaminants used as markers for particle-phase constituents of the smoke [56].

The substances most commonly utilised as markers of ETS are repairable suspended particulate matter, nicotine, CO, 3-ethenylpyridine (3-EP) and solanesol [57, 58]. All these substances are associated, however, with potential problems, when used as markers. Both CO and repairable suspended particulate matter are not unique to ETS. The use of nicotine as a marker of ETS presents a problem because [59] (1) nicotine is found primarily in the gas phase (90%), making it a relatively poor particle marker, (2) gas phase nicotine is strongly basic and is removed from indoor environments at a faster rate than particle-phase nicotine or the particle portion of ETS and (3) the fraction of nicotine in ETS varies with measurement conditions. For example, 5–10% of ETS nicotine was found in the particle phase in a controlled atmosphere, while 20% was found in field environments. 3-Ethenylpyridine (3-EP) and solanesol are currently considered the best available ETS markers for the vapour and particulate phases, respectively [58]. The problems, however, are that airborne concentrations of 3-ethenylpyridine do not increase with source strength (i.e. with the number of cigarettes smoked) and that the methods for 3-EP determination do not possess adequate sensitivity [58].

In addition to the discussed above limitations in using various substances as ETS markers, consideration should also be given to the degree of complexity associated with using a particular substance as a marker and also, whether it could be measured in real time (to enable immediate mitigation actions if necessary). Respirable suspended particulate matter, CO and NO_x are the only markers for which measurements can be performed on a real-time basis with existing commercial equipment. However, as was explained above, none of them are specific to ETS. Often the majority of CO and NO_x comes from sources other than ETS [60]. The utilisation of solanesol presents a limitation because it cannot be measured in real time and what is more, its determination is not simple. It involves extraction from a filter to a solution with a recovery of about 60–90% and sample analysis by gas chromatography (GC) and supercritical fluid chromatography [61].

In summary, the chemistry of cigarette smoke is very complex with a vast number of elements and compounds generated in particle, vapour and gaseous phases. Some of these elements and compounds are generated in very small quantities, while others are in larger, easily measurable quantities. Some of the combustion products are specific to tobacco smoking, such as nicotine; others are also emitted by different, particularly combustion, sources. These compounds, which are specific to ETS, have been used as a marker of the smoke, but also compounds that are nonspecific to the smoke have been used for this purpose as well. Applicability of a compound as a marker of cigarette smoke does not necessarily point out the health impact of this particular compound.

Changes Occurring to Cigarette Smoke After Generation

After generation, the highly dynamic and reactive mixture of combustion products undergoes substantial changes resulting from complex physical and chemical processes taking place. Most substantial changes occur immediately after generation when the concentration of combustion products is still high. With time, the concentration decreases due to dilution with atmospheric air and the rate of most of the processes decreases. Removal of the combustion products from the air is mainly due to ventilation and filtration processes, with other processes, for example, surface deposition, playing a smaller role. This applies both to the particle and gaseous phases. The particles generated from cigarette combustion change their size distribution mainly due to coagulation, which shifts the peak of the distribution

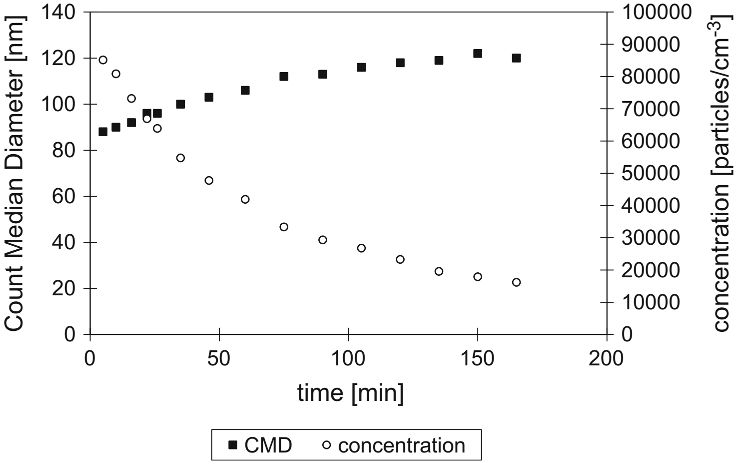


Fig. 6 The dependence of particle CMD and concentration on time for minimum ventilation in a brick house [54]

towards larger sizes. They can remain suspended in the air for long periods of time, up to several hours, as the process of gravitational deposition is not efficient for particle in this size range.

Variations in peak shape and location between the measurements taken at 10 and 180 min after cigarette smoke introduction to an experimental chamber are also presented in Fig. 5 [54]. It can be seen that during the latter measurement, the ETS peak is still clearly distinguishable from the background environmental aerosol spectrum and its location is shifted towards a larger size compared with the initial location.

Changes occurring the ETS in real indoor environment were investigated by Morawska et al. [54] who measured ETS decay in residential houses for conditions of minimum and normal ventilation. For minimum ventilation (all the windows and doors closed), air exchange rate was estimated to be in the range of 0.55–0.79 h^{-1} for brick houses and 1.05 h^{-1} for wooden house and for normal ventilation (defined as all the windows which are normally opened), from 1.93 to 4.48 h^{-1} and 4.73 h^{-1} for brick and wooden houses, respectively. The dependence of particle CMD and concentration on time for minimum ventilation in a brick house is presented in Fig. 6. Inspection of Fig. 6 reveals that for minimum ventilation in a brick house, a distinctive ETS peak is present in the air, even 3 h after one cigarette was smoked. Measurements conducted in a wooden house (higher air exchange range) showed that the peak is present in the air for up to 2 h. Li and Hopke [51] drew similar conclusions as to the concentration changes in the submicrometre ETS peak in a residential location of low air exchange rate. For normal ventilation, the ETS peak was distinguishable for short periods of time, which was about 1 h for a brick house and less than half an hour for the wooden house. When concentration remained high for longer time (as in the brick house at minimum ventilation), the CMD of the peak increased from about 90 nm to about 150 nm. At lower concentrations, the size distribution did not markedly change, which can be explained by a slowing down of the coagulation process and particle growth at lower concentrations.

Measurements of ETS characteristics in a large, naturally ventilated university club showed that at most times when the number of smokers was small (<10), the smoke concentration decreased rapidly with time and the ETS peak was not detectable even 10–15 min after a cigarette was smoked [54]. However, when the number of smokers was large, for example, during a rock concert, ETS concentration in the room was very high, up to 5×10^4 particles cm^{-3} and characterised by the stable, well-defined broad peak.

4.3.2 Biomass Burning

Biomass burning in the indoor environment is most commonly conducted for the purpose of cooking or heating, using a fuel wood and in developing countries, also animal dung, crop residues or charcoal. Household cooking stoves and space heaters are counted in billions throughout the world, providing the very basic household needs of heat; however, they are a source of significant levels of pollutants, when dirty biomass fuels and inadequate technologies are used. The levels of particles, PAHs, CO, and other air pollutants found in the kitchens of developing country villages are orders of magnitude above Western urban levels or relevant standards [5, 9, 62, 63]. In general, because the household sources emit directly into the spaces and at the times of human occupancy, exposures to certain air pollutants derived from these small sources are often greater than those derived from large outdoor sources [64–66].

The majority of particles emitted from biomass burning are ultrafine, with only a small fraction in the larger size range and with most of the mass present in particles less than 2.5 μm in aerodynamic diameter [67]. For example, Raiyani et al. [68] investigated emissions from burning several biomass fuels in cooking stoves. The fuels tested included cattle dung, wood, crop residues, low-quality charcoals and also coal. The study revealed that 50–80% of TSP emissions from these cooking stoves were ≤ 2 μm and also that a large amount of the PAHs ($>75\%$) were found in this size fraction. A number of studies investigated characteristics of emissions from residential wood burning stoves, which are common in the USA for space heating or aesthetic purposes. These studies reported that (1) the particle mass distribution from wood (pine, oak, eucalyptus) combustion has a single mode at approximately 0.1–0.2 μm [20], (2) the particles are compact structures with fractal-like dimensions close to three and contained low mass fractions of volatile compounds [69] and (3) that operating conditions such as amount of air supply had a strong impact on the particle-size distribution and the emission of particle-bound PAHs [69].

Vegetation burning results in emission of a large number of compounds including retene, phytosterols, ligmens, phenolic compounds from lignins and diterponoids from resins [53]. The emissions from wood burning stoves were found to be acidic (pH 2.8–4.2) [70]. Organic acids, of which the major constituents are monocarboxylic (emitted from combustion of fossil fuels and biomass) and dicarboxylic acids [71], have been linked to health effects. High concentrations of PAHs have been found in soot generated from wood burning stoves [72], with some of them specific to biomass burning. This makes it possible to use certain PAHs as source signatures in receptor modelling for residential wood combustion [73]. For example, guaiacol and its derivatives (e.g. 4-methylguaiacol, 4-ethylguaiacol) result solely from the pyrolysis of wood lignin, are relatively stable in the atmosphere and therefore can serve as unique tracers of wood [74]. In terms of elemental composition, noticeable elemental carbon was found in wood smoke as well as measurable quantity of Na, K, Fe, Br, Cl, nitrate, sulphate and ammonium. Statistically significant amounts of Na, Al, K, Sr, Ba, Cl, nitrate, and sulphate were found in meat charbroiling emissions [20].

Health effects of wood smoke were reviewed by Naeher et al. [75] who examined several topics: the chemical and physical nature of wood smoke; the exposures and epidemiology of smoke from wildland fires and agricultural burning and related controlled human laboratory exposures to biomass smoke; the epidemiology of outdoor and indoor wood smoke exposures from residential wood burning in developed countries; and the toxicology of wood smoke, based on animal exposures and laboratory tests. In addition, the review provided a short summary of the exposures and health effects of biomass smoke in developing countries.

4.3.3 Vehicle Emissions

As discussed above, outdoor air has a significant impact on indoor air, with up to 100% penetration efficiency of pollutants from outdoor air to indoor air for naturally ventilated buildings. For mechanically ventilated buildings, the fraction of pollutants penetrating indoors is smaller and depends on a number of factors including the type and operation of the filtration and ventilation system, type of filters used and thus their efficiency for particles in different size range, location of the air intake and tightness of the building. Due to the significant effect of motor vehicle emissions on outdoor air, and efficient penetration of outdoor pollutants to the indoor environments, the impact of vehicle emissions on indoor concentration levels of particles and other pollutants could be substantial.

Vehicle emissions, like other combustion products, are comprised of pollutants in gaseous and particle forms, which are complex in chemistry, and contain many compounds, which have been shown to affect human health. The main gaseous emissions include hydrocarbons (HC), CO, NO_x, CO_2, SO_2 and water vapour. Particles generated from vehicle emissions are generally small. A significant proportion of diesel emission particles have diameters smaller than 0.1 μm [76]. Gasoline particles are mostly carbonaceous spherical submicrometre agglomerates ranging from 10 to 80 nm. Particles from compressed natural gas (CNG) emissions are smaller than from diesel or even petrol emissions and range from 0.01 to 0.7 μm, with majority being between 0.020 and 0.060 μm. [77]. Associated with particles (especially fine and ultrafine) are many toxins, trace elements and also carcinogenic compounds. An example of these is 3-nitrobenzanthrone, a nitrated polycyclic aromatic hydrocarbon (nitro-PAH) originating from diesel emissions which has been shown to have high cancer-causing potential.

The chemistry of particles originating from vehicle emissions varies and depends on engine technology, test conditions, type of fuel on which the vehicle operates, its specific composition and on other characteristics, as well as lubricating oil used and its composition. There are thus differences between particles originating from diesel or spark ignition vehicles, the latter including petrol, compressed natural gas (CNG), liquid petroleum gas (LPG) or, for example, ethanol-fuelled vehicles. Diesel emission particles are primarily elemental carbon, but they also contain adsorbed or condensed hydrocarbons, hydrocarbon derivatives, sulphur compounds and other materials [78]. Solvent-extractable organic components of diesel aerosols represent 5–40% of the particle mass. Particles emitted from spark ignition vehicles are mostly carbonaceous spherical submicrometre agglomerates, consisting of a carbon core with various associated organic compounds. The main components of the particle phase include soot and ash, which consist of trace elements such as lead, iron, chlorine and bromine, organic compounds and a low-to-medium boiling fraction of engine oil [79]. Lubricating oil and other fuel hydrocarbons are the main contributors to emissions of particles of nanometre size [80]. The sulphate particles present in gasoline engine emissions are mainly from catalyst-equipped vehicles utilising unleaded gasoline [81].

In general, motor vehicle emissions contain various organic compounds including hopanes and steranes (present in lubricating oil for diesel and gasoline vehicles and in diesel) and black elemental carbon (present in a higher fraction in diesel emissions) [53]. Semi-volatile aliphatic hydrocarbons present in emissions from diesel and gasoline engines consist of a narrow band of $C_{15\text{–}27}$ n-alkanes maximising at $C_{20\text{–}21}$, a very similar pattern to lubricating oils n-$C_{13\text{–}27}$, maximising at C_{19}. The signal of diesel fuel has a broader spectrum extending to n-C_{33}, with a higher proportion of lower molecular weight components (n-$C_{10\text{–}22}$, maximising at C_{19}) [82]. Common organic compounds are polycyclic aromatic hydrocarbons (PAHs), such as pyrene, chrysene, benzo[*a*]pyrene and BaP. The semi-volatile fraction of the emissions can be associated either with vapour or with particle phases.

There have been a range of different fuels introduced over the past years and their emissions have been tested. Results reported in literature on emissions from engines and vehicles operating on these fuels are somewhat inconsistent because the emission process is sensitive to many factors, such as engine-operating conditions, the type and strength of biofuel blends and the specific vehicles used. However, in general, combustion of biodiesel leads to the reduction in particle mass, hydrocarbon and CO emissions, but to the increase in particle number and NO_x by comparison with conventional diesel engines [83, 84]. It should be also noted that the composition of the products emitted by these new biofuel engines is different compared to tradition engines [85]. Thus, combustion of these fuels by modern engines yield products that are very different to those emitted 20 years ago and profoundly changing the composition and dynamics of urban atmospheres.

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Volatile Organic Compounds in Indoor Environments

Godwin A Ayoko and Hao Wang

Abstract This chapter provides an overview of the types, sources and current techniques for characterising volatile organic compounds (VOC) in nonindustrial indoor environments. It reviews current knowledge on the levels of volatile organic compounds in indoor environments, discusses concepts for regulating indoor levels of volatile organic compounds and appraises current efforts to understand the links between VOCs and building-related health/sensory effects. It also provides an up-to-date outline of new trends in and perspectives for indoor air VOC research.

Keywords Characterisation, Purification, Regulation, Sampling, Volatile organic compounds (VOCs)

Contents

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Abbreviations

AFoDAS/ AVODAS	Automated formaldehyde data acquisition system/automated VOC data acquisition system
AM	Arithmetic mean
BRI	Building-related illness
BTEX	Benzene toluene, ethylbenzene and xylene
DMF	Dimethylformamide
DMSO	Dimethylsulphoxide
ECA	European Collaborative Action
ECD	Electron capture detector
ETS	Environmental tobacco smoke
EXPOLIS	Air pollution exposure distributions of adult urban populations in Europe
FID	Flame ionisation detector
GC	Gas chromatography
HPLC	High-performance liquid chromatography
I/O	Indoor/outdoor
IAQ	Indoor air quality
MCS	Multiple chemical sensitivity
MS	Mass spectrometry
OCIA	Organic compounds in indoor air
PAS	Photoacoustic spectroscopy
PDMS	Polydimethylsiloxane
SBS	Sick building syndrome
SER	Area specific emission rate
SSVs	Safe sampling volumes
SVOC	Semi-volatile organic compounds
TVOC	Total volatile organic compounds
US EPA	United States Environmental Protection Agency
VOC	Volatile organic compounds
VVOC	Very volatile organic compounds

1 Introduction

There is a long history of interest in volatile organic compounds (VOCs) in indoor environments. This is evidenced by the large number of national and regional studies/campaigns that have been undertaken to model, identify or quantify indoor VOCs or relate indoor levels of VOCs to indoor materials, indoor activities and some perceived health/sensory effects. The main interest in such studies lies in the fact that most people spend up to 80% of the day in one indoor environment or another, where pollution levels can be higher, pollutant sources are more varied and exposures are more important than those found in outdoor microenvironments. Many novel insights have emerged from the studies, and some of the main features of these insights are outlined in this chapter. In particular, the types of VOCs commonly found in indoor air, sources/source characteristics of indoor VOCs, measurement techniques for profiling indoor VOCs, typical results from indoor air VOC studies, health effects of VOCs, concepts for reducing indoor VOCs and new trends in indoor VOC studies, particularly in the last decade, are discussed in the following sections.

To put the concepts discussed in the chapter in the right context, distinction must first be made among the terms very volatile organic compounds (VVOCs), VOCs, semi-volatile organic compounds (SVOCs) and particulate organic matters (POMs), which are commonly used to describe organic compounds in indoor air. According to WHO [1], VVOCs, VOCs, SVOCs and POMs are compounds with boiling ranges between 0°C and 50–100°C, 50–100°C and 240–260°C, 240–260°C and 360–400°C and higher than 380°C, respectively.

2 Types of Indoor VOCs

Hundreds of VOCs are found in a typical nonindustrial indoor environment. Many of these compounds are aromatic hydrocarbons, alkenes, alcohols, aliphatic hydrocarbons, aldehydes, ketones, esters, glycols, glycolethers, halocarbons, cycloalkanes and terpenes [2] but amines like nicotine, pyridine, 2- picoline, 3-ethenylpyridine and myosmine are also widespread, especially in smoking microenvironments [3]. Moreover, low molecular weight carboxylic acids, siloxanes, alkenes, cycloalkenes and Freon 11 are also frequently encountered in typical nonindustrial indoor air [1].

3 Sources of Indoor VOCs

VOCs are ubiquitous in indoor environments. They are widespread in household and consumer products, furnishing and building materials, office equipment, air fresheners[4], paints [5, 6], paint strippers, household solvents and in microorganisms found in indoor environments. In addition, humans and their indoor activities such as cooking, cleaning, building renovation and tobacco smoking generate high levels and wide varieties of VOCs. Apart from these indoor sources, intrusions of VOCs from attached garages [7–9], outdoor traffic as well as biogenic and industrial emissions contribute significantly to indoor VOC levels. Furthermore, indoor air reactions are now recognised as sources of indoor VOCs [10], as exemplified by the reaction of ozone with primary VOC emissions from building products to generate appreciable amounts of aldehydes [11].

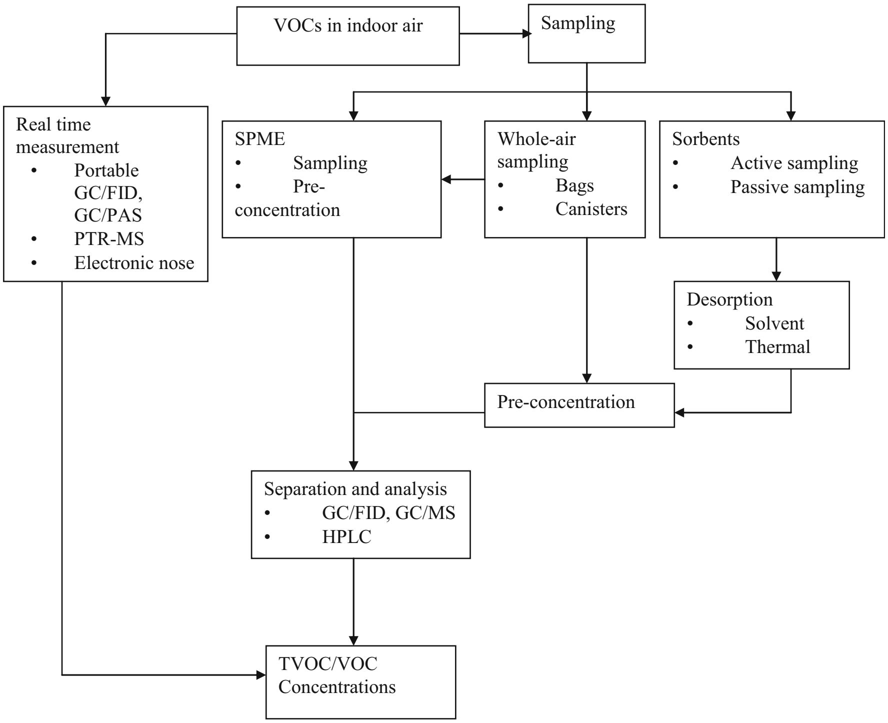
While some common indoor VOCs originate exclusively from indoor sources, others have multiple indoor and outdoor sources. Consequently, the indoor level of a particular VOC is the summation of the contributions of its various indoor and outdoor sources. In recent years various authors have undertaken comprehensive reviews of indoor VOC sources [4, 10, 12–16], and it is apparent from these reviews that the main sources of the typical indoor VOCs together with the major chemical classes associated with the sources are as summarised in the following:

- *Outdoor sources*: traffic, industry, agriculture, green plants and waste storage and treatment (aliphatic and aromatic hydrocarbons, aldehydes, ketones, esters)
- *Building materials*: insulation, paint, plywood and adhesives (aliphatic and aromatic hydrocarbons, alcohols, ketones, esters)
- *Furnishing materials*: furniture and floor/wall coverings (aliphatic and aromatic hydrocarbons, alcohols, halocarbons, aldehydes, ketones, ethers, esters)
- *Consumer products*: cleaning agents, polishes, incenses, air freshener, personal care products and newspaper (aliphatic and aromatic hydrocarbons, alcohols, halocarbons, aldehydes, ketones, terpenes, ethers, esters)
- *Equipment*: laser printers, photocopiers, computers, other office equipment and vehicles parked inside (aromatic hydrocarbons, aldehydes, ketones, esters)
- *Indoor activities*: cooking, tobacco smoking and use of candle and solvents (amines, aliphatic and aromatic hydrocarbons, aldehydes, halocarbons)
- *Ventilation systems*: filters of HVAC systems (aliphatic and aromatic hydrocarbons, alcohols, halocarbons, aldehydes, ketones, terpenes, ethers, esters)
- *Biological sources*: humans, moulds, bacteria and indoor plants (terpenes, glycoesters, alcohols, esters, aldehydes)

4 Sampling and Characterisation of Indoor VOCs

Interest in indoor air monitoring is driven by a wide variety of reasons [17]; the most prominent ones include the desire to:

- Undertake baseline measurements in order to set limits.
- Identify the presence of specific pollutants (e.g. formaldehyde).
- Apportion indoor VOC sources.
- Evaluate levels of compliance with legislations.
- Assess contaminated buildings.



Scheme 1 Summary of the steps involved in the characterisation of indoor VOCs

- Apply and validate sampling/analysis methods.
- Validate models.
- Evaluate ventilation systems.
- Evaluate the strength of a specific source.
- Relate sick building syndrome (SBS)/health effects to VOC levels.
- Understand the mechanisms of VOC transport from source to receptor sites.

While specific details may differ, the general analytical procedures described below and summarised in Scheme 1 apply to most monitoring exercises. Firstly, the purpose of the monitoring exercise must be clearly set out, then an appropriate method of sampling must be chosen, followed (where applicable) by the choice of suitable methods for sample storage, sample preparation or preconcentration and sample separation. Lastly, identification and/or quantification of the components are performed [18].

Assessment of VOC levels in an indoor microenvironment may be accomplished by direct measurements or by collection of a sample of air followed by subsequent laboratory analysis of the sample. Both of the approaches can be devised to answer the basic questions: what is present and how much is present?

4.1 Online Measurements

In general, direct measurement is achieved through the use of proton transfer reaction mass spectrometry (PTR-MS), portable gas chromatography (GC), photoacoustic spectroscopy (PAS), photoionisation detectors (PIDs) and infrared spectroscopy and by the so-called electronic noses [13, 19, 20] . Zhang and Mo [21] recently reviewed direct reading instruments used in monitoring indoor organic compounds and provided comprehensive knowledge on their detection or measurement principle, sensitivity and accuracy as well as application illustrations. Such real-time measurement instruments facilitate rapid data acquisition and are especially useful for rapid assessment of contaminated sites and for screening purposes. However, because logistics demand that the equipment involved is portable, some of them are relatively expensive and do not always afford detection limits that are as low as those obtained by conventional laboratory instruments [22]. In addition, it is often necessary to 'calibrate' or 'train' the equipment with the analytes of interest. For example, 'electronic noses' are specially 'trained' through extensive chemometrics procedures [23], while PIDs are calibrated with a particular VOC (e.g. toluene), and the other components of the air sample are determined as equivalents of that VOC. Measurements obtained in this way give little or no qualitative information about the constituent of the air sample. For example, Li et al. [24] measured the total volatile organic compounds (TVOCs) in indoor microenvironment continuously with photoacoustic Multi-Gas monitor, but apart from formaldehyde, the other constituents of the samples were unknown.

PTR-MS has also been recently [19, 25–31] developed to allow real-time online measurement of VOCs in indoor and outdoor environments. In the past 10 years, it has been successfully applied in a number of studies to continuously measure the VOC concentration in various indoor environments, such as animal buildings [31, 32], aircraft cabins [29, 30] and workplace [33]. It has also been used in laboratory experiments. For example, Mo et al. investigated by-products resulting from photocatalytic oxidation of toluene by PTR-MS [27]. Zhang et al. used the PTR-MS technique to characterise VOC emission signatures of individual building materials, resulting in the establishment of a database which can be utilised for indoor emission source identification [19, 26]. Recently, Jordan et al. further improved the resolution and sensitivity of PTR-MS by using a time-of-flight mass spectrometer, which is capable of measuring VOCs at ultra-low concentrations (as low as a few pptv) under high mass resolution with a mass range beyond 100,000 amu [28].

Various types of portable gas chromatographs are now available for the direct measurements of VOCs. These include gas chromatographs with high-speed temperature and pressure programming and a GC-ion mobility spectrometer [22]. In addition, portable GC time-of-flight (TOF) mass spectrometers [34] are available. But GC–MS is not routinely used for indoor air field measurements because of size, vacuum and energy requirements [22]. According to Santos and Galceran [22], portable GCs provide near real-time measurements, interactive sampling and quick solution to the problem faced at the time of the investigation. Nevertheless, they are usually expensive and are only able to achieve detection limits of the order of micrograms per cubic metres. However, a commercial portable GC–MS is now available to provide fast analysis (with less than 10 min turnaround time) and low detection limits (usually less than 1 μg/m^3) for indoor VOCs [35, 36].

4.2 *Offline Measurement*

Sampling can be done by passive or active techniques. Irrespective of the sampling technique adopted, subsequent laboratory analysis can be time-consuming and labour-intensive. Some of the common techniques used to collect and analyse indoor air samples are outlined below.

4.2.1 Active Air Sampling

This technique entails moving a predetermined volume of air at a controlled flow rate into a container or onto a sorbent. In its various forms, it is the most common technique used for the sampling of indoor VOCs.

Whole-Air Sampling

In whole-air sampling, a sufficient quantity of air is pumped into a container such as a polymer bag (Tedlar, Teflon or Mylar) [37] or a passivated stainless canister (e.g. SUMMA® or Silocan® canisters) [38–40]. The attraction in using whole-air sampling is that sample collection is relatively simple and rapid, especially when time-weighted sampling is not required. In addition, the analyst has the opportunity to monitor the presence of a wide variety of polar and nonpolar VOCs from one sample and to carry out replicated analysis on the sample. Furthermore, there is no sample breakthrough (i.e. some of the analytes do not pass through the sampler without being held). However, loss of VOCs due to chemical reactions within the container, physical adsorption by the walls of the container and dissolution in the water condensed in the container is not uncommon [41]. To minimise these, Tedlar bags should be protected from light by covering them with black bags and the internal surfaces of canisters should be electroplated or covered with siloxane [41]. Other shortcomings associated with the use of this sampling method include the high cost involved in purchasing and the inconveniences in transporting canisters. Despite these drawbacks, it is the method of choice for sampling and storing very volatile hydrocarbons (e.g. C_2–C_4 compounds) and reactive compounds such as terpenes and aldehydes [42]. Hsieh et al. [41] showed that the half-lives of 56 VOCs, including several highly reactive alkenes in SUMMA® canisters, Silocan® canisters and Tedlar, were generally in excess of 30 days.

Sampling onto Sorbent Tubes

Excellent reviews on the use of sorbents for sampling air in general [43, 44] and indoor VOCs in particular have appeared in the literature [45]. The most popular sorbents for sampling indoor VOCs can be classified into three broad categories: porous polymer-based sorbents (e.g. Tenax, Chromosorb), carbon-based sorbents (activated charcoal, graphitised carbon blacks, Carbotraps, Anasorb, Carboxens and Carbosieve) and silica gels. Of these, porous polymers and carbon-based sorbents are the most widely used for indoor VOC sampling.

The choice of the sorbent material employed for a specific sampling depends on the absorption and desorption efficiencies of the sorbent for the target VOCs as well as the stability of the VOCs on the sorbent. Additionally, the amount of VOCs retained on a sorbent is determined to a large extent by the sorbent bed length and sorbent mass. Thus, a typical sorbent tube has a length of 90 mm and an outer diameter of 6 mm and contains 0.1–1 g of the sorbent(s) [46, 47]. Some parameters that should be considered when choosing the most appropriate sorbent method for a particular study include the 'hydrophobicity', the 'thermostability' and the 'loadability' of the sorbent [48]. The less water is retained by the sorbent, the less interference is experienced during analysis; the more stable the sorbent is, the more robust it is during thermal desorption of the analyte. Lastly, the more the air that can be sampled onto a sorbent without sample breakthrough, the lower the detection limit that can be achieved.

Tenax TA, poly (2,6-diphenyl-*p*-phenylene oxide), is highly thermally stable and does not retain water. In addition, it affords high desorption efficiency for a wide range of VOCs. Consequently, it is the most widely used sorbent for sampling multicomponent indoor VOCs in the carbon size range C_5–C_6 to C_{18}. The literature on indoor air is filled with examples of measurement studies conducted with this sorbent as the VOC trapping medium [47, 49–54]. However, care must be exercised when using Tenax TA as a sorbent since it reacts with ozone and NO_x to form compounds which may facilitate the degradation of the sorbent [55]. To avoid this, ozone scrubbers must be used in conjunction with the sorbent, particularly when sampling is carried out in environments with high ozone concentrations [56].

When a single sorbent is not sufficiently efficient in capturing a wide suite of VOCs, combinations of sorbents are employed to increase the range of compounds that can be confidently sampled. Consequently, multi-bed sorbents made up of Anasorb®GCB1, Carbotrap® and CarbopackB® have been employed in some validated methods [56]. Similarly, multi-bed sorbents consisting of Carbotrap, Carbopack X and Carboxen 569 [57] and CarbopackB® and Carbosieve SIII® [17] have been used to trap a wide diversity of indoor VOCs.

Baltussen et al. [58] described the versatility of liquid polydimethylsiloxane (PDMS) as a sorbent material for volatile organic compounds. Unlike other common solid sorbent materials, retention on PDMS occurs as a result of dissolution rather than adsorption. In addition, it also has several advantages over the other forms of sorbents that are commonly used for indoor air sampling. For example, Baltussen et al. [58] showed that (1) it is more inert than other common sorbents and, therefore, it undergoes less reactions with the analytes and forms less artefacts, (2) it is more efficient in trapping polar compounds like organic acids, and (3) it requires lower thermal desorption temperatures than other sorbents. Also, Stefan et al. [59] showed that it does not absorb or introduce water into the analytes. Despite these advantages, PDMS is not as widely used in sorbent tubes for indoor VOC monitoring as Tenax. However, it is becoming more frequently employed in headspace sampling of VOCs and as a fibre coating material in solid-phase microextraction (SPME) [60–62].

Active sampling onto sorbents entails storing known amounts of sorbent material(s) in glass or stainless steel tubes and drawing the sample through the tube by means of small battery-powered pumps. Since sorbents do not possess unlimited capacities to hold samples, caution must be exercised not to sample too much air onto the sorbent; otherwise, 'sample breakthrough' will occur. Representative samples are only obtained when the appropriate volume of air and size of sorbent that minimises breakthrough are employed. To minimise errors due to sample breakthrough, the total volume of sample collected must be scrupulously monitored and a second bed of sorbent arranged in series with the first must be analysed. When the percent of a particular VOC in the second bed is greater than 5% of the amount in the first, sample breakthrough is implied [56]. While the safe sampling volumes (SSVs) suggested by US EPA Method TO-17 for various VOCs are a useful sampling guide, care should be taken in applying the SSVs since breakthrough volumes (BTV) are influenced by environmental factors like humidity, temperature and other factors. In keeping with this, US EPA Method TO-17 [56] suggested that sampling volume should not be greater than approximately 66% of the breakthrough volume [56].

Most classes of VOCs found in the indoor environment are sampled onto sorbents by adsorption, but highly reactive VOCs like carbonyl compounds are sampled by chemical reactions with the sorbent. Thus, aldehydes and ketones are sampled by their reactions with sorbent gels coated with 2,4-dinitrophenylhydrazine (2,4-DNPH) to form stable hydrazones [49, 63]. Similarly, formaldehyde has been sampled by its reaction with *N*-benzylethanolamine to give 3-benzyloxazolidine [64] or with hydroxymethyl piperidine to form oxazolidine derivative [65, 66].

Despite its widespread use in indoor VOC sampling, sorbent trapping provides no information about (1) all of the VOCs present in the sampled air since some VOCs are either not trapped by the sorbent(s) or are too reactive to remain on the sorbent surface and (2) the temporal variations in the concentrations of the VOCs that are being monitored.

4.2.2 Passive Air Sampling

Solid-Phase Microextraction

Ouyang and Pawliszyn [67] reviewed the application of this combined sampling and sample preconcentration procedure to indoor air VOC measurement. Typically, an SPME sampler consists of a fused silica fibre that is coated by a suitable polymer (e.g. polydimethylsiloxane (PDMS), PDMS/divinylbenzene, carboxene/PDMS) and housed inside a needle [67]. The fibre is exposed to indoor air, and after sampling is complete, it is retracted into the needle until the sample is analysed. Compared to other sampling methods, it is simple to use and reasonably sensitive. However, samples collected by the procedure are markedly affected by environmental factors such as temperature. Therefore, they cannot be stored for extended periods of time without refrigeration [68].

Koziel and Novak's review of SPME [69] is replete with examples of its use for (1) indoor VOC sampling followed by off-site laboratory analysis, (2) on-site sampling and analysis of indoor VOCs, (3) preconcentration of samples collected into canisters as well as (4) headspace sampling of the solvents extracted from samples collected by sorbent tubes. In addition to its ability to sample chlorinated VOCs [70], *n*-alkanes, aromatic hydrocarbons [61] and oxygenated hydrocarbons [61, 62, 70], the fibres of SPME can be doped with derivatising agents to make them amenable to sampling reactive VOCs such as formaldehyde [71]. Despite its virtues, relatively few examples of the application of this technique for indoor air sampling have been described in the literature.

Passive Sampling onto Sorbents

The sorbents used for passive sampling are identical to those described for active sampling. The only difference is that while samples are pumped through the sorbents in the latter, they diffuse into the sorbents in the former. Woolfenden [44] has shown that the diffusive uptake rates of VOCs commonly found in indoor air on different sorbents vary from about 0.8–15 (ng/ppm/min). Consequently, passive sampling is generally relatively slower than active sampling and may occur over several hours or days. Nevertheless, it is a popular sampling method, particularly for the evaluation of personal exposure. Raw et al. [72] used diffusive tubes packed with Tenax TA to monitor VOCs in 876 English homes; Missia et al. [73] employed Radiello passive samplers with charcoal/carbograph to monitor indoor and outdoor VOC levels in five European cities, while Schieweck et al. [74] used it to measure VOC levels in museum showcases in Germany. As in active sampling, chemical coated sorbents are also employed for the passive sampling of carbonyl compounds [73].

4.2.3 Sample Desorption/Preconcentration

Whole-Air Samples

Preconcentration of samples collected into canisters and polymeric bags is accomplished by passing known quantities of the samples through narrow capillary tubes held at very low temperatures by means of liquid cryogens [42, 75]. The tubes are then rapidly heated to release the analytes into a cryofocussing unit and eventually to the GC. The procedure affords excellent recoveries for many VOCs, but recoveries from samples stored in Tedlar bags are generally lower than those stored in canisters [41]. The main drawbacks of this procedure include the high cost of the cryogen and the susceptibility of the transfer tube to blockage.

SPME Samples

Extraction occurs when the needle of the syringe is exposed to fast moving, hot streams of gas within the injection port of the GC [76].

Samples Collected onto Sorbents

Depending on the sorbents used, solvent desorption or thermal desorption may be applied to the sampled analytes. For silica gel and carbon-based sorbents, solvent desorption [56, 77] and microwave desorption [48] are the preconcentration methods of choice.

Solvent Desorption

Acetonitrile is frequently used for the desorption of 2,4-dinitrophenylhydrazones of carbonyl compounds collected on silica gel [78, 79], while CS_2 is used for samples collected onto charcoal and dichloromethane for samples collected onto Anasorb 747 [79]. Carbon disulphide is particularly suitable for the desorption of nonpolar compounds but gives less satisfactory outcomes for polar compounds. To overcome this shortcoming, polar co-solvents such as DMF, DMSO and EtOH are added to CS_2 to increase the recovery of polar analytes [68]. In addition, the use of CS_2 suffers from a number of other drawbacks, including the facts that (1) it reacts with amines and volatile chlorocarbons, (2) it is unsuitable when electron detectors (e.g. electron capture detectors) are used, (3) it is toxic and (4) it has an unpleasant odour [68].

Compared to thermal desorption, solvent desorption is plagued by a number of shortcomings. For example, very volatile organic compounds are lost when the liquid sample is reconcentrated prior to its analysis. Moreover, solvent peaks may overlap with the peaks of very volatile organic compounds. According to Wolkoff [48], solvent desorption leads to loss of analytical sensitivity. Similarly, a comparison of the efficiencies of thermal and solvent desorption techniques showed that with few exceptions, solvent desorption consistently underestimates various classes of VOCs found in typical indoor air [79].

Thermal Desorption

This is a very popular method of transferring indoor VOC samples trapped by polymeric and carbon-based sorbents into analytical instruments. It usually entails running a stream of hot carrier gas (usually helium or argon), through the tubes in a direction opposite to that used for the sample collection. Typically, thermal desorption is carried at ca. 250°C [80]. After desorption, the compounds are reconcentrated by cryotrapping and then transferred directly by heat into the GC column. Although it affords greater sample desorption efficiency than solvent desorption, the desorbed sample can only be analysed once. Therefore, the only way to test the reproducibility of the method is to analyse multiple samples [68].

4.2.4 Characterisation of Indoor VOCs

Laboratory-based analyses of indoor VOCs are usually performed with gas chromatography, which are coupled with flame ionisation detectors (FIDs), electron capture detector (ECDs) or mass spectrometry (MS). Alternatively high-performance liquid chromatography (HPLC) is used. Of these techniques, GC–MS provides the most conclusive qualitative and quantitative information, although a combination of FID and ECD has also been reported to permit the identification of compounds with widely different properties [18, 81]. Nonetheless, GC–MS remains the most widely used technique for the characterisation of indoor VOCs [7, 47, 49, 82, 83]. Total ion chromatogram (TIC) is usually conducted to obtain global information on the ranges of compounds present, and selected ion monitoring (SIM) is performed to identify and monitor particular analytes. To facilitate the acquisition of quantitative information, the response factors of individual VOCs are often calculated against that of toluene, which is present in many indoor air samples and also acts as an internal standard. Typically, splitless injection technique is employed [22, 84] to ensure the detection of compounds that are present at low levels. Various validated US EPA Methods recommend the use of dimethyl polysiloxane capillary column for the speciation and quantification of a suite of VOCs [56, 85]. Similarly, the ECA Report No 19 recommended a column with a polarity not exceeding that of 8% diphenyl polysiloxane [2]. Such nonpolar columns are widely used in indoor air studies [74, 86–88].

In order to increase the number of compounds that can be separated in a single analysis, it is not unusual to use a combination of GC columns with different polarities [81]. Temperature programming is also often required to achieve acceptable separation of analytes. A typical temperature programme, which has been used to separate different classes of indoor VOCs, is summarised below: (1) hold at 40°C for 1 min, (2) raise at 15°C/min to 105°C, (3) hold at 105°C for 5 min, (4) raise at 20°C/min to 245°C and (5) hold at 245°C for 5 min [17]. Column diameters ranging from 0.25 to 0.53 mm and lengths from 25 to 100 m have been employed for indoor VOC measurements [83, 86, 89]. The choice of column dimensions depends on the properties of the compounds to be separated.

In their review of the application of GC in environmental analysis, Santos and Galceran [22] suggested that future perspectives of GC analysis include increasing the use of:

- GC–MS with positive and negative ion capabilities and sensitivities as low as ppq (parts per quadrillion)
- HSGC (high-speed GC) – with reduced sizes and capabilities for providing near real-time monitoring
- GC x GC (multidimensional GC) – which remarkably increase the separation capabilities of the two columns used
- GC–TOF–MS – with scanning capabilities of the order of 500 scan/s

Such developments are beginning to affect indoor VOCs measurement markedly.

HPLC is only used for the analysis of the derivatives of low molecular weight carbonyl compounds such as formaldehyde [49, 56, 90]. However, formaldehyde is also quantified by a variety of other procedures, including spectrometric acetylacetone method [91] and chromotropic acid procedure [92].

4.2.5 Quality Assurance/Quality Control

Caution must be exercised to minimise errors at every stage of the characterisation. Therefore, quality assurance/quality control principles must be applied to: sampling, sample storage, sample reconcentration and sample analysis [56]. For testing and calibration laboratories, the standard ISO/IEC 17025 has been published. Accreditation bodies have been established worldwide on national levels and which follow up the implementation of this standard. In many countries certification or notification procedures and strict quality control measures are required for laboratories working in the field of indoor air quality.

Sampling

Short-term samplings are subject to temporal variations because of changes in source strength and ventilation conditions, while long-term measurements may show diurnal and seasonal variations [48]. These facts should be considered when planning sampling. Prior to sampling, sorbent tubes should be conditioned using a stream of carrier gas and temperatures that are higher than those that will be used for the analysis [47, 50]. Similarly, canisters need to be cleaned by repeated cycles of evacuation, flushing with humidified zero air and analysis for any trace levels of undesired gases [39, 41, 93]. As part of the quality control, sample breakthrough must be checked when sorbent tubes are used. Breakthrough may be a serious problem resulting in underestimation of concentrations in indoor air. Uhde has discussed this in more detail [45]. In addition, field and method blanks as well as field duplicates must be collected and analysed.

Sample Storage

As a general rule, samples should be analysed as soon as possible after sampling and when immediate analysis is not feasible, they must be stored and transported under conditions that minimise artefact formation. Thus samples trapped onto sorbent tubes are commonly covered with Swagelok type of screw caps fitted with ferrules and stored in clean containers filled with nitrogen gas or activated charcoal [44, 80]. Tedlar bags must be protected from direct exposure to UV radiation, while canisters must be sealed airtight and transported to the laboratory in cool containers [17, 41].

Sample Desorption

Regardless of the desorption method used (solvent or thermal) it is essential to ascertain the recovery efficiency of the VOCs of interest by spiking sorbent tubes and canisters.

Calibrations

Pumps should be calibrated with a rotameter prior to and after sampling. Analytical instruments must also be calibrated before measurements. For example, GC–MS must be calibrated for mass and retention times using reference standard materials [89] and comparison made with the fragmentation patterns of known standards, usually a deuterated compound like toluene-d8. Similarly, the method detection limit must be determined by finding the standard deviation of seven replicate analyses and multiplying it by the *t*-test value for 99% confidence of seven values [56, 93]. It is also usual for internal standards to be added to the samples and to evaluate the correlation coefficients of each standard used when multilevel calibration is employed. For automatic thermal desorption tubes, external and internal standardisations are achieved by injecting solutions of standards into the tubes [57]; for canisters, solutions of standards are injected into the canisters followed by zero air.

In addition, the identity of each species must be obtained by comparing its retention time with that of an authentic standard sample or to inter laboratory set of established retention times and by comparing its mass spectrum with that contained in a National Bureau of Standards (NBS) or National Institute of Standards and Technology (NIST) library installed on most modern instruments. It is usual to assign positive identification to a compound if its retention time is within 1% that of the corresponding standard and the ratio of its quantifying ion to the target ion is not more than 10 times the standard deviation of the analogous ratio for an associated standard [94].

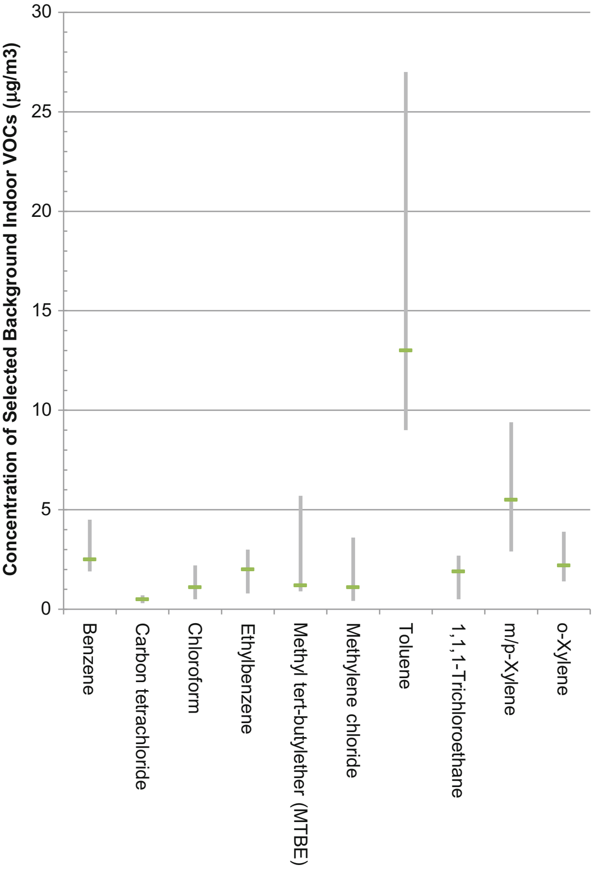
5 Current Knowledge on the Levels of VOCs in Indoor Microenvironments

Several studies have been undertaken to measure the level of indoor VOCs, in dwellings and offices in the past two decades, and some results from such studies have been reviewed [12, 95, 96]. For example, Dawson and McAlary (2009) reviewed VOCs from post-1990 indoor air concentration studies in North American residences unaffected by subsurface vapour intrusion [95]. They found that eleven compounds were detected in more than 50% of samples collected and for several compounds (benzene, carbon tetrachloride, chloroform, ethylbenzene and tetrachloroethene), the lower and upper quartiles of the indoor air concentrations are within the range of typical risk-based target levels (Fig. 1).

A survey of two reference databases, CAPLUS and MEDLINE on the web version of SciFinder Scholar (American Chemical Society, 2013), confirmed that interest in the characterisation of VOCs in various environments has not abated. Although the survey did not capture all of the research done on indoor VOC, it gave a good indication of what has been done and where. Three thousand one hundred and thirty-four hits (3,134) were recorded when 'VOC, indoor air' was searched on SciFinder. Further searches conducted to find out where these studies were done revealed that most have been conducted in the USA and European countries, although some significant results have also emanated from China, Japan, Canada, Hong Kong, Australia and Brazil in the past 10 years. Representative examples of some of the studies are presented in Table 1, while the concentration levels of BTEX (benzene, toluene, ethylbenzene and xylenes) in selected nonindustrial indoor air are presented in Table 2. The salient features of the studies reviewed reiterate some facts that have previously been known while others emphasise current trends.

1. GC–MS and GC–FID were used in most of the studies and are clearly the most popular detection methods used for VOCs quantification. Nevertheless, for reactive carbonyl compounds such as aldehydes and ketones, HPLC analysis of their derivatised products is still the method of choice.
2. Compared to whole-air sampling into Tedlar bags and canisters, active sampling onto sorbent materials is used more widely in these IAQ studies. Only a few studies made use of OVM passive samplers. Of the sorbent materials used, Tenax is the most frequently employed, possibly because of its virtues, which are mentioned in Sect. 4.2.1. It has been used for the characterisation of aromatics, alkenes, cycloalkanes, aldehydes, ketones, esters, alcohols, terpenes, glycol derivatives and even amines [79, 94].
3. For samples collected onto sorbents materials, thermal desorption is used, except for a few instances where CS2 desorption [97] and isooctane desorption [64] were preferred.
4. Generally, more studies have been conducted in residential indoor microenvironments than in offices. Although most of the studies were conducted in established rather than new buildings, many VOCs found in the former were

Fig. 1 The 25th, 75th and mean background concentrations for selected indoor VOCs measured in North American Residences between 1990 and 2005. Data from Dawson and McAlary [95]



also present in the latter but at higher concentrations. This is consistent with the thinking that VOC emission rates from building materials decrease with the age of the building [92]. Based on measurements in 243 homes, Herbarth and Matysik showed that a waiting period in the range of 60 days after renovation of a building should be observed to allow a decrease of VOC concentrations to a reference level [98].

5. Only a few of the studies estimated the TVOC of the microenvironment reported [64, 99] or focussed on complaint buildings [100] [101]. Because different definitions and methods of TVOC have been employed to estimate the quoted values, it is difficult to make direct inter-study comparisons; nevertheless, it appears that TVOC can range from 10 µg/m^3 to several thousand µg/m^3 in indoor environments.
6. Direct comparison of the concentration levels found in each study is difficult since the sampling was conducted over different times, with different sampling techniques and different sample treatments, and methods of analysis were used.

Table 1 Selected indoor air VOC studies conducted in the last two decades

Type of indoor	Compounds	Country	Collection medium	Sample treatment	Analytical instrument	References
Office	11	Singapore	CarbopackB and Carbosieve SIII	Thermal desorption	GC–MS	[17]
Homes	10	Korea	OVM 3500 passive sampler badges	Carbon disulphide (chemical desorption)	GC–MS	[77]
Office	60	Europe	Tenax tubes	Thermal desorption	GC–FID, GC–MS, PAS	[145]
Homes	6 Chlorinated VOCs	Japan and Sweden	Diffusion sampler packed with activated charcoal	Toluene (chemical desorption)	GC–ECD	[223]
Homes	30	Germany	OVM 3500 passive sampler badges	Carbon disulphide (chemical desorption)	GC–FID, ECD	[224]
Homes	25	Belgium	Tenax tubes	Thermal desorption	GC–MS	[50]
Furniture: showcase	45	Germany	Tenax tubes	Thermal desorption	GC–MS	[74]
Shopping mall	14	China	Tenax tubes	Thermal desorption	GC–MS	[54]
Homes	8	China	3L stainless steel evacuated canisters	Cryogenic preconcentrator	GC–MS	[75]
Public sites and homes	4 Aldehydes	France	2,4-DNPH cartridges	Acetonitrile (chemical desorption)	HPLC–UV	[225, 226]
Flemish homes	25	Belgium	Diffusion sampler packed with Tenax tubes	Thermal desorption	GC–MS	[50]
Public building/schools/kindergartens	23	European	Diffusion sampler packed with charcoal for hydrogens and DNPH for carbonyls	ISO/FDIS 16200-2 method	GC–FID, LC–DAD	[87]
Ships	5	South Korea	Tenax tubes	Thermal desorption	GC–MS	[227]
Homes	23	USA	Perkin Elmer tubes packed with Supelco Carbopack B	Thermal desorption	GC–MS	[127]
Museum	46	Germany	Tenax tubes	Thermal desorption	GC–MS	[228]
Apartment	240	Finland	Tenax tubes	Thermal desorption	GC–MS/FID	[229]
Homes	8 Chlorinated VOCs	USA	‘Tri-bed’ sorbent trap	Thermal Desorption	Portable GC–MS	[35]

Table 2 Arithmetic mean of the concentrations (μg/m^3) of BTEX in selected countries

Compound	Australia [92]	Finland [84]	Hong Kong [40]	USA [17]	Seoul, Korea [77]
Benzene	7.0	1.66	15.0	4.1	43.71
Toluene	14	5.62	206.3	15.3	170.67
m, p-Xylene	6.9	3.12	25.1	34.9	27.49[b]
Ethylbenzene	1.8	0.99	50.4	9.71	1.33
o-Xylene	8.9[a]	1.26	17.3	11.2	33.45

[a] *o*-Xylene/nonane
[b] *p*-Xylene only

However, the frequency with which the 64 VOCs that are of interest to the ECA are encountered in the different microenvironments can be classified as shown in Table 3.

The web-based Japanese automated formaldehyde data acquisition system/ automated VOC data acquisition system (AFoDAS/AVODAS) [102] showed that toluene was detected in 78% of the 1,422 homes monitored and that *p*-xylene, styrene, limonene and *α*-pinene were present in more than 50% of the homes. The data corroborates the classification in Table 3. However, most of the VOCs are not frequently quantified. Therefore, comparison between different studies is complicated.

5.1 The Total Volatile Organic Compounds (TVOCs) Concept

Hundreds of VOCs are present in some typical indoor environments. It is therefore not practically possible to identify and quantify every compound, even with the most sensitive and selective techniques [48]. Consequently, different techniques have been used to express the total volatile organic compounds (TVOCs) (for reviews of the methods, see [2, 18, 48, 103]).

To redress problems caused by the different approaches to TVOC estimation, a uniform procedure was proposed [2, 18]. The procedure, which is based on sampling of VOCs on Tenax tubes followed by thermal desorption and GC–MS analysis (with nonpolar columns), proposed that TVOC be defined as:

$$\mathrm{TVOC} = S_{\mathrm{id}} + S_{\mathrm{un}}$$

where S_{id} is the sum of identified VOCs expressed in milligram per cubic metre and S_{un} is the sum of unidentified VOCs relative to the response factor of toluene.

The procedure further recommends that as many VOCs as possible should be quantified in the analytical window bounded by the retention times of hexane and hexadecane and that these VOCs should as far as possible include the 64 VOCs that are of special interest to the European Community [2]. A major shortcoming of

Table 3 Frequency with which the 64 ECA VOCs were monitored in selected studies

Always[a]	Frequently[b]	Normally[c]	Occasionally[d]
Toluene, benzene	Ethylbenzene, styrene, *o*-xylene, limonene, tetrachloroethylene, *α*-pinene, *n*-hexane, trichloroethene, *n*-decane, hexanal, 1,4-dichlorobenzene benzaldehyde	1-Butanol, *n*-undecane, 1,1,1-trichloroethane, 1,2,4-trimethylbenzene, *n*-nonane, *n*-heptane, *n*-dodecane, naphthalene, 1,3,5-trimethylbenzene, nonanal, *n*-octane, cyclohexane, 2-ethyl-1-hexanol, 2-propanol, pentanal, methylisobutylketone (MIBK), *n*-tridecane, *β*-pinene, butanal, *n*-propylbenzene, *n*-pentadecane, 3-carene, 2-butoxyethanol, *n*-hexadecane, methylcyclohexane, hexanoic acid, acetophenone, 2-ethyltoluene, cyclohexanone, *m*/*p*-xylene, butylacetate, methylcyclopentane, 2-methylpentane, ethylacetate, 2-pentylfuran, 1-octene, texanolisobutyrates (TXIB), *n*-tetradecan, 2-ethoxyethanol 4-phenylcyclohexene, 3-methylpentane, tetrahydrofuran (THF), 2-butoxyethoxyethanol	1-Decene, 1-methoxy-2-propanol, isopropylacetate, methylethylketone, 2-methoxyethanol, 2-ethoxyethylacetate

Note: Three thousand one hundred and thirty-four references searched through SciFinder with key words of 'Indoor' and 'VOC' on 9 May 2013 were used as a database for this analysis

[a] Monitored in >40% of the studies

[b] Monitored in 10–39% of the studies

[c] Monitored in 1–9% of the studies

[d] Monitored in <1% of the studies

the recommendation is that not all VOCs present in indoor air are included in the approach. For example, important indoor VOCs like 2-propanol, 2-methylpentane, 3-methylpentane and butanal elute before hexane, while texanolisobutyrate (TXIB) elutes after hexadecane [104]. It was also expected that the definition would enhance interlaboratory of TVOC values, classification and screening of indoor materials and the identification of problems with ventilation design, indoor activities or materials [18]. However, De Bortoli et al. [105] observed large variances in interlaboratory studies performed with the approach. Nevertheless, it has been adopted in many indoor air studies [17, 104, 106].

5.2 Trends in Indoor VOC Concentrations

A couple of excellent papers have reviewed the trend of indoor VOC levels in the past several decades. Indoor VOC concentrations measured in North American offices and homes since 1990 were reviewed by Hodgson and Levin [107]. They concluded that there was a significant decrease in the average concentrations of some toxic VOCs, including benzene, 111-trichloroethane and tetrachloroethylene by comparing the dataset from 1990 to 2003 with two published reviews of data obtained in the 1980s [107].

Weschler [12] also examined the changes in indoor VOCs since the 1950s. He found that while average indoor concentrations of most VOCs decreased, some aldehydes and terpenoids increased due to increased indoor chemistry and use of solvents and scents, respectively. In addition, it was observed that the use of dimethyl and diethyl phthalates in personal care products and cosmetics, as well as the use of cyclopentasiloxane in antiperspirants, has increased during the past several decades [12].

6 Concepts for Regulating Indoor VOCs

Wolkoff [108] reviewed initiatives taken in Europe to reduce indoor air pollution by VOCs. Initiatives mentioned in the review include:

- Source control
- Control of emission from building materials
- Establishment of a Europe-wise database of outdoor and indoor VOC levels through EXPOLIS programme
- Indoor air quality audit projects
- Labelling scheme
- Establishment of guidelines for TVOC/VOCs
- Avoiding nonessential VOC

In addition to the above initiatives, various schemes aimed at reducing formaldehyde emission from building products, sensitising people to the effects of the presence of unsaturated fragrances in indoor air, use of labelled or low VOC, low isocyanate and acid anhydride emitting products have been introduced [108]. Because of their vast contribution to indoor VOCs, particularly in newly constructed buildings, a lot of the efforts highlighted above have focussed on the reduction of emissions from building products.

Bluyssen [109] reviewed more recent initiatives to improve and/or regulate IAQ, including:

- Development of WHO guidelines for indoor air quality which covers VOCs such as formaldehyde and benzene [110]
- European Parliament and the Council of Europe’s REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals) and ECHA (European Chemical Agency) initiatives [111]
- Implementation of mandate 366 for CPD (Construction Products Directive) [112]
- EnVIE [113]
- BUMA [114]
- Harmonisation of labelling schemes for construction and furnishing products [115]
- Establishment of the European Commission’s Scientific Committee on Health and Environmental Risk [116]
- Healthy Air Initiative [117]

She also identified nine facts and problems associated with IAQ. In the light of those facts and problems, she proposed an integrative approach to improving indoor air quality that incorporated the occupant(s), the air and the sources of pollution [109]. Kephalopoulus et al. critically reviewed the existing indoor materials labelling systems in the European Union [118], while Yu and Time [119] discussed the requirements for investigation of sick buildings with a particular focus on emissions of VOCs and formaldehyde in indoor environments, as well as some guidelines for assessment of exposure risks.

The US EPA [120] suggests, among other steps, that using household products as directed by the manufacturers and increasing ventilation when using household products could reduce indoor VOCs. In Japan, a database system for indoor formaldehyde and VOC (AFoDAS/AVODAS) has been established [102]. This should facilitate direct access to vital information needed by building designers, engineers and occupants to implement control measures [102]. In the USA, Hodgson et al. [78] have suggested the use of low VOC latex paints and carpet systems and decreased infiltration of unconditioned air. Mesaros [121] described the construction of ‘a low VOC house’ in Australia, in which materials with low VOC emission factors like ceramics are used in preference to those with high VOC emission factors such as carpets. The house provides a good illustration of the use of source control in eliminating indoor VOCs. Another low VOC emission house, which employed low VOC emission materials and high ventilation rates, was independently reported by Guo et al. [122]. The marked difference in the TVOC levels in the house and those in normal houses provide support for the fact that indoor air quality can be improved through a combination of source control and building designs that minimise the negative impact of uncontrollable sources. Consequently in Scandinavian and other

European countries recent standards for residential houses have combined strict energetic and ventilation requirements [123].

6.1 Source Identification

Unlike ambient VOCs, which originate predominantly from natural, vehicular and industrial emissions, indoor VOCs have numerous and diverse origins. Therefore, source identification is an important factor in source control and the prime driving force for many IAQ studies. It can be accomplished by many methods, including:

6.1.1 Comparison of Indoor-to-Outdoor Concentration Ratios

This method assumes that indoor/outdoor pollutant ratio depends on indoor and outdoor pollutant sources as well as the ventilation rates of the source and the sink, as shown in the following equation [124]:

$$C_{\mathrm{I}}/C_0 = 1 + 1/C_0[(S_{\mathrm{source}} - S_{\mathrm{sink}})/(q_{\mathrm{source}} - q_{\mathrm{sink}})]$$

where q is the rate of ventilation, S_{sink} is the indoor pollutant sinks, S_{source} is the indoor pollutant sources and C is the pollution concentration level.

When the indoor-to-outdoor pollutant ratio is approximately 1 for a VOC, it has comparable indoor and outdoor sources, and when the ratio is greater than 1, it has dominantly indoor sources [54, 77, 87, 125–127]. Typical indoor-to-outdoor pollutant ratio values for some VOCs are presented in Table 4, and these suggest that some VOCs have predominantly indoor sources while others have mainly outdoor sources. The method is often used in combination with statistical methods like Kruskal–Wallis, Wilcoxon *W* and Kolmogorov–Smirnov *Z* tests [84, 126].

6.1.2 Multivariate Data Analysis

These techniques reduce a large number of indoor VOCs to a few factors that can account for most of the cumulative variance in the VOCs data [126, 128, 129]. A factor loading matrix, which shows the correlation between the factors and the variables, is often obtained. Edwards et al. [129] have used this method to reduce 23 indoor VOCs in ETS-free microenvironments to 6 factors and to identify the most likely sources of the VOCs. A summary of the VOC classes that loaded on each factor and their probable sources are presented in Table 5. It is, however, noteworthy that UNMIX and positive matrix factorisation, both of which are based on factor analysis and have been applied frequently to ambient air quality data [130], have not featured prominently in indoor VOC source identification reports.

Table 4 Comparison of the ratios of the arithmetic mean of indoor concentration/arithmetic mean of outdoor concentration for selected VOCs in six homes

Compound	Living room	Kitchen
Benzene	2.35	2.05
Toluene	1.30	1.45
m,*p*-Xylene	0.62	0.84
o-Xylene	1.00	0.97
Ethylbenzene	0.54	0.75
1,3,5-Trimethylbenzene	0.82	0.77
Trichloroethane	1.06	1.24
Tetrachloroethene	0.52	0.48
1,4-Dichlorobenezene	0.90	1.03
Chloroform	2.00	2.38
Methylene chloride	0.92	0.84

Data was extracted from Lee et al. [125]

Table 5 The use of factor analysis for source apportionment of VOCs identified in residential indoor air

% variance accounted for	Component	Associated VOC classes	Assigned source
18	1	Alcohols and alkanals	Cleaning products, fragrances, consumer products, particle board
18	2	*n*-Alkenes, substituted aromatics, hydrocarbons	Traffic emissions
17	3	Aromatics	Long-range transport
9	4	Alcohols and alkanals	Carpets, rubber, adhesives
6	6	Mainly 2-butoxy ethanol	Cleaning products

Deduced from the data of Edwards et al. [129]

In addition, a number of studies have used principal component analysis (PCA) to recognise the sources of indoor VOCs [128, 131–136]. For example, Santarsiero and Fuselli investigated indoor and outdoor carbonyl compounds sources in Rome residential area as well as their mutual interrelations by means of PCA [135]. They identified six different sources of which four have a seasonal variation and observed a strong indoor/outdoor carbonyl compounds exchange.

6.1.3 Chemical Mass Balance Modelling

Watson et al. [137] undertook a review of the application of chemical mass balance modelling as a source identification technique for volatile organic compounds. The model assumes that the concentration of a chemical pollutant in a given sampling site is the summation of the contributions of all of the sources of the pollutant at the site. Thus, the concentration of the pollutant at the site can be predicted using the following equation [138]:

$$X_i = \sum_{j}^{P} a_{ij} S_j \quad i = 1, \ldots, m$$

where X_i is the predicted concentrations of pollutants at the site, a_{ij} is the source signature for the pollutant i from source j, S_j is the contribution of source j, m is the number of pollutants (VOCs in this case) and p is the number of sources.

Although the model is frequently used to identify contributions of ambient VOCs from different sources [137], it has not been widely used for source identification of indoor VOCs. Won et al. [138] used the model to show that wall adhesive, caulking, I-beam joist and particle board were the dominant sources of 24 indoor VOCs that were measured from a newly constructed building. However, similarities in the signatures of the various sources were observed. Such high correlations (collinearity) among measured chemical species could lead to large uncertainties in the estimated source contributions [137].

6.1.4 Recognition of Source Emission Signature

With the development of online measurement technology for indoor VOCs, some researchers have started to build up a database of emission signature of individual materials using PTR-MS. This database can be used for source identification of indoor VOC by comparing the emission profile of mixed VOC samples using the method of multiple regression least squares (MRLS) and a normalisation technique [26].

6.1.5 Instruments Used for Source Identification

Field and laboratory emission cell (FLEC) affords a portable, nondestructive method of testing the surfaces of potential VOC sources. In addition to its utility as a climatic chamber, it provides valuable information on source strength, which can be used for source identification and to formulate strategies for emission control. Wolkoff et al. [139] used it to identify emission processes in a number of building materials, while Jarnström and Saarela [140] utilised it to show that the dominant source of TXIB (2,2,4-trimethyl-1,3-pentadiol-butyrate) in the indoor air of some problem apartments was the floor surface.

Apart from FLEC, other instruments that have been used for source identification and apportionment include:

- Direct measurements by portable instruments [141]
- Passive samplers [142]
- Headspace samplers [143]
- Multisorbent tubes [144]

6.2 Understanding Emissions from Indoor Sources

Indoor VOC levels are influenced by a large number of factors [17, 48, 145]. The most prominent ones include: (1) air exchange rate; (2) source characteristics; (3) ventilation systems; (4) meteorology (temperature and relative humidity); (5) age of a building; (6) building design; (7) type of indoor activities (e.g. cooking, smoking and photocopying); (8) sorption, desorption and deposition rates; (9) mixing and distribution of pollutants and (10) removal rate.

Of these factors, source characteristics, particularly characteristics of building materials, have been the most explored in the literature. Thus, various studies have attempted to link emission rates and sink effects of building and furnishing materials with indoor VOC levels [5, 138, 146, 147]. Many studies indicate that emission levels in new buildings are much higher than those in established buildings [78, 92]. This is possibly because emissions from building materials generally exhibit a decaying profile that is illustrated by the following equation [101]:

$$\mathrm{EF} = M_0 k_1 \exp(k_1 t)$$

where EF is the emission factor for source material (usually expressed in μg/m^2/h), M_0 is the quantity of pollutant on the surface of the material (usually expressed as μg/m^2) and k_1 is the decay constant (h).

Alternatively, when there are multiple decay processes, the equation shown below is useful:

$$\mathrm{EF} = \mathrm{EF}_{01} \exp(-k_1 t) + \mathrm{EF}_{02} \exp(-k_2 t)$$

where EF_{01} and EF_{02} are the initial decay constants for two simultaneous decay processes. Thus, decay is initially fast and VOC level is higher in new buildings [92] as illustrated by Fig. 2.

Another approach that is commonly used to evaluate the emission rates of VOCs in indoor microenvironments is to estimate an area specific emission rate (SER). This approach assumes that the VOCs are homogeneously mixed in the environment and that SER can be calculated with the following equation [17]:

$$\mathrm{SER} = NV(C_1 - C_0)/A,$$

where the SER is in micrograms per square metre per hour, V is the volume of the space (cubic metres), N is the air exchange or infiltration rate (per hour), A is the floor area space (square metres), C_1 is the indoor concentration (micrograms per cubic metre) and C_2 is the outdoor concentration (micrograms per cubic metre).

Van Winkle and Scheff [148] used a variant of this equation to show that indoor VOCs have predominantly indoor sources, while Hodgson et al. [78] reported the SER values for a wide range of VOCs in manufactured and site-built homes in the

Fig. 2 Dependence of TVOC (in μg/m^3) on the age of a newly constructed home. Data from [92]

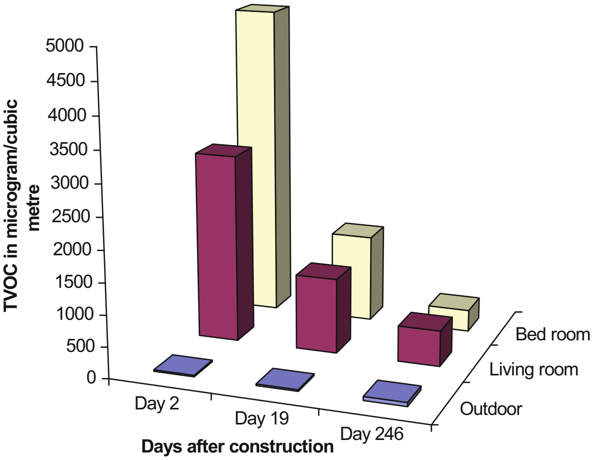
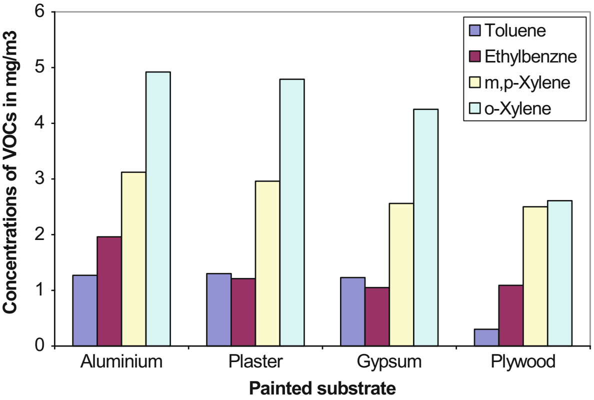


Fig. 3 Effect of substrate on VOC emissions from indoor building materials. Constructed from the data of Kwok et al. [152]



USA. As expected, many of the VOCs monitored by Hodgson et al. [107] showed decreased emission rates with the age of the building.

It is noteworthy that the VOC emission rates of building materials vary widely [92, 149, 150]. Thus Mølhave [151] reported over a few decades ago that the emission rates of wall/flooring glue, water-based PVA glue and gypsum board are of the order 2.7 × 10^5, 2.1 × 10^3 and 30 μg/m^2/h, respectively. Kwok et al. [152] reported VOC emission rates for varnish-painted aluminium, plaster, gypsum and plywood for toluene, *o*-xylene, *m*/*p*-xylene and ethylbenzene are also significantly different, with that of aluminium being approximately 65% higher than that of plywood as illustrated by Fig. 3.

It is also worth mentioning that emission rates vary with different indoor activities. Van Winkle and Scheff [153] associated 1,1,1-trichloroethane emission factors of 353, 522, 988, 1,419 and 2,790 μg/h with the presence of washer/drier in a utility room, storage of hair products, storage of chemicals, periodic dry cleaning and storage of moth balls, respectively. Occupants of air-conditioned offices and

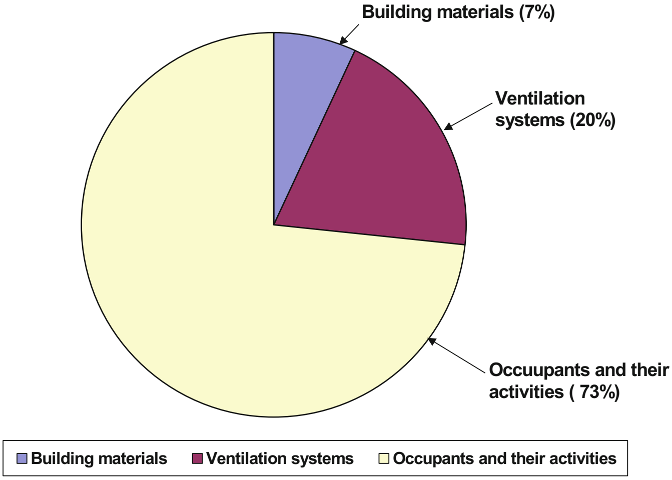


Fig. 4 Source apportionment of indoor aromatics. Data from Zuraimi et al. [144]

their activities have also been shown to contribute more to VOC levels in commercial offices in Singapore than ventilation systems and building materials [17], as illustrated by Fig. 4. This result corroborates the findings of Hodgson et al. [78], which suggested that occupants of new office buildings contributed more VOCs to the indoor air than other sources.

6.3 *Understanding the Interaction of VOCs with Indoor Materials*

In the past two decades, over 250 peer-reviewed publications addressing chemical reactions among indoor pollutants have appeared [10]. Diffusive interactions of VOCs with the surfaces of building walls, floors and household materials have been the subject of several experimental investigations, modelling and simulation [11, 29, 154–156]. Such interactions regulate peak levels of indoor VOCs, while subsequent desorption of the adsorbed VOCs delays their disappearance from the indoor environment. In order to predict and model the VOC emission rates of indoor materials, it is essential to know the diffusion and partition coefficients of individual VOCs. These coefficients can be simultaneously estimated through inverse analysis [157]. In addition, a couple of extraction methods have been developed to determine the initial concentration emitted and the partition coefficient of VOC in dry building materials [158, 159].

Secondly, VOCs react with indoor ozone to produce submicron particles [160–163]. Thus, terpenes, which is commonly found in many household consumer products, interacts with ozone, which is also widespread in indoor air through outdoor infiltration and use of office equipment like laser printers and photocopiers, to form particles [164–166]. Such reactions can markedly increase the number and mass concentrations of sub-micrometre particles. In addition, styrene and skin lipids react with ozone to generate appreciable amounts of aldehydes [33, 167].

6.4 Indoor VOC Guidelines

According to Mølhave [168], *"A guideline is a set of criteria (*i.e. *standards for making judgements) specifically assembled to indicate threshold levels of a harmful or noxious agent consistent with the good health."* The first notable attempt to provide some guidelines for indoor VOCs was made by Seifert [169] in which he classified indoor VOCs into alkenes, aromatic hydrocarbons, terpenes, halocarbons, esters, aldehydes and ketones (excluding formaldehyde) and proposed that:

- TVOC should not exceed 300 μg/m^3.
- No individual compound should have a concentration greater than 10% of TVOC or 10% of the concentration apportioned to that class of VOC.

Pluschke [170] reviewed Seifert's paper [171] and showed that only a few countries have guidelines for indoor TVOC. The USA has a value of 200 μg/m^3 [172], Germany 300 μg/m^3 [171] and Australia 500 μg/m^3 [173]. Pluschke [170] also stated that Seifert modified his original concept to include a target value defined as 200–300 μg/m^3 and a recommendation for official intervention if the TVOC concentration exceeds 1,000–3,000 μg/m^3. In Germany the federal and state authorities have developed a joint concept for the evaluation of indoor air contaminants by means of reference and guideline values and some guiding principles for the assessment of TVOC concentrations [174, 175].

Guidelines for individual VOCs are also available in some countries. In Poland, the maximum allowable concentrations for some VOCs have been set at 10 μg/m^3 for benzene, 200 μg/m^3 for toluene, 100 μg/m^3 for butyl acetate, 100 μg/m^3 for ethylbenzene, 100 μg/m^3 for *m*-xylene, 20 μg/m^3 for styrene and 30 μg/m^3 for *p*-dichlorobenzene [176]. In the context of the 64 VOCs of interest to the European Commission [177], only toluene, 2-ethoxyethanol, 2-butoxyethoxylethanol and 1-methoxy-2-propanol have readily available guideline values [178]. The odour threshold, sensory irritation exposure limit and health-based indoor exposure limits of these VOCs are presented in Table 6.

Despite the ubiquitous nature and importance of VOC in the indoor environment, it is surprising that no international indoor VOC guideline has emerged. Nielson et al. [178] have linked the difficulty experienced in evaluating sensory and health effects of indoor VOCs with the absence of indoor VOC standards and guidelines. Although the European Commission Report No 19 [2] recommended that indoor VOCs should be kept as low as reasonably achievable, more concerted efforts should be made to formulate a universally acceptable set of guidelines for as many indoor VOCs as possible.

Table 6 Guideline levels for some VOCs

Compound	Odour threshold (mg/m$^{3)}$	Sensory irritation exposure limit estimate (mg/m^3)	Health-based indoor air exposure limit estimate (mg/m^3)
Toluene	1	8	8
2-Ethoxyethanol	4.6	10	0.4
2-Butoxyethoxylethanol	0.0092	–	9
1-Methoxy-2-propanol	0.7	10	10

According to Pluschke [170] and Nielsen et al. [178]

7 Health Effects of Indoor VOCs

Mølhave [168] suggested that the health effects of VOCs could be grouped into:

- Immune effects and other hypersensitivity effects (e.g. asthma and allergy)
- Cellular effects (e.g. cancer)
- Cardiovascular effects
- Neurogenic and sensory effects (e.g. odour and irritation)
- Respiratory effects other than immunological

The US EPA website [120], assessed in May 2012, provides more details, suggesting that the health effects of indoor VOCs include: eye, nose and throat irritations; headaches; loss of coordination; nausea; damage to liver, kidneys and central nervous system; and cancer. Similarly, the health effects of environmental tobacco smoke (ETS), which contains several VOCs as well as nicotine, 3-ethenylpyridine, carbon monoxide and particulate matter, include: eye, nose and throat irritation, carcinogenic effects, activation of the immune system and exacerbation of asthma and respiratory tract illnesses [179].

While individual VOCs like benzene and toluene have been linked with acute myeloid leukaemia and neurotoxicity, respectively [180, 181], epidemiological studies of the health effects of indoor VOCs have related TVOC rather than individual VOC levels to exposure. The outcomes of such studies have been mixed [182]. In some cases, positive associations between sick building syndrome (SBS), building-related illness (BRI) or multiple chemical sensitivity (MCS) syndromes and TVOC levels were observed [183, 184], while negative associations were reported by Sundell et al. [185]. Yet, no association was found in a few studies [186].

Several reasons may be adduced for the inconsistent association between SBS complaints and TVOC levels [18, 48]. Some these are outlined below:

- Indoor air chemistry leads to the formation of VOCs and other species that are different from those monitored. For example, ozone reacts with VOCs to give secondary products that could be responsible for the observed SBS [187].
- Only compounds in a narrow chromatographic window are normally monitored; low molecular weight aldehydes, which may play a significant role in SBS, are not routinely monitored as part of TVOC [188].

- Ventilation systems are significantly associated with SBS complaints [189, 190].
- Particles present in indoor environment might contribute significantly to SBS systems [48].
- Environmental tobacco smoke is associated with many SBS-type symptoms [191].
- Measurements are usually carried out as mean time-integrated concentrations at the centre of the room rather than in the breathing zones of the subject [191].
- Self-reporting questionnaires are subjective means of assessing SBS [183].
- The influences of psychosocial factors are being ignored [184].
- TVOC is not biologically important [188].
- Biologically important VOCs have not been found and are not being monitored [188].

Thus, the role of VOCs in SBS complaints is far from being fully understood. More research is particularly required in the:

- Development of validated methods for TVOC and dose–response relationship
- Risk indicators for multiple exposures [182]
- Evaluation of TVOC and SBS/health effects from carefully designed epidemiological studies
- Development of universal guidelines for evaluating exposures [192]

Since TVOC does not permit consistent exposure–dose relationship, Mølhave [168] suggested that it should be treated as an indicator of the presence of VOCs and used only for source identification, indoor air quality assessment and as a screening tool for exposure assessment rather than a guideline or an official recommendation.

However, despite the statistically insignificant difference between the TVOC values in buildings with and without SBS problems, the underlining difference among such buildings was manifested in Coomans plot and partial least squares discriminant analysis plots [193]. Similarly, principal component analysis has been used to separate buildings with low and high prevalence of SBS [194]. It therefore appears that multivariate projection methods could play significant roles in the identification of causality in indoor VOC exposure studies.

8 Purification of Indoor VOCs

Although technologies for particle removal are rather well established, there are no satisfactory methods for VOC control because removing indoor VOC sources or increasing ventilation rates is often not feasible or economical [195]. Nevertheless, new technologies for the removal of indoor VOCs are emerging and some of them are outlined below.

8.1 Biological Treatment

Guieysse et al. reviewed the potential and challenges of biological treatment approaches in the removal of indoor VOC pollutants [195]. Although biological methods have shown some potential for indoor VOC removal, specific characteristics of indoor air and the indoor air environment pose numerous challenges, which may include the (1) maintenance of suitable and diverse catabolic ability under conditions that do not sustain microbial growth and (2) purification of large amounts of air in confined environments with minimal nuisances and release of microorganisms [195]. Liu et al. screened 73 ornamental plant species for their ability to remove 150 ppb benzene from indoor air [196]. The results showed that over 10 species, including Crassula portulacea and Hydrangea macrophylla, have the greatest capacity to remove benzene from indoor air. Wang and Zhang developed and evaluated a dynamic botanic air filtration system (DBAF), a fan-assisted with controlled airflow activated carbon/hydroculture-based potted plant unit [197]. They concluded that the DBAF effectively removed both formaldehyde and toluene under 5–32% volumetric water content of the root bed.

The removal efficiencies of formaldehyde by common indoor plant species were evaluated under various growing media, including three porous materials (growstone, expanded clay and activated carbon) [198]. It was found that the formaldehyde removal by the root zone was more rapid than the removal by the aerial plant parts. Lu et al. studied four bacterial strains isolated from the biotrickling filter for indoor VOC removal [199], demonstrating high removal efficiencies for formaldehyde, benzene, toluene and xylenes.

8.2 General Air Cleaning Technology

A multidisciplinary panel of experts has recently undertaken a thorough and critical review [200] of the existing fan-driven air cleaning technologies. They concluded that (1) none of the reviewed technologies was able to effectively remove all indoor pollutants and many were found to generate undesirable by-products during operation; (2) particle filtration and sorption of gaseous pollutants were among the most effective air cleaning technologies, but there is insufficient information regarding long-term performance and proper maintenance; (3) the existing data make it difficult to extract information such as Clean Air Delivery Rate (CADR), which represents a common benchmark for comparing the performance of different air cleaning technologies; (4) to compare and select suitable indoor air cleaning devices, a labelling system accounting for characteristics such as CADR, energy consumption, volume, harmful by-products and life span is necessary. For that purpose, a standard test room and condition should be built and studied; (5) although there is evidence that some air cleaning technologies improve indoor air quality, further research is needed before any of them can be confidently recommended for use in indoor environments.

Air cleaners can remove indoor organic compounds, but also generate ozone, another indoor air pollutant [201]. Yu et al. [202] evaluated the effective ozone emission rates and the VOC removal efficiencies of six selected air cleaners by combinations of chamber experiments and modelling studies. They found that the removal of toluene and formaldehyde might have resulted from the adsorption on the filters and the decomposition by the high-voltage electric discharge or the ionisation that generated ozone.

8.3 Bake-Out

The effect of bake-out using a radiant floor heating system on reducing VOC emissions and indoor concentrations in a residential housing unit has been investigated recently [203]. It was shown that there were differences in the time for each material to reach the desired surface temperature, which resulted in different reduction ratio of VOC emissions and concentrations.

8.4 Optimised Ventilation

Indoor air quality is significantly affected by ventilation parameters, such as building recirculation rates (BRR) [204] and airflow profile [205]. A Singapore study recently investigated the impact of BRR on formation of SOAs [204]. They found that number and mass concentrations of SOA decreased significantly at higher recirculation rates primarily due to the lower precursor concentrations, which is confirmed by the study conducted by Fadeyi et al. [206].

8.5 Adsorption

A number of materials have been selected to absorb indoor VOCs, including porous clay heterostructures (PCH) [207], carbon nanotubes [208], metal-organic frameworks [209], activated carbon fibre [210] as well as packed-bed absorber using triethylene glycol [211] and other porous materials [198].

8.6 Photocatalytic Oxidation

In the past decade, a number of studies have been conducted to assess indoor VOC removal by photocatalytic oxidation techniques [27, 212–220]. An excellent review was undertaken by Mo et al. on photocatalytic purification of indoor VOCs [218]. In the paper, the preparation and coating of various photocatalytic catalysts, different kinetic experiments and models, novel measurement methods and reaction mechanisms are reviewed and discussed in detail. However, the by-products resulting from photocatalytic oxidation of indoor VOCs may bring new health risks to exposed people. Mo et al. investigated the by-products of toluene (at ppb level) during photocatalytic oxidation by using PTR-MS. The identified main by-products included benzaldehyde, methanol, acetaldehyde, acetone/propionaldehyde, formic acid/ethanol and acetic acid. They calculated the health-related index and concluded that these by-products may not have negative effects to human health due to their low concentrations.

9 Trends/Perspectives

It is evident from the above sections that:

- The TVOC concept has limited use and must be used with caution [188].
- A compound-to-compound approach of evaluating the health effects of OCIA as suggested by Wolkoff [108] should be explored in place of the TVOC approach.
- More research is required on the role of reactive chemistry in the causation of SBS.
- The roles of ionic species, hydroxyl and peroxide radicals as well as substances absorbed onto particles in the causation of SBS should be researched [221].
- Attempts have been made to estimate OH radicals through modelling and indirect measurements [221], but limited direct measurements have been made to date. Future work is required in this area.
- More research is required in order to understand the health effects of secondary products like ketones, PAN (peroxyacetyl nitrate) and organic acids, which are generated from the reactions of VOCs in indoor air [221].
- More indoor air audits are required in developing nations.
- Need for collaborative approach from environmental scientists and health agencies.
- Need for practical applications of the knowledge gained in the various areas of IAQ.
- Need for development of appropriate indicators for IAQ, such as indoor fungi [222].
- Need for further work on emerging technologies designed to remove indoor VOCs.

10 Concluding Remarks

The past decades have witnessed tremendous growth in indoor air audits, development of a quasi-uniform definition of VOC levels in indoor microenvironments and improvement of concepts for screening indoor air quality and assessing exposure. Significant advancements have been made in emission modelling, source identification, source control, source characteristics and interaction of indoor VOCs with indoor materials, particularly the formation of indoor secondary organic aerosols. Some progress, albeit slow, have occurred in the development of universally acceptable exposure guidelines. The entrance of smaller, faster and smarter instrumentation into the market could enhance fieldwork markedly. However, the link between health/sensory effects and indoor VOC levels is still largely unclear. Further work is urgently required in this area and in the search for insights on the role of reactive chemistry in the generation, degradation and transformation of indoor VOCs.

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The Shift in Human Health from Infection-Related Diseases to Chronic Illnesses and the Importance of Indoor Chemical Exposure

Carl-Gustaf Bornehag

Abstract It has been recently estimated that the pattern of the global burden of diseases – expressed as disability-adjusted life years (DALY) – has changed over the last 20 years and a shift from communicable disorders to noncommunicable disorders has been observed. This shift is more pronounced in high-income countries. Even though there is lack of knowledge regarding the cause(s) behind the increase in chronic diseases/disorders, there are scientifically based suspicions that environmental factors do play an important role in interaction with genetic predisposition. Especially diffuse emissions of endocrine-disrupting compounds (EDCs) from consumer products are a matter of concern. The four reasons for concern with human EDC exposure are:

- The low-dose effect and a non-monotonic dose-response relationship
- Early life sensitivity and the persistency of effects
- The large number of EDC sources in our daily life
- The wide range of health effects

A full chain model is proposed which is following chemicals from their sources over environmental exposures in food, air, and dust over to human uptake and finally to human health effects. The model also includes modifying factors for environmental exposures, different pathways for human uptake, and biological mechanisms involved in health effects. With scientific information in this model risk management should be possible and result in preventive actions in order to reduce children's exposure to health relevant factors.

Keywords Chronic disorders, Consumer products, Endocrine disrupting chemicals, Indoor chemical exposure, Phthalates, Bisphenol A

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Abbreviations

ADHD	Attention deficit hyperactivity disorder
ASD	Autism spectrum disorders
BBzP	Butyl benzyl phthalate
BPA	Bisphenol A
DALY	Disability-adjusted life years
DEHP	Di(2-ethylhexyl) phthalate
DES	Diethylstilbestrol
DiBP	Di-iso-butyl phthalate
DiDP	Di-iso-decyl phthalate
DINP	Di-iso-nonyl phthalate
DnBP	Di-*n*-butyl phthalate
DnOP	Di-*n*-octyl phthalate
EDCs	Endocrine-disrupting compounds
HIV	Human immunodeficiency virus
NCD	Noncommunicable disease
PFC	Perfluorinated chemicals
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate
PVC	Poly vinyl chloride

1 The Shift in Human Health from Infection-Related Diseases to Chronic Illnesses and the Importance of Indoor Chemical Exposure

Infection-related diseases, also called communicable diseases, have always been a burden for humans and in particular for children, and such health effects are estimated to be the major burden of disease and mortality globally [1]. Important examples of such diseases are HIV/AIDS and tuberculosis, diarrhea and lower respiratory infections, neglected tropical diseases and malaria, neonatal disorders, and nutritional deficiencies. People in low-income countries do suffer more from such diseases than people in high-income countries [2]. It is also well known that important actions for decreasing the burden from infection-related diseases are improvements of hygiene where cleaner food and water, better sanitary, and better environment and housing are known to be effective [3].

In a recently published article in Lancet the global burden of disease has been estimated – expressed as disability-adjusted life years (DALY) – for 291 causes in 187 countries [4]. It is estimated that communicable disorders were responsible for 47% of the global burden of disease in 1990, while noncommunicable diseases stood for 43% and injuries stood for 10%. However, one conclusion from this study is that during 20 years, this pattern of diseases has changed. In 2010 the communicable diseases stood for 35%, while noncommunicable disorders were 54% and injuries 11%. The shift from infection-related diseases to chronic diseases is more pronounced in high-income countries where there is an increasing body of evidence documenting a historic shift in the epidemiology of children's health [5].

A noncommunicable disease (NCD) is defined as a noninfectious and non-transmissible disease. NCDs are often chronic diseases with long duration and slow progression, but they may also be a result of more rapid death such as some types of sudden stroke. Important examples of noncommunicable diseases are cancer, chronic respiratory diseases, mental and behavioral disorders, cardiovascular and circular diseases, diabetes, and urogenital disorders.

The fact that children in the richer part of the world do suffer more from different kind of chronic disorders will have a large impact on their entire life and on the public health situation in general. Important examples of increased chronic diseases/disorders among children during the last decades are immunological diseases such as asthma and allergy [6], metabolic disorders including overweight/obesity and diabetes [7], neurodevelopmental disorders such as autism spectrum disorders (ASD), behavior-related problems and intelligence [8], and reproduction-related disorders including malformation of genitals [9]. These different chronic adverse health effects have all increased during recent decades, and the incidence seems to be higher in children than in adults.

Such an increase in childhood diseases and disorders is indicating that early life is of the greatest importance for understanding the causal factors. It is further obvious that developing a chronic disorder in early life has a major impact of the burden of disease for a society and consequently impact on society's costs but also on the life quality for the individuals since it will last for the rest of the life.

In general, we don’t know the reason(s) for the dramatically increase in chronic diseases and disorders in children that have been observed the recent decades. However, there is a scientific understanding that this is a matter of environmental factors in a sophisticated interaction with the individual’s genetic background also including epigenetic mechanisms, meaning changes in gene expression rather than changes in the underlying DNA sequence.

2 Factors of Concern Contributing to an Increase in Chronic Diseases/Disorders

2.1 Environmental Factors of Concern

Even if there is lack of knowledge regarding the cause(s) behind the increase in chronic diseases/disorders, there are scientifically based suspicions that environmental factors do play an important role in interaction with genetic predisposition. Environmental pediatrics, the branch of pediatric medicine that studies the influence of the environment on children’s health, has in the past decade grown exponentially [10]. Rising rates of pediatric chronic disease and growing recognition of children’s extensive exposures and great vulnerabilities to toxic hazards in the environment have catalyzed this expansion. One focus here is on environmental chemicals that have increased dramatically during the last decades, which in fact are during the same period of time that the chronic diseases have increased.

2.2 Shift from Industrial Exposures to Leakage of Chemicals from Every Day Consumer Products

In high-income countries there has during the last decades been a progress in the societal work focusing on reduction of environmental chemicals from large local point emissions such as industrial plants, wastewater systems, and occupational settings including farming, i.e., the “old” kind of chemicals. Because of this development the concerns faced today are more related to the diffuse emissions of chemicals from articles or products that may be harmful for the environment, wildlife, and humans. Most of these new consumer products – such as plastics, cleaning products, cosmetics, packaging materials, toys, textiles, and building materials – include chemicals that permanently leak to the surrounding environment and can be found in food, air, dust, soil, water, etc., and therefore of relevance for human uptake. These kinds of products and articles are related to our daily life and often used indoor, meaning that our homes and nonindustrial workplaces (such as offices, schools, hospitals, and day-care centers) are important places for exposure. Since these sources can be found in our daily life environments, the general population is exposed including more vulnerable groups such as pregnant women and fetuses, infants, and children. Many of these “new” chemicals have been shown to be toxic in experimental studies on animals and cell system as well as in epidemiological investigations, and an increasing body of evidence suggests that they also have impact on human health.

2.3 Indoor Environments Are Important

People in high- and medium-income countries spend more and more time indoors. Studies have shown that adults can be up to 75–80% in nonindustrial indoor setting such as homes, offices, and schools, and the situation for infants and children is that they spend even more time indoors. Furthermore, as said, most of the modern consumer products that includes chemicals of concern are used indoors and most often in our homes. This is true for a huge number of products and building materials as well which are known to be a strong source for modern chemicals in indoor air and dust. Taken together, the amount of time we spend indoors and the fact that the exposures occur indoor mean that indoor environments are of the greatest importance for human exposure to modern chemicals.

2.4 Endocrine-Disrupting Chemicals

Among all the many thousands of chemicals that have been introduced to the environment during the last decades, the endocrine-disrupting compounds (EDCs) are of particular concern. The human endocrine system uses chemical signals or hormones for steering development and reproduction, regulation of body function and metabolism, and influence behavior and immunity [11]. Endocrine disruption occurs when an agent alters hormone signaling or the response to hormone signaling and in so doing alters some aspect of the organism under hormonal control [12]. According to the Endocrine Society, the world’s authoritative scientific association of clinical and research endocrinologists, an endocrine-disrupting chemical (EDC) is an exogenous chemical, or mixture of chemicals, that can interfere with any aspect of human hormone action [13].

Chemicals with proven or suspected endocrine-disrupting properties have been introduced to our environment mainly after Second World War and can be found as constitutes in a huge number of commonly used consumer products and building materials. Examples are a number of phthalates in soft polyvinyl chloride (PVC) and other plastics, bisphenol A in polycarbonatic plastics, flame retardants in textiles and electronical equipments, perfluorinated compounds in textiles, and nonstick cookwares.

3 Children in Low-Income Countries Are More Exposed

In a global perspective there is a difference in exposure for children from low- to high-income countries. In many low-income countries children do suffer from the traditional infection-related diseases, mainly due to the fact that there are limited resources available for improving the hygiene. On top of that they are also exposed to occupational exposures and the "old" industrial chemicals. This is due to the fact that much of the global production of consumer articles and products has been phased out in rich countries (in order to cut salary costs but also due to environmental problems) and moved to low-income countries. A good example is the different kinds of plastics where very much of the production has moved to Southeast Asia. The situation is furthermore problematic since it is not uncommon that children do such industrial work in these countries. Furthermore, children in low-income countries are also exposed to a huge number of "new" chemicals including EDCs. The reasons for this are several. People in low-income countries long for changing their way of life to a western style which in many cases is a matter of consumption of modern articles and products including modern chemicals. Another problem is that the health risks with chemical exposures don't have as much of attention in low-income countries as in high-income countries in Europe, North America, and Japan. This means that children in low-income countries are more exposed to health risks including environmental chemical hazards. They do suffer from infectious diseases due to bad hygiene. On top of that they and their families are exposed to old occupational-related exposures, and they are more and more exposed to modern chemicals related to articles and products with endocrine-disrupting properties. However, all children globally have in common that they are exposed to modern chemicals including compounds with proven or suspected endocrine-disrupting properties which is an issue for concern.

4 Four Topics for Concern for EDC Exposure and Children's Health

Large-scale bio-monitoring programs around the world have provided extensive information on human exposure to industrial chemicals in general and more specifically for EDCs. The overall picture is that the entire population is exposed. It is also more and more evident that the exposure of such new chemicals continues to climb, both in doses and in number of chemicals. A related problem is that humans (and animals) normally are exposed to a number of EDCs in complicated mixtures. The knowledge regarding the health risks with such multiple exposures is limited, and an important issue for future is to understand the health risks with multiple exposures to compounds that exist below their limit values, a knowledge that is limited today.

A major concern is that there are an increasing number of scientific studies – both in epidemiology and in in vitro/in vivo experiments – reporting that there might be a relation between EDC exposures and adverse health effects in the general population. Such recent data can be exemplified with one of the most studied chemical with endocrine-disrupting properties, bisphenol A [14]. Such scientific activities during the last decades have identified at least four circumstances as topics for concern for EDC exposure, which makes EDC exposure different than other types of health risks related to chemicals. The four reasons for concern with human EDC exposure are:

- The low-dose effect and a non-monotonic dose-response relationship
- Early life sensitivity and the persistency of effects
- The large number of EDC sources in our daily life
- The wide range of health effects

4.1 The Low-Dose Effect and a Non-monotonic Dose-Response Relationship

Studies of endocrine-disrupting chemicals during the last decade have challenged the traditional concepts in toxicology, namely the very old statement that “the dose makes the poison” which is also built upon the idea of a monotonic dose-response relationship. Today we have scientific evidence that EDCs can have effects at low doses, i.e., doses that are typical for the general population and doses that are below those used in traditional toxicological studies [11]. Additionally, more and more evidence shows that there might be non-monotonic dose-response relationships [15–17]. In the recent review by Vandenberg and others – which is an update of the 10-year-old National Toxicology Report on endocrine disruptors [18] and in part an update of the Chapel Hill document on bisphenol A [19] – a weight-of-evidence approach is provided to conclude the current state of the art [11]. In this review they provide hundreds of studies from cell cultures and animal models as well as human epidemiological investigations and show that low-dose effects as well as non-monotonic relationships are rather common reported. One strong evidence for a low-dose effect is the increasing number of epidemiological studies showing that existing (low) levels in humans can be related to adverse health effects such as overweight and obesity [20], infertility [21], behavioral development in children [22], and immunological dysfunctions [23]. These evidences indicate that there is a strong need for changes in the risk assessment procedures which means that the effect of low doses cannot be predicted by effects observed at high doses and that we have to consider non-monotonic dose-response relationships. These phenomenons are not covered in the risk assessment used today.

4.2 *Early Life Sensitivity and the Persistency of Effects*

There is also a growing knowledge indicating that EDC exposure is more important during development in the prenatal and infancy period [11]. Furthermore, scientific reports during the last decade have shown that effects related to EDCs can be observed long time after the exposure did occur, where animal models and human studies have shown effects in offspring; see dozens of studies reported in the Vandenberg et al. [11] paper. This has been known for a long time due to the experiences from the use of diethylstilbestrol (DES) and thalidomide (man-made estrogens) as drugs for pregnant women where no adverse effects could be seen in the pregnant women who took the drug but in their offspring, sometimes decades later as in the case with DES [24]. Such delayed effects are now indicated even for other compounds with the same properties. This fact is also stressing for new thinking about how to do risk assessment of chemicals with endocrine-disrupting properties.

4.3 *The Large Number of EDC Sources in Our Daily Life*

Man-made chemicals with suspected or proven endocrine-disrupting properties are used in a very large number of commonly used products, and they may leak into the surrounding environment [25–27]. Important examples are phthalates, widely used as plasticizer in polyvinyl chloride (PVC) and in numerous consumer products such as baby infant care products, toys, cosmetics, food packing materials, and cleaning products [25, 26, 28]. Recent data have further shown an increase in exposure for several phthalates, e.g., di-iso-nonyl phthalate (DINP) that have replaced di (2-ethylhexyl) phthalate (DEHP) and bisphenol A, a high-volume production chemical used in polycarbonate plastic and resins. Of particular concern is the use of bisphenol A (BPA) in food containers, baby bottles [19], and many other products. Another example is perfluorinated chemicals (PFCs), a group of chemicals known for their bioaccumulating properties and high persistency beside the risk for endocrine effects. PFCs (e.g., perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS)) are used in many products such as stain-resistant carpets, clothes and floor wax, and products that resist heat, oil, stains, grease, and water [25]. These examples show that there are numerous sources in daily life, meaning that "everyone" is exposed, including pregnant women, fetus, infants, and children.

4.4 *The Wide Range of Health Effects*

Epidemiological and experimental in vitro/in vivo studies have reported a possible relationship between EDC exposure and multiple chronic illnesses such as reproduction and infertility [29], asthma and allergy [30], overweight/obesity

[31], and neurodevelopmental disorders such as autism spectrum disorders (ASD) and attention deficit hyperactivity disorder (ADHD) [32]. These are examples of chronic adverse health effects that have increased among children during the last 5–6 decades, which is about the same period of time as an increase in the use of EDCs has happened. A more comprehensive description of the literature is provided in the mentioned review by Vandenberg et al. [11].

5 The Full Chain Model Following EDCs from Their Sources to Health Effects

As pointed out earlier we know that humans are surrounded by articles and products including chemicals with suspected or proven endocrine-disrupting properties and that such compounds leak to the surrounding environment. We also know that such compound routinely can be found in food and in the environment including indoor air and dust as well as outdoor in air, soil, water, etc. It is furthermore evident that humans are exposed since EDCs routinely can be measured in fluids such as blood, urine, breast milk, and amniotic fluids, in the entire population.

When establishing new scientific knowledge on the importance of environmental toxicants for human health and how to reduce such health risks, it might be fruitful to use a “full chain model” described in Fig. 1 [33].

The base of the model is divided into three main parts. The most triangle to the very left describes the environmental exposure (A) focusing on sources and environmental exposures in food, air, and dust, etc. In order to understand the environmental exposures we should also focus on factors that may modify the environmental exposures. Examples of such modifiers are air exchange rate, temperature, relative humidity, and not at least the presence of surfaces indoors with an adsorption potential for these kinds of compounds. The triangle to the very right (C) is focusing on different health effects that can be related to environmental toxicants. Of the greatest importance for understanding how chemicals impact on human health is the biological mechanisms in action. Finally, the triangle in the middle (B) describes the interface between the environment and humans with focus on human uptake of chemicals and different pathways. There is a strong need for new information on how these kinds of chemicals reach our bodies and on which pathways they act. Data from such a full chain is essential input for risk governance activities (D) described in the top of the model which may lead to prevention activities focusing on the sources in order to reduce or elimination of health relevant exposures.

Taking the example of phthalates – which is one of the most studied chemical groups with endocrine disrupter properties – there is scientific data for following up such compounds through the entire chain in Fig. 1. Phthalates have been used in a large variety of industrial and consumer applications for more than 5 decades. They are the most common compounds used as plasticizers worldwide, primarily in soft

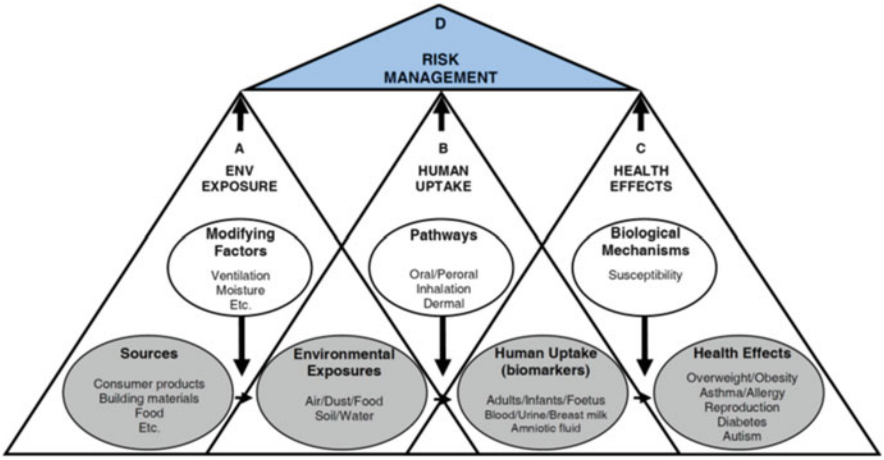

Fig. 1 The full chain model following chemicals from their sources over environmental exposures in food, air, and dust over to human uptake and finally to human health effects. The model also includes modifying factors for environmental exposures, different pathways for human uptake, and biological mechanisms involved in health effects. With scientific information in this model, risk management should be possible and result in preventive actions in order to reduce children's exposure to health relevant factors

polyvinyl chloride (PVC), where they can account for up to 40% within the final product, but are also constitutes in a huge number of other commonly used consumer products. The annual production volume of phthalates in Western Europe alone is currently around one million tons.

Exposure to chemicals like phthalates can occur through a variety of sources, such as foodstuff, water, air, dust, and the use of a huge number of consumer and personal care products. Phthalates are constantly released to the surrounding environment since they most often are not bound to the chemical structure of the article including them. Such release can go on for the entire life for the product. From all these sources, phthalates end up in the human body via three pathways: ingestion, inhalation, and/or dermal absorption.

Foodstuff is reported to be the major source of phthalate exposure, particularly for the long-chain phthalates such as di(2-ethylhexyl) phthalate (DEHP) and di-iso-nonyl phthalate (DINP). These kinds of phthalates are primarily used in PVC polymer applications. For short or moderate chain length phthalates such as butyl benzyl phthalate (BBzP), di-*n*-butyl phthalate (DnBP), and di-iso-butyl phthalate (DiBP), additional pathways may be of relevance. These compounds are often used also in non-PVC applications such as personal care products, paints, adhesives, or enteric-coated tablets. Of the greatest importance is that more and more data are showing that children in general are exposed to higher phthalate doses than adults which could be a result of different behavior in that children is mouthing, e.g., plastics, and may ingest dust [27]. Furthermore, recent data is suggesting that the dermal uptake of phthalates and other semi-volatile organic compounds has been underestimated and that transdermal permeation of compounds such as phthalates may have been under estimated [34, 35].

In the European Union, the use of phthalates in materials and articles intended to come into contact with food is restricted and standard migration limits apply, e.g., 0.3 mg DBP/kg food simulant or 1.5 mg DEHP/kg food simulant. DEHP, DnBP, and BBzP are banned from toys and in childcare articles. Di-n-octyl phthalate (DnOP), DINP, and di-iso-decyl phthalate (DiDP) are approved only for such toys that cannot be placed in the mouth of children. All substances classified as toxic to reproduction categories 1 and 2 (which applies to DEHP, DnBP, DiBP, and BBzP) are banned in cosmetic products and restricted in preparations such as paints and varnishes for end-consumers [27].

5.1 Following Phthalates from Sources to Health Effects

Regarding *sources* we now know that humans are surrounded by articles and products including phthalates [25, 27] as well as food [36] and that these consumer goods emit phthalates to the surrounding environment. We further know from dozens of studies that phthalates can be routinely found as *environmental exposures* in air and dust indoor [37] which are not strange since the sources are there. *Human uptake* is furthermore evident since EDCs routinely can be measured in blood and urine (but also in breast milk and amniotic fluids) in the entire population supported by data from, e.g., the US NHANES study described in a large number of scientific papers, and there are recent data showing that children are more exposed than adults [27]. Finally, we have experimental data from animal models and cell studies as well as epidemiological data showing evidence for a possible relationship between phthalate exposure and several *health* effects in humans [11]. Major health endpoints are reproduction-related disorders, cancer, neurodevelopmental disorders including behavior problems, metabolic disorders including overweight and obesity (and diabetes), and immunological responses such as asthma and allergy.

We now also have scientific data showing that products/articles in fact emit phthalates to indoor air and dust. PVC as flooring material in the home [26] and the use of different polishing products [38] have been shown to increase phthalate concentrations in indoor dust. There is data showing that what's in the air and dust indoor can be related to human uptake of phthalates. American data showed that phthalates in the air of the homes could be related to phthalate metabolite levels in the urine of pregnant women [39, 40], and in German data [41] as well as in Taiwan data [42], it has been reported that dust concentrations of selected phthalates could be related to urinary concentrations of phthalate metabolites in adults and children. Articles and products including phthalates have also been related to uptake in humans as in the case of medical equipment for premature children [43], baby skin care products [44], food packaging products [36], and PVC as flooring material [45]. Finally, both consumer products and building materials including phthalates

[30] as well as environmental exposures of phthalates in indoor air and dust [42, 46, 47] and urinary levels of phthalates [42, 48] have been associated to asthma and allergy in children.

6 Do We Have Enough of Evidence for Actions in Order to Reduce EDC Exposure?

As shown we currently have scientific data making it possible to follow several EDCs from sources to possible health effects from such exposure. We now know quite a lot about the sources, we know that these compounds can be found in the environment that surround us, we know that all humans have these compounds in their bodies including fetuses and infants, and we have finally indications for chronic health effects in humans and specifically for chronic diseases/disorders in children. In the case of phthalates, we also have scientific indications for human uptake of butyl benzyl phthalate from PVC flooring materials, a compound that is banned in toys for children of health risk reasons. With such information from a full chain model, one important question is if we are enough prepared for taking action in order to reduce the exposure. If not, we have to more precisely declare what more information we need.

7 Conclusions

- Communicable diseases, also called infection-related diseases, are a burden for humans and specifically for children and are recently estimated to be the major reason for children's burden of disease and mortality globally.
- There is an ongoing global shift (mainly driven by the situation in high-income countries) in the epidemiology of children's health with a change from infection-related diseases to chronic illnesses during the last 50 years.
- The reason for the increase in chronic diseases in children is not clear, but environmental chemicals are suspected, and there is a particular concern related to the exposure for chemicals with endocrine-disrupting properties.
- Today we know that fetuses and children are exposed to EDCs globally and that such exposure is related to consumer products and articles that most often are used in our nonindustrial indoor environments such as homes, day-care centers, and schools.
- There are at least four reasons for concern about the exposure for EDCs including that the health effects can be related to low-dose exposure and non-monotonic dose-response relationships; that early life is of the greatest importance for exposure and that the effects may be delayed and may go over generations downstream; and that everyone is exposed since the sources for

EDC exposure is found in our everyday life in consumer products, and if there are endocrine mechanisms involved, we can expect multiple health effects as a result of such exposure.
- There is a concern expressed by many scientists that the current methods for risk assessment of chemicals doesn't cover the abovementioned problems with low-dose exposure and non-monotonic dose-response relationship as well as delayed effects.
- We have today scientific data and evidence for following up several EDCs (such as phthalates) from the sources over to environmental exposures and further to human uptake of such compounds, and finally there is data showing associations to adverse health effects from such exposure.
- On this background, maybe the most important question is how much more of evidence do we need before we act and reduce the exposure significantly for children globally?

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Household Air Pollution Related to Solid Cookfuel Use: The Exposure and Health Situation in Developing Countries

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Abstract About half of the world's population relies on traditional fuels such as biomass (wood, agricultural residues, animal dung, and charcoal) as the primary source of domestic energy. Nearly 2 billion kilograms of biomass is burnt everyday in developing countries. Use of open fires for cooking and heating exposes an estimated 2.8 billion people in the world to enhanced concentrations of particulate matter and gases, greatly in excess of the respective, current WHO air quality guidelines for 24-hour and annual mean concentrations. Household air pollution (HAP) from solid cook-fuel use is now estimated to be responsible for nearly 3.5 million deaths annually, with some of the highest disease burdens experienced by countries in South Asia and sub-Saharan Africa.

An understanding of the linkages between household fuel use and human health is especially crucial for developing strategies to improve the household environment and the status of public health as they form an important prerequisite for all subsequent economic development. This chapter is devoted to describing the sources, emissions, and patterns of exposure and consequent health risks for solid cookfuel use in developing country settings. Potential intervention options for addressing household air pollution and household energy issues within the mainstream of environmental health and public health policies of the region are also described.

Keywords Biomass fuels, Burden of disease, Coal, Developing countries, Household air pollution, Wood

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Contents

Abbreviations

ALRI	Acute lower respiratory infection
AQG	Air quality guidelines
ARI	Acute respiratory infection
CO	Carbon monoxide
COPD	Chronic obstructive pulmonary disease
CRA	Comparative risk assessment
DALYs	Disability-adjusted life years
ESMAP	Energy Sector Management Assistance Program
GBD	Global burden of disease
GM	Geometric mean
HAP	Household air pollution
IAP	Indoor air pollution
IARC	International Agency for Research on Cancer
m^3	Cubic meter
mg	Milligram
PAHs	Polycyclic aromatic hydrocarbons
PM_{10}	Particulate matter less than 10 μm in aerodynamic diameter
$PM_{2.5}$	Particulate matter less than 2.5 μm in aerodynamic diameter
ppm	Parts per million
USEPA	Unites States Environmental Protection Agency
WHO	World Health Organization
μg	Microgram
μm	Micrometer

1 Background

Around 2.8 billion people, mostly in the least developed and developing countries, rely on solid fuels such as biomass (wood, agricultural residues, and animal dung), charcoal, and coal for cooking and other household energy needs [1] (Fig. 1). Use of such solid fuels in inefficient devices, often in kitchens that are poorly ventilated, results in very high exposures to multiple toxic products of incomplete combustion [2], exposing families, in particular women and children, to toxic smoke for several

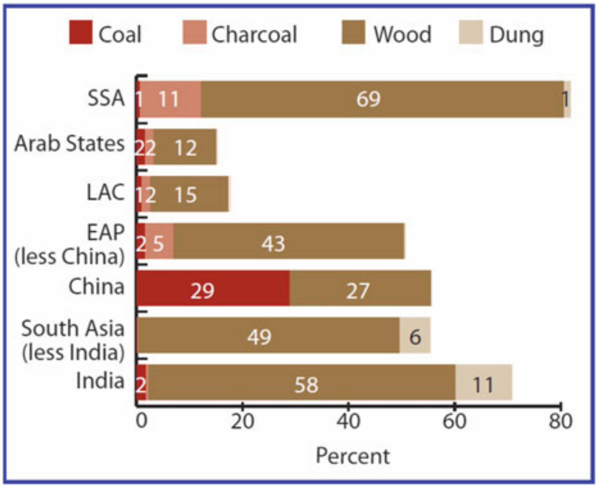

Fig. 1 Household fuel use across world regions [81]

hours every day over their lifetimes. The comparative risk assessment (CRA) exercise conducted by the World Health Organization (WHO) in 2002 estimated that exposure to indoor smoke from solid fuels may be annually responsible for about 1.6 million premature deaths in developing countries and 2.6% of the global burden of disease (GBD) [3]. This estimate has been recently revised as part of the GBD 2010 exercise [4]. Household air pollution (HAP) from solid cookfuel use is now estimated to be responsible for 3.5 million premature deaths annually, with some of the highest burdens experienced by countries in South Asia and sub-Saharan Africa.

Given the wide spread prevalence of solid fuel use and the strength of the scientific evidence of health impacts associated with exposures to emissions from solid fuel use, HAP issues in rural households of developing countries are of tremendous significance from the stand point of public health. An understanding of these linkages is especially crucial for developing strategies to design interventions that can improve the household environment and facilitate subsequent economic development. The following sections of this chapter are devoted to describing the nature of emissions, patterns of exposure, and consequent health risks from solid cookfuel use (primarily biomass) in developing country settings. Although coal use is fairly widespread in some parts of the world (especially China), much of the exposure and health evidence presented in this chapter has been drawn from studies conducted in biomass cookfuel using settings. A brief discussion on intervention options and future research directions is also included.

2 Characteristics of Solid Cookfuel Smoke

Air pollutants derived from solid cookfuels are the result of incomplete combustion (conditions for efficient combustion of these fuels are difficult to achieve in the typical household scale stoves). Hundreds of different chemical substances are

Table 1 Toxic pollutants from biomass combustion and potential for toxicity

Pollutant	Known toxicological characteristics
1. Particulates (PM 10, PM 2.5)	Bronchial irritation, inflammation increased reactivity, reduced mucociliary clearance, reduced macrophage response
2. Carbon monoxide	Reduced oxygen delivery to tissues due to formation of carboxyhemoglobin
3. Nitrogen dioxide	Bronchial reactivity, increase susceptibility to bacterial and viral lung infections
4. Sulphur dioxide	Bronchial reactivity (other toxic end points common to particulate fractions)
5. Organic air pollutants	Carcinogenicity or cocarcinogenicity
Formaldehyde	Mucus coagulation, cilia toxicity
Acetaldehyde	Increased allergic sensitization, increased airway reactivity
Phenols	
Phenols pyrene	
Benzo(a)pyrene	
Benzopyrenes	
Dibenzopyrenes	
Dibenzocarbazoles	
Cresols	

Source: [5, 6, 10]

emitted during the burning of solid cookfuels in the form of gases, liquids (suspended droplets), or solids (suspended particulates) [5, 6]. However, the amount and characteristics of pollutants produced during the burning of solid cookfuels depend on several factors including composition of original fuel, combustion conditions (temperature and air flow), mode of burning, and even shape of the fireplace [6, 7]. Gases and vapors are mainly emitted as a combination of carbon monoxide, water vapor, small amounts of nitrogen dioxide, sulfur dioxide, and low/high molecular weight organic compounds. The aerosol (mixture of very fine solid and liquid particles) is made up of organic and inorganic matter. The organic matter is often referred to as "tar" in tobacco literature and "suspended particulates" in air pollution literature. A significant fraction of the particulates are in the respirable range (2–10 μm in aerodynamic diameter) and can penetrate deeply into the lungs [8]. The inorganic matter is mostly ash and is produced in very small amounts during regular stove combustion.

Unlike biomass, coal often can contain intrinsic contaminants such as sulfur, arsenic, silica, fluorine, lead, and mercury. During combustion these inorganic contaminants are not destroyed but are released into the air in their original or oxidized form. Also, because coal burns at a substantially higher temperature than biomass, substantially higher emissions of gases have been shown to result from coal combustion [9].

Wood smoke has been shown to contain 17 pollutants designated as priority pollutants by the USEPA because of their toxicity in animal studies, up to 14 carcinogenic compounds, 6 agents toxic to cilia and mucous-coagulating agents, and 4 cocarcinogenic or cancer-promoting agents (Table 1) [10]. Carcinogenic PAHs, methylated PAHs, and nitrogen-containing heterocyclic aromatic compounds have been reported in the particles emitted from bituminous (smoky) coal combustion [11].

3 Household Air Pollution Levels in Solid Cookfuel Using Households

3.1 Approaches to Estimate Household Concentrations and Exposures

Several methods and technologies have been used to estimate exposures to HAP. In the absence of a regulatory framework in most countries (stipulating for example a specific protocol for routine monitoring), the choice of the method has been dictated by research needs such as characterization of exposures for a population or landscape, linking exposures with specific health risks, assessing the impact of an intervention, deriving dose–response relationships, or others. In developing country settings, approaches used to estimate exposure have varied from simple reporting of household fuel use on questionnaires to multiyear monitoring efforts with thousands of direct exposure measurements with methods varying by precision and geographic scale, as well as cost/resource intensity. Figure 2 shows a hierarchy of such approaches used in HAP exposure assessment which is briefly described below.

3.1.1 Qualitative Exposure Assessment

As direct quantitative measurements of HAP exposure are difficult to perform, a common approach to making broader estimates of exposure has been to use qualitative indicators as proxies or predictors of exposure levels. Previous burden of disease estimates, for example, have relied on this approach [13]. Commonly used indicators include fuel/stove type, housing type, kitchen type and location, cooking location (indoor/outdoor), and others. The benefit of such an approach is that data on these many indicators are often available from census or other large-scale demographic surveys (tier #1; Fig. 2) or can be relatively easily obtained with additional simple surveys at modest costs (tiers #2, 3; Fig. 2).

3.1.2 Microenvironmental Monitoring and Exposure Reconstruction

Air pollution measurement studies with devices set in stationary positions in the house or in the ambient environment (tier #4, Fig. 2) afford more accuracy but are much higher in cost as compared to studies using survey-based data. Since these studies provide only household concentrations, exposures are reconstructed by using a combination of microenvironmental concentrations measured in locations where the study participant spends most of his/her time (or where significant contributions are expected to come from) and integrating across the time spent in those environments [14–17].

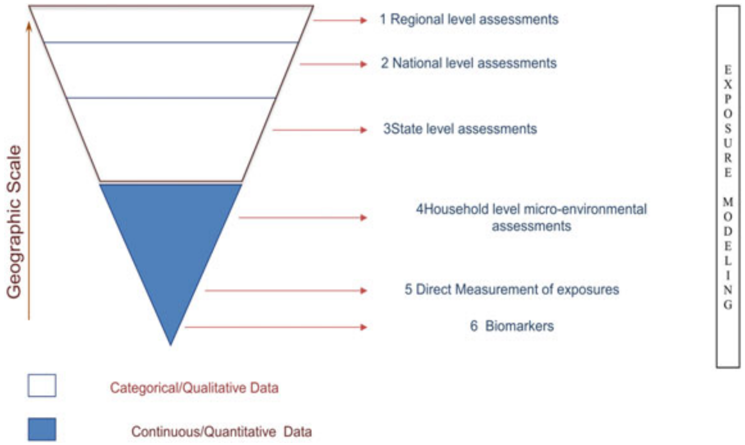

Fig. 2 The exposure assessment pyramid: a hierarchy of exposure assessment methods used by HAP studies in developing countries with modeling methods straddling across multiple or all levels (Adapted from [12])

3.1.3 Direct Measurements of Personal Exposures

The most direct method for monitoring personal exposure is to use instruments which can be placed on a participant, preferably in the breathing zone (tier #5, Fig. 2). Direct measurement of personal exposure is generally considered to provide the most accurate estimate and has been employed in several household energy studies [18–21]. Direct measurement can be especially critical for understanding exposure patterns typical of the household energy sector, which are characterized by highly variable contributions from a variety of factors.

3.1.4 Short- and Long-Term Exposure Assessment

Direct measures of exposure to household air pollution are usually limited to specific time-points, which can provide a reasonable indication for short-term exposure. In such cases where exposure monitoring is limited to single cross-sectional or before/after measurement of a sample group, the temporal changes in exposure in relation to time-varying exposure determinants may not be adequately addressed and fail to capture representative exposures for the sample or population.

Larger epidemiological studies looking at health impacts of household air pollution often need to rely on long-term exposure assessment with enough direct measurements over sufficiently long time periods together with models to address heterogeneity within or between individuals and groups as has been done in recent studies [22].

3.1.5 Measurement of Biomarkers

Biomarkers are often considered to be a more reliable measure of absorbed dose (tier# 5, Fig. 2), due to interindividual differences that may be caused by factors such as ventilation volume and breathing rate from similar environmental concentrations. Some common biomarkers that have been evaluated include hydroxylated polycyclic aromatic hydrocarbons [23, 24], methoxyphenols (MPs) [25], and levoglucosan [26, 27]. However, these exposure assessment procedures are subject to limitations. These include the cumbersomeness and inconvenience associated with deploying monitoring equipments, difficulties in obtaining biological samples, storage and preservation of samples, and accounting for confounding exposures. Because of these limitations few biomarkers have been assessed with regard to cookfuel smoke exposure as compared to environmental exposure measurements and their routine use remains to be validated.

3.2 Household Concentrations and Exposures Related to Solid Cookfuel Use

Well over 100 studies over the last two decades have assessed HAP levels in relation to cookfuel use. Table 2 provides a listing of select studies that illustrate the breadth of exposure measurements that have been performed across world regions. As may be seen in Table 2, over the last several years, there has been a continuous evolution of methods and protocols for assessing exposure to HAP. The quantity and quality of information collected has also become considerably more detailed, with single pollutant, cross-sectional studies measuring area concentrations being supplemented by multi-pollutant, longitudinal exposure monitoring studies as part of intervention trials.

Combining results from individual studies to develop consistent exposure estimates has however been challenging due to differences in measurement protocols, types of summary measures reported, the types or nature of household level determinants explored, and quality control criteria used for sampling and analysis. To overcome these challenges, a global database was developed in 2000 to extract results of quantitative measurements reported from approximately 110 studies in China and 70 studies from developing countries in Asia, Latin America, and Africa [63, 82]. In view of the large number of new studies, this database has been recently updated to include studies published through 2011 (Balakrishnan et al., “Global Household Air Pollution Database: Household concentrations and exposures from cooking fuels” Version 2.0, September 2012, Sri Ramachandra University, University of California, Berkeley, World Health Organization; url to come). While a comprehensive description of the studies in the database is beyond the scope of this paper, Figs. 3 and 4 provide a representative range of household concentrations and exposures encountered in these settings.

Table 2 Summaries from select exposure assessment studies reporting detailed results from measurements of household air pollution related to solid fuel (primarily biomass) use

Author references together with study locations	Study designs/methods	Implications of findings
Balakrishnan (India) Andresan (India) Colbeck (Pakistan) Begum (Bangladesh) Kurmie (Nepal) Gao (Tibet) Albalak (Mexico)	● Cross-sectional studies that measured 24 h household area concentrations and/or personal exposures ● Many studies collected samples from multiple villages/rural habitations to generate representative exposure profiles for the population at hand	● Provided estimates of daily average area concentrations across a wide range of household configurations using a variety of cooking fuels including biomass (such as wood, dung, crop residues) kerosene, LPG, and electricity ● Few also estimated exposures for women, children, and men for PM and/or CO
Saksena (India) Jin (China) He (China) Lan (China)	● Repeat cross-sectional studies to address seasonal and/or spatial variation and contributions from heating ● Colocated area measurements for multiple pollutants	● Provided estimates of daily average concentrations for multiple pollutants while addressing spatial and seasonal variations across multiple household configurations including estimates for coal use for cooking and/or heating ● Intra and inter-household variations recognized to be influenced by multiple household level variables
Balakrishnan (India) Dasgupta (Bangladesh) Baumgartner (China)	● Cross-sectional studies that combined household level area measurements with questionnaire-based categorical information on multiple household level variables for inclusion in models	● Provided modeled estimates of household concentrations and/or exposures on the basis of household level characteristics for use in long-term exposure reconstruction ● Modest levels of prediction from models
Ezzati (Kenya) Dionisio (The Gambia) Smith (Guatemala) Bautista (Dominican Republic)	● Longitudinal studies that monitored 24–48 h personal exposures and area concentrations to address spatial and temporal variations	● Provided estimates of variability in daily average/peak personal exposures to individual or multiple pollutants as well as correlations between pollutant exposures for women and infants
Jiang (China) Khalequzzamman (Pakistan)	● Cross-sectional studies that combine data on household and community level measurements to address rural–urban source/exposure differentials	● Provided evidence of community level exposures, including exposures in urban low-income communities
Neher (Guatemala) Clark (Honduras) Pennise (Ghana, Ethiopia) Zuk (Mexico)	● Cross-sectional or paired before–after studies to evaluate reductions in area concentrations after introduction of improved biomass cookstoves	● Provided estimates for percent reductions in household concentrations of PM and/or CO with use of improved cookstoves together with reductions in fuel consumption
Smith (HEH project – India, Mexico) Edwards (China)	● Paired before–after study design with a common protocol across multiple sites using improved biomass cookstoves to assess reductions in household concentrations of PM and CO	● Evaluated multiple improved cookstove programs for emission and exposure reductions under field use conditions and provided important insights into contributions from user preference/behavior for efficacy of interventions

(continued)

Table 2 (continued)

Author references together with study locations	Study designs/methods	Implications of findings
Smith (Guatemala) Thompson (Guatemala) Dix-Cooper (Guatemala) McCracken (Guatemala) Baumgartner (China)	• Longitudinal monitoring of area concentrations and personal exposures to CO and/or PM to obtain individual estimate of exposure for control and intervention arms of improved biomass cookstove RCT in Guatemala	• Provided personal exposure estimates for use in generation of continuous exposure–response functions for outcomes including birth weight, ALRI, BP, ST segment depression, and neurodevelopment
McCracken (Guatemala) Cynthia Armendariz Arnez (Guatemala)	• Modeling studies that used data from longitudinal monitoring studies to make comparisons of single vs. multiple and/or group vs. multiple measures of exposures	• Highlighted the need for repeated long-term measures to address uncertainties in long-term exposure reconstruction
Cynthia Armendariz Arnez (Guatemala)	• Cross-sectional studies that compare particle size distributions in relation to kitchen concentrations and personal exposures among and traditional and improved biomass cookstove users	• Highlighted the potential for significant bias if the shift in size distribution and the change in relationship between indoor air concentrations and personal exposure concentrations are not accounted for between different stove types
Northcross (Guatemala) Kumie (Ethiopia) Lisouza (Peru) Ansari (India) Colbeck (Pakistan) Fullerton (Malawi)	• Cross-sectional studies that measure different air toxics	• Provided estimates of area concentrations and/or exposures to PAHs, VOCs, NO_2 dioxins, metals, and other air toxics

Table references: [14–20, 28–62]

The available exposure studies collectively provide measurement results for short-term and 24-hour concentrations and exposure estimates for a wide cross section of *rural* homes using a variety of household fuels under a range of exposure conditions across multiple countries. Collectively, these studies have shown HAP exposures to be consistently in excess of pollutant-specific air quality guideline values provided by WHO (WHO-AQGs). The exposures are also quite heterogeneous with multiple household level variables such as fuel, fuel quantity, stove or meal type together with household layout, family size, and location of cooking and ventilation directly influencing household concentrations with additional contributions from time–activity profiles of individual household members for personal exposures. In addition variables such as household income and education, together with energy market structures, sociocultural preferences, and geographical location, affect HAP exposures indirectly through their influence on household energy choices. While most of solid fuel use-related emissions occur indoors, it is important to recognize that the exposures may occur indoors and outdoors [14, 15, 18]. Further, solid fuel use and accompanying exposures are not uncommon among the urban poor [64]. Limited information is currently available, however, to assess the scale and levels of such exposures.

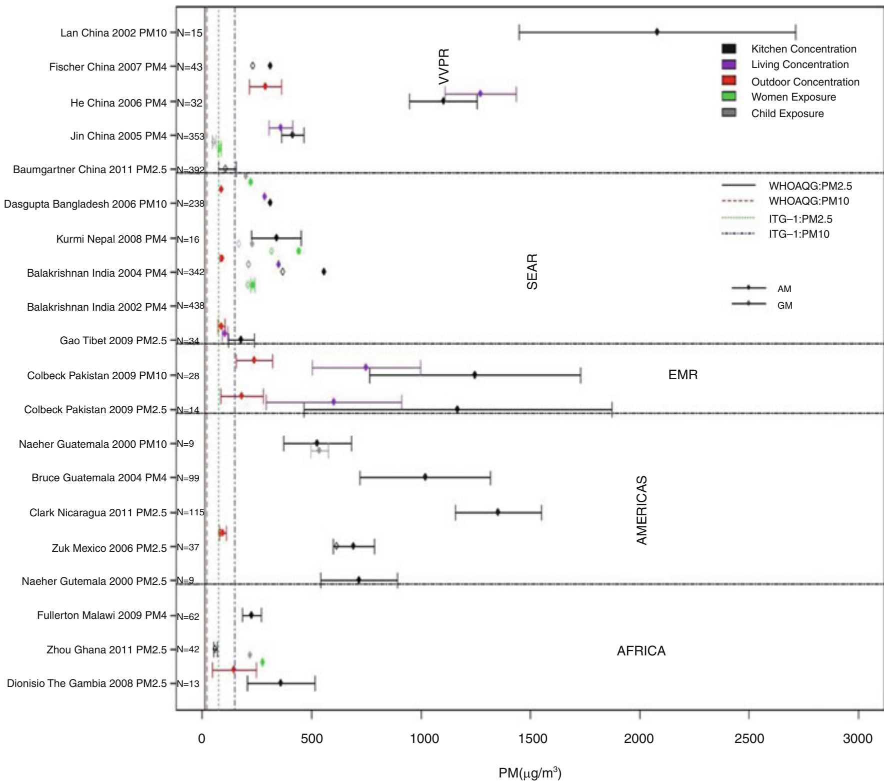

Fig. 3 Reported 24 or 48 h means and standard deviations for area concentrations and personal exposures for PM among households using solid cookfuels in traditional stoves in relation to the annual WHO AQG and ITG levels for PM 10 and PM 2.5 from select studies. *Note*: PM expressed in μg/m$_3$

4 Health Effects Associated with Solid Cookfuel Smoke Exposure

Evidence for health effects associated with exposure to smoke from combustion of biomass fuels was provided initially by studies on outdoor air pollution as well as by studies dealing with exposure to environmental tobacco smoke. Criteria documents for outdoor air pollutants published by the USEPA [8] for example detail the effects of many components including particulate matter, carbon monoxide, oxides of sulfur and nitrogen, and PAHs.

Considerable scientific understanding now exists about aerodynamic properties of the particles that govern their penetration and deposition in the respiratory tract. The health effects of particles deposited in the airway depend on the defense mechanisms of the lung such as aerodynamic filtration, mucociliary clearance, and in situ detoxification. Since most particulate matter in biomass fuel smoke is

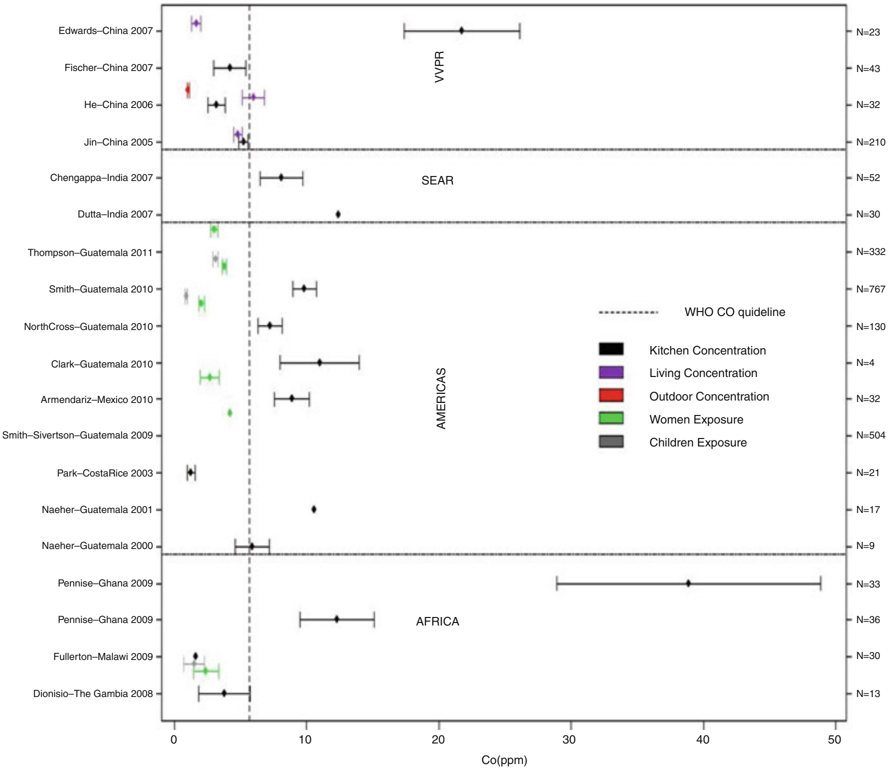

Fig. 4 Reported mean and S.D of 24-h area concentrations and/or 24 exposures to CO among households using solid cookfuels in traditional stoves, in relation to WHO AQG levels for CO from select studies

less than 3 μm in diameter, it is possible that such particulate matter may reach the deepest portions of the respiratory tract and alter defense mechanisms. Several biomass fuel combustion products may also impair the mucociliary activity and reduce the clearance capacity of the lung resulting in increased residence time of inhaled particles including microorganisms and favors their growth. In situ detoxification, the main mechanism of defense in the deepest non-ciliated portions of the lung may also be compromised by exposure to components of biomass fuel smoke [65].

Gases such as carbon monoxide are known to bind to hemoglobin that reduces oxygen delivery to key organs and may have important implications for pregnant women, with developing fetuses being particularly vulnerable. Although emissions of other gases such as sulfur dioxide and nitrogen dioxide are of lesser concern in biomass combustion (very high levels of sulfur dioxide may be reached with other solid fuels such as coal), they are known to increase bronchial reactivity. Polycyclic aromatic hydrocarbons such as benzo[a]pyrene are known carcinogens.

On an epidemiological platform, the earliest evidence linking biomass combustion, indoor air pollution, and respiratory health came from studies carried out in Nepal and India in the mid-1980s. Since then, there has been a steady stream of studies linking HAP and a range of health effects especially in women who cook with these fuels and young children. Recent systematic reviews describe the evidence for acute lower respiratory infections, chronic obstructive lung disease, cataracts, and lung cancer in adults [66, 67]. Recently, the International Agency for Research on Cancer [68] concluded that indoor emissions from household combustion of coal are carcinogenic to humans (group 1) and that indoor emissions from biomass, primarily wood, were classified as probable human carcinogens (group 2A). Although cardiovascular disease has not been studied in relation to HAP, studies concerning combustion particles in relation to outdoor air pollution, environmental tobacco smoke, and active smoking strongly suggest a similar impact from household fuels as well [69–72]. Additional evidence for other impacts is also now emerging, including low birth weight, child cognitive function, and tuberculosis [37, 73, 74].

The global burden of disease (GBD) attributable to solid cookfuel use was initially estimated by the World Health Organization in 2002 [3]. Revised estimates for 2010 have been performed by a global consortium of investigators lead by the Institute for Health Metrics and Evaluation, USA [4]. The approaches used in the two estimations although similar involved some important additions. The 2010 burden of disease was calculated using a stepwise process that involved:

1. Estimation of distribution of exposure to solid cookfuel smoke in the population (using global solid fuel use models that provided the proportion of households using solid fuels for cooking, i.e., coal, wood, charcoal, dung, and agricultural residues from world regions).
2. Choosing health outcomes with strong epidemiological evidence for association with HAP (this included lower respiratory infections; trachea, bronchus, and lung cancers; interstitial heart disease (IHD); cerebrovascular disease; chronic obstructive pulmonary disease (COPD); and cataracts).
3. Estimation of the relative risk per unit of exposure for each outcome (using integrated exposure–response curves for lower respiratory tract infection, IHD, and stroke; meta-analysis estimates for cataracts, COPD, and lung cancer).
4. Choosing an alternative (counterfactual) exposure distribution (defined as when all households use clean fuels for cooking (vented gas, electricity) termed as the theoretical-minimum-risk exposure distribution.
5. Calculation of the attributable disease burden by comparing the present distribution of exposure to the theoretical-minimum-risk exposure distribution in the population.

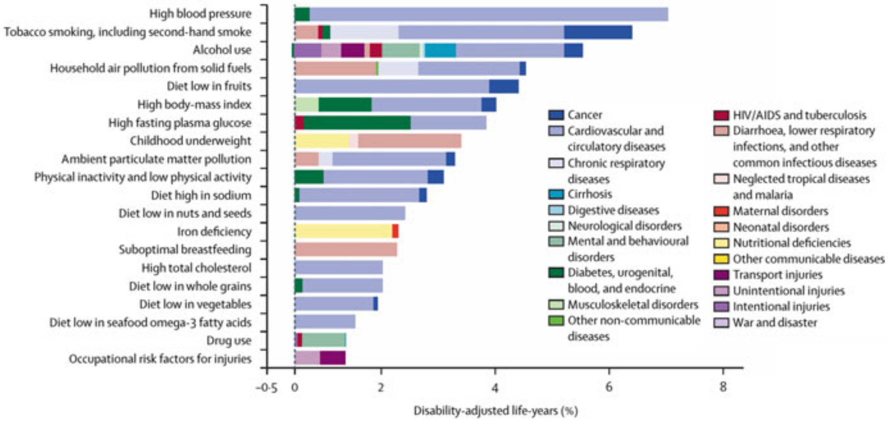

Fig. 5 Global burden of disease attributable to 20 leading risk factors in 2010 expressed as percentage of global disability adjusted life years. Reproduced with permission from Elsevier [4]

The 2002 WHO led GBD exercise estimated up to two million excess deaths to be attributable to HAP accounting for 4% of global DALYs. These risks were comparable to risks from tobacco and only exceeded by malnutrition (16%), unsafe water and sanitation (9%), and unsafe sex (4%). In 2010[4], these risks were estimated to be substantially higher. This came from being able to account for additional health outcomes (the 2002 effort included only ALRI in children under 5, COPD in women, and lung cancer from coal) and choosing a lower counterfactual level for comparison (equivalent to that of levels achieved in gas using households of developed countries). Thus in 2010, HAP accounted for 3.5 million (2.7–4.4 million) deaths and 4.5% (3.4–5.3) of global DALYs (Fig. 5). In poorer regions of the world such as South Asia and much of sub-Saharan Africa, HAP ranked as a leading risk factor exceeding disease burdens attributable to not only tobacco smoking and blood pressure but also child undernutrition, water and sanitation, and ambient (outdoor) air pollution in addition to being the most important risk factor for women. The underlying epidemiological transition from communicable to noncommunicable diseases and the overall improvements in childhood mortality rates have decreased the burden on children from HAP, although it continues to be among the leading risk factors for children in sub-Saharan Africa.

5 Options for Interventions

Fuels such as LPG, ethanol, and biogas when used as household cookfuels are much more efficient from a fuel consumption perspective and much cleaner from an emissions/exposure perspective. However, accessibility and affordability pose major barriers for such cleaner fuels to be a viable solution for many current solid fuel users. Household energy interventions have thus largely been focused on reducing fuel use through more energy-efficient stoves [1]. Emissions[1] and health risks from associated exposures have however seldom been a part of technological considerations in the design of interventions thus far. In India and China where on national level improved stove programs have been operational for the last two decades, the impact of the programs has largely been assessed in terms of number of units disseminated and the impact on health risk reduction remains poorly understood. The residual pollutant levels even among those in regular use are often high [7, 39].

Over the last decade, a variety of solid fuel-based cookstove models that improve combustion efficiency and reduce emissions when compared with open fires and traditional stoves have entered the market. Laboratory testing has shown nearly 50% reductions in PM2.5 emissions from typical natural draft stoves and over 90% reductions from some forced draft stoves, which employ a fan to increase combustion efficiency [75, 76]. These reductions may however be less robust in field settings [33, 38, 52, 60, 77]. Clean stoves used with chimneys have been shown to further reduce indoor PM2.5 exposure (e.g., [36]) and produce improvements in health as demonstrated in a recent randomized control trial involving such stoves ([22, 53, 78, 79].

Experience with well-performing stoves with high rates of adoption however has been quite limited. Many considerations have been shown to determine the acceptability of cookstove interventions among communities including market environments, people's sociocultural preferences, capital and running costs, and access to fuel wood. People living in rural settings often are under limited pressure to conserve wood (except in arid areas) and are not willing to spend any additional resources on alternative fuels or stoves. Indeed when fuel wood becomes scarce people move down the energy ladder shifting to agricultural produce and animal dung, known to be more polluting. Perception of health risks thus currently plays no (or a very limited) role in determining energy choices that households make in their daily lives. Innovations that target user behavior (such as design of biomass stoves to mimic the proven attractiveness and user satisfaction of gas stoves [80]) and alternative financing mechanisms (such as using the international carbon market for cleaner cookstoves and/or promoting subsidies for cleaner fuels such as natural gas or electricity) would be needed to implement interventions. Additional efforts to not only build cleaner stoves but also eliminate residual use of traditional stoves and open fires for cooking and non-cooking tasks (that can often offset the exposure reductions gained from the cleaner cookstoves/fuels) are also needed. Intervention efforts thus need to integrate cleaner technologies with community preferences and cost in order to achieve significant improvements in HAP exposures and consequent health benefits.

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[1] Improved efficiency is not always accompanied by reduced emissions. Increase in efficiencies may be accomplished by increasing merely the heat transfer efficiency, in which case emissions (which are directly related to fuel combustion) are not reduced. Further, expected gains in efficiency (set in laboratory conditions) are seldom realized under field conditions with most improved stoves resulting in less than 25% savings in fuel consumption.

6 Challenges and Opportunities for the Future

The burden of environmental health risks is just beginning to catch the attention of health policy makers in developing countries. Developing countries are faced with several new environmental challenges against a backdrop of traditional public health risks. Allocation of scant resources for risk reduction will necessarily demand that the weight of evidence for each of these risk factors be built on a strong scientific foundation. The preceding account of studies that contribute towards better estimating the health risks associated with household air pollution from solid cookfuel use points out the imminent need for augmenting intervention efforts.

In developing country households using solid cookfuels, exposures to household air pollution happen every day and in widely varying exposure configurations. With multiple household level determinants of exposure that vary across world regions and within countries, there is a need to collect information on both exposures and their determinants on a regional basis. The currently available techniques for the conduct of such assessments are laborious and expensive and there is a need for developing newer methods that are suited for being scaled up to local, national, and regional levels. The challenge in future studies would be to identify a key set of exposure indicators that, while being easy to collect information on, would provide sufficient resolution to classify populations into exposure subcategories.

The exposure and the health studies on this issue have largely remained separate from each other. While financial constraints may be responsible for some studies not being able to address them simultaneously, it is also in some measure a reflection of lack of capacities in performing quantitative environmental health assessments in developing country settings. Even in instances where health-based environmental standards are available (criteria outdoor air pollutants for example) they are based on underlying exposure–response relationships that are largely derived from developed country studies. Risk perception and risk communication mechanisms within research/policy communities are therefore significantly handicapped either due to the lack of locally derived relationships that reduce acceptability or due to lack of understanding on methodologies that limits transferability across settings. With household air pollution being a largely developing country issue with strong regional differences, it is anticipated that health-based standards will have to rely on studies largely executed in individual countries. The strengthening of local technical capacities through academic and interagency partnerships is thus crucial not only to enhance the cost-effectiveness of research initiatives but also to ensure sustainability of subsequent environmental management initiatives and supporting policies.

The issue of indoor air pollution associated with household fuels in developing countries is deeply embedded in a matrix of environmental, energy, health, and economic/developmental considerations. While it is unlikely that health risks alone would determine the future course of how it is managed, an in-depth understanding of the potential for health risks is crucial for ensuring that the most vulnerable poor communities among us are not required to endure years of suffering, before development can “catch up” with them. Indeed if human development is the goal, addressing health risks is an important mechanism of ensuring equity in quality of life among populations and it is hoped that the information presented here represents a small incremental step towards achieving the same.

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Indoor Air Pollution and Its Control in China

Jiming Hao, Tianle Zhu, and Xing Fan

Abstract The status of indoor air pollution and its control in China are reviewed by introducing the pollution characteristics of major indoor air pollutants, the strategies and measures adopted to control indoor air pollution, as well as the major problems existing in the current indoor air pollution control. Although indoor pollution of formaldehyde and benzene has been effectively alleviated in recent years in China, indoor pollution of toluene and xylenes is still serious. Besides, studies show that indoor pollution of particulate matter (PM), biological pollutants, and semi-volatile organic compounds (SVOCs) might also be serious in China. The establishment and implementation of indoor air quality (IAQ)-related regulations and standards, the research on indoor air pollution and its control, and the development of indoor environmental monitoring and cleaning industry have played significant roles in preventing and controlling indoor air pollution in China. However, problems such as lack of mandatory standards for IAQ, lack of regulation and labeling of pollutant emissions from indoor decorating and refurbishing materials, lack of an effective performance evaluation system for air cleaning products, and lack of proper maintenance of air cleaners remain to be solved for further improvement of IAQ.

Keywords Characteristics, China, Countermeasures, Indoor air pollution

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Contents

1 Introduction

Indoor air quality (IAQ) problems have always existed in China due to the extensive use of solid fuels (coal and biomass) in simple stoves for household heating and cooking (in the vast rural and also many urban areas) as well as some traditional but unhealthy living habits, such as cooking using stir frying and deep frying methods, smoking in homes, using commodes with no flushing systems (in most rural areas), and living together with livestock (in some rural areas). According to studies of relationship between IAQ and human health conducted in the 1980s, the main indoor air pollutants at that time were fuel combustion products, cooking fumes, and environmental tobacco smoke (ETS); poor IAQ was associated with a variety of negative health outcomes, the most notable being lung cancer [1–5].

In order to alleviate indoor air pollution from household energy use and unhealthy lifestyle, many interventions have been introduced in China, including technological interventions (e.g., stove and ventilation improvement, development and use of clean energy), behavior interventions (e.g., cooking methods change, indoor smoking avoidance), health education, and social mobilization [4–6]. The China National Improved Stove Program (CNISP), which resulted in the installation of 129 million new stoves in rural homes (representing around 65% of all rural households) between 1982 and 1992, has proved to greatly improve the IAQ in rural China [4, 5, 7].

Since the late 1990s, more and more residents in China have started to own apartments or townhouses with the deepening of city and township housing reform, and as personal incomes have rapidly increased with the high-speed economic growth, indoor decoration and refurbishment of newly constructed or existing apartments have become very popular, which has brought not only great prosperity to the indoor decoration and refurbishment industry but also extremely serious IAQ problems to the country. On the one hand, due to the lack and the lax enforcement of regulations and standards for indoor decorating and refurbishing materials, some materials and products containing large amounts of harmful substances have entered the market and then the indoor environments, releasing various pollutants such as formaldehyde and volatile organic compounds (VOCs) during and after the decoration and refurbishment. On the other hand, air-conditioning systems have been installed in almost all modern buildings with the improvement of living standards. In order to save energy during the use of air-conditioning, most buildings have been constructed to be tightly sealed, leading to a significantly reduced indoor-outdoor air exchange. As a result, pollutants released from inferior decorating and refurbishing materials build up in indoor air, severely deteriorating the IAQ and thus posing serious hazards to human health. In June 2001, the Beijing Changping District People's Court judged China's first damages lawsuit for indoor air pollution. Formaldehyde, in a concentration of 1.56 mg/m^3 (surpassing the indicated standard by 19.5 times) in the owner's bedroom, resulted in the lawsuit.

Excessive complaints relevant to indoor air pollution caused by interior decoration and refurbishment and the prevalence of sick building syndrome (SBS) have aroused deep concern of IAQ problems from the government and the public. From June to September 2001, Vice-Premiers Wen Jiabao and Li Lanqing made important instructions on interior decoration pollution three times in succession, calling for great attention to the pollution and requesting relevant departments to develop effective pollution control measures. In order to increase public awareness of the importance of IAQ, propaganda and education of indoor environmental knowledge, especially the causes and effects of indoor air pollution, have been greatly strengthened.

The government's guidance and the increasing public requirements for improving IAQ have significantly promoted the establishment of IAQ-related regulations and standards, the research on indoor air pollution and its control, and the development of indoor environmental monitoring and cleaning industry in China, which provide solid foundations for prevention and control of indoor air pollution. However, the existing pollution control measures mostly focus on the pollutants originating from indoor decorating and refurbishing materials (mainly formaldehyde and VOCs). Other indoor air pollutants such as particulate matter (PM), biological pollutants, and semi-volatile organic compounds (SVOCs) have not obtained adequate attention in China.

In this chapter, the current status and future trends of indoor air pollution in China are introduced first. Then the strategies and measures adopted to control indoor air pollution are reviewed. Finally, major problems existing in the current indoor air pollution control are analyzed.

2 Current Status and Future Trends of Indoor Air Pollution in China

In order to get an insight into the current status and future trends of indoor air pollution in China, pollution characteristics of major indoor air pollutants, including formaldehyde, VOCs, PM, biological pollutants, and SVOCs, are introduced as follows.

2.1 Formaldehyde

Formaldehyde is the most common and the best-known indoor air pollutant. In homes, the most significant sources of formaldehyde are likely to be engineered wood products made by using adhesives that contain urea-formaldehyde (UF) resins. Engineered wood products made for indoor use include particleboard (used as subflooring and shelving and in cabinetry and furniture), hardwood plywood paneling (used for decorative wall coverings and used in cabinets and furniture), and medium-density fiberboard (used for drawer fronts, cabinets, and furniture tops).

Indoor formaldehyde pollution was very serious in the early 2000s in China due to the extensive use of engineered wood products with high formaldehyde emission in home decoration and refurbishment. A survey conducted in 2003 in six cities of China indicated that the percentage of recently renovated homes with indoor formaldehyde concentrations above the national standard (0.10 mg/m^3) reached 82.3% (Table 1) [8].

In recent years, formaldehyde pollution has been effectively alleviated by forbidding the use of substandard engineered wood products in indoor decoration and refurbishment via implementing the mandatory national standard, GB 18580-2001 “Indoor decorating and refurbishing materials – Limit of formaldehyde emission of wood-based panels and finishing products.” Table 2 shows the passing rates of formaldehyde emission of medium-density fiberboard and blockboard on the market in different years [9]. It can be seen that for both kinds of wood-based panels, the passing rates of formaldehyde emission in 2009–2011 were much higher than those in 2003 or 2004.

As the awareness of the importance of IAQ has been increasing in these years, public demand for healthy/green/nontoxic decorating and refurbishing materials is growing. Predictably, the indoor formaldehyde pollution will be further reduced or completely eliminated with the development of wood-based panels with lower or zero formaldehyde emission in the near future.

2.2 Volatile Organic Compounds

VOCs have become major indoor air pollutants in China since the prevalence of interior decoration and refurbishment. Indoor sources of VOCs mainly include

Table 1 Indoor formaldehyde concentrations in recently renovated homes in six cities of China[a] [8]

City	Sample number	Formaldehyde concentration (mg/m^3) Mean ± SD	Formaldehyde concentration (mg/m^3) Range	Percentage above standard[b] (%)
Beijing	530	0.210 ± 0.152	0.025–1.382	75.5
Tianjin	164	0.267 ± 0.170	0.025–1.100	89.6
Shanghai	182	0.205 ± 0.135	0.025–0.869	79.1
Chongqi	198	0.142 ± 0.084	0.025–0.461	64.6
Shizuishan	212	0.610 ± 0.311	0.104–1.712	100
Changchun	201	0.412 ± 0.208	0.025–1.243	96.0
Total	1,487	0.290 ± 0.238	0.025–1.712	82.3

[a] Sampling time: January to April 2003; time since renovation: <6 months

[b] The "Indoor air quality standard" of China (GB/T 18883-2002) stipulates the indoor limit of 0.10 mg/m^3 (1-h average) for formaldehyde

Table 2 Passing rates of formaldehyde emission of medium-density fiberboard and blockboard[a]

Year	Medium-density fiberboard Sample number	Medium-density fiberboard Passing rate (%)	Blockboard Sample number	Blockboard Passing rate (%)
2003	41	75.6	–	–
2004	–	–	91	80.2
2009	93	86.0	188	89.4
2010	119	93.3	1,206	90.8
2011	189	96.3	200	88.0

[a] Calculated from inspection results by the General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China [9]

solvent coatings for woodenware, interior architectural coatings, adhesives, wood-based furniture, carpets, and carpet cushions. Generally, various kinds of VOCs coexist in indoor environments, in concentrations decreasing with time after renovation.

Similar to formaldehyde, indoor pollution of VOCs was also very serious in the early 2000s in China. An investigation conducted from 2002 to 2004 on VOCs concentrations in 1,241 recently renovated residences in China showed that the average concentration of total volatile organic compounds (TVOC) was as high as 2.18 mg/m^3 and benzene, toluene, and xylenes (BTX) were the primary VOCs pollutants in indoor air, with average concentrations of 124.04, 258.90, and 189.68 $\mu g/m^3$, respectively (Table 3) [10].

The high concentrations of BTX in newly renovated buildings could be mainly attributed to the widespread use of BTX as solvents and diluents for coatings and adhesives used in interior decoration and refurbishment. Considering the high toxicity of benzene to human beings, the use of benzene has been forbidden since 2002 by implementing mandatory national standards, including GB 50325-2001 "Code for indoor environmental pollution control of civil building engineering," GB 18581-2001 "Indoor decorating and refurbishing materials – Limit of harmful substances of solvent coatings for woodenware," and GB 18583-2001 "Indoor

Table 3 Indoor concentrations of benzene, toluene, xylenes, and TVOC in recently renovated residences in China[a] [10]

		Concentration (BTX: μg/m^3; TVOC: mg/m^3)	
Pollutant	Sample number	Mean ± SD	Range
Benzene	843	124.04 ± 272.60	0.18–4,284.20
Toluene	901	258.90 ± 671.98	0.04–9,093.70
Xylenes	958	189.68 ± 561.16	0.05–10,413.70
TVOC	982	2.18 ± 12.94	0.005–394.44

[a] Sampling area: Beijing, Tianjin, Shanghai, Dalian, Shizuishan, Pingliang, Zhuhai, Changchun, and Chongqing; sampling time: May 2002 to November 2004; time since renovation: <12 months

Table 4 Indoor concentrations of benzene, toluene, and xylenes in a newly renovated hotel in Beijing[a] [11]

			Concentration (μg/m^3)		
Pollutant	Sampling site	Sampling times	Mean	Range	Indoor limit[b] (μg/m^3)
Benzene	Guest room	6	107.0	45.7–178.8	110 (1-h average)
	Conference room	6	87.0	21.7–191.5	
Toluene	Guest room	6	400.7	159.0–639.7	200 (1-h average)
	Conference room	6	174.3	47.1–330.8	
Xylenes	Guest room	6	1,623.4	1,185.7–2,484.9	200 (1-h average)
	Conference room	6	402.4	42.1–1,037.0	

[a] Sampling time: April to June 2010

[b] Stipulated by GB/T 18883-2002 "Indoor air quality standard"

decorating and refurbishing materials – Limit of harmful substances of adhesives." Meanwhile, the use of toluene and xylenes has been limited although they are less toxic compared to benzene. Field survey data prove that implementation of these standards has indeed reduced indoor pollution of benzene; however, indoor pollution of toluene and xylenes is still serious (Table 4) [11]. In order to further control the pollution of toluene and xylenes, the abovementioned national standards have been revised successively in recent years to strengthen the limits of toluene and xylenes.

Besides promoting the use of less toxic organic solvents, great efforts have also been made in China to develop coatings and adhesives with no organic solvents, such as water-based coatings and adhesives, powder coatings, radiation curable coatings, and hot melt adhesives. Increasing use of these environment-friendly products in interior decoration and refurbishment would greatly reduce the indoor pollution of VOCs.

2.3 Particulate Matter

Compared with gaseous pollutants (formaldehyde, VOCs), indoor particulate matter (PM) has gathered much less concern in China, although inhalable PM

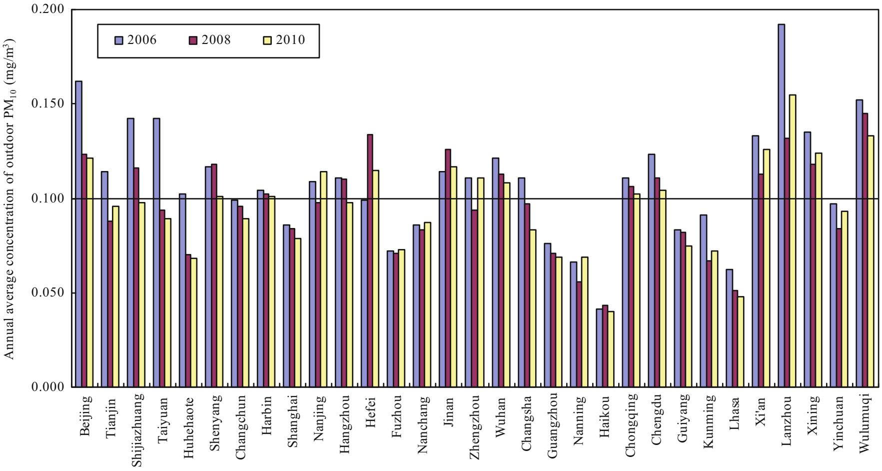

Fig. 1 Annual average concentrations of outdoor PM_{10} in 31 cities in China [14–16]

(PM_{10}, particles smaller than or equal to 10 μm in aerodynamic diameter) is included in the "Indoor air quality standard" (GB/T 18883-2002) as one of the 13 controlled chemical pollutants.

In China, indoor PM mainly originates from outdoor sources such as fuel burning (mainly coal), vehicle emissions, and transformation of gaseous emissions in the atmosphere [12]. According to the air quality monitoring results of major cities in China since 2000, PM_{10} has been the most important primary pollutant (among PM_{10}, SO_2, NO_2, CO, and O_3) in ambient air [13]. Figure 1 illustrates the annual average concentrations of PM_{10} in 31 cities in China for the years 2006, 2008, and 2010 [14–16]. It can be seen that the overall PM_{10} pollution was still serious in 2010 although an alleviation trend was observed for most cities. In 2006, 2008, and 2010, 19, 14, and 14 monitored cities had annual average concentrations of outdoor PM_{10} exceeding the limit value (0.10 mg/m^3, annual average) stipulated by GB 3095-1996 "Ambient air quality standard" (Fig. 1).

On the other hand, the frequent occurrence of haze in many cities (especially mega cities such as Beijing, Shanghai, and Guangzhou) of China since the autumn of 2011 indicates that the concentration of fine particles ($PM_{2.5}$, particles smaller than or equal to 2.5 μm in aerodynamic diameter) in ambient air has been increasing in recent years although the PM_{10} concentration tends to decrease. It is well known that fine particles are more hazardous to human health than coarse particles since they can travel deep into the lungs, enter the bloodstream, and penetrate into cells. In the newly revised "Ambient air quality standard" (GB 3095-2012), $PM_{2.5}$ has been included as another common pollutant besides PM_{10}, SO_2, NO_2, CO, and O_3.

Since outdoor PM can migrate into indoor environments through fissures and cracks in the building structures, high concentration of PM in ambient air can not only deteriorate the quality of outdoor air but also negatively influence the quality of indoor air. Shi et al. [17] investigated the concentrations of PM in indoor and

Table 5 Concentrations of PM_{10} and $PM_{2.5}$ in indoor and outdoor air of residences in Taiyuan [17]

			Average concentration (mg/m^3)	
Sampling time	Sampling site	Sample number	PM_{10}	$PM_{2.5}$
Heating period	Indoor	438	0.327	0.107
	Outdoor	319	0.547	0.123
Non-heating period	Indoor	485	0.239	0.089
	Outdoor	457	0.321	0.109

Table 6 Concentrations of PM_{10} in indoor (I) and outdoor (O) air of residences and the I/O ratios [18]

		Average concentration of PM_{10} (mg/m^3)		
Residence	Sampling times	Indoor	Outdoor	I/O ratio
Without indoor PM source	3	0.093	0.107	0.869
With smoking	3	0.109	0.060	1.817
With cooking	3	0.067	0.060	1.117

outdoor air of 541 homes in Taiyuan (the capital of Shanxi province) during both heating and non-heating periods of 2004–2006. Results showed that the PM concentration in outdoor air was always higher than that in indoor air and both the indoor and outdoor concentrations of PM during the heating period were higher than those during the non-heating period (Table 5). Correlation analysis proved that the outdoor PM pollution contributed greatly to the indoor PM pollution, especially during the heating period.

In addition to outdoor sources, indoor PM can also originate from indoor sources such as fuel-burning, cooking, smoking, and sweeping activities. For modern buildings with low ventilation rates, the presence of indoor sources may result in higher PM concentrations in indoor environments than in outdoor air (Table 6) [18].

In view of the severe outdoor pollution of PM, the significant influence of outdoor pollution on indoor environments as well as the serious health risks from PM exposure, strengthening research on indoor PM pollution and its control, has become imperative in China.

2.4 *Biological Pollutants*

Biological pollutants in indoor environments mainly include bacteria (including endotoxins from bacteria), fungi (including spores and cell fragments of fungi), viruses, dust mites, and animal dandruff. These pollutants exist in the air mainly as bioaerosols (biological particles). Major indoor sources of biological pollutants at residential homes include human occupants, pets, house dust, organic waste, as well as the heating, ventilation, and air-conditioning (HVAC) system [19–23]. Adverse health effects/diseases related to biological pollutants exposure can be divided into two categories: infectious diseases such as influenza, viral pneumonia (e.g., severe acute respiratory syndrome, SARS) and bacterial pneumonia (e.g., Legionnaires' disease) and allergic diseases such as allergic asthma, allergic rhinitis, and allergic alveolitis [24].

Although studies on concentrations and health effects of indoor biological pollutants started in the 1950s in China [25–31], control of indoor biological pollution had not obtained adequate attention until the outbreak of SARS in 2003. Since bad-designed ventilation systems were considered to have played important roles in the rapid and widespread dissemination of the SARS virus, air-conditioning and ventilation systems have become the focus of concern in the control of indoor biological pollution.

In 2004, a national inspection on central air-conditioning and ventilation systems in 937 public places, including hotels, restaurants, shopping malls, and supermarkets, was performed by the Ministry of Health of the People's Republic of China. The report showed that around half of the samples were heavily polluted with high concentrations of dust, bacteria, and fungi and the passing rate was only 6%. The maximum concentrations of dust, bacteria, and fungi on the inner surface of air ducts reached 486 g/m^2, 277×10^4 cfu/(g dust), and 480×10^4 cfu/(g dust), respectively [32]. During the use of air-conditioning, the dust containing bacteria and fungi can be transferred into indoor environments by the airstream, causing serious biological pollution and then adverse health effects.

In order to prevent and control the pollution caused by air-conditioning and ventilation systems, the Ministry of Health of the People's Republic of China issued a comprehensive hygiene management approach and three supporting hygienic norms (hygienic norm, hygienic assessment norm, and cleaning norm) for central air-conditioning and ventilation systems in public places, which came into effect on March 1, 2006. Tables 7 and 8 present some survey data on hygienic conditions of central air-conditioning and ventilation systems in public places of China after implementing the management approach and hygienic norms [33–45]. It can be seen that the overall concentration-passing rates of typical pollutants both on the inner surface of air ducts (Table 7) and in the supply air (Table 8) have been significantly improved in recent years, indicating an effective control of biological pollution related with central air-conditioning and ventilation systems.

Compared with public places, however, much less concern has been given to control of biological pollution in residences, which should also be addressed in the future to protect residents from hazards of ubiquitous biological pollutants.

2.5 Semi-volatile Organic Compounds

SVOCs are organic compounds with boiling points in the range of 240–400°C [46]. They are considered as new-type chemical pollutants in indoor air and have become a hot research topic in the field of indoor environment and health in recent years. Major sources of indoor SVOCs include materials and products containing

Table 7 Concentration-passing rates of dust, bacteria, and fungi on inner surface of air ducts of central air-conditioning and ventilation systems in public places of China[a]

			Dust		Bacteria		Fungi		
Area	Sampling time	Number of public place	Sample number	Passing rate[b] (%)	Sample number	Passing rate[c] (%)	Sample number	Passing rate[d] (%)	References
Hebei (Shijiazhuang)	2010	22	110	70.0	110	88.2	110	80.0	[33]
Hunan	2010	24	24	58.3	24	75.0	24	100.0	[34]
Pearl River Delta	2009–2010	25	170	68.2	170	99.4	170	100.0	[35]
Guangdong (Zhongshan)	2009	18	183	95.6	183	94.5	183	88.0	[36]
Fujian (Xiamen)	2009	43	398	98.5	398	97.2	398	95.0	[37]
Zhejiang	2009	240	237	75.5	236	94.9	236	70.8	[38]
Jiangsu	2009	97	312	67.3	458	90.6	456	89.7	[39]
Liaoning (Dalian)	2008	14	14	64.3	69	97.1	69	97.1	[40]
Beijing	2008	12	56	67.9	56	76.8	56	53.6	[41]
Hunan (Changsha)	2008	15	61	50.8	61	50.8	61	59.0	[41]
Tianjin	2006	16	–	–	113	89.4	113	71.7	[42]
Henan (Jiaozuo)	2006	16	96	59.4	96	47.9	96	41.7	[43]

[a] Evaluated based on the “Hygienic norm for central air-conditioning and ventilation systems in public places” of China

[b] Percentage of samples with dust concentrations $\leq$ 20 g/m^2

[c] Percentage of samples with bacteria concentrations $\leq$ 100 cfu/cm^2

[d] Percentage of samples with fungi concentrations $\leq$ 100 cfu/cm^2

Table 8 Concentration-passing rates of PM_{10}, bacteria, fungi, and β-hemolytic streptococcus in supply air of central air-conditioning and ventilation systems in public places of China[a]

			PM_{10}		Bacteria		Fungi		β-Hemolytic streptococcus		
Area	Sampling time	Number of public place	Sample number	Passing rate[b] (%)	Sample number	Passing rate[c] (%)	Sample number	Passing rate[d] (%)	Sample number	Passing rate[e] (%)	References
Zhejiang	2011	180	179	79.3	179	62.6	179	74.3	179	100.0	[44]
Hebei (Shijiazhuang)	2010	22	–	–	110	95.4	110	99.1	110	100.0	[33]
Pearl River Delta	2009–2010	25	112	75.9	112	73.2	112	97.3	112	100.0	[35]
Guangdong (Zhongshan)	2009	18	–	–	183	86.3	183	76.5	183	100.0	[36]
Fujian (Xiamen)	2009	43	31	93.6	31	90.3	31	96.8	398	100.0	[37]
Zhejiang	2009	240	239	64.4	240	71.7	240	76.7	240	100.0	[38]
Liaoning (Dalian)	2008	14	14	57.1	66	47.0	66	71.2	–	–	[40]
Beijing	2008	12	56	33.9	56	58.9	56	100.0	–	–	[41]
Hunan (Changsha)	2008	15	33	27.3	33	39.4	33	48.5	–	–	[41]
Tianjin	2006	16	–	–	164	40.2	164	59.1	164	100.0	[42]
Heilongjiang (Harbin)	2006	20	20	45.0	20	0	20	75.0	–	–	[45]

[a] Evaluated based on the “Hygienic norm for central air-conditioning and ventilation systems in public places” of China

[b] Percentage of samples with PM_{10} concentrations ≤ 0.08 mg/m^3

[c] Percentage of samples with bacteria concentrations ≤ 500 cfu/cm^3

[d] Percentage of samples with fungi concentrations ≤ 500 cfu/cm^3

[e] Percentage of samples with no detectable β-hemolytic streptococcus

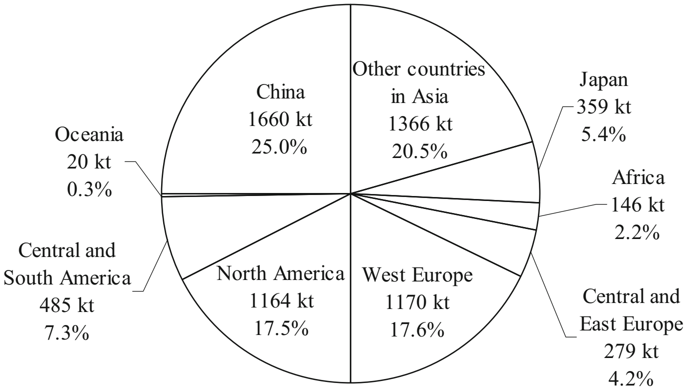

Fig. 2 Consumption of plasticizers in the world in 2006 by region [47, 48]

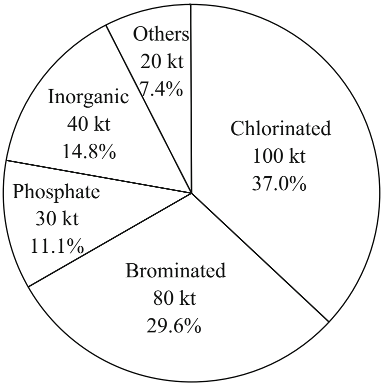

Fig. 3 Production of flame retardants in China in 2006 by type [47]

plasticizers (additives in plastics to enhance their flexibility and extensibility) and flame retardants (additives in materials to reduce their combustibility), household pesticides, and human activities such as smoking, incense burning, and cooking [47].

China produces and consumes the largest amounts of plasticizers in the world. As shown in Fig. 2, 25% of plasticizers in the world were consumed by China in 2006 [47, 48]. Phthalates are produced and consumed in the largest amounts among all plasticizers, with diethylhexyl phthalate (DEHP) and dibutyl phthalate (DBP) being the two main types [47, 48].

The production and consumption of flame retardants are also very high in China. In 2006, China produced 270-kt flame retardants, 66.6% of which were halogen-based (chlorinated and brominated) (Fig. 3) [47]. The largest production and consumption among halogen-based flame retardants belong to decabromodiphenyl ether (DecaBDE) due to its lowest price and highest performance [47].

China also produces and consumes great amounts of pesticides, the main active ingredients of which are SVOCs [47]. Besides, field measurements show that smoking, coal combustion, and Chinese cooking produce large quantities of polycyclic aromatic hydrocarbons (PAHs) [49–51].

Epidemiological and toxicological studies have proved that SVOCs exposure can cause serious harm to human health, including harming the endocrine and reproductive systems [52–55]. In recent years in China, indoor SVOCs pollution has gathered escalating concern from the researchers. Several studies on pollution and exposure levels of indoor SVOCs have been conducted, and the results show that indoor SVOCs pollution might be very serious in China [56–60]. In order to prevent and control this new type of pollution, more concern from the government as well as the public is required and more systematic and in-depth research should be carried out.

3 Strategies and Measures for Control of Indoor Air Pollution

In order to control indoor air pollution, a series of strategies and measures have been adopted in China since the 1980s, including enacting regulations and standards, strengthening research on indoor air pollution and its control, and developing indoor environmental monitoring and cleaning industry.

3.1 Formulating and Improving Regulations and Standards

Legislation concerning IAQ in China started in the late 1980s based on broad surveys and studies on health effects of indoor environments. In 1988, the Ministry of Health of the People's Republic of China issued a set of hygienic standards for public places, in which concentration limits of carbon monoxide (CO), carbon dioxide (CO_2), inhalable particulate matter (PM_{10}), and bacteria were stipulated. These standards played important roles in improving sanitation in public places as well as controlling the propagation of diseases. Revised versions of these standards, GB 9663–9673-1996 and GB 16153-1996, were promulgated by the General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China in 1996. In the revised standards, the concentration limits of formaldehyde were also included.

The first hygienic standard for residences, GB/T 16127-1995 "Hygienic standard for formaldehyde in indoor air of house," was issued by the General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China in 1995. After that, a series of hygienic standards for other pollutants in indoor air were successively promulgated by the General Administration of Quality Supervision, Inspection and Quarantine and/or the Ministry of Health of the

People's Republic of China, including GB/T 17093-1997 (bacteria total), GB/T 17094-1997 (carbon dioxide), GB/T 17095-1997 (inhalable particulate matter), GB/T 17096-1997 (nitrogen oxides), GB/T 17097-1997 (sulfur dioxide), WS/T 182-1999 (benzo(a)pyrene (B(a)P)), GB/T 18202-2000 (ozone), GB/T 18203-2000 (*Streptococcus hemolyticus*), and GB 18468-2001 (*p*-dichlorobenzene). These standards laid good foundations for the establishment of IAQ standard in China.

The prevalence of indoor decoration and refurbishment of existing and new buildings since the late 1990s has caused serious indoor air pollution in China due to the extensive use of indoor decorating and refurbishing materials containing large amounts of harmful substances. In order to control indoor air pollution effectively, the Chinese government has greatly accelerated the pace of establishing IAQ-related regulations and standards since 2000.

GB 50325-2001 "Code for indoor environmental pollution control of civil building engineering," jointly issued by the Ministry of Construction and the General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China on November 26, 2001, took effect on January 1, 2002. This code is applicable for the newly constructed, extended, or renovated civil building engineering, which is divided into two groups: Group I includes residential house, hospital, home for the elderly, kindergarten, and classroom and Group II includes office building, shopping center, hotel, public place of entertainment, bookstore, library, exhibition, gymnasium, waiting room of public transit means, dining room, and barbershop. The code prescribes the requirements for building and decorating materials (inorganic nonmetallic main materials for buildings and decorating materials such as wood-based panels, coatings, adhesives, and water-based treatment agents), survey and design (site investigation on soil radon concentration and material selection), construction, inspection, and acceptance to control the concentrations of radon, formaldehyde, benzene, ammonia, and TVOC in civil buildings.

GB 50325-2001 played important roles in preventing and controlling the indoor air pollution caused by building and decorating materials. Considering the development of construction, decoration, and refurbishment industry as well as the changes in characteristics of indoor air pollution in China, the code was revised first in 2006 and then in 2010. GB 50325-2010, which was brought into effect on June 1, 2011, strengthens the original requirements for pollution control and includes new requirements as well. For instance, for solvent-based coatings used in civil building engineering, the content limits of TVOC and benzene have been strengthened, and a limit value for the total content of toluene, xylenes, and ethylbenzene has been added in GB 50325-2010 (Table 9) [61, 62]. GB 50325-2010 also strengthens the concentration limits of formaldehyde and ammonia for Group II civil building engineering (Table 10) [61, 62].

In order to improve the quality and regulate the production of indoor decorating and refurbishing materials in China, the General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China issued ten mandatory national standards for limits of harmful substances in indoor decorating and refurbishing materials on December 10, 2001, which came into effect on

Table 9 Limits of harmful substances in solvent-based coatings used in civil building engineering [61, 62]

	TVOC (g/L)		Benzene (%)		Toluene + xylenes + ethylbenzene (%)	
Solvent-based coatings	GB 50325-2001	GB 50325-2010	GB 50325-2001	GB 50325-2010	GB 50325-2001	GB 50325-2010
Alkyd coatings	≤550	≤500	≤0.5	≤0.3	–	≤5
Nitrocellulose coatings	≤750	≤720	≤0.5	≤0.3	–	≤30
Polyurethane coatings	≤700	≤670	≤0.5	≤0.3	–	≤30
Phenolic antirust coatings	≤270	≤270	≤0.5	≤0.3	–	–

Table 10 Limits of environmental pollutants in civil building engineering [61, 62]

	Group I civil building engineering		Group II civil building engineering	
Pollutant	GB 50325-2001	GB 50325-2010	GB 50325-2001	GB 50325-2010
Radon (Bq/m^3)	≤200	≤200	≤400	≤400
Formaldehyde (mg/m^3)	≤0.08	≤0.08	≤0.12	≤0.10
Benzene (mg/m^3)	≤0.09	≤0.09	≤0.09	≤0.09
Ammonia (mg/m^3)	≤0.2	≤0.2	≤0.5	≤0.2
TVOC (mg/m^3)	≤0.5	≤0.5	≤0.6	≤0.6

January 1, 2002 (Table 11). These standards provide mighty technical and legal support for ultimately improving IAQ and safeguarding people's health by explicitly limiting the contents and emission intensity of harmful chemicals of indoor decorating and refurbishing materials. Materials and products that fail to meet the standards were forbidden to enter the market from July 1, 2002. Inspections of the product quality have been conducted frequently, and punishment for production, sale, and use of substandard products has been enforced to ensure the quality of construction, decoration, and refurbishment. Consumers can take the mandatory national standards as authoritative references when they address any dispute arising from indoor decoration and refurbishment. Some specifications in these standards, such as those for VOCs in interior architectural coatings, conform to those standards in the European Union and the USA.

In recent years, the Standardization Administration of the People's Republic of China has been revising these standards by strengthening some of the limits as well as including more limited harmful substances and materials taking into account the increasing requirements for better IAQ and the development of indoor decoration and refurbishment industry (Table 11).

The first national indoor air quality standard (GB/T 18883-2002), released jointly by the General Administration of Quality Supervision, Inspection and Quarantine, the Ministry of Health, and the Ministry of Environmental Protection of the People's Republic of China, came into effect on March 1, 2003. The standard

Table 11 Standards for indoor decorating and refurbishing materials

Standard number	Standard name	Revised version (implementation date)
GB 18580-2001	Indoor decorating and refurbishing materials – Limit of formaldehyde emission of wood-based panels and finishing products	–
GB 18581-2001	Indoor decorating and refurbishing materials – Limit of harmful substances of solvent coatings for woodenware	GB 18581-2009 (June 1, 2010)
GB 18582-2001	Indoor decorating and refurbishing materials – Limit of harmful substances of interior architectural coatings	GB 18582-2008 (October 1, 2008)
GB 18583-2001	Indoor decorating and refurbishing materials – Limit of harmful substances of adhesives	GB 18583-2008 (September 1, 2009)
GB 18584-2001	Indoor decorating and refurbishing materials – Limit of harmful substances of wood-based furniture	–
GB 18585-2001	Indoor decorating and refurbishing materials – Limit of harmful substances of wallpapers	–
GB 18586-2001	Indoor decorating and refurbishing materials – Limit of harmful substances of poly(vinyl chloride) floor coverings	–
GB 18587-2001	Indoor decorating and refurbishing materials – Limit of harmful substances emitted from carpets, carpet cushions and adhesives	–
GB 18588-2001	Limit of ammonia emitted from concrete admixtures	–
GB 6566-2001	Limit of radionuclides in building materials	GB 6566-2010 (July 1, 2011)

stipulates the requirements for IAQ parameters, including physical, chemical, biological, and radioactive parameters relevant to human health (Table 12) [63], and the relevant testing methods. Besides the measurable indices, the feeling index of "no odor in indoor air" is also included in this standard. The standard is suitable for residential and office buildings. The built environments of other buildings can be designed or evaluated according to it as well. GB/T 18883-2002 provides a scientific basis for IAQ evaluation and has played important roles in preventing and controlling indoor air pollution and safeguarding people's health in China.

Air-conditioning and ventilation systems can be important sources of indoor pollutants and major propagation pathway of airborne diseases. In order to strengthen the management of central air-conditioning and ventilation systems in public places to protect the public health, the Ministry of Health of the People's Republic of China promulgated "Hygiene management approach of central air-conditioning and ventilation systems in public places" and three supporting hygienic norms in 2006.

The "Hygienic norm for central air-conditioning and ventilation systems in public places" prescribes the requirements and testing methods of hygienic conditions of central air-conditioning and ventilation systems in public places. The hygienic requirements for air supply and inner surface of air ducts are listed in

Table 12 Indoor air quality standard (GB/T 18883-2002) [63]

Parameter category	Parameter	Unit	Standard value	Note
Physical	Temperature	°C	22–28	With air-conditioning (summer)
			16–24	With heating (winter)
	Relative humidity	%	40–80	With air-conditioning (summer)
			30–60	With heating (winter)
	Air velocity	m/s	0.3	With air-conditioning (summer)
			0.2	With heating (winter)
	Amount of fresh air	$m^3/(h \cdot person)$	30[a]	
Chemical	Sulfur dioxide (SO_2)	mg/m^3	0.50	1-h average
	Nitrogen dioxide (NO_2)	mg/m^3	0.24	1-h average
	Carbon monoxide (CO)	mg/m^3	10	1-h average
	Carbon dioxide (CO_2)	%	0.10	24-h average
	Ammonia (NH_3)	mg/m^3	0.20	1-h average
	Ozone (O_3)	mg/m^3	0.16	1-h average
	Formaldehyde (HCHO)	mg/m^3	0.10	1-h average
	Benzene (C_6H_6)	mg/m^3	0.11	1-h average
	Toluene (C_7H_8)	mg/m^3	0.20	1-h average
	Xylenes (C_8H_{10})	mg/m^3	0.20	1-h average
	Benzo(a)pyrene (B(a)P)	ng/m^3	1.0	24-h average
	Inhalable particulate matter (PM_{10})	mg/m^3	0.15	24-h average
	Total volatile organic compounds (TVOC)	mg/m^3	0.60	8-h average
Biological	Bacteria total	cfu/m^3	2,500	Depending on instrument
Radioactive	Radon (Rn)	Bq/m^3	400	Annual average (action level[b])

[a] Requirement for the amount of fresh air: ≥ standard value; requirements for other parameters except temperature and relative humidity: ≤ standard value

[b] Recommended to introduce interventions to reduce the indoor concentration of radon when the standard value is reached

Tables 13 and 14 [64]. The "Hygienic assessment norm for central air-conditioning and ventilation systems in public places" prescribes the requirements of hygienic assessment of the new, reconstructed, extended, or running central air-conditioning and ventilation systems in public places. The purposes, references, contents, and methods of preventive and regular hygienic assessment are provided in the norm. The "Cleaning norm for central air-conditioning and ventilation systems in public places" prescribes the requirements of cleaning methods, cleaning process, performance and security of cleaning, and cleaning agencies and devices for the main parts of air-conditioning and ventilation systems in public places. Implementation of these norms has greatly improved the hygienic conditions of air-conditioned indoor environments in China.

Table 13 Hygienic requirements for air supply [64]

Parameter	Requirement
PM_{10}	≤ 0.08 mg/m^3
Bacteria	≤ 500 cfu/m^3
Fungi	≤ 500 cfu/m^3
Pathogenic microorganisms	Not detectable

Table 14 Hygienic requirements for inner surface of air ducts [64]

Parameter	Requirement
Dust	≤ 20 g/m^2
Pathogenic microorganisms	Not detectable
Bacteria	≤ 100 cfu/cm^2
Fungi	≤ 100 cfu/cm^2

On the whole, a primary regulation and standard system for preventing and controlling indoor air pollution has been established through many years of endeavors in China. Indoor air pollution can be controlled from the beginning of building design, during building construction and decoration, and through proper maintenance of air-conditioning and ventilation systems. Meanwhile, the increasing requirements for better IAQ as well as advancement in IAQ research will help improve the regulation and standard system continuously during practical applications.

3.2 Strengthening Research on Indoor Air Pollution and Its Control

Besides promoting the establishment and improvement of IAQ-related regulations and standards, the Chinese government has also been very active in organizing and supporting the research on indoor air pollution and its control. Under the 10th National Five-Year Plan, the Ministry of Science and Technology of the People's Republic of China developed grants for three projects under the National Key Technologies R&D Program: "Evaluation technologies of health hazards of key indoor air pollutants," "Control technologies of health hazards of key indoor air pollutants," and "Control technologies of indoor air pollution from coal burning." These studies provided important technical support for evaluation and control of indoor air pollution.

As the "Outline of the National Program for Mid- and Long-Term Science and Technology Development (2006–2020)" requires to develop technologies for improving urban living and indoor environments, research on indoor air pollution and its control has been greatly strengthened and conducted in a more systematic way in China since 2006. The research contents and objectives of three major projects implemented during the 11th Five-Year Plan period are briefly described as below.

1. Research and development of environment-friendly building materials and products

   As a source control strategy, the use of environment-friendly building materials and products can prevent indoor air pollution caused by building materials. In order to guide, regulate, and promote the research, development, production, and application of environment-friendly building materials and products, the Ministry of Science and Technology of the People's Republic of China organized and supported the "Research and development of environment-friendly building materials and products" as a key project of the National Key Technologies R&D Program.

   The project mainly focused on research of production and application technologies, regulations and standards, and technical and economic policies related with environment-friendly building materials and products, aiming at mastering key common technologies of building material industry, developing key technologies and equipment with independent intellectual property rights for production and application of environment-friendly building materials and products, and achieving "leapfrog" development by widely promoting the application of environment-friendly building materials and products. It also aimed to establish a research, development, and production base and a technological innovation system for environment-friendly building materials and products by promoting the partnership of industry-university-research institute, integrating the technologies, and building demonstration production lines. Implementation of this project would provide technical support and safeguard measures for research, development, production, application, and operation management of green building materials and products in China.

2. Key technologies for improving and ensuring the quality of urban living environments

   The living environment greatly affects people's quality of life, health, and productivity. It is an important indicator of a country's or region's economic development and living standards and also an important part of the key technology system for sustainable development of construction industry. In order to improve the quality of living environments to improve people's quality of life, a major project of the National Key Technologies R&D Program, "Key technologies for improving and ensuring the quality of urban living environments," was launched by the Ministry of Science and Technology of the People's Republic of China.

   The major goal of the project was to realize the "leapfrog" development of the key technologies for improving and ensuring the quality of urban living environments by developing key technologies and complete sets of equipment with independent intellectual property rights and formulating and improving regulations and standards related with urban living environments. Based on technology integration and demonstration, the project was expected to establish a research and development base and an independent innovation system for technologies of improving and ensuring the quality of urban living environments.

3. Key technologies and equipment for removal of typical indoor air pollutants

   As one of the most serious environmental problems in China, indoor air pollution in public places has gathered growing concern in recent years due to its adverse effects on human health and quality of life and work. In order to meet major demands of indoor air pollution control for large public places (e.g., supermarkets, shopping malls, places of entertainment), the Ministry of Science and Technology of the People's Republic of China provided financial supports for a key project under the National High-Tech R&D Program (863 Program), i.e., "Key technologies and equipment for removal of typical indoor air pollutants."

   The overall objective of the project was to develop technologies for analyzing and regulating typical indoor air pollutants (organic pollutants and harmful microorganisms) in public places, provide new cleaning materials and technologies with independent intellectual property rights, manufacture cleaning function modules, develop and demonstrate new independent air cleaners and air cleaning prototype units for central air-conditioning systems, establish an indoor air cleaning technology system, and provide support for boosting the technical level of indoor air cleaning technologies and equipment and improving the IAQ of public places in China.

In summary, China has conducted large amounts of research work on indoor air pollution and its control over the past two "Five-Year Plan" periods and has realized "leapfrog" progress in improving IAQ by developing and applying new technologies, materials, and equipment. According to the "National 12th Five-Year Program for the Science and Technology Development," China will further push forward the research, development, and industrialization of indoor air cleaning technologies and equipment during the 12th Five-Year Plan period.

3.3 Developing Indoor Environmental Monitoring and Cleaning Industry

The indoor environmental monitoring and cleaning industry emerged in the 1980s in China and has grown at a rapid pace since 2000 along with the occurrence of serious indoor air pollution due to interior decoration and refurbishment, the increasing public awareness of the importance of IAQ, the promulgation and implementation of several national standards concerning IAQ, and the progress in research on indoor air pollution and its control. A relatively complete industrial system of indoor environmental monitoring and cleaning has formed in China, including air cleaners, air cleaning materials, air cleaning systems, indoor decorating and refurbishing materials with cleaning function, fresh air ventilation systems, and indoor environmental pollution monitoring, control, and evaluation systems. Statistics indicate that the scale of the indoor environmental protection (mainly monitoring and cleaning) industry has been growing with a compound annual growth rate of 28% in recent years, reaching 30 billion yuan in 2008 [65].

In view of the current status and future trends of indoor air pollution in China, demand for indoor environmental monitoring and cleaning will continue to grow in the coming years, requiring further development of the indoor environmental monitoring and cleaning industry. On the other hand, as an industry with high technology content, technical level of the indoor environmental monitoring and cleaning industry determines its capability of providing new types of services and products quickly in response to market needs. In order to boost the innovation capacity of enterprises engaged in indoor environmental monitoring and cleaning to facilitate technical upgrading, the Chinese government has been greatly promoting the partnership of industry-university-research institute in research, development, and industrialization of new technologies and equipment, which will undoubtedly help enhance the overall development of the industry.

4 Major Problems in Current Indoor Air Pollution Control

Despite the great progress in indoor air pollution control in China, there are still some problems which limit the effectiveness of pollution control measures, including lack of mandatory standards for IAQ, lack of regulation and labeling of pollutant emissions from indoor decorating and refurbishing materials, lack of an effective performance evaluation system for air cleaning products, and lack of proper maintenance of air cleaners.

4.1 Lack of Mandatory Standards for Indoor Air Quality

GB/T 18883-2002 “Indoor air quality standard” has been implemented for nearly 10 years in China and has played significant roles in preventing and controlling indoor air pollution and safeguarding people’s health. As a standard with no mandatory enforcement mechanism, however, GB/T 18883-2002 is not binding for either building developers or renovation contractors. Although GB 50325 “Code for indoor environmental pollution control of civil building engineering” stipulates mandatory acceptance criteria for indoor environmental quality of newly constructed, extended, or renovated civil building engineering, it does not apply to civil buildings delivered to users. Moreover, passing the acceptance inspection required by GB 50325 does not mean or provide assurance for good IAQ during the normal use of civil buildings. On the one hand, inadequate ventilation and/or introduction of new pollution sources such as furniture and chemical household products can increase the indoor pollutant concentrations. On the other hand, many other pollutants besides those included in GB 50325 (radon, formaldehyde, benzene, ammonia, and TVOC) can also deteriorate the IAQ and cause adverse health effects.

4.2 Lack of Regulation and Labeling of Pollutant Emissions from Indoor Decorating and Refurbishing Materials

Although GB 18580 and GB 18587 include pollutant emission limits for wood-based panels and finishing products and carpets, carpet cushions, and adhesives, national standards for other indoor decorating and refurbishing materials (GB 18581–18586) prescribe content limits of the regulated harmful substances. The contents of harmful substances determine the potential of materials (products) to cause indoor air pollution but not the actual indoor air pollution level due to the lack of correlation between the contents and the emission features of harmful substances. As a result, the current labeling system for indoor decorating and refurbishing materials, mainly based on the contents of harmful substances, is not effective enough in guiding the selection of indoor decorating and refurbishing materials to control indoor air pollution at its source, explaining why the IAQ of some newly renovated buildings fails to meet the state standards although all the decorating and refurbishing materials (products) used conform to the corresponding national standards.

4.3 Lack of an Effective Performance Evaluation System for Air Cleaning Products

Both the type and quantity of air cleaning products on the market have been significantly increasing in recent years in China due to the rapid development of indoor air cleaning industry. Effective performance evaluation of air cleaning products is the base for regulating the market and promoting the development of better performing products. At present, however, no performance evaluation for air cleaning materials is being conducted due to the lack of relevant regulations and standards. Although a recommendatory national standard GB/T 18801-2002 “Air cleaner” was promulgated by the General Administration of Quality Supervision, Inspection and Quarantine of the People’s Republic of China in 2002 and revised in 2008 (GB/T 18801-2008), they have played very limited roles in regulating the air cleaner market and guiding the consumers to choose appropriate air cleaners because the involved performance evaluation indices, cleaning efficiency of pollutants (GB/T 18801-2002), and clean air delivery rate (CADR) per unit of energy consumption (GB/T 18801-2008), cannot reflect the applicability or the actual CADR of air cleaners. Besides, the current evaluation of air cleaners has not taken the production of possible hazardous byproducts except O_3 (included in the building industry standard JG/T 294-2010 “Test of pollutant cleaning performance of air cleaner”) into consideration.

4.4 Lack of Proper Maintenance of Air Cleaners

As the most important type of air cleaning products, air cleaners have become more and more popular in urban Chinese households. The domestic sales volume of air cleaners increased from 400 thousand units in 2006 to 1 million units in 2010 [12]. Since filtration and adsorption are the commonly used air cleaning technologies in air cleaners, periodical cleaning and/or replacement of the filtration and adsorption materials are required to ensure the cleaning performance of cleaners. Non-/improperly maintained equipment will lose its cleaning capability or even cause secondary indoor air pollution by releasing trapped pollutants back into the air. At present, however, neither the air cleaner production enterprises nor any special agencies in China provide specialized maintenance service for air cleaners, which absolutely discourages the promotion use of air cleaners and obstructs the development of air cleaner industry.

5 Concluding Remarks

Indoor formaldehyde pollution has been effectively alleviated in recent years in China by limiting the formaldehyde emission of wood-based panels and finishing products used in indoor decoration and refurbishment. Although indoor pollution of benzene has been effectively reduced through forbidding the use of benzene as solvents and diluents for coatings and adhesives that are intended for indoor use, indoor pollution of toluene and xylenes still exists. Serious outdoor pollution of PM, poor maintenance of air-conditioning systems, and the increasing use of materials and products containing plasticizers and flame retardants are major causes of indoor pollution of PM, biological pollutants, and SVOCs in China, to which more attention should be paid in the future.

Great progress in indoor air pollution control has been achieved in China through many years of efforts on formulating and improving IAQ-related regulations and standards, strengthening research on indoor air pollution and its control, and developing indoor environmental monitoring and cleaning industry. However, there are still some problems in the current control of indoor air pollution, including lack of mandatory standards for IAQ, lack of regulation and labeling of pollutant emissions from indoor decorating and refurbishing materials, lack of an effective performance evaluation system for air cleaning products, and lack of proper maintenance of air cleaners. These problems should be addressed as soon as possible to ensure the effectiveness of pollution control measures.

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