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Ecotechnologies for the Treatment of Variable Stormwater and Wastewater Flows



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Ecotechnologies for the Treatment of Variable Stormwater and Wastewater Flows



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Preface

Wastewater and stormwater flows are inherently variable, responding to rainfall events and fluctuations in loading. This variability has significant effects on treatment systems performance, largely related to changes in hydraulic residence times and fluctuations in pollutant loadings. How much this variability influences treatment performance has rarely been specifically addressed in design manuals or performance assessments and modelling. This volume aims to start the process of filling this gap by focusing on ecotechnologies, in particular various types of wetlands and ponds, which are generally considered to be relatively resilient to such variations in inflow.

The initial impetus for this book was a 2¹/₂ day workshop on *Ecotechnologies for* treatment of variable wastewater and stormwater flows held at RWTH Aachen University in Germany in October 2014. The workshop was a joint collaboration between the RWTH Aachen University's Institute of Environmental Engineering and The National Institute of Water and Atmospheric Research (NIWA) in New Zealand, funded through the New Zealand-Germany Scientific and Technological Co-operation Agreement. The workshop attracted approximately 30 researchers and practitioners from Germany, France, England, Belgium, Denmark and New Zealand, including research organisations and a range of private sector and other supporting organisations working on treatment wetlands and related ecotechnologies.

A core group from the workshop showed interest in developing and further extending the ideas discussed at the workshop. Despite our initial enthusiasm for the task, other commitments inevitably intervened and threatened to derail the endeavour. However, with perseverance (and including some supplementary assistance along the way to extend and bolster coverage), it has finally come together.

At the beginning, we introduce the range of treatment ecotechnologies considered. Then, we discuss the general impact of flow variability on treatment processes in these systems. Subsequent chapters cover the main contaminant categories of interest: nutrients, faecal microbes, metals and emerging contaminants, spanning urban and agricultural applications. Finally, we review modelling tools that have the potential to improve our understanding and ability to simulate and predict responses to flow variability.

We would like to take this opportunity to thank the funding agencies—the German Federal Ministry of Research and Education and the Royal Society of New Zealand (on behalf of the Ministry of Business, Innovation and Employment)—for providing the means to run the initial workshop and strengthen our collaboration; and our respective institutes for the support that has enabled us to bring this publication to fruition. We hope the information and ideas contained in the book prove useful to both researchers and practitioners, and inspire others to better understand the extent to which flow variations influence how well treatment ecotechnologies function, and promote development of improved modelling tools for design and assessment.

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Chapter 1 Introduction

Katharina Tondera, Chris C. Tanner, Florent Chazarenc and Godecke-Tobias Blecken

Abstract The occurrence of variable stormwater and wastewater flows, mostly precipitation driven, brings with them the challenge of both peak flows and pollutant loads. Wastewater treatment systems can be divided into those that are specifically designed and operated to deal with variable flows, and those that presume more steady-state operation, only coping with peak flows as anomalies for short periods of time. To date, the different types and scales of variability and the impact of this variability on functioning and treatment performance have neither been well characterised nor properly dealt with for the design of suitable treatment systems. In this book, *ecotechnologies* are defined as processes for the treatment of variable wastewater flows that

- harness ecological processes involving microbes, plants, animals, natural soils and media or recycled materials;
- have a low reliance on mechanical machinery or external energy sources; and
- have a positive impact on the quality and biodiversity of the surrounding environment.

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This book focuses on treatment systems compliant with these definitions, but which are also specifically designed for variable flows.

General Scope

Stormwater treatment systems are, by definition, subject to the vagaries of climate, especially variations in rainfall patterns and intensity, and snowmelt runoff. Highly variable wastewater flows often appear when such climate events occur, but also when there are major fluctuations in waste water generation, for example, in tourist and recreational facilities and seasonal processing industries. Wastewater treatment systems can be divided into those that are specifically designed and operated to deal with variable flows, and those that presume more steady-state operation, only coping with peak flows as anomalies for short periods of time.

To date, the different types and scales of variability and the impact of this variability on functioning and treatment performance have neither been well characterised nor properly dealt with for the design of suitable treatment systems. Nevertheless, there are some examples of treatment systems that are specifically constructed for such extreme conditions, e.g. treatment wetlands for combined sewer overflows (CSOs) and urban stormwater (Uhl and Dittmer 2005; Fassman-Beck et al. 2014). Based on monitoring of actual performance in practice, many existing systems for highly variable flows require improvements to enhance their efficacy, and associated design and operational guidelines need refinement.

Combined wastewater flows containing surface runoff do not follow regular and predictable patterns. Their operation is defined by the irregularity of precipitation and runoff. Although much effort has been put into developing models based on regular patterns of return period and intensity and real-time data from sewer installations, it is still hard to get reliable predictions on events causing overflows (Löwe et al. 2016).

Guidelines for dimensioning sewer systems are based on assumed standardised rainfall patterns; however, every rainfall event creates its own characteristic pattern. Moreover, many regions of the world are experiencing climatic changes resulting in a shift in the return periods and intensities of storms, a phenomenon which is likely to escalate in the future (IPCC 2014). In some areas, once-in-a-hundred-year events based on measurements from former decades have occurred repeatedly in the last 15 years (Larsen et al. 2009; Arnbjerg-Nielsen et al. 2013). Changing climates can also result in extended droughts, leading to higher first flush concentrations in combined sewer systems and elevated pollutant loads, especially during summer storm events.

On the other hand, warmer temperatures in winter and steady rains lead to decreased wastewater temperatures in combined sewer systems and increased sewer overflows at this time of the year. In separate sewer systems, wastewater treatment systems face higher pollutant loads in wet periods due to groundwater ingress and illicit connections (Panasiuk et al. 2015; Ellis and Bertrand-Krajewski 2010).

In terms of strongly varying sediment, nutrient and microbial loads, farmland is one of the main contributors to surface water pollution outside of urban areas (Carpenter et al. 1998; Howard-Williams et al. 2011; McDowell 2008; Schreiber et al. 2015). Especially in highly intensified farming areas, manure application in combination with (heavy) rainfall events leads to increased nutrient concentrations in rivers that can result in algal blooms and fish kills (Magaud et al. 1997).

Variation Driven by Precipitation

The concentrations found in stormwater and stormwater-driven wastewater flows such as CSOs or agricultural diffuse runoff often vary

- within single events (e.g. due to first flush effects or varying rain intensities during the event which transport different fractions),
- between different events (e.g. due to varying antecedent dry periods, seasonal variations, varying rain characteristics),
- between seasons, and
- between different catchments (due to different catchment characteristics).

These variations interact with each other which makes it difficult to define standard values for certain pollutants and/or assess stormwater quality by grab sampling.

Often, a greater proportion of the pollution is flushed off at the beginning of a runoff event as a first dirt pulse, which is often described as 'first flush'. The concentrations then subside with the ongoing runoff (Marsalek 1976). There is no rigid definition of the proportion of particles in a first flush. Notwithstanding, first flush effects are presented in many studies (Sansalone and Buchberger 1997; Lee et al. 2002; Wicke et al. 2012), while in others, this phenomenon only occurred in a small part of the analysed sampling events (Saget et al. 1996; Deletic 1998; Tondera et al. 2013) or not at all (Bertrand-Krajewski et al. 1998). Also, the remobilisation of sediments in sewers can contribute to first flush effects. The release from sewer sediments themselves is again dependent on the chemical composition of the runoff, e.g. the pH value (Li et al. 2013). However, a first flush is not always observed. In larger catchments, different 'first flushs' from sub-catchments can overlap, resulting in other distributions of the pollutant concentrations during the runoff event. Furthermore, varying rain intensities during the event can transport different contaminant fractions at different time steps of the event.

Important factors determining the pollutant concentrations in storm runoff are the **precipitation depth, intensity** and **duration** (Borris et al. 2014). Additionally, the **length of the dry period** preceding the storm event has a significant impact on most of the pollutants present in the water (Pitt et al. 1995).

Significant **seasonal variations** of stormwater quality have been observed. In general, in cold and temperate climates, pollutant concentrations are higher and more variable in winter runoff, e.g. after snowmelt.

Ecotechnologies

Before modern wastewater treatment systems were implemented, all wastewater was essentially treated 'ecologically', although it was often discharged at rates greatly exceeding the natural assimilation capacity of the environment. Even the modern activated sludge process is driven by natural microorganisms; hence, a distinction between 'ecotechnologies' and 'conventional treatment' is not clear-cut.

The idea behind the first developments of so-called ecotechnologies was to harness the capacity of nature, particularly plants and soil, to degrade and cleanse impurities. Engineered systems were developed that mimic the treatment processes occurring in natural ecosystems, particularly wetlands (Seidel 1966). This emerged from rising awareness of the consequences of unchecked growth of industry and the human population on the environment, ecosystems and human health. Ecotechnologies also offer a wide range of other potential benefits including cost-effectiveness, energy efficiency, often simple operation and maintenance, robustness and resilience. However, they often require larger land areas than conventional mechanised approaches.

In urban drainage, green infrastructure—as defined by the terms Sustainable Drainage Systems (SuDS), Water Sensitive Urban Design (WSUD) or others¹ (Melbourne Water 2005; Woods Ballard et al. 2015)—was developed to move from a focus on 'end-of-pipe' approaches to management of stormwater close to its source and adaption to the natural water cycle. Approaches employed include harvesting of rainfall runoff from roofs, maximising infiltration to soil and using ecotechnologies such as bioretention filters, water gardens, swales and wetlands to buffer and treat flows.

Unfortunately, comprehensive SuDS/WSUD approaches are not always possible, especially in densely populated areas; however, end-of-pipe treatment of discharge events that cannot be managed at source, or in conventional infrastructure, can often be accomplished using ecotechnologies such as constructed wetlands (Seidel 1966; Kadlec and Wallace 2008).

In this book, *ecotechnologies* are defined as processes for the treatment of variable wastewater flows that

- harness ecological processes involving microbes, plants, animals, natural soils and media or recycled materials;
- have a low reliance on mechanical machinery or external energy sources; and
- have a positive impact on the quality and biodiversity of the surrounding environment.

¹An overview of terminology for green infrastructure in Urban Drainage is provided by Fletcher et al. (2015).

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Chapter 2 Treatment Techniques for Variable Flows

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Abstract A wide range of ecotechnologies has been applied to treatment of variable stormwater and wastewater flows. Stormwater ponds and basins were already introduced as common 'end-of-the-pipe' treatment solutions in the 1960s, almost parallel to the first attempts to develop structured wastewater treatment with the help of plants, inspired by natural wetlands. Constructed wetlands specifically designed for the treatment of variable flows emerged in the 1990s and were joined by a growing group of vegetated filter systems, named bioretention filters, raingardens or retention soil filters, all following the principle of gravity-driven wastewater filtration. This chapter provides a general overview of these treatment facilities, including swales and buffer strips. Although the latter ones are gravity-driven filtration systems, they are commonly used for the treatment of road runoff and are highly adapted to fit into their landscape structure, they are described in a separate section. Each section includes references to detailed design and operation guidelines.

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Treatment Systems

Ecotechnologies for the treatment of variable flows, especially for those driven by stormwater, combined sewer overflows (CSOs) and agricultural runoff, come in various designs and definitions. As described in Chap. 1, several concepts exist to group these techniques by their purpose or design. This chapter provides an overview of principle design components and operational challenges of the most common and widely used techniques as background information for the following chapters. Each section includes references to detailed design and operation guidelines.

Stormwater Ponds and Basins

Stormwater ponds (also called wet detention basins and sedimentation basins/ ponds) and other sedimentation-based treatment facilities are common 'end-of-the-pipe' treatment solutions for the storage and treatment of large stormwater volumes.

Stormwater ponds have been implemented since the 1960s in the USA (Clar et al. 2004) and their number has increased constantly since then (Marsalek and Marsalek 1997; Starzec et al. 2005; Karlsson et al. 2010).

During the last three to four decades, design and dimensioning of ponds have been improved by research and practical experience. Their main design elements are the different hydraulic structures (inlet and outlet, overflow structures) and their volume (extended detention volume, storage volume for sediment). Furthermore, hydraulic efficiency has to be considered to ensure that flows are distributed as evenly as possible throughout the pond to ensure efficient sedimentation.

Outlets, which are frequently designed to detain fractions of runoff for multiple days after a storm, are prone to clogging, which can affect the water level in the pond and, thus, its function. Hence, regular (at least annual) inspections of the key structures of ponds are required.

Usually the whole runoff volume is captured in the facility and released over time (sometimes up to several days), a process that enables settling of suspended sediments and associated pollutants. These ponds can provide treatment mostly through sedimentation when designed, constructed and maintained to this purpose. However, field experience shows that, in practice, sediment settling is a rather complex process which is affected by a range of factors (e.g. disturbance by turbulence generated at high flow rates, waves or currents).

Accumulated sediment must be removed regularly from the pond to maintain its treatment volume and guard against remobilisation during high flow events. How often sediment needs to be excavated depends on the catchment to pond ratio, the sedimentation efficiency and the sediment load from the catchment, but an interval of five years is reasonable. Ponds must thus be accessible for personnel (regular inspection) and machinery to ensure a sufficient long-term function (excavation of accumulated sediment).

Pollutants such as metals often occur as very small particles. Xanthopoulos (1990) investigated the size distribution of particulate matter for several heavy metals and found 67–87% were bound to particles with a grain size of less than 60 μ m. Boogaard et al. (2014) confirmed these results in runoff from the Netherlands, showing that approximately 50% of the investigated particle mass is bound to particles <90 μ m. Accordingly, how effectively stormwater ponds remove pollutants depends heavily on their association with settleable solids. Healthy Waterways (2006) proposes targeting sediment that is 125 μ m and larger in ponds and choosing alternative treatment technologies to remove finer material and/or dissolved pollutants from urban stormwater. However, in practice, many ponds also remove considerable loads of finer sediment (Al-Rubaei et al. 2016).

Often stormwater ponds are combined with a smaller upstream pretreatment basin or a forebay which provides an initial deposition area for coarse and larger soil particles. These coarser particles represent a relatively large volume of the total sediment, but carry only a minor portion of the total pollutants. Thus, forebays which are typically sized to comprise 10% of a pond's surface area facilitate maintenance of the whole system.

Theoretically, a gradient from coarse to fine sediment will form as the flows pass through the pond, since the settling velocity decreases with the sediment diameter (i.e. gravel and sand settle close to the inlet, Fig. 2.1). The theoretical sediment settling efficiency can then be easily calculated with empirical equations.

However, field experience shows that sediment settling in practice is a rather complex process affected by various factors (e.g. disturbance by turbulence generated at high flow rates, waves or currents). Al-Rubaei et al. (2016) showed in a performance survey of 30 municipal ponds in Sweden that in some ponds, the percentage of fines (<125 μ m) was below 5% at both inlet and outlet while in others it was already above 90% close to the inlet. Some ponds also showed a decreasing content of fine solids from the inlet to the outlet. These variations underline how various factors influence the settling performance in practice. Due to this settling, ponds remove the pollutants attached to the sediments.

Often, the percentage of settled sediment is used as a parameter to describe a pond's treatment efficiency. However, Marsalek et al. (2005) argue that this measure is insufficient since it does not take into account the particle sizes of settled and



Fig. 2.1 Simplified sketch of sedimentation of different particle size fractions in a stormwater pond including a forebay (*Scheme* G.-T. Blecken)

Reference	TSS (mg/L)		Removal
	In	Out	efficiency (%)
Pettersson et al. (1999)	55.2	16.6	70
	153	25	84
Comings et al. (2000)	16.2	2.9	82
	22.8	8.9	61
Mallin et al. (2002)	10.5	4.4	58
Vollertsen et al. (2009)	276	43	84
Istenič et al. (2012)	48	15	69
	53	5	91
	37	2	95

*Table partially based on the work of Al-Rubaei (2016) and Søberg (2014)

discharged sediment; even with a substantial removal of 70%, a pond may be poor at removing fines (Greb and Bannerman 1997). This is important for the overall treatment capacity since the fine particles commonly exhibit relatively high pollution loads (Sansalone and Buchberger 1997; Liebens 2002) and, along with dissolved forms, tend to be the most bioavailable and toxic to aquatic life (Luoma 1983).

There are a large number of studies evaluating removal efficiencies in stormwater ponds. Since the removal rates vary considerably, Table 2.1 gives an overview of total suspended solids (TSS) removal in a range of studies. A larger International Stormwater Best Management Practices (BMP) Database has been compiled with performance data summarised for a wide range of different stormwater treatment devices and contaminants by Leisenring et al. (2014).

In general, ponds only remove dissolved pollutants to a limited extent since sedimentation is the main treatment process. Dissolved pollutants can be removed by biological processes associated with emergent vegetation planted in shallow parts of ponds (Van Buren et al. 1997). Under favourable conditions (e.g. large vegetated shallow areas), relatively high removal rates can be achieved. Nonetheless, ponds are not a sufficient treatment solution if removing dissolved substances is a high priority even though some ponds can achieve relatively high removal rates.

During typical temperate climate winters (Fig. 2.2), high variability in flows, characterised by extended periods with no runoff followed by snowmelt events with large stormwater volumes over a short period, may result in reduced removal efficiencies (German et al. 2003). Due to density differences compared to pond water, salt-laden and/or cooler inflows from roads may pass through the pond as an underflow or sinking jet (Marsalek et al. 2005). This can generate flow shortcuts, with higher flow velocities disturbing and resuspending already accumulated sediment. Conversely, in warm regions, hot inflow water may pass in the top water layer only.

 Table 2.1
 Mean inflow and outflow concentrations, with nominal removal efficiencies (%) of TSS (mg/L) in nine stormwater wet ponds*



Fig. 2.2 Stormwater pond in winter (Photo G.-T. Blecken)

Roseen et al. (2009) evaluated the seasonal variation of removal efficiencies in stormwater treatment facilities in New Hampshire, USA. While nitrate removal in ponds was less efficient during winter, no significant differences of TSS, phosphorus and zinc removal were detected. Neither did German et al. (2003) observe direct temperature effects on the removal of TSS, phosphorus and nitrogen. Kadlec and Reddy (2001) conclude that the physical treatment processes (mainly sediment settling) are not directly affected by cold ambient temperatures. Since sedimentation is the main treatment process in ponds, their overall treatment performance during winters is likely to be primarily influenced by flow dynamics rather than low temperatures. Conversely, under warm conditions with minimal flushing, phytoplankton and filamentous algal may proliferate in wet retention ponds causing increases in particulate loads to receiving waters once flow resumes (Gold et al. 2017).

Constructed Wetlands

Constructed wetlands (CWs) are being used for the treatment of wastewater and stormwater worldwide, but are also increasingly becoming a recognised system for treating agricultural wastewater and drainage water. CWs have the potential to deal with fluctuations in usage and loading because they harness robust natural treatment processes and have extended residence times.

Based upon flow routing, there are two basic types of CWs: surface- and subsurface-flow wetlands. Four variants are dominantly used for the treatment of variable flows:

- surface-flow wetlands,
- floating treatment wetlands, a variation of the surface-flow wetlands,
- subsurface-flow wetlands with horizontal flow and
- subsurface-flow wetlands with vertical flow, which are summarised with the bioretention filters in this book due to their similar design and function.

Surface-Flow Constructed Wetlands

In surface-flow constructed wetlands (SFCWs), especially in Australasia referred to as constructed stormwater wetlands (CSWs), the deeper pools facilitate sedimentation, while the diverse water-vegetation-soil matrix in the shallower, extensively vegetated zones of SFCWs provide complex multiple pollutant treatment mechanisms. These include sedimentation, flow detention, filtration, adsorption, precipitation, microbial decomposition and plant uptake. Vegetation within a pond/ wetland system reduces flow velocities and allows suspended solids to settle out of the water column. In addition, nutrients and metals can be taken up by vegetation (Fig. 2.3).

In contrast to large detention/sedimentation facilities like wet ponds, which are dominated by large open water areas, SFCWs include various zones with different water depths, thus improving flow retention and providing more diversified high quality treatment mechanisms, particularly with respect to more effective removal of dissolved pollutants and nutrients. Moreover, CSWs are commonly equipped with a forebay to minimise the sediment load and facilitate maintenance. In general, it is preferable to choose native plant species, since the introduction of foreign species via CWs led to spreading of neophytes with severe consequences for native species in some cases (Albert et al. 2013) (Fig. 2.4).

Suspended solids serve as pollutant transport vectors from the input source to the downstream receiving environment. Phosphorus and metals adhere to solids surfaces as they travel along the route. Removal of suspended solids from the water columns in pond and wetland systems is primarily achieved by sedimentation and filtration. Stormwater ponds are primarily designed to provide sufficient removal of TSS with absorbed pollutants from stormwater by sedimentation (VanLoon et al. 2000).



Fig. 2.3 Surface-flow constructed wetland during rain (left), in summer (middle) and in winter (right) (*Photos* G.-T. Blecken)



Fig. 2.4 Simplified sketch of a surface-flow constructed wetland (figure courtesy of Tom Headley)

Table 2.2 Mean inflow and outflow concentrations with nominal removal efficiencies (%) of TSS in surface-flow constructed wetlands*

	1		
	TSS (mg/L)		
	In	Out	Removal (%)
Carleton et al. (2001)	-	-	-300-99.6
Bulc and Slak (2003)	42	11	69
Birch et al. (2004)	48–154	33–172	-97-56
Terzakis et al. (2008)	203	22	89
Yi et al. (2010)	282.8	33.4	84.7
Lenhart and Hunt (2011)	23.6	32.7	-39
Merriman and Hunt (2014)	9.89	8.37	15

*Table partly based on the work of (2016) and Søberg (2014)

However, this removal process can be disturbed by solids scouring in ponds and chemical releases from the deposited sediments (Marsalek and Marsalek 1997).

In practice, high variations of CSWs' treatment efficiencies have been observed. Commonly, CSWs are combined with a preceding forebay or pond to reduce sediment loads entering the wetland itself.

Table 2.2 gives an overview of total suspended solids (TSS) concentrations in the inflow and outflow of different SFCWs.

Floating Treatment Wetlands

Floating treatment wetlands (FTWs), also known as Constructed Floating Wetlands and a wide range of alternative names, consist of buoyant artificial rafts or islands



Fig. 2.5 Floating Treatment Wetlands in a residential stormwater treatment pond in Illinois (USA) (*Photo* C. C. Tanner)



Fig. 2.6 Generalised sketch of a Floating Treatment Wetland (figure courtesy of Tom Headley)

vegetated with emergent macrophytes. They are ideal for systems that experience large variations in flow because their buoyancy allows them to rise and fall with fluctuating water levels, therefore avoiding submergence stress on the emergent plants (Headley and Tanner 2012). They also have the advantage that they can be retrofitted into existing pond systems to augment conventional pond treatment processes (Fig. 2.5).

The floating island matrix (Fig. 2.6) is often made of post-consumer plastics with the aid of synthetic foam sections in combination with organic material such as coconut fibre. The islands are anchored to avoid drifting.

The design of FTWs has been adapted from naturally occurring floating vegetated islands, which can be found in freshwater lakes and ponds, and are comprised of a matrix of floating organic material and plant associations growing at the water surface. Buoyancy is provided by gaseous emissions from organic decomposition (mainly CH_4 and N_2) trapped beneath the organic mat and the air spaces (aerenchyma) within the roots, rhizomes and stolons of vegetation (Hogg and Wein 1988; Mitsch and Gosselink 1993). In contrast, most artificial FTWs rely primarily on buoyant structures to keep them afloat, likely aided by plant tissue buoyancy as vegetative biomass increases.

Recognising the habitat value of floating islands, particularly for birds, the UK Royal Society for the Protection of Birds constructed artificial islands for the conservation of threatened species in as early as the 1960s (Hoeger 1988; Burgess and Hirons 1992). Following these early successes, FTWs have since been used for a variety of purposes including treatment of stormwaters, mine and landfill leachates, CSOs, domestic, industrial and agricultural wastewaters, and eutrophic ponds, reservoirs, lakes, drains, streams and rivers (Chen et al. 2016; Headley and Tanner 2012; Pavlineri et al. 2017).

The plant roots and attached biofilms that extend into the water beneath the floating mats are considered to be crucial to the functioning of FTWs (Headley and Tanner 2012). This root mass reduces flow velocities beneath the FTWs, promoting settlement and physical filtering of suspended solids (Fig. 2.6). Biofilms attached to the suspended root mass promote adhesion of fine particulates, adsorption and nutrient transformations (Borne 2014; Borne et al. 2013a, b, 2014; Tanner and Headley 2011; Winston et al. 2013). Plant detritus can act as metal biosorbent (Southichak et al. 2006), and, along with roots and biofilms, contribute organic exudates, extracellular polymeric substances and humic compounds that promote floc formation that may enhance settling of fine particulates (Borne et al. 2015; Kosolapov et al. 2004; Tanner and Headley 2011).

FTWs may also indirectly affect contaminant removal processes by modifying the physicochemical environment in ponds. FTWs shade the water surface, moderating temperatures (Strosnider et al. 2017) and reducing growth of phytoplankton and submerged macrophytes (Jones et al. 2017). Ponds with a significant cover of FTWs generally show deoxygenation beneath the beds and within the root mass, due to the respiratory demand of the large root and microbial biomass and restriction of atmospheric exchange (Tanner and Headley 2011; Strosnider et al. 2017). Such anaerobic conditions can promote microbial processes such as denitrification (Borne et al. 2013b) and sequestration of metals in underlying sediments (e.g. as metal sulphides) (Borne et al. 2013a, 2014).

The plants growing on FTWs, of course, also take up a range of nutrients, metals and organic compounds directly from the water column via their roots. However, the importance of such plant assimilation compared to other removal processes varies depending on relative nutrient loading rates, pond coverage, plants species, stage of growth, season, etc. (Chen et al. 2016; Headley and Tanner 2012; Pavlineri et al. 2017). Where plant uptake is a quantitatively important removal mechanism, harvesting of emergent biomass is a potential way to permanently remove nutrients from the system and sustain ongoing uptake (Keizer-Vlek et al. 2014; Wang et al. 2014).

Although FTWs are mainly applied for treating stormwater from separate sewer systems, there are also a few examples of FTWs used for CSO treatment. The first system described was a system in Belgium (Van de Moortel et al. 2011). As is common for CSO treatment, a preliminary sedimentation basin lined with hardened bitumen reduces the energy of the incoming water and minimises the resuspension of settled sediments. When entering the second treatment stage, a floating baffle retains large floating debris. The second stage consists of a long basin that is almost completely covered with FTWs and is designed to enhance plug flow.

Another system in the USA combines a FTW, serving as the preliminary stage, with a vertical-flow wetland as the secondary and a SFCW as the final stage (Tao et al. 2014).

Plant species for FTWs have to be chosen according to the environment where the treatment systems are applied, e.g. stormwater ponds or lagoons for CSO treatment. In general, the species should be able to provide the aforementioned root system which removes fine suspended solids and dissolved substances from the inflowing water. For the removal of nutrients, a strong plant uptake without extensive growth on the mat surface is favourable.

The knowledge base on FTW performance treating a wide range of different stormwaters and wastewaters is increasing rapidly (see reviews by Chen et al. 2016; Headley and Tanner 2012; Pavlineri et al. 2017). However, most quantitative studies were conducted on relatively small and immature experimental systems, and so long-term experience is missing. This is especially important for understanding and optimising the scale-dependent indirect effects of FTWs and managing possible unintended consequences on the biogeochemistry and ecology of water bodies. For instance, high covers of FTWs under certain circumstances could result in excessive deoxygenation of the water column, stimulating processes such as phosphorus and methylmercury release from sediments or impacting on resident or downstream aquatic fish and invertebrates (Fig. 2.7).

Headley and Tanner (2012) proposed a conceptual design for incorporating FTWs into a stormwater treatment train. However, at this stage, there are still no established guidelines for optimal coverage, distribution or configuration of FTWs in ponds, and reliable estimates of their performance remain a significant engineering need. A simple first-order model to predict treatment performance for the water body plus the additional treatment provided by different coverages of FTW has recently been developed (Wang and Sample 2013). An expert panel convened by the Chesapeake Stormwater Network on the eastern seaboard of USA has recently assessed the evidence base for FTW stormwater treatment performance and, for regulatory purposes, determined expected enhancements of sediment and nutrient removal rates for FTW retrofits in the region (Schueler et al. 2016). Preliminary guidance on implementation and maintenance of FTWs for urban stormwater treatment has also been developed based on experience in USA and New Zealand by Borne et al. (2015).

Fig. 2.7 Extracted section of a Floating Treatment Wetland treating road runoff showing root mass extending beneath floating mat (Karine Borne, Auckland, New Zealand) (*Photo* C. C. Tanner)



Subsurface-Flow Constructed Wetlands

Subsurface-flow constructed wetlands (SSFCWs) can be designed as horizontal or vertical-flow systems (Kadlec and Wallace 2008). A porous sand or gravel media is generally used to provide adequate hydraulic conductivity. Emergent wetland plants grow hydroponically in the media providing for at least partial interaction with the plant root zone (Brix 1997; Tanner 2001). Inflow is either introduced passively at one end of a saturated bed, promoting horizontal flow through the media, or dosed intermittently to the top of the media promoting percolation down through unsaturated media. SSF systems have the advantage that contaminated water is generally retained below the surface and so avoid potential for human contact or proliferation of mosquitos or other insect pests. The media also provides a physical filtering role, enhanced solids retention and a stable substrate for biofilm development.

SSFCWs are able to retain a large number of pollutants and to partially degrade them. The relevant treatment mechanisms have been investigated for saturated soils



Fig. 2.8 Principles of surface filtration (left), straining (middle) and adsorption (right) in vertical-flow systems (adapted from Seidemann 1997)

and unsaturated sand as well as on laboratory and large-scale systems. The major mechanism for particle retention is filtration, which can be divided into straining and surface filtration. Surface filtration retains all particles that cannot pass the surface, which applies to particles with a size >5 μ m when the filter sand is chosen with the characteristics of the one used in Germany (grain size 0/2 mm). Figure 2.8 illustrates the principles of filtration, straining and adsorption in a vertical-flow system.

Straining occurs when a particle in suspension flows through a pore opening that is too small for it to pass through so microorganisms become entrapped and accumulate on the surface of substrate media.

Suspended particles are adsorbed when their diameter is much smaller than the diameter of the filter material. Corapciogliu and Haridas (1984) found diffuse trajectories of the particles due to Brownian motion and gravitation forces on a particle as drivers for this phenomenon. There is a difference between the sorption capacity of organic and inorganic substances present in soil (abiotic sorption) and the one of microbial structures such as biofilm: the so-called biotic adsorption increases as a biofilm grows in the filter. The sum of exchangeable cations defines the overall sorption capacity of the soil or sand in question.

Low temperatures generally decrease soil biological activity, which may impair biological treatment processes (e.g. biofilm growth, plant uptake). They also result in reduced organic matter decomposition, possibly leading to lower dissolved organic matter (DOM) concentrations in the outflow. Other than the overall treatment performance of ponds, the treatment performance of bioretention filters relies on temperature-dependent biogeochemical processes to a larger extent and, thus, varies with seasons.

Only few studies specifically addressed the problem of clogging in CWs for stormwater treatment, although the phenomenon is well described for systems with relatively constant inflow, e.g. systems for domestic wastewater treatment (Knowles et al. 2011). The main factors leading to clogging—accumulation especially of fine solids, biofilm development, vegetation and chemical decomposition —can be reduced by intermittent operation and sufficient dry periods (Knowles et al. 2011), which is the general nature of stormwater treatment. Insufficient sizing and an overload with fine solids, constant infiltration inflow and the choice of inadequate filter material remain major risk factors for clogging of the systems (Laber 2000; Grotehusmann et al. 2017). However, this is often reversible either by eliminating the cause of the clogging, e.g. by replanting, introducing pretreatment or redirecting infiltration inflow (Laber 2000; Grotehusmann et al. 2017).

Both systems are used to treat fluctuating wastewater and combined sewer flows (Griffin 2003), and more rarely urban, industrial and rural stormwaters (e.g. Laber 2000; Shutes et al. 1997).

The **vertical-flow constructed wetland (VFCW)** is most commonly used in the treatment of variable flows. However, for the treatment of stormwater and wastewater flows—the latter limited to the treatment of CSOs in this book—the system will be described in the Section 'Bioretention Filters'.

Alternatively, in the so-called French vertical-flow systems, raw wastewater is applied directly to the wetland creating a sludge layer on the surface through which inflows are initially filtered (Molle et al. 2005). Such systems are operated in sequence with extended rest periods to maintain the porosity of the media and require periodic removal of the surface deposits after 10–15 years. They have been shown to be able to maintain functioning with stormflows of up to 10-fold normal hydraulic loadings (Molle et al. 2006). Another system based on vertical-flow wetlands is described by Hasselbach (2013): two VFCWs operating in parallel treat the dry weather flow after having been pretreated in a pond. In case of a rainfall event, a third VFCW is fed as well, so that a total flow of two times the dry weather flow and additional infiltration inflow can be treated.

Lucas et al. (2015) report of 67 CWs for stormwater treatment in the UK, most of which are designed as **horizontal-flow constructed wetlands** (HFCWs), used



Fig. 2.9 Generalised sketch of a horizontal subsurface-flow wetland (figure courtesy of Tom Headley)

also for combined sewer systems, separate sewer systems and road runoff. The authors hereby present the largest study on HFCWs for stormwater treatment, including comparisons of design guidelines and a ratio of the required CW area to the catchment of 1–5%. A generalised sketch of the principle is shown in Fig. 2.9.

Some of the systems were already addressed by Ellis et al. (2003) and Rousseau et al. (2005). The removal efficiencies presented by Ellis et al. (2003) were comparably low with regards to vertical-flow systems (see Section 'Bioretention Filters'): the performance of six sites was presented, of which three reached removal efficiencies of -4-75% for TSS, whereas the other three reached 95–99%. Rousseau et al. (2005), who presented the results of a survey on seven HFCWs, suggested that accumulated sludge can be washed out of the system and lead to low or even negative removal rates. Pollutant traps such as settling tanks or ponds could reduce this risk. However, Ávila et al. (2013) described something similar when using a horizontal-flow constructed wetland as part of a treatment train (hybrid wetland): the authors investigated a system treating combined sewage both during dry and wet weather flow, which consists of a pretreatment via screens, sand and grease trap and an Imhoff Tank, followed by a VFCW, a HFCW and a SFCW. During wet weather conditions, the TSS concentrations in the HFCW increased compared to the influent, which the authors led back to a washout of material retained in the gravel bed.

The filter media in the CWs is not only the main treatment media, but also decides the hydraulic retention time. Its porosity determines the water storage capacity; however, it can also be the cause of scouring of filter media (Ellis et al. 2003). In general, vertical-flow systems are preferred over horizontal-flow systems due to their shorter hydraulic retention time, which is crucial especially for the treatment of highly fluctuating stormwater flows.

Bioretention Filters

A wide range of filter technologies is available for stormwater treatment including among others: unvegetated sand filters, vegetated biofilters and compact filters facilitating reactive filter materials for targeted treatment of dissolved pollutants.

The planted gravity flow system—based on slow sand filtration with retention volume on top of the filter level—has proved to be relatively stable in terms of treatment performance, operation and sustainability. It is analogous to the vertical subsurface wetlands used for wastewater treatment (see above), but is only operated during rain periods. In dry weather, the bed drains and is aerated through the drainage pipes. The conditions in the filter sand during operation, change from unsaturated to saturated and back to unsaturated after draining (Dittmer 2006).

Vegetated vertical-flow bioretention filters (also known as rain gardens, biofilters or retention soil filters) typically consist of a vegetated swale or basin, underlain by a filter medium. The water infiltrates and percolates through the filter and during its passage it is filtered by the filter media, plants and microbes via a combination of mechanical and biochemical processes. The treated water is either infiltrated into the surrounding soil or collected in a drainage pipe at the bottom of the filter and then discharged to a recipient or the existing sewer system.

Depending on region, historical background—or as Fonder and Headley (2013) humorously put it, 'the author's desire to give the impression that their design is new or innovative'—the system is called vertical-flow constructed wetland for the treatment of stormwater, CSOs or highway runoff, biofilter, bioretention filter or cells, rain gardens or vegetated sand filter (further names to be continued). In Germany, the term 'Retention Soil Filter' (RSF) is used and accepted for the system. Though this term is used for constructions that treat CSOs, stormwater from separate sewer systems and for highway runoff, international literature commonly uses the term only for application in combined sewer systems.

When such systems were first implemented in Germany in the late 1980s, cohesive material such as soil was used as filter material for CSO treatment. Around the same time, Prince George's County (1993), Maryland, USA, started developing stormwater biofilters as stormwater treatment systems. Since bioretention filter is the most common name for the system, it will be used in the following (Fig. 2.10).

The overall design for all constructions is the same: a preliminary pretreatment stage protects the filter surface from clogging and erosion. In separate sewer systems and for highway runoff, it can be a simple grit chamber, while in combined sewer systems, retention tanks are often used. The bioretention filter itself typically consists of a vegetated swale or basin underlain by a filter medium. A ponding zone



Fig. 2.10 Bioretention filter (2200 m^2) for the treatment of pre-settled CSOs in Germany (*Photo* K. Tondera)



Fig. 2.11 Sketch of a general bioretention filter/vertical-flow CW for stormwater treatment design. The treated water can be either infiltrated (lower left section) or discharged into surface water (right) (*Scheme* K. Tondera)

(height: from approx. 0.2 m for stormwater in separate sewer system and highway runoff to 2.0 m for CSOs) allows temporary storage of water since the stormwater inflow commonly exceeds the infiltration capacity. The filter material consists of either natural soil or engineered media ('technical sand'), typically in a 0.5–1.0 m layer and has a surface area of approximately 0.5–6% of the impervious catchment area.

The treated water is commonly collected in a drainage pipe and discharged to the surface water body, sewer system or infiltrated directly into the surrounding soil, especially in case of highway runoff treatment (Fig. 2.11).

Bioretention filters are not designed to infiltrate high flows in general; these are commonly bypassed directly using an overflow pit or via a retention bed overflow. Thus, bioretention filters are not fully applicable for stormwater retention in the event of intense rain events and have to be combined with retention facilities when flood protection is targeted. Different to the systems treating highway runoff or stormwater in separate sewer systems, those for CSOs are not being loaded during each rain event, but only when a certain storage capacity of the sewer system is exceeded. In Germany, the storage capacity usually includes a certain 'design storm' (r = 15, n = 1) before the overflow feeds the filter bed. However, in first pilot systems built in Italy, 5 mm of the first flush are caught in storage tanks and in case of ongoing rains, the tanks are bypassed and the filter systems fed (Meyer et al. 2014). In Sweden, commonly a retention volume corresponding to 10 mm precipitation is required.

Plants are important for the system to achieve a sufficient performance since they not only contribute to erosion control by stabilising the filter material and lowering water flow velocities, but also support infiltration capacity, provide conditions for microbiological treatment processes (e.g. in the rhizosphere) and aesthetic values.

When designing bioretention filters in public space, engineers have to pay particular attention to landscape design without compromising their primary purpose of handling urban stormwater runoff. Systems for CSO treatment are planted rather monoculturally: in Europe, common reed (*Phragmites australis*) has become state of the art for the filter bed and grass for planting the bank since this helophyte

has proved to be most resilient to water stress during dry phases and shock loading during feeding events. In other regions of the world, local species should be chosen in order to prevent neophytes from spreading. The helophytes need to be able to deal with the extreme conditions of long lasting droughts, temporal impounding after shock loading and low nutrient availability. At the same time, they should not produce much biomass, which would clog the filter over time.

The choice of filter material is crucial to the hydraulic conductivity of the system. Fassman-Beck et al. (2014) describe effects of filter media on the hydraulic conductivity of systems' bioretention filter cells such as New Zealand's rain gardens. The media were mixed with organic material (compost). One of the results showed that the use of a proportion of incompressible sand has a positive effect on unwanted compaction of the filter material. Long-term large-scale applications in Germany also showed that inorganic materials are more resilient to clogging, which led to a shift from using cohesive material to technical sand (0/2 mm) with a steep sieving curve (Dittmer et al. 2016; DWA-A 178 2017). An organic layer which builds up during several years of operation serves as a secondary filter layer. Over time, secondary layers form on top of the filter material from the surface filtration process and mostly contain suspended solids which accumulate on the filter surface and organic material. These secondary filter layers themselves contribute to the overall sorption capacity of the filter. However, accumulation of fines can lead to clogging of the filter surface. Hence, hydraulic conductivity and the retention of substances with no renewable adsorption capacity are in competition. In cold climates and separate sewer systems, an excessively fine-grained filter material with low hydraulic conductivity can also lead to clogging in winter: the pre-freezing soil water content at the time of freezing might lead to the soil becoming an impervious layer with none or close to zero infiltration (e.g. no pollutant removal) referred to as concrete frost. Using a coarser filter material with a higher hydraulic conductivity, thereby minimising the soil water content, might lead to granular or porous frost instead. The latter will maintain and might even exceed the infiltration capacity of the unfrozen soil, thus maintaining proper filter function regarding water quantity.

A coarser grained filter material might jeopardise pollutant removal due to an excessively short retention time in the biofilter. However, the use of a filter material with coarser grain size (e.g. higher sand and lower silt and clay content) than the normally recommended sandy loam soils has been successfully tested in several studies (Blecken et al. 2011; Muthanna et al. 2007a; Søberg 2014). These results were similar to what has been found in other biofilter studies where winter conditions were not taken into account.

A study about seasonal climatic effects on the hydrology of stormwater biofilters (Muthanna et al. 2007b) found a strong correlation between the hydrologic performance of stormwater biofilters and temperature and antecedent dry days. Their results indicate that below zero temperatures and snowmelt can be expected to lower stormwater biofilter hydrology. However, pilot-scale stormwater biofilters have been shown to treat roadside snowmelt efficiently (Muthanna et al. 2007a).



Fig. 2.12 Cross section of a roadside swale (Mangangka et al. 2016)

Swales and Buffer Strips

Swales (or buffer strips) are shallow, vegetated (generally grassed) channels with gentle side slopes (often 1V:13H or more) and longitudinal slopes (typically <1.5%) conveying runoff downstream (Kachchu et al. 2014). Swale and buffer strip use is particularly prevalent along roadways. A low-profile kerbing system is often used to allow water to discharge freely from the road surface into the swale or filter strip. Figure 2.12 shows a cross section through a schematic roadside swale. Buffer strips for runoff from agricultural fields are not treated in this book.

Swales are simple and cost-effective stormwater treatment devices for controlling runoff volumes and pollutants yielded from impervious surfaces (Deletic and Fletcher 2006). The ability of swales to reduce total runoff volumes and for flow attenuation has been reported in the literature, particularly in low to medium storm events (Deletic and Fletcher 2006; Davis et al. 2012). However, the majority of the research done on swales appears to have focused on their water quality improvement capabilities rather than their flow reduction and attenuation benefits.

Water quality treatment in a swale occurs through the process of sedimentation, filtration, infiltration and biological and chemical interactions with the soil (Winston et al. 2012). Swale performance studies by Deletic and Fletcher (2006) demonstrated average pollutant reduction efficiency of 72% for TSS, 52% for total phosphorus (TP) and 45% for total nitrogen (TN). Simulated runoff tests on nine swales by Bäckström (2002) demonstrated TSS removal rates between 79 and 98%. He also observed more particles were trapped when a swale had dense and fully developed turf.

Bäckström (2003) reported that a 110 m long grass covered swale removed sediments of particle sizes greater than 25 μ m. He also found that small particles (between 9 and 15 μ m in diameter) were exported from the swale. The sediment capturing performance of swales was found to reduce exponentially with the length of the swale, often reaching a constant value (Deletic 2005; Deletic and Fletcher 2006). Deletic (2005) also observed that large particles settled out within the first few metres of the swale, while smaller particles travelled further downstream. These results showed that the runoff sediment concentration is rapidly reduced after entering the swale.

Kachchu et al. (2014) investigated the effectiveness of using grass swales as pretreatment devices for permeable pavements in order to reduce clogging and extend the lifespan of these systems. While swales were effective at removing TSS from stormwater runoff, they found that they were only of limited effectiveness in the removal of nutrients. The results of their simulated runoff experiments demonstrated that between 50 and 75% of the TSS was removed within the first 10 m of the swale length. They concluded that installation of excessively long swales to reduce stormwater TSS pollution may not be the most cost-effective solution. The authors also found that swales can be used successfully to pre-treat stormwater for other stormwater treatment devices to increase the effective life of the systems.

Thus, swales can be used as an alternative to, or an extension of pipe systems, or as a pretreatment system for other treatment devices. They not only provide a stormwater retention function due to the relatively low flow velocities, but they also provide treatment opportunities through sedimentation and can promote (sometimes modest) infiltration (depending on the in situ soil characteristics). Low-intensity rainfall events can often be fully infiltrated in swales (depending on the infiltration capacity of the in situ soil) while more intense rains are generally conveyed through the swale to the downstream stormwater system or receiving waters.

The stormwater runoff from swales may be discharged through the underground stormwater pipe system when outlets are installed at the base of the swale. It is often good practice to place the outlets between 50 and 100 mm above the base of the end of the swale's lower end to encourage low-level ponding which can enhance water retention and sedimentation processes. However, prolonged ponding should be avoided. The in situ soil must therefore be suitable for infiltration.

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Chapter 3 Nutrient Removal from Variable Stormwater Flows

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Abstract When nutrient loads are discharged into surface waters with variable stormwater and wastewater flows, surface water pollution is impaired. Nutrients can lead to oxygen depletion and eutrophication of surface waters, including excessive plant and algae growth. Popular examples of structures harmed by excessive nutrient inflow are the Baltic Sea or the Great Barrier Reef in Australia. Hence, removing nutrients, especially nitrogen and phosphorus compounds, is a major target when variable flows should be treated. This chapter gives an overview of the available removal mechanisms and the potential efficiencies of different treatment facilities. While particle-bound nutrients can be removed via sedimentation processes, dissolved nitrogen and phosphorus compounds cannot as they differ in their biochemical degradation: the adsorption capacity for nitrogen compounds is often renewable, whereas the uptake of phosphorus compounds is limited over time. Hence, treatment facilities need to be able to address the different requirements.

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Treatment Mechanisms for Nutrient Removal

Nutrient exports from anthropogenic activities in urban areas or agricultural fields contribute to water resource degradation considerably. The sum of all nitrogen contributions defines the Net Anthropogenic N Inputs (NANIs, Howarth et al. 1996) expressed in kgN/(km² year) and thus characterises how intense human activity perturbates the natural N cycle. During the period between 1970 and 2000, according to GlobalNEWS model, NANIs transferred directly from watersheds to ocean increased by 16% (Seitzinger et al. 2010; Billen et al. 2013).

Nutrients can lead to oxygen depletion and eutrophication of surface waters, including excessive plant and algae growth (Taylor et al. 2005). In freshwater ecosystems, the nitrogen to phosphorus (N:P) ratios commonly exceed 15–30 and, thus, phosphorus is the limiting nutrient for eutrophication (Berge et al. 1997). In large eutrophic systems with long turnover times, however, nitrogen may be limiting (Murray and Parslow 1999; Taylor et al. 2005).

Phosphorus enters ponds and wetlands in inorganic or organic form, either particulate or dissolved (Reddy et al. 1999; Vymazal 2007). Particulate P is primarily removed from the water column through sedimentation and filtration processes, while the dissolved form is removed by chemical and biological processes, including soil/peat accumulation, media adsorption, precipitation, plant and microbial uptake, leaching and mineralisation (Vymazal 2007). The dissolved inorganic form of P is bioavailable, whereas organic and particulate forms must be converted to inorganic forms to become so.

The mainly particle-bound P can be removed effectively by settling or mechanical filtration, and is hence correlated to total suspended solids (TSS) removal. Dissolved P can be adsorbed by filter media in sand- or soil-based systems (Henderson et al. 2007; Hsieh et al. 2007; Blecken et al. 2010). Plant uptake can also be a significant sink for phosphorus in different kinds of constructed wetlands (CWs) (Vymazal 2007).

P compounds accumulate in filter materials over time, depending on the inflow concentrations (Grotehusmann et al. 2017). Although several authors predicted a re-dissolution of phosphorus after some years of operation, the boundary conditions for such an effect have not yet been clearly determined (Felmeden 2013; Grotehusmann et al. 2017).

Removal of **nitrogen** is a complex process which passes through several stages of reactions (Vymazal 2007). Nitrogen enters wetland systems in both organic and inorganic forms, with the relative proportion of each based on the characteristics of the input source (Collins et al. 2010). Inorganic forms include ammonium nitrogen (NH_4 –N), nitrate nitrogen (NO_3 –N) and nitrite nitrogen (NO_2 –N), and both organic and inorganic forms exist in dissolved fractions or are bound to suspended particles (Vymazal 2007; Collins et al. 2010). The nitrogen cycle in CWs encompasses microbiological activities such as denitrification, and vegetation uptake (Fig. 3.1).



Fig. 3.1 The nitrogen cycle in wetlands (adapted from Tournebize et al. 2017)



Fig. 3.2 Range of driving factors of nutrient removal from CW (adapted from Fisher and Acreman 2004)

Fisher and Acreman (2004) established a list of driving factors controlling nutrient removal in surface-flow CWs, as listed in Fig. 3.2, which can be generalised for the application of ecotechnologies described in this chapter. The effective removal is strongly dependent on different biotic and abiotic factors (Kadlec 2012). Additionally, flow regime is a key factor influencing nitrate N reduction in free surface-flow CWs.

However, many of these processes do not readily remove nitrogen, but only convert it into its various forms. Real removal of nitrogen is provided by denitrification under anoxic conditions, plant uptake (only if biomass is harvested), ammonia adsorption and organic nitrogen burial (Vymazal 2007).

Denitrification rates are positively influenced by nitrate loading (Ayyasamy et al. 2009) and negatively affected by oxygen concentration (Beutel et al. 2009). Thus, in vertical-flow CWs and bioretention systems, a permanently water-saturated submerged zone (also known as Internal Water Storage) has been introduced to enhance nitrogen removal by promoting denitrification in some countries (Dietz and Clausen 2006). Organic carbon, mostly provided by the decay of macrophytes, also influences it (Burchell et al. 2007; Hernandez and Mitsch 2007; Lu et al. 2009). However, how much organic carbon for denitrifying bacteria is available differs greatly depending on the characteristics of the macrophyte species. The effects of different carbon sources on these rates were investigated by adding fructose (Lin et al. 2002), methanol (Huett et al. 2005), dredged organic rich sediments (Burchell et al. 2007), acetic acid (Rustige and Nolde 2007), glucose (Lu et al. 2009), ethanol and woodchips (Domingos et al. 2009), as well as eucalyptus wood mulch (Saeed and Sun 2011) to the sediment.

In another study, carbon was added, either as easily bioavailable carbon in specific macrophytes [watercress (Nasturtium officinale) and reed (Phragmites australis)] (Pulou et al. 2012). Similar investigations were performed for bioretention filters, testing the effect of different decomposing macrophytes as sources of organic carbon (Kim et al. 2003; Zinger et al. 2013). The efficiency of nitrate removal varied between the species. Watercress, for instance, supplied more available organic carbon and was more efficient for nitrate removal than reed (Fig. 3.3).

Heterotrophic denitrification is often the dominant N removal process in CWs, although plant uptake—combined with vegetation harvesting to permanently remove N from the system—can significantly contribute to it (Vymazal et al. 2006).



Fig. 3.3 N removal in laboratory reactors as function of carbon source [sediment, reed (Phragmites australis), watercress (Nasturtium officinale)] from sediment/water column sampled in situ (adapted from Pulou et al. 2012)

Nevertheless, plant uptake is an active process during the vegetated period, while denitrification processes occur throughout the course of a year. Lin et al. (2002) reported that the uptake of various emergent and floating plants contributed only 4-11% to nitrate removal in CWs treating groundwater. A study from Pulou (2011) concluded that macrophyte uptake in CWs removes nitrate at about 10% compared to 90% by denitrification.

During filtration in media-based systems, ammonium is retained until the sorption capacity is reached, which is mostly the case in systems for combined sewer overflow (CSO) treatment. After filtration has ended and the filter is aerated through the drainage pipes, the ammonium is nitrified. Hence, nitrate will be washed out at the beginning of the next event. The degradation of organic substances in the dry phase leads to the additional production of ammonium and a further increase in nitrate as microbial metabolite if the dry phase lasts longer than approximately five days (Dittmer 2006). Organic compounds can already be degenerated under aerobic conditions during the loading regime, but this depends on how much dissolved oxygen is available in the retention layer and the filter material. A significant impact of dry periods between two and four weeks on nitrogen removal in bioretention filters was shown by Hatt et al. (2007): while the mean total nitrogen (TN) outflow concentration during wet periods (inflow twice weekly) was 1.89 mg/L, it significantly increased after dry periods (5.2 mg/L). This difference between wet and dry periods was also observed for outflow concentrations of NO_x (1.3 and 4.0 mg/L, respectively) and total dissolved nitrogen (1.7 and 4.9 mg/L).

For natural contexts, nutrient removal processes are known to occur more rapidly with increasing temperature. An optimal temperature for denitrification is 20 °C. For bioretention filters, Blecken et al. (2010) observed significantly increasing NO_x net production rates with increasing temperature, which were well described by the Arrhenius equation. Additionally, biological activity and, thus, the related removal mechanisms (e.g. plant uptake) are commonly higher in warmer temperatures (Weis and Weis 2004). Cold temperatures during winter can especially decrease nitrogen removal since associated processes are temperature-dependent (Bachand and Horne 1999; Wu et al. 2014). In contrast to nitrogen, phosphorus sorption reactions and physical processes involved in the removal of particulate pollutants are not significantly affected by temperature changes (Kadlec and Reddy 2001).

The main function of the plants is provided by the roots: they help keep the filter material open for hydraulic flow, infiltrate oxygen and provide a surface for biofilm to attach (Brix 1997). However, the selection of vegetation can also be an important factor for a sufficient nutrient removal by uptake processes: significant variations in removal rates between bioretention filters with different plant species have been reported (Read et al. 2008). The **vegetation**, which supports biological processes as well as biochemical processes in biofilms, needs time to establish itself before it reaches full treatment performance (Merriman and Hunt 2014). Phosphorus may be re-released to planted systems due to plant decay if the biomass is not harvested

(Chimney and Pietro 2006). **Plant harvesting** is not a common maintenance measure, but it has been suggested by Lenhart et al. (2012), who observed that 20% of the inflow nitrogen to a sub-tropical CW was harvestable. In contrast, it is common practice in Germany to leave the plants on the filter beds unharvested in a way that the dried leaves and root stems provide a bulked surface which helps to prevent clogging (Born et al. 2000).

Concentrations of Nutrients from Stormwater, CSOs and Agricultural Diffuse Pollution

The main source of nutrients in variable stormwater flow comes from leaching of organic matter and fertilisers. Nutrients can be found in various forms including.

- soluble ions, e.g. NH_4^+ , NO_3^- , NO_2^- , PO_4^{3-} ;
- precipitates, e.g. with metals, carbonates,
- colloids and
- particulate matter (particles larger than 0.45 μm).

Their importance differs depending on the specific nutrient, flow and concentration patterns.

Stormwater from Separate Sewer Systems and Highway Runoff

The nutrient concentration in stormwater runoff is relatively low and usually not of main interest in research studies, unless local necessities require a very low inflow concentration into the surface waters. However, illicit connections in separate sewer systems can lead to comparably high pollutant concentrations in the discharged stormwater (Panasiuk et al. 2015), which is often a problem of relatively old systems, as can be seen in a direct comparison of the values from Lü (2011) and Yin et al. (2017) in Table 3.1.

The phosphorus concentrations in highway runoff are even lower than those in separate sewer systems because there are no cross-contaminations by illicit connections and the organic debris on highways is comparably low.

Combined Sewer Overflows

The major oxygen compound that needs to be removed from CSOs is ammonium. It is chemically in a pH and temperature-dependent equilibrium state with fish-toxic

able 3.1 Measured values for phosphorus and	l nitrogen compoun	ds in stormwater out	lets					3
Catchment (number of sampled events)	Value(s)	TP	NT	NO _x	$\rm NH_{4}-N$	NH_3	References	Nut
		[mg/L]						rien
Urban (8 catchments), Melbourne, Australia	Mean, coefficient of	1	2.13, 0.79 1.63^{**} ,	0.74, 0.76		$0.29, \\ 1.37$	Taylor et al. (2005)	ıt Ren
	variance		0.82**					nova
	EMC	0.03-0.106	0.6–3.2	1	I	I		al fr
Inner city of Shanghai, PR China	EMC	0.4-1.1	1	1	-6.0	I	Lü (2011)	om
					2.6			Va
New development areas in Shanghai, PR	EMC	0.2–0.4	I	I	0.8-	I		aria
China					1.5			ble
Storm drain with illicit connection in	EMC	0.6–8.16	5.9-42.2	I	I	1.4-	Yin et al.	Sto
Shanghai, PR China						33.6*	(2017)	orm
Low-density residential area under	min-max	0.3–2.2	0.6 - 3.2	0.01 - 0.36	I	0.01 -	Walker et al.	wat
development, Queensland, Australia $(n = 15)$	median	0.0	1.1	0.08		0.31	(2017)	er l
						0.05		Flo
Commercial, agricultural and residential runoff	min-max	0.09-0.144	0.567-	I	Ι	I	Hartshorn	ws
(n = 3)			1.480				et al. (2016)	
Highway runoff, Germany (10 different	min-max	0.25-0.49	I	I	0.2 -	I	Kasting	
campaigns)	median	0.31			2.31 0.6		(2002)	
Highway runoff, Auckland, New Zealand	EMC	~ 0.04 to	0.349	0.006*	0.002	I	Borne et al.	
(n = 17)		0.22/~0.09					(2013, 2014)	
		(min-max/median)						
Highway runoff (13 ha, 88% impervious area),	mean \pm std dev	0.19 ± 0.10	1.17 ± 0.34	$0.34 \pm 0.21^{**}$	I	I	Winston et al.	
Florida, USA $(n = 16)$	median	0.18	1.25	0.34			(2013)	
							(continued)	

Table 3.1 Measured values for phosphorus and nitrogen compounds in stormwater outlets

(continued)
3.1
Table

Table 3.1 (continued)							
Catchment (number of sampled events)	Value(s)	TP	IN	NOx	NH ₄ -N	NH ₃	References
		[mg/L]					
Highway, forest and residential runoff $(n = 3)$	min-max	0.777-1.105	0.570-	I	I	Ι	Hartshorn
			0.831				et al. (2016)
University campus, Queensland, Australia	EMC	0.047-0.636	0.432-	I	I	Ι	Nichols and
			2.068				Lucke (2016)
University campus, Florida, USA $(n = 3)$	min-max	0.017-0.020	0.375-	I	1	I	Hartshorn
			0.840				et al. (2016)
Parking lot, maintenance building, picnic area,	mean \pm std dev	0.41 ± 0.52	3.49 ± 4.70	0.17 ± 0.11	I	Ι	Winston et al.
Florida, USA $(n = 18)$	median			0.20			(2013)
NN							

*as NO₂-N **as NO₂₋₃-N

Discharge structure	Value(s)	ТР	PO ₄	NH ₄	References	
		(mg/L)				
Overflow of two storage	min–max	0.3–7.6	0.1–3.9	0.1–29.3	Waldhoff (2008)	
sewers	mean	2.0	0.6	2.8		
	median	1.4	0.4	1.6		
	min–max	0.1–1.5	0.1-0.6	0.2–2.0		
	mean	0.5	0.2	0.8		
	median	0.4	0.1	0.7		
Overflow of a CSO storage	min–max	0.5-3.0	0.2-1.8	1.0-14.2		
tank	mean	1.3	0.7	3.3		
	median	1.3	0.6	2.5		
Overflow of a storage sewer	min–max	0.2–1.9	0.1-0.5	0.1–2.3	Felmeden (2013)	
	mean	0.6	0.2	1.0		
	median	0.2	0.2	1.0		
Overflow of a CSO storage tank	min–max			~0.5-7.7	Dittmer (2006)	
CSOs in inner city, Shanghai	EMC	2–4		25-35*	Lü (2011)	
CSOs in inner city, Shanghai	EMC	2.76-6.88			Zhang and Li (2015)	
CSO outlet, Paris				3.3–9.3	Gasperi et al. (2012)	

Table 3.2 Measured values for phosphorus and nitrogen compounds in CSOs

*as NH₄-N

ammonia. Nitrification of ammonium takes almost four times more oxygen out of the surface waters than the complete oxidation of chemical oxygen demand (COD). Ammonium occurs in higher concentrations in CSOs than in stormwater from separate sewer systems, since it mainly comes from urine and faeces in the wastewater portion of CSO, but also from residues of, for instance, food production or slaughterhouse effluents.

Due to their potential to accelerate eutrophication, phosphate and nitrate are the other nutrient compounds of major interest. Values from different studies worldwide are given in Table 3.2.

Agricultural Diffuse Pollution

Nutrient removal for agricultural diffuse pollution has been investigated in several studies since nutrient leaching from crop areas and livestock farming significantly contributes to surface water pollution (De Paula Filho et al. 2015) and can result in eutrophication with massive algal bloom in surface waters (Brandenburg et al. 2017).



Fig. 3.4 Cumulative average monthly contribution (%) of drained water flow and nitrate fluxes over the period 1987–2005 in North of France. (Average annual flow: 200 mm annual nitrate leaching: 37 kg $ha^{-1} y^{-1}$) (adapted from Tournebize et al. 2017)

Water flow and nutrient concentrations at agricultural watersheds are typically highly variable in time. In the case of subsurface drainage, water flow is seasonal: starting from autumn, there is a peak flow period during winter, sparse flow during spring and no flow during summer in central Europe. Nitrate fluxes at drain pipe outlets generally show the following pattern in this climate zone: a period of high concentrations in autumn, a base concentration during winter and fertiliser application dependent concentration during spring (Fig. 3.4).

The discharge and concentration variation at pilot-scale should also be analysed at different scales. A nested watershed study (Fig. 3.5) showed a scale-nondependent effect: the range of nitrate concentrations at different spatial levels—outlet of agricultural plots, sub-catchment (basin) and the watershed—was quite similar. Thus, given their nitrogen application levels, all of the agricultural plots of a watershed contribute to nitrate leaching.

In addition to seasonal variation due to crop and nitrogen cycle in soil, the detail of a high-frequency flow/concentration chronicle show in Fig. 3.6 that nitrate concentrations could be positively correlated or anti-correlated to drain discharges due to farmers' management. This hydrological nitrate behaviour at watershed scale influences the nitrate mitigation's strategy. Monitoring revealed that:

- water flows are highly variable due to rainfall regime;
- basically, all water flow contains nitrate; and
- nitrate concentrations are highly variable over time.

Boreal water regime may desynchronize and moderate wetland treatment efficiency.



Fig. 3.5 Scale dependency of watershed size to nitrate concentration (adapted from Billy et al. 2013; Tournebize et al. 2017)



Fig. 3.6 Subsurface drainage flow from a 335 ha agricultural watershed and inlet/outlet nitrate concentration of a constructed wetland described in Tournebize et al. (2012) (unpublished data, experimental field of Rampillon, France)

Treatment Systems

Stormwater Sedimentation Ponds and Basins

Since ponds and basins mainly rely on sedimentation, they are generally less effective in removing nitrogen and nitrate than other treatment technologies and, thus, not recommended when nitrogen treatment is targeted (Collins et al. 2010). To a considerable extent, nitrogen is not particle-bound (Taylor et al. 2005). Further,

anoxic conditions to support sufficient denitrification are often missing in ponds (Taylor et al. 2005). Improved performance can be achieved by extended detention capacity in ponds and when second stage denitrification zones are incorporated in the system (Marsalek et al. 2005).

Given that phosphorus is, however, particle-bound to large extent, total phosphorus (TP) removal in ponds can be relatively high: Marsalek et al. (2005) report removal percentages varying between 60 and 75%.

Constructed Wetlands

Surface-Flow Wetlands

Given that CWs, in comparison to, e.g. sedimentation ponds, utilise a wider range of biological and chemical (biogeochemical) treatment processes, varying ambient conditions affect the nutrient removal in CWs (Wu et al. 2014).

Most phosphorus and nitrogen treatment processes described beforehand (see Fig. 3.1) can take place in surface-flow CWs (SFCWs). These complex treatment processes are affected by many factors and, thus, also nutrient removal can vary significantly between different studies as summarised in Fig. 3.7. For instance, in a CW in Sydney, Australia, relatively low removal efficiencies for NO_x–N (22%), TN (16%) and TP (12%) were measured (Birch et al. 2004). In studies done in 1997 and 2012/13 in a stormwater wetland in Växjö, Sweden, N removal of 50 and 45% as well as P removal of 86 and 79% was observed (Al-Rubaei et al. 2016; Semadeni-Davies 2006). The latter is far higher than in most studies included in Fig. 3.7 while the nitrogen removal is in the same range. Another Swedish study



evaluated the performance of a stormwater wetland in Kalmar. Median concentration removals were 50% for nitrogen, 38% for phosphorus and 50% for TSS resulting in load reduction of 43% for nitrogen and 35% for phosphorus (Herrmann 2012). Only evaluating concentration removal may, however, be misleading: Lenhart and Hunt (2011) found increased nutrient concentrations in the effluent of a stormwater wetland in North Carolina, USA, i.e. a negative removal. However, due to an effective volume reduction, the nutrient load removal was efficient (36% for TN and 47% for TP).

Due to upstream hydrological dependency, the hydraulic load of the CWs receiving agricultural flow is highly variable. Two performance time scales should be explored: inter-annual and seasonal assessment.

During dry days (baseflow conditions), denitrification in CWs may increase given the relatively low water exchange and, thus, increase the chance that anaerobic conditions can develop. In contrast, during wet days, the prevailing aerobic conditions decrease the denitrification (Guerra et al. 2013).

Measured nitrate nitrogen removal in CWs receiving fluctuating diffuse water flows from agricultural watershed has been shown to be inherently variable from year to year (Fig. 3.8). Despite similar annual water and nitrate inlet fluxes (in 2007–2008 and 2010–2011), the removal efficiency was different (20 and 55%, respectively). Nevertheless, the long-term monitoring over 8 years led to an average of 50% as inter-annual removal percentage.

Similar seasonal and year-to-year variability in performance have also been measured for a range of other sites treating nitrate-rich agricultural runoff (e.g.



Fig. 3.8 Annual nitrate removal efficiency of CWs (2005–2013) * and ** indicate automatic sampling or direct online measurements of nitrate concentration content in drained water (adapted from Tournebize et al. 2015)

Crumpton et al. 2006; Kovacic et al. 2006; Tanner and Sukias 2011; Diaz et al. 2012). Tanner and Kadlec (2013) found that a simple first-order, hourly time-step dynamic model calibrated to multi-year data from their study wetland was able to closely simulate flow-proportional nitrate–N outlet concentrations. The model predicted that aerial mass removal rates for the wetland would increase gradually as wetland size was increased from 0.5 to 5% of contributing catchment, but with diminishing returns. Application of the model to different simulated flow regimes predicted markedly reduced wetland nitrate–N removal efficiency under highly variable flow compared to that under moderate flow variability, which in turn was poorer than under stable flows. This suggests that seasonal patterns and variability of flow need to be seriously considered when predicting wetland treatment performance.

At seasonal time scale, Fig. 3.6 allows comparison of inlet and outlet concentrations during peak flow periods. The hydrological regime impacts the removal efficiency. When discharge decreased, the gap between inlet and outlet concentrations increased, and inversely. In several cases, hydrological response (water flows) are more important during winter, and consequently removal processes appear to be depressed compared to the summer season.

Removal processes also require time and contact between water flow and substrate. Three factors manage hydraulic residential time: the volume of the CW congruent with hydrological regime, a distributed water pathway which can be designed having baffles of optimised length/width ratio, and the surface rugosity of the CW, controlled by vegetation cover density. As based on residential time values, some experiments in the laboratory (controlled temperature, static hydraulic) show that to reach a removal rate of 80% of inlet nitrate fluxes, the storage time should last at least four days. Under natural conditions, including hydrological regime, temperature variation, the design of the CW should lead to a longer storage time.

Merriman and Hunt (2014) compared the performance of a SFCW in North Carolina directly after construction and after 4 years of operation. The nitrogen outflow concentrations were lower after 4 years (1.1 versus 0.85 mg/L). Similarly, treatment performance improved over time due to the maturation of the treatment processes, as observed by Al-Rubaei et al. (2016) when evaluating the treatment performance development of the 19-year-old Bäckaslöv wetland in Växjön (Sweden). In contrast, Kadlec and Knight (1996) noted that nutrient treatment in CWs may be enhanced during the first years of operation due to a higher availability of sorption sites, the increasing expansion of the vegetation and the biological activity while in later stages a steady state is achieved.

Floating Treatment Wetlands

Floating treatment wetlands (FTWs) are often used to retrofit existing ponds. A study comparing stormwater pond effluent quality before and after a FTW was

installed found that nutrient removal can be improved. Borne et al. (2013) found that N removal improved moderately in stormwater ponds with FTWs compared to those without, while the TP removal was significantly enhanced (P outflow concentrations were 27% lower in the pond equipped with FTWs). P removal was mainly enhanced due to sorption of dissolved P, since the roots trapped particulate P and lead to settling of suspended solids (Borne 2014). Results from a study in Florida, USA, show that N removal was significantly enhanced by plant uptake (Chang et al. 2013). Headley and Tanner (2006) describe a system for CSO treatment in Belgium with removal rates of 24–61% for TP. However, this system showed signs of anaerobic zones due to missing external aeration.

The results of the aforementioned studies are listed in Table 3.3. As can be seen in Table 3.1 which gives the inflow values to the studies listed in Table 3.3, the variation among the studies is quite high; however, there were generally low inflow concentrations for TN (median below 1.3 mg/L) and TP (median below 0.2 mg/L).

Borne et al. (2013) identified seasonal variations in the mineralisation of nitrogen compounds in a 1-year pilot study in New Zealand. During the summer months, the authors observed larger mineralisation and/or settling of organic nitrogen particles compared to the control pond than they observed in winter. They concluded that these processes were increased due to a higher temperature and biomass availability. The biomass resulted from plant parts dying off in summer and did not only provide a carbon source, but might also enhance chemical flocculation and particle sedimentation.

However, not all removal efficiencies presented in Table 3.3 are statistically significant when inlet and outlet concentrations are compared or the removal efficiencies of retrofitted stormwater ponds with non-retrofitted ones.

Several studies investigated the plant nutrient uptake and the accumulation in the sludge deposit under FTWs (Van de Moortel et al. 2011; Borne et al. 2013; McAndrew et al. 2016). Harvesting the vegetation shoots was recommended by Wang et al. (2014) to ensure long-lasting removal of nutrients (and other pollutants) and prevent re-release due to organic matter breakdown. Since FTWs can also be used to retrofit stormwater tanks (Ruppelt et al. 2017), sludge removal is also an option to permanently remove nutrients from the system and to avoid re-suspension.

However, given that FTWs are a rather new technology, most of these studies have been conducted on fairly new installations, and results from long-term operation of these systems have not yet been collected.

Subsurface-Flow Constructed Wetlands

For subsurface-flow constructed wetlands and bioretention filters, the major influences on the sorption capacity for nutrients are the filter material, biofilm development and aeration. The type of sorption is decisive for whether a retained substance is degraded or accumulated in the filter body. Simplified, there is a

Location	Catchment	Pond size/FTW	Removal effic	References	
(number of sampled events)	specifics	coverage	TN	TP	
Auckland,	Highway runoff	100 m ² /50%	MRE ^a		Borne
New Zealand $(n - 17)$	(3.7 ha)		median	min–max (median)	et al. (2013,
(II = 17)			29.1	~ -25 to ~ 70 (25)	2014)
North Carolina, USA			mean		Winston et al. (2013)
(n = 16)	Highway runoff (13.07 ha, 87.7% impervious area)	3600 m ² (incl. forebay of 11.7% of the surface area)/9%	48	39	
(n = 18)	Parking lot, a maintenance building, picnic area (2.37 ha, 54.3% impervious area)	500 m ² (incl. forebay of 18% of the surface area)/18%	88	88	
Florida,	Multi-unit 340 m ² /8.7%		CRP ^b		Chang
USA (n unclear)	residential area (0.066 ha)		29	34	et al. (2015)
Florida, USA $(n = 3)$	orida, SA (n = 3)		Average reduction (lower/upper bounds of confidence interval)		Hartshorn et al. (2016)
	Highway, woods, residential	2363 m ² /5%	36.5 (12.5/60.5)	71.2 (67.0/75.4)	
	Highway, commercial, tomato field	1263 m ² /5%	33.1 (-25.2/91.4)	-35.9 (-99.8/25.0)	
	Commercial, woods, grassy area	2792 m ² /6.4%	4.2 (0.4/8.0)	-28.6 (-59.9/2.7)	
Queensland, Australia	Low-density residential under development	5048 m^2 pond size, separated treatment are of $101 \text{ m}^2/100\%$ of the treated surface area	CRP mean ± 7 ± 48 (18)	std (median) 33 ± 33 (33)	Walker et al. (2017)

Table 3.3 Comparison of removal efficiencies in seven different stormwater ponds retrofitted with floating treatment wetlands

^aMRE—mass removal efficiency ^bCRP—concentration reduction percentage

regenerative and non-regenerative sorption capacity in the filter material. In case of ammonium and COD (Dittmer 2006; Woźniak et al. 2007), the sorption capacity between two loading events regenerates. In contrast, the capacity to adsorb metals and phosphates dissipates (Felmeden 2013; Grotehusmann et al. 2017). Degradation of adsorbed substances results in a negative balance of inflow and outflow for the degradation products, e.g. nitrate, which only occurs in low concentrations in the feed, but is formed by the nitrification of adsorbed ammonium (Dittmer 2006). In contrast, under certain circumstances, retained phosphate can be desorbed after reaching the sorption capacity (Felmeden 2013).

Davis et al. (2006) reportedly observed 70–80% total phosphorus (TP) removal in biofilter box experiments, but Li and Davis (2009) observed strong leaching (0.1 and 0.35 mg/L TP in influent and effluent, respectively). Similarly, the efficacy of TN treatment is highly variable, ranging from effective removal to significant leaching (Kim et al. 2003; Blecken et al. 2010).

Commonly, net P leaching from CWs has been observed due to wash-out of fine materials with associated P (Hatt et al. 2009; Hunt et al. 2006; Li and Davis 2009; Read et al. 2008). P release can be observed during extreme rain events, leading to peak flow, associated with sediment re-suspension in the CW. That is the reason why, over the last years, specific designs have been favoured which enable buffer capacity in the CW associated with a constant outflow weir to promote laminar flow.

In newly constructed filters, P release is often observed, but decreases over time due to media stabilisation (e.g. repacking, settling) and/or depletion of the reserves (Hsieh et al. 2007). Thus, to achieve low P concentrations in the effluent, it is essential to select appropriate filter media (Hunt et al. 2006), and filter media with high P concentrations should be avoided (FAWB 2008).

In addition to stormwater runoff, eroded sediments are important non-point sources of P (Brady and Weil 2002); thus, biofilters might indirectly reduce P discharge to recipients since they diminish surface runoff flows by reducing erosion losses in urban catchments.

The **vertical-flow system** is most commonly used to treat variable flows. However, for the treatment of stormwater and wastewater flows—the latter limited to the treatment of CSOs in the sense of this book—this system is covered in the section Bioretention filter.

Horizontal subsurface-flow systems are predominantly anaerobic systems with limited oxygen availability, so processes such as ammonium N removal are limited by low nitrification potential, reducing the potential for removal of N via subsequent denitrification. This makes them often part of treatment train solutions.

As single application in the treatment of variable stormwater and wastewater flows, the system is mainly used in Great Britain for the treatment of CSOs, as described in Chap. 2. The number of publications on nutrient removal is, thus, limited: Griffin (2003) investigated the treatment efficiency of two different setups, one treating only the CSOs in a horizontal-flow wetland (sizing: 0.5 PE/m²) and one treating both overflows and tertiary treated wastewater (sizing: 1.0 PE/m²). Since the combined sewage was diluted with tertiary treated wastewater in the second

wetland, the inflow concentrations were lower for all measured parameters [biological oxygen demand (BOD), TSS, NH_4-N , total oxidised nitrogen (TON)]. The CW treating only CSOs (referred to as "storm only reed bed" in the publication) removed 41–57% of ammonium during three monitored events, and 70–77% of the TON (sum of nitrate and nitrite). Although the effluent concentrations for the wetland that treated both tertiary treated wastewater permanently and stormwater during overflow events was lower at two monitored events than for the storm-only reed bed, the removal efficiency was only approximately 20% of these events due to the low inflow concentrations. However, there was no explanation given why the total oxidised nitrogen was not removed at all during these events. Griffin (2003) also reported how these types of wetlands performed over a decade; however, effluent values or removal efficiencies for nitrogen compounds were not provided for this period.

In the hybrid systems described by Ávila et al. (2013) (see Chap. 2), the horizontal-flow wetland removes TN to $56 \pm 19\%$ during dry weather conditions, but less (~35 ± 27%) during wet weather. The concentrations of NO_x–N followed the same trend. The removal of TP was low in the overall system (22%).

Bioretention Filters

Principally, bioretention filters use vegetation and a filter material through which the water percolates to treat stormwater and wastewater. Between events, the filter bed is aerated through the drainage pipes. Thus, adsorbed ammonium is nitrified and nitrate may be released into the surface waters with the first flush of the next heavy rainfall event. Due to this, the filter's capacity to absorb ammonium is regenerated (Dittmer 2006).

Figure 3.9 shows the results of a single event in a large-scale bioretention filter for CSO treatment in Germany (Tondera et al. 2013). The beginning of the inflow (first dashed vertical line) is followed by a delayed rise of the ammonium inflow as well as of the nitrate outflow. The filter with a surface area of 2200 m^2 was monitored for a period of 1 year and retained ammonium at 88% based on the mean values of the event samples. Each event showed a nitrate peak in the beginning due to nitrification processes between filter events with negative nitrate removal rates (Tondera et al. 2013). These dynamics and resulting removal rates are described in detail in Chap. 7.

 NO_x removal is generally low since denitrification is not favoured in bioretention filter conditions in separate sewer system (low carbon content, low anoxic conditions, etc.). Low denitrification has been identified as the main reason for the N net leaching commonly observed in bioretention filters (Davis et al. 2006; Hatt et al. 2009; Li and Davis 2009; Kim et al. 2003; Blecken et al. 2010).

In order to enhance denitrification in bioretention systems, a submerged zone (also known as Internal Water Storage) has been introduced in the stormwater bioretention filter systems (Kim et al. 2003; Dietz and Clausen 2006; Davis et al. 2009).



Fig. 3.9 Recorded ammonium and nitrate data for inflow and outflow of a bioretention filter for CSO treatment (five-minute intervals) (Tondera et al. 2013)

This zone can be created by elevating the outlet, thus maintaining a permanent water level at the base of the filter system. The recommended depth of a saturated zone is 45–60 cm (Zinger et al. 2013). Denitrification can be further improved with the addition of a carbon source such as newspaper shredding, straw, woodchips and saw dust (Peterson et al. 2015; Kim et al. 2003).

One of the most comprehensive research studies on the removal of phosphorus from CSOs was performed by Felmeden (2013). The author investigated, among others, the retention of TP on two large-scale bioretention filters. In the first 2 years of their operation, a filter with a surface of 430 m² and one with a surface of 1275 m² had a mean removal efficiency of approximately 76% (median 81%) and 69% (median 77%), respectively.

Grotehusmann et al. (2017) showed that phosphorus removal generally decreased from the beginning of operation to 6 years later, based on the data of a large-scale bioretention filter for CSO treatment. Tondera (2017) combined data of several bioretention filters: for plants with at least three data sets per group for the operation periods of 0 to 2 years, 3 to 5 years and 6 to 7 years (inflow and outflow values), the mean inflow and outflow values for each period and the removal efficiency were calculated via Monte Carlo simulation. A weak negative correlation was found between the theoretically accumulated phosphorus load in the filter in relation to the removal efficiency. Hence, the total inflow loads rather than filter age play the most important role for the filter service life.

Swales and Buffer Strips

Nutrient removal in swales and buffer strips can be attributed mainly to the retention of particle-bound substances, partially also to plant uptake of dissolved substances.

However, the results on nutrient uptake presented in literature vary extremely. According to a study done by Yu et al. (2001), removal varies from 14–99% for TSS, N and TP. Deletic and Fletcher (2006) report removal efficiencies in swale depend on the flow rate between approx. 10 and 70% for TP, and between 40 and 70% for TN. In contrast, Lucke et al. (2014) show some P removal due to P being mostly particle-bound while N, to a large extent commonly dissolved is only retained to lesser degree in swales. Winston et al. (2012) show that vegetated filter strips at comparable catchment areas can both remove TN and TP or even release it. Additionally, they showed a higher TN removal in wetland swales than in dry swales, which is led back to the more complex processes in wetlands, including denitrification. Other reasons for the extremely varying performances can be the percentage of the vegetation cover of swales (less coverage led to less removal according to Winston et al. 2012), the use of fertilisers close to the treatment sites which impacts the removal negatively (Lucke et al. 2014), and the length of swales (Deletic and Fletcher 2006).

Common swales for stormwater management cannot be regarded as a complete nutrient treatment system and have to be complemented with other facilities if quality treatment is targeted.

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Chapter 4 Microbial Loads and Removal Efficiency Under Varying Flows

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Abstract A variety of ecotechnologies have shown promising yet variable results in reducing faecal microbial contaminants under challenging operational conditions. But relatively limited work has been conducted to investigate and understand faecal microbe removal in these systems under highly fluctuating hydraulic and contaminant loading. In most instances, ecotechnology-based systems such as sedimentation ponds, constructed wetlands and bioretention filters have proved effective for treating episodic discharges and demonstrated performance resilience removing faecal microbial contaminants with modest to good efficiency particularly where inflow concentrations are high. However, microbial removal may depend greatly on the type of microorganism, treatment system design and operational factors. Design characteristics such as type of filter material and depth, presence of a submerged zone, type of vegetation and operational conditions such as inflow concentration, and antecedent dry periods in combination with temperature changes can all affect the removal of faecal microbes. Factors influencing survival, fate and behaviour of retained faecal microbes are still poorly understood. These knowledge gaps need addressing in order to fully evaluate microbial removal from fluctuating

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contaminated flows and more accurately interpret faecal indicator bacteria-based water quality and potential health risks associated with discharge from these ecotechnology-based systems.

Introduction

This chapter considers microorganisms occurring in different kinds of rain-event triggered wastewater and runoff and their removal in ecotechnological systems treating these variable flows with possibly highly varying concentrations. Since analysing water for potentially pathogenic microorganisms is both complicated and expensive, microbial water quality is typically assessed based on the enumeration of faecal indicator organisms (FIO) which are cheaper to assay and more abundant in faecal material. Moreover, FIO are used to indicate the presence of faecal contamination and to assess the potential risk of water contamination with faecal pathogens. Typical indicator organisms are the faecal indicator bacteria (FIB) E. coli, thermotolerant coliforms (faecal coliforms) and enterococci as well as bacteriophages such as F-RNA or somatic coliphages. Bacteriophages are considered better indicators for viruses than FIB as they behave more conservatively and more adequately resemble the morphological characteristics of human enteric viruses (Havelaar et al. 1993). Both FIB and bacteriophages can occur in high concentrations in wastewaters and environmental waters as a result of sewage overflows and non-point sources of human and animal wastes. In view of this, this chapter focuses on faecal microbial indicators to evaluate microbial loads and to discuss treatment mechanisms and removal efficiencies of ecotechnologies under varying flows.

Treatment Mechanisms

The simplest kind of treatment is sedimentation of microorganisms, either of the microorganisms themselves due to their size (e.g. parasites), or because they are bound to settable particles. In stormwater, Characklis et al. (2005) concluded that 30-55% of faecal indicator bacteria (*E. coli*, intestinal enterococci and coliforms) were associated with settleable solids. Attachment behaviour can vary between the types of microorganism. Jeng et al. (2005) found greater attachment of *E. coli* to stormwater particles than of enterococci which ranged from 22 to 30% and 8 to 12%, respectively, whilst Krometis et al. (2007) found a higher proportion of FIB (on average 40%) associating with settleable particles compared to bacteriophages (13%). *E. coli* in artificial stormwater was found to be predominantly (>90%) associated with particles smaller than 3 µm or unassociated with particles, i.e. the bacteria were freely suspended (Chandrasena et al. 2012).

Since findings of Characklis et al. (2005) and Chandrasena et al. (2012) are from stormwater from separate sewer systems, the proportion of microbes attached to solids may be greater in high strength organic wastewaters which contain greater



Fig. 4.1 Size of pathogens found in wastewater

solid loads. In a wastewater study, the majority of *E. coli* (91%) and enterococci (83%) were found attached to particles $\leq 12 \ \mu$ m in wastewater and 8 and 16% respectively attached to particle fractions ranging between 12 and 63 μ m which contributed to around 50% of the overall total suspended solids load in the wastewater (Walters et al. 2014). Thus, the removal of fine particles is an important factor to consider for reducing microbial contaminants during treatment of stormwater or higher strength wastewater, particularly as microbial survival can be greater when associated with particles (Davies and Bavor 2000).

Microorganisms attached to settleable solids and those microbes larger than 5 μ m, such as some bacteria and parasites in free phase, can be filtered out and retained on the filter surface, if the principles of surface filtration apply as described in Chap. 2 (Fig. 4.1).¹

Gargiulo et al. (2007) investigated the effect of straining, which occurs when particles in suspension are retained at pore openings that are too small for it to pass through (as described in Chap. 2), in unsaturated sand columns with a grain size in a range of middle sand (330–607 μ m). In this study, straining was considered responsible for the retention of up to 80% of the *Rhodococcus rhodochrous* bacteria used in the experiment. The deposition rose exponentially as the filter depth increased, and the majority of the retained bacteria was found in the first five centimetres of sand depth.

Similarly, Waldhoff (2008) reported a mean retention of 40% of bacteria in the first 10 cm of the sand filter layer of a bioretention filter. Orb (2012) conducted lab scale investigations on filter columns in order to evaluate the retention of *E. coli* as a function of the filter depth and found that the biggest proportion is retained at a depth between 9 and 43 cm. This study also analysed the effect of unsaturated and saturated zones on the removal efficiency, finding that saturated conditions led to poorer retention of *E. coli* than unsaturated conditions.

Suspended particles are adsorbed or removed by 'physico-chemical filtration' (Chap. 2, Fig. 2.8), when their diameter is much smaller than the diameter of the filter material. Thus, a predominant retention mechanism for viruses in porous media is adsorption (Corapciogliu and Haridas 1984) with adsorption increasing with the growth of the biofilm mass in the filter media (Waldhoff 2008). Similarly, adhesion and entrapment of microorganisms within biofilms is also influenced by biofilm biomass and structure (Stott and Tanner 2005).

Ecotechnologies can physically remove faecal microbes from contaminated flows by sedimentation (Karim et al. 2004), filtration and adsorption to organic

¹See also Chap. 2, Fig. 2.8.

material and subsurface substrate (Gerba et al. 1999). However, microorganisms removed by these physical processes have the potential to be eluted or re-entrained later unless other attenuating mechanisms inactivate or permanently remove them.

UV radiation can significantly inactivate microorganisms where open water areas are present for sufficient light penetration such as waste stabilisation ponds (Davies-Colley et al. 1999) and in areas within surface-flow wetlands (Nguyen et al. 2015). Other attenuating mechanisms include predation (Decamp et al. 1999, Stott et al. 2003), biofilm entrapment (Stott and Tanner 2005) and inhibition (Stevik et al. 2004).

Concentrations of Pathogens from Stormwater, Combined Sewer Overflows and Agricultural Diffuse Pollution

Stormwater from Separate Sewer Systems

In general, stormwater from separate sewer systems can be heavily contaminated with faecal microorganisms at concentrations similar to that of partially treated wastewater. Comparing indicator bacteria levels in stormwater with microbiological water quality regulations show that stormwater runoff could be a potential threat if the receiving water is intended for raw drinking water extraction and/or recreational purposes (Galfi et al. 2016). Based on an analysis of the US stormwater database, Pan and Jones (2012) report that the event mean concentrations (EMCs) of faecal indicator bacteria in stormwater were above water quality criteria. They conclude that these bacteria are of major concern, and efficient treatment facility design is thus required.

Stormwater runoff can be contaminated with faecal microorganisms with highly varying concentrations from numerous sources of contamination. These include wash-off from roof or sealed areas containing animal faeces (wildlife or domestic) and organic waste (Schares et al. 2005), but also wastewater originating from illicit sewer connections (Mertens et al. 2017).

Concentrations of faecal coliforms and enterococci from 10^2 to 10^5 CFU²/100 mL have been detected in stormwater within separate sewer systems (Qureshi and Dutca 1979). Similar concentrations of *E. coli* and enterococci have been reported in the outflow from a rain basin of a separate sewer system with median concentrations of 2.4×10^4 MPN³/100 mL and 3.4×10^4 CFU/100 mL, respectively, although the rainwater has already been pretreated by sedimentation (Mertens et al. 2014;⁴ Schreiber et al. 2016).

 $^{^{2}}$ CFU = Colony Forming Unit.

 $^{^{3}}MPN = most probable number.$

 $^{^{4}}$ Runoff from sealed surface area of 62.8 ha, volume of storage tank 3650 m³, max. outflow 1100 l/s and median outflow at discharge point 158 l/s.



Fig. 4.2 Range of Escherichia coli concentrations in stormwater from separate sewer systems

Bacteria concentrations in stormwater vary significantly depending on various factors. Significant variations between the sampling sites/catchment characteristics and between events have been reported, as summarised for the indicator bacteria *E. coli* in Fig. 4.2. In addition, intra-event variability has been reported with higher loading rates of settleable microbes occurring during the rising limb (Krometis et al. 2007). Many variables (e.g. flow rate, suspended solids or sediments, water/air temperature, pH, catchment land use and faecal sources in catchments, etc.) can all influence the contamination of stormwater with faecal indicator bacteria to varying degrees.

Bacteria concentrations in high-density residential and central urban catchments tend to be higher than those in low-density residential and/or 'green' urban catchments suggesting a relationship between bacterial loading and existing land use (Selvakumar and Borst 2006; Galfi et al. 2016). However, McCarthy et al. (2012) reported higher bacteria concentrations in runoff from a low-density compared to a medium-density catchment. Thus, factors other than catchment characteristics may affect bacteria levels in stormwater such as antecedent dry periods and rainfall intensity affecting microbial persistence in and transport from a variety of faecal sources prevalent in the catchment.

Significant seasonal variation in microorganism concentrations is commonly observed. A season with commonly lower concentrations is winter (Selvakumar and Borst 2006, Pan and Jones 2012; Hathaway et al. 2014). A similar effect has been observed in cold climates (Östersund, Northern Sweden) where indicator bacteria concentrations were typically lower (Galfi et al. 2016) than those from warmer climates (Selvakumar and Borst. 2006), which might be attributable to the considerably lower air and water temperatures in the colder climate. Pan and Jones (2012)

reason that seasonal differences in FIB concentrations in stormwaters depend on the indicator bacteria and/or the extent of seasonal differences in the actual region.

Whilst a number of studies have investigated the variability in microbial concentrations between different storm events, there is limited data on the faecal microbial dynamics during a runoff event. Galfi et al. (2016) evaluated the temporal variability in microbial indicator concentrations within events in four urban catchments in Östersund, Sweden. In more than half of the cases, a first flush was observed for total coliforms and also (to a lesser degree) for *E. coli*. Stormwater runoff in North Carolina, USA, also showed a significant first flush effect for thermotolerant (faecal) coliforms whereas *E. coli* and enterococci did not (Hathaway and Hunt 2011). In contrast, no consistent first flush phenomenon was observed but rather the opposite (an 'end flush') was detected for *E. coli* in a study from Melbourne, Australia (McCarthy 2009).

Typically, median concentrations of faecal indicator bacteria such as *E. coli* in stormwater are two to three \log_{10} lower than those found in combined sewer overflows (CSOs) (see Fig. 4.3) (McCarthy 2009; Dickenson and Sansalone 2012; McCarthy et al. 2012; Mertens et al. 2017) with median concentrations of *E. coli* in stormwater around $10^3-10^4/100$ mL⁵ compared to 10^6 *E. coli*/100 mL³ in CSO (e.g. Kistemann et al. 2004; Tondera et al. 2013; Christoffels et al. 2014). Median concentrations for enterococci in stormwater are also approximately one \log_{10} lower than in CSOs which generally contain enterococci levels of around $10^5/100$ mL (Brownell et al. 2007; Dickensen and Sansalone 2012). Similarly, somatic coliphages, often used as faecal indicators of human pathogenic viruses, occur in stormwater from separate sewer systems in concentrations up to 7.4×10^4 PFU⁶/100 mL with median concentrations of 1.1×10^3 PFU/100 mL (Mertens et al. 2017). This is about up to $2 \log_{10}$ lower compared to concentrations in CSO (Christoffels et al. 2014).

Combined Sewer Overflows

Interest in the microbial contamination of CSOs has been increasing since the 1970s when the first studies were reported on faecal and total coliforms (Diaper and Glover 1971). Load calculations within a catchment area showed that the annual impact of event-based discharge of CSOs on surface water contamination can be comparable to or even higher than that of continuously discharged effluent from WWTP with secondary treatment. On the other hand, CSOs may contribute even higher annual loads depending on the management of other contributing sources of contamination within the catchment (Rechenburg et al. 2006; Kistemann et al. 2008; Schreiber et al. 2016). Concentrations of faecal indicator bacteria in CSOs are

⁵MPN or CFU depending on reference.

 $^{^{6}}$ PFU = plaque forming unit.



Fig. 4.3 Concentrations of Escherichia coli in CSOs at the overflow of CSO settling tanks

illustrated in Figs. 4.3 and 4.4, and show highly variable concentrations of *E. coli* and enterococci. Generally, median values of *E. coli* and *Enterococcus* at the outlet of overflow or retention tanks range from 10^4 to 10^6 MPN or CFU/100 mL.

Agricultural Diffuse Pollution

Discharges of faecal microbial contaminants from agricultural land can vary depending on land use and management. Surface runoff and subsurface drainage can contain concentrations of faecal microbes from animal waste of public health concern including faecal virus indicators such as somatic or F-RNA coliphages and zoonotic pathogens such as *Campylobacter, Cryptosporidium* and *Salmonella* from farm animals or wildlife (Kistemann et al. 2007; Franke et al. 2009; Dufour et al. 2012; Schreiber et al. 2015, 2016).

In addition, concentrations of faecal indicator bacteria can be at levels comparable to those in waste- and stormwaters. Up to $10^7 E. coli$ MPN/100 mL have been reported in overland and subsurface-flow from grazed pastoral land (Collins et al. 2005). Thus, if not intercepted and treated, these flow pathways can transport substantial loads of faecal microbes to waterbodies. Not surprisingly, agricultural diffuse pollution can be a significant cause of impaired water quality (Wilcock et al. 2013) and a potential human health risk (Dufour et al. 2012). Major disease outbreaks have occurred as a result of contamination of surface and ground waters with livestock wastes. The largest outbreak of Campylobacteriosis to date was associated


Fig. 4.4 Concentrations of Enterococcus at the overflow of CSO settling tanks

with drinking water supplies contaminated with *Campylobacter* from sheep as the likely source (DIA 2016; ESR 2017).

Fenlon et al. (2000) reported that around 10% of *E. coli* and *E. coli* 0157 in livestock slurry applied to land was transported in overland and subsurface-flow associated with rainfall events.

High microbial loads up to 2×10^6 *E. coli* CFU/100 mL are found in runoff from farmland areas following heavy rainfall events. Contamination of the receiving river occurred especially after dry weather periods after manure was applied on fields. High concentrations of *E. coli* and enterococci up to 10^5 CFU/ 100 mL have also been reported in surface runoff from grassland being extensively used as pasture (Schreiber et al. 2015). In contrast, low concentrations of up to 10 somatic coliphages PFU/100 mL were found in runoff from the same catchment (Franke et al. 2009). Typically, subsurface drainage flow is less contaminated than surface runoff. The reduction determined as difference between the median concentrations of surface runoff and subsurface runoff varied up to 3 log₁₀ for *E. coli* and enterococci depending on several parameters such as slope, soil type, amount of rainfall and land cover (Schreiber et al. 2015).

Elevated discharge of drainage waters from grazed pastures associated with storm events mobilised *E. coli* almost 20-fold higher than at low flow conditions (Collins et al. 2005).

Microbial concentrations can remain high in overland flow long after manure is used. Concentrations greater than 10^4 *E. coli* MPN/100 mL were sustained in overland flow over 40 days in a field trial on saturated soil (McDowell et al. 2006).

Similar concentrations of 2.5×10^4 *E. coli* CFU/100 mL were sustained for more than 2 months from sheep-grazed pastures (Vinten et al. 2004).

Microbial discharges from agricultural land can exhibit great spatial and temporal variability in concentrations and loads. Mitigation strategies for environmental protection particularly of surface waters from agricultural diffuse pollution are required to cope with fluctuating flows that are often highly contaminated. Ecotechnologies such as constructed wetlands (CWs) can provide an effective management approach to intercept and treat such variable overland and subsurface-flows.

Treatment Systems

Urban and diffuse runoff, CSO, and wastewater are all influenced by rain events. Interception and mitigation of this episodic point and diffuse contaminating flows require systems capable of treating these highly variable loads of faecal microbes. The public health risks associated with pathogens found in these storm- and wastewaters have been well established (Fleisher et al. 1998; Haile et al. 1999; Donovan et al. 2008; McBride et al. 2013). It is, therefore, necessary to ensure that these contaminating flows can be adequately treated and managed before being discharged into the receiving environment. The performance resilience of the treatment system to highly fluctuating flows is particularly important because health risk implications are associated with the discharge of treated outflows.

Stormwater Ponds and Basins

Stormwater ponds have the potential to remove microorganisms considerably since significant percentages of bacteria are associated with particles as discussed previously (Characklis et al. 2005). A performance summary of stormwater retention ponds in the International Stormwater BMP⁷ database shows the greater removal of *E. coli* (median removal 96%) compared to other systems (median removal 65 to 80%) (Clary et al. 2017). However, comparing the bacteria removal capacity of ponds and CWs, Davies and Bavor (2000) showed that ponds may have a relatively low ability to remove bacteria since fine particles < 2 μ m to which bacteria are predominantly adsorbed are not often retained well in ponds. Additionally, Hathaway et al. (2009) found highly varying indicator bacteria removal in ponds and, thus, do not recommend ponds as a first choice when bacteria removal is targeted. Similarly, Pan and Jones (2012) reported relatively efficient enterococcus removal by a detention basin in Houston, TX, USA whilst a similar facility in

 $^{^{7}}BMP = Best Management Practise.$

Dover, DE, USA showed increased enterococci concentrations illustrating the high variability in bacteria removal. Similarly, other detention basins have also shown net export of indicator bacteria (Clary et al. 2010). However, some detention basins/ ponds may provide significant volume reduction for storm events and so may be effective in reducing bacterial loading to receiving waters despite minimal reductions in concentrations.

Constructed Wetlands

Surface-Flow Constructed Wetlands

Studies of CWs treating regular and continuous inflow of contaminated waters have generally reported effective removal of microbial contaminants such as faecal indicator bacteria and bacterial and parasite pathogens (Rivera et al. 1995; Stott et al. 1999; Stott and Tanner 2005), particularly when influent concentrations are high (Kadlec and Wallace 2009). There is less information on the performance of these passive systems to treat highly variable flows.

Surface-flow constructed wetlands (SFCWs) treating variable stormwater flows have demonstrated significant removal of faecal indicator bacteria with removal rates exceeding 75% for influent concentrations of almost 10^8 CFU/100 mL (Davies and Bavor 2000). However, the performance of SFCWs can be highly variable: Birch et al. (2004) reported that faecal coliforms were removed at rates varying between 26 and 98% during eight storm events. Removal rates may also vary depending on the type of FIB with greater removal of faecal coliforms reported for stormwater wetland basins compared to *E. coli* and enterococci (median removal 93, 64, and 61%, respectively) (Clary et al. 2017).

As discussed previously, a key factor for effective microbial removal from stormwater by ecotechnological treatment systems is the ability to trap fine particles $<2 \mu$ m to which microbes such as bacteria are predominantly absorbed. SFCWs can be more efficient in removing such fine particles from stormwater compared to ponds because emergent vegetation can impede water flow thereby reducing water velocity to enhance fine particle settling but also provide sites for biofilm growth and microbial attachment so their bacteria removal performance is commonly higher (Davies and Bavor 2000).

SFCWs are one tool being used to reduce diffuse microbial contaminant losses from intensifying pastoral agriculture particularly dairy farming by intercepting and treating drainage flows (Tanner and Sukias 2011). Wetlands are also known to be important sites in the landscape for attenuating pollutants from land runoff. CWs have demonstrated they can play an effective role in attenuating dairy farm faecal pollution spill events by intercepting effluent contaminated overland and subsurface-flow (Sukias et al. 2007). A two-stage SFCW demonstrated considerable buffering capacity and was effective in attenuating a significant pulse of dairy parlour wastewater as overland and subsurface-flow. Passage through the wetland



Fig. 4.5 Episodic rainfall-driven flow and *E. coli* concentrations for inflow and outflow from a two-cell surface-flow wetland treating agricultural drainage waters (time-based sampling) (data from Sukias et al. 2011)

reduced faecal indicator bacteria concentration by 5 to 6 \log_{10} , similar to rates observed in waste stabilisation ponds. The median concentration of around 10^8 *E. coli* MPN/100 mL in overland flow and 4×10^6 *E. coli* MPN/100 mL in subsurface drainage was reduced to a median of 528 *E. coli* MPN/100 mL by the wetland system with a hydraulic retention time (HRT) of around 2 weeks.

For wetland systems receiving more frequent episodic (rain-event driven) subsurface drainage from grazed dairy pastures, higher concentrations and yields of faecal indicator bacteria are found in inflows on the rising limb of hydrographs. A study following the performance of a drainage treatment horizontal-flow CW (HFCW) during seven storm events found peak E. coli concentrations in drainage inflows to the wetland, ranging from 10^2 to 10^4 MPN/100 mL. E. coli yields on rising limbs for inflows were up to four times higher than falling limbs, ranging from 4×10^6 to 7.6×10^9 MPN *E. coli*/100 mL (Sukias et al. 2011). However, the study also found a surprising net export of E. coli with increases in total yield ranging up to 34 fold in the wetland outflow (Fig. 4.5). Inlet drainage water concentrations for this wetland system are typically low (median 23 E. coli MPN/ 100 mL), but higher concentrations were found in the outlet (median 98 E. coli MPN/100 mL), typically by more than an order of magnitude. The increase in E. coli is probably due partly to wildlife deposition, but complimentary genetic evidence suggests that in situ growth of environmentally adapted strains of E. coli may be a major contributing source within the wetland (Stott et al. 2014a). However, increases in E. coli observed during passage through the wetland do not necessarily mean that potential pathogen levels and associated health risks have increased.

Subsurface-Flow Constructed Wetlands

Wastewater treatment systems may also be impacted by stormwater flows and sewer infiltration and, hence, can experience variable wet weather flows. Although diverting stormflow will protect WWTPs from flows exceeding peak design flow and treatment capacity, WWTPs may receive the first flush from storm events, which can be a major source of faecal microbe pollution (Hathaway and Hunt 2011). Wastewater treatment systems, thus, also need to cope with periodic fluctuations in loading driven by rainfall events. However, there are few studies and only limited information on the performance on how sensitive passive systems such as CWs are to abrupt increases to their hydraulic and/or pollutant load (i.e. shock loading) from wastewater.

The microbial removal performance of a wide range of different subsurface-flow constructed wetlands (SSFCWs) under constant flow has been compared by Headley et al. (2013) who found E. coli removal was a function of design and loading rate. The resilience of CWs to variable hydraulic and faecal microbial contaminant loads has been investigated by Stott et al. (2014b). Their studies followed the effects of shock loading on the microbial disinfection treatment performance for different configurations of SSFCW systems and denitrifying bioreactors by increasing the hydraulic loading of primary treated wastewater fivefold for 5 days. Horizontal- and vertical-flow wetland systems both responded quickly to changes in flow with a decline in median removal of *E. coli* and F-RNA phages by 0.1-2.1 log₁₀. Vertical-flow wetland systems followed by carbonaceous bioreactors demonstrated greater treatment resilience under shock loading (with regard to disinfection performance) with higher microbial removal rates of up to $3 \log_{10}$ in comparison to less than 2.3 log₁₀ for *E. coli/*F-RNA removal in HFCW. With the resumption of normal flow, all systems recovered quite quickly (within 5 days) to around 70-95% of pre-shock loading performance. The HFCW showed the greatest recovery, but the least removal for both microbial indicators. The removal of E. coli was more affected by shock loading, with a greater decline in removal of up to $2 \log_{10}$ in comparison to a smaller loss in removal of $1 \log_{10}$ for F-RNA. With the continuation of normal flow conditions, E. coli and F-RNA removal in all systems continued to improve such that after 13 days, removal rates approached or exceeded that of baseline conditions at the start of the trial.

Bioretention Filters

Bioretention systems (biofilters) have been employed globally to treat storm- and wastewaters prior to discharge. In Germany, bioretention filters [called retention soil filters (RSFs)] have been used for treatment of CSOs in full-scale applications for more than two decades. Waldhoff (2008) conducted a comprehensive investigation into the removal efficacy of *E. coli* and intestinal enterococcus in pilot- and large-scale bioretention filters for CSO treatment. Reduction of faecal indicator

bacteria ranged between 1 and 2 \log_{10} with removal considered to be mainly due to filtration and adsorption. After a week, no culturable bacteria were detected in outflows indicating potentially greater treatment efficiency. Waldhoff (2008) attributed this mainly to natural death caused by starvation and drought stress. However, it is known that bacteria can enter a viable but non-culturable state (VBNC) in which they are not dead, but in a kind of dormant stage from which they can return back to a culturable and infectious one (Oliver 2005; Li et al. 2014).

Merkel and Schaule (2010) investigated the performance of four established but sparsely loaded large-scale bioretention filters for CSO treatment in North Rhine-Westphalia (Germany), in operation for 3.5–4 years. The removal of *E. coli* varied between a median of 1.4 \log_{10} to almost 3 \log_{10} for the different facilities. The observed retention time was up to 8 days, with removal performance improving during longer lasting events.

Long-term monitoring of a large-scale bioretention filter [hydraulic filter performance of 0.015 L/(s m²)] by Christoffels et al. (2014) showed a greater mean reduction capacity, of at least 3 log₁₀ for FIB, *Giardia lamblia* cysts and somatic coliphages, in CSOs than results from previous studies of Waldhoff (2008) and Merkel and Schaule (2010). The mean reduction capacity in case of (faecal) pathogenic bacteria investigated (e.g. *Campylobacter* spp., *Salmonella* spp. or *Clostridium perfringens*) was similar than those of FIB. Additionally, higher maximum reductions were observed of 4.7 log₁₀ for *E. coli* and of 4.9 log₁₀ for enterococci (Mertens et al. 2014).

Studies on stormwater biofilters suggest that adsorption and straining processes retain indicator bacteria within the filter material during wet weather events where they can survive during dry weather periods, depending on the moisture content of the filter media (Stevik et al. 2004). Temperature is also a critical factor during dry phases with survival of trapped bacteria prolonged under low-temperature conditions (Zhang et al. 2012). Bacteria may then desorb from filter media following subsequent wet weather events (Chandrasena et al. 2012) and be present in relatively high concentrations in the outflow (e.g. 10^3-10^4 *E. coli* MPN/100 mL, Li et al. 2012). However, the presence of a submerged zone and the type of vegetation can enhance the removal of faecal microbes particularly indicator bacteria in stormwater bioretention filters (Chandrasena et al. 2014a, b).

Generally, lower removal rates are observed for faecal indicator bacteria in comparison to viral indicators. Li et al. (2012) reported mean removal rates of 2 log₁₀ for *E. coli* during wet periods compared to removal rates for F-RNA bacteriophages exceeding 3 log₁₀ for inflow concentrations of 10^5 *E. coli* MPN/ 100 mL and 10^4 F-RNA PFU/100 mL; attachment and straining were considered to be significant processes involved in the removal of the F-RNA phages. Microbial indicators may also behave differently from pathogens as Chandrasena et al. (2016) reported higher removal of *E. coli* compared to *Campylobacter* though this may reflect the lower pathogen concentration in the stormwater inflow.

Bioretention filters may, thus, exhibit relatively low and variable removal rates, particularly at lower temperatures and after dry periods. However, Li et al. 2012 reported that, whilst antecedent drying significantly decreased *E. coli* removal

efficiency, maintaining a saturated zone and introducing a carbon source reportedly eliminated the impact of drying on *E. coli* removal.

Carbonaceous bioreactors have been used successfully to supplement SSFCWs and small package plants to achieve improvements in wastewater effluent quality and facilitate resilience to loading fluctuations. Pilot-scale carbonaceous bioreactors contributed up to 2 \log_{10} removal of *E. coli* following hydraulic shock loading of vertical-flow treatment wetlands with sand media; overall *E. coli* removal rates exceeded 3.5 \log_{10} for the combined wetland and bioreactor systems (Stott et al. 2012). Similarly, a full-scale saturated horizontal-flow woodchip denitrifying bioreactor receiving variable flow ranging from 0 to 30 m³/day demonstrated consistent removal of *E. coli* (2.9 \log_{10} reduction) and F-RNA phages (3.9 \log_{10} reduction) despite receiving highly fluctuating inflow concentrations of up to 10^5 *E. coli* MPN and F-RNA PFU/100 mL (Rambags et al. 2012). Outlet concentrations of 20 and 30 MPN/100 mL for *E. coli* and 3 and 23 PFU/100 mL for F-RNA phages demonstrating the resilience of these systems for microbial contaminant removal.

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Chapter 5 Metals: Occurrence, Treatment Efficiency and Accumulation Under Varying Flows

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Abstract Metals were the first priority pollutants to be widely investigated in stormwater. In solid phase, they are often attached to very fine particles. The dissolved fraction creates considerable environmental problems as it is the most bioavailable fraction. Hence, removal of both fine and dissolved particles plays a major role in the treatment of polluted runoff. Ecotechnologies specifically designed to remove metals should be able to address different treatment mechanisms. However, the exhaustion of sorption capacity reduces the lifespan of treatment facilities. Additionally, metal concentrations fluctuate extremely—spatially, seasonally and over time—which poses another challenge for further increasing removal efficiencies. While soil- or sand-based systems should be designed in a way that the filter material can be exchanged, newer developments such as Floating Treatment Wetlands show promising removal capacities as the installations bind metals in sludge sediments, which can be removed from time to time. The different treatment mechanisms, aforementioned developments and techniques as well as their removal capacities will be discussed in this chapter.

Introduction

Metals from various sources are commonly found in stormwater (and to a lesser extent in wastewater) discharges and have long been in focus when stormwater impacts on receiving water bodies and/or water quality treatment demands are assessed and discussed. Early research evaluating stormwater quality has recog-

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nised metals to be of certain importance. Wilber and Hunter (1975) emphasise that 'heavy metal concentrations (in stormwater runoff) were found to vary significantly throughout runoff events and from storm to storm'. This chapter will describe and discuss these variations and treatment technologies, which have been extensively evaluated with a focus on their metal treatment capacity.

Treatment Mechanisms

One main characteristic of metals which significantly affects removal processes is their distribution between the dissolved and particulate phase. A common way to estimate this distribution is by passing them through filters with a pore size of 0.45 μ m and dividing them into the fractions as shown in Fig. 5.1. In a geochemical context, the dissolved fraction is commonly divided into colloidal and genuine dissolved fractions (Ingri 2012). Colloids, unlike particles, do not settle but remain in solution. The surface of colloids is often negatively charged, causing positively charged metal ions to bind to it. The ions and molecules present in free form without binding to colloids or particles are referred to as a true or authentic dissolved fraction. This is also the most bioavailable fraction since it can be taken up by aquatic plants and organisms, which also means it has increased toxicity (Ingri 2012; Campbell 1995).

Important factors that affect the solubility of metals and mobility are pH values and dissolved organic matter (DOM, such as humic and fulvic acids) and the access to particle surfaces for them to attach to. Generally, the solubility is higher at low pH values (Ingri 2012).

In stormwater quality studies, often both total and dissolved (i.e. <0.45 μ m) fractions are analysed. This enables researchers to calculate the particulate fraction by subtracting the dissolved from the total concentration. The distribution between these fractions can vary substantially, not only between different metals but also within a runoff event and between different sites and seasons. For instance, investigations on the distribution of metals from runoff of five German highways by Dierkes (1999) revealed that

- 51–90% of Cd (mean 70%),
- 28–55% of Cu (mean 42%) and
- 14–51% of Pb (mean 36%).

are in the dissolved phase (<0.45 μ m). Boogaard et al. (2014) found even broader ranges and mean values of approximately 60% for Cd and for Cu

Fig. 5.1 Metal fractions in stormwater (simplified	Diameter	0.1 j	μm 0.4	5μm Ι
scheme: H. Österlund)	Truly dissolv	ved	Colloid	
		Particulate		

(range ~20–90%), 55% for Ni (20–95%), 70% for Pb (10–99%) and 80% for Zn (10–90%). As already mentioned in Chap. 2, these particles are bound in large proportions to particles with a grain size of less than 90 μ m or even 60 μ m (Xanthopoulos 1990; Boogaard et al. 2014). Hence, treatment installations should ideally be at least capable of retaining fine suspended solids.

Metals in Stormwater from Separate Sewer Systems and Combined Sewer Overflows

One important source for metals in stormwater is vehicular traffic. Further, metals also leach from surfaces in the urban environment, such as roofs, lampposts, barriers, facades, etc. or are of geogenic origin. Their composition in runoff changes over time, e.g. substitutes used in industrial products, such as the replacement of lead in fuel over the last two decades.

As listed in Chap. 1, the concentrations found in stormwater often vary during single events (e.g. due to first flush effects or varying rain intensities during the event that transports different fractions), between different events (e.g. due to varying antecedent dry periods, seasonal variations and varying rain characteristics), seasons and between different catchments (due to different catchment characteristics).

Seasonal Variations

In a study in northern Sweden, significantly higher concentrations in snowmelt runoff have been observed in March and April (Cu: 37–199 mg/L, Pb: 16–80 mg/L; Zn 105–791 mg/L) compared to runoff from rain events in May and June (Cu: 30–45 mg/L, Pb: 14–19 mg/L, Zn 130–169 mg/L) (Westerlund et al. 2003). In this study, concentrations of both metals and suspended matter in stormwater are higher in snowmelt runoff than during rainy periods. In snowmelt runoff, relatively high concentrations of Cu, Pb and between 16 and 80 mg/L were measured. This can be explained by the long period of contaminant accumulation in the snow; these contaminants are then released during a relatively short period. In a study in Germany, similar results have been confirmed by Helmreich et al. (2010), who showed significantly higher metal and total suspended solids (TSS) concentrations in winter runoff compared to the summer season. Reasons given by the authors were the use of sand and gravel for anti-slip applications, which increases wear and tear on road surfaces and vehicles.

Besides concentration variations, metal characteristics may also change. During winter in cold or temperate climates, de-icing salts are applied regularly, which, for instance, affects metal partitioning towards the dissolved phase (Marsalek et al. 2003). Higher percentages of dissolved pollutants can affect the performance of treatment technologies (Søberg et al. 2017).

Variation Between Catchments

The quality of stormwater depends on the surface characteristics of the catchment and the anthropogenic activities in or around the catchment (Eriksson et al. 2007). The contamination of stormwater with metals in urban catchments largely depends on the use of building materials, on the one hand, and the presence of frequently used roads, on the other hand. Studies have shown that runoff from metal roofs may have higher concentrations of, e.g. Cu and Zn than road runoff while other metals such as Cd, Pb, Ni and Cr are higher in road runoff (Göbel et al. 2007). In general, areas with direct connection to traffic and runoff from industrial and commercial areas often exhibit relatively high pollutant concentrations (Pitt et al. 1995; Czemiel Berndtsson 2014).

Although a correlation between the traffic density and the concentration of metals in road runoff is often assumed, Kayhanian et al. (2012) could not prove such a correlation in a literature review on road runoff worldwide. They showed, however, differences between the concentrations in North America, Europe and Asia, which prove that a local aspect has to be considered. The authors also mention the influence of preceding dry phases and the catchment area, as mentioned in Chap. 1.

Site variations may also vary for different pollutants. Gasperi et al. (2014) analysed pollutants in stormwater from three different areas. They found different Cu, Cr, Ni and Zn concentrations in these areas while they did not detect any differences in Cd and Pb concentrations, although the land use in the areas was different.

Variations Over Time

In general, it is quite difficult to compare the ranges of concentrations found in road runoff since the sampling points vary in the different studies, as can be found in literature reviews. However, many publications refer to investigations made in the middle of the 1990s or even earlier. The age of these studies is important, since metal concentrations have shifted over the decades; the ban on leaded gasoline in most countries has reduced lead concentrations in runoff significantly (Kayhanian et al. 2012; Ayrault et al. 2014). Future trends of stormwater quality changes depend on how treatment facilities perform during their lifespan. However, simulating these developments over time involves quite high uncertainties (Borris et al. 2016). Changes in climate, building materials and building design, environmental regulations and the use of unknown substances today may affect stormwater quality in the future.

Table 5.1 shows ranges of road runoff concentrations from different sources, which only include values published after 2005 given the fact that runoff composition has changed during the next last decades. In general, all values vary over two

Table 5.1 Concentrations in urb	oan runoff (μg/L), ₁	published after 20	600				
Source/Catchment	Value(s)	Cd	Cu	Ni	Pb	Zn	Reference
Highway runoff							
Road high density, 57,000	Min-max	<0.5-4.8	24-604	4.2-403	<5.0-405	128–3470	Helmreich et al.
vehicles7d-1, Munich, Germany (63 sampled events	Median/mean	<0.5/<0.5	155/191	35/55	43/56	592/847	(2010)
over a period of 2 years)							
Road load density	Mean	1	11	1	34	315	Li et al. (2012)
Separate sewer system							
Residential area	Min-max	0.1-0.4	3-15	1-10	1-8	5-140	Hvitved-Jacobsen et al. (2010)
	Mean \pm SD	1	38 ± 28.4	2.9 ± 2.0	1	212.4 ± 145.1	Gasperi et al.
	Mean \pm SD	1	14.9 ± 11.3	3.1 ± 2.3	I	126.3 ± 87.1	(2014)
	Min-max	1	11-87	0.9–13	0.2-10	33-400	Valtanen et al.
	Min-max	1	0.3–28	0.4–30	0.1 - 9.8	0.02-156	(2014)
Industrial	Mean ± SD	1	34.6 ± 29.2	6.6 ± 4.5	I	239.8 ± 169.8	Gasperi et al. (2014)
Commercial	Min-max	0.1-0.5	4-31	1–11	1–19	8–92	Hvitved-Jacobsen et al. (2010)
Mixed	Min-max	1	1.8-129	0.7-125	0.2–68	7.3-703	Valtanen et al.
	Min-max	1	1.8-656	0.7-122	0.2–98	7.3-1937	(2014)
	Min-max	0.1-0.5	6-120	3–190	1–33	10–300	Hvitved-Jacobsen et al. (2010)
Roofs	Mean	1	8	1	31	778	Li et al. (2012)
Combined sewer system							
Mixed area	Median ± SD	0.24 ± 0.02	27.0 ± 2.6	18.0 ± 1.3	26.0 ± 3.1	220 ± 15	Raclavska et al. (2015)
	Min-max	1	86–134	1	46–175	658–1137	Gasperi et al. (2014)

to three magnitudes. Due to the high variations, no clear overall trend can be derived, even if similar sampling locations are compared.

In terms of CSO, to date, there are only few discharge measurements available before the flow volume enters the river. In most cases, researchers concentrate on an increase of the pollutants in the receiving surface water body by measuring upstream and downstream of a discharge point or in the river sediment. Table 5.1 presents results of some measurements taken in combined sewer systems or at their outlets. Since the sampling points and the catchment are not completely comparable, it is only possible to deduct general trends. Variations between minimum and maximum are within one magnitude. The values are in general also comparable to those from separate sewer systems and highway runoff despite the different composition of CSO.

Treatment Systems

Stormwater Ponds and Basins

As described in Chap. 2, the removal mechanisms of stormwater ponds rely mainly on sedimentation. Given that fine sediments show relatively higher metal concentrations (Sansalone and Buchberger 1997; Liebens 2002), sediment close to the inlet tends to have lower metal concentrations (Karlsson et al. 2010). As mentioned above, the coarser forebay sediment may show a lower toxicity.

Table 5.2 gives an overview of data on metal concentrations in stormwater pond sediments published between 2010 and 2017. As can be seen, similar to the metal concentrations in the stormwater itself, the range found in the dry matter (DM) is quite high.

Dissolved substances are only reduced in shallow, planted areas, comparable to ponds with FTWs (see Chap. 2). For instance, a Swedish study demonstrated considerable removal of dissolved metals in a stormwater pond (Cd: 73%, Cu: 58%, Pb: 41% and Zn: 64%). However, this is still far lower than the removal of particulate metal [between 85 and 92%, (Al-Rubaei et al. 2016)]. In studies from USA and Sweden, stormwater pond influent and effluent concentrations of dissolved metals were in the same range (Stanley 1996; Pettersson 1998).

Also, when the metal contamination is assessed in sediment accumulated in ponds, only looking at the total metal content can be misleading since metals may be present in different fractions and, thus, potentially available to different degrees. Sequential extraction procedures reveal the metal fractionation by distinguishing between the five fractions: exchangeable (I), carbonate-associated (II), Fe–Mn oxide-associated (III), organic matter/sulphide-associated (IV) and residual (V). Metals in fractions I to IV are potentially bioavailable since they can be released

Source/Catchment	Value(s)	Cd	Cu	Pb	Zn	Reference
Highway/nature	Min-max	0.4–0.6	200–250	40-60	800-100	Karlsson et al. (2010)
Residential/ industrial	Min-max	0.8–1	50–150	60-80	20–700	Karlsson et al. (2010)
Commercial/ residential	Min-max	1.1–1.7	403–581	133–179	579-825	Karlsson et al. (2010)
Commercial/ residential	Min-max	0.5–1.7	138–406	47–109	427–1069	Karlsson et al. (2010)
Industrial	Mean for inlet; middle; outlet	<0.5; <0.5; <0.5	3293; 3137; 1625	220; 198; 83	1361; 1051; 760	Isteniç et al. (2012)
Residential	Mean for inlet; middle; outlet	<0.5; <0.5; <0.5	133; 171; 129	10; 10; 6	234; 240; 190	
Residential	Mean for inlet; middle; outlet	<0.5; <0.5; <0.5	45; 6; 4	22; <2; <2	378; 82; 26	
25 Swedish municipal ponds	Min-max	0.1–2.3	3–109	3-60	14–597	Al-Rubaei et al. (2017)

Table 5.2 Overview of metal concentrations measured in sediment from stormwater ponds (mg/kg DM), published after 2009 (partly based on Søberg 2014)

from the sediment if the ambient conditions change (e.g. after excavation during maintenance, see the following text). For instance, a recent study from Sweden (Karlsson et al. 2016) shows—for sedimentation ponds and tanks as well as for storm drain sediment—that the majority of Cd, Cu, Pb and Zn and a significant amount of Ni were in potentially mobile forms. This fact must be considered during pond maintenance (sediment removal, drying/de-watering/disposal) to prevent metal from being released. Similar results were reported in various studies (Marsalek and Marsalek 1997; Camponelli et al. 2010; Lee et al. 1997).

In winter, an ice cover on the sediment pond reduces oxygenation of the pond water (e.g. by wind) (German et al. 2003), which can affect metal partitioning. Additionally, road salt used in cold climates affects the metal partitioning between particulate and is dissolved: if road salt is present in stormwater, a higher percentage of the metals is in the dissolved phase (Søberg 2014). Since ponds mainly remove metals in particulate form, the overall metal treatment performance may decrease.

Constructed Wetlands

Surface-Flow Constructed Wetlands

Since metals are often bound to particles and since wetlands capture such particles to a great extent (Sansalone and Buchberger 1997), wetlands remove a significant reaction of total metals thanks to their sedimentation process. Resuspension of the captured metals has to be avoided (Zhang et al. 2012). In comparison to ponds, wetlands provide more heterogeneous morphology including dense vegetation; therefore, treatment of fine particles and/or dissolved metals is potentially more effective than in ponds.

In an extensive literature study and meta-analysis, Carleton et al. (2001) investigated factors affecting the stormwater quality treatment performance of constructed wetlands (CWs). The review published data from 49 wetlands in 35 studies. In combination with results of other studies, the removal rates achieved are illustrated in Fig. 5.2.

The figure underlines that, similarly as for ponds, the metal treatment efficiency reported in different studies varies significantly depending on a wide range of factors. In general, however, the figure corroborates the assumption of Birch et al. (2004), who conclude that a mean removal of Cd, Cu, Pb and Zn of approximately 60% can be achieved. In studies done in 1997 and 2012/13 in Bäckaslöv, Växjö (Sweden), metal removal exceeding 80% was observed (Semadeni-Davies 2006; Al-Rubaei et al. 2016), which is in the upper range of the data included in the meta-analysis done by Carleton et al. (2001). The study of Al-Rubaei et al. (2016)



Fig. 5.2 Interval plot (95% confidence interval bar) of removal percentages achieved in constructed stormwater wetlands

also included dissolved metals. Their outflow concentrations were significantly below the inflow concentrations. Removal rates were between 55 and 80%. In the combined pond–wetland system evaluated in this study, the wetland increased the removal of the dissolved metals significantly compared to the removal in the pond only, underlining the importance of more advanced treatment processes for dissolved contamination removal.

Floating Treatment Wetlands

Since the treatment in stormwater ponds relies on sedimentation to large extent, the treatment of dissolved metals (and other compounds) in stormwater ponds may be insufficient. Accordingly, retrofitted Floating Treatment Wetlands should improve their treatment performance. After such pond retrofitting, the metal and sediment removal significantly increased [TSS, particulate Cu, and particulate Zn by 40% and dissolved Cu by 16% (Borne et al. 2013)]. Reasons for that are an increased direct plant uptake (Borne et al. 2013; Ladislas et al. 2013, 2015), bacterial/biofilm uptake (Borne et al. 2014), increased sorption [e.g. to organic matter (Borne et al. 2014)] and precipitation processes due to higher humic content, lower dissolved oxygen and more neutral pH value (Borne et al. 2013). In a study in New Zealand, some release of metals was observed in the spring, especially of Cu, due to organic matter degradation and, and thus the export of dissolved organic matter from the pond (Borne et al. 2014).

Subsurface-Flow Constructed Wetlands

Since most metals entering media-based systems are particle-bound, mechanical filtration of the incoming stormwater sediment also removes substantial loads of metals (and other particle-bound pollutants). Thus, the efficiency of TSS and particle-bound metal removal is correlated which was shown by Hatt et al. (2008) for vertical-flow stormwater wetlands (see section Bioretention filters). Studies on systems for horizontal-flow wetlands used for stormwater or CSO treatment are missing; however, the general processes in media-based systems are the same. Dissolved metal removal varies more since it is affected by diverse factors that influence soil and metal interactions. The main metal retention processes in soil are adsorption (including metal-OM complexation and cation exchange), surface precipitation and fixation (mainly to clay minerals) (Alloway 1995). Key soil properties controlling these processes are, among others, pH, OM content, clay mineral content and oxidation reduction potential (Bradl 2004). Besides these geochemical processes, plant metal uptake plays a less significant role (Read et al. 2008; Søberg et al. 2014a; Muthanna et al. 2007a) and is less important since the plants are not usually harvested. However, vegetation in media-based systems plays an important role in maintaining the infiltration capacity, facilitating treatment indirectly (e.g. by

effects on microbial communities in the filter) and providing aesthetical values and/or (urban) biodiversity.

Bioretention Filters

From approximately 2000 onwards, numerous studies have been published on how well stormwater bioretention filters remove pollutants. A summary of inflow and outflow metal concentrations reported in selected studies is given in Table 5.3. Cadmium concentrations were only investigated in two studies with inflow values between 4.6 and 5.6 mg/L and removal efficiencies between 66 and >99.5%.

Table 5.3 Biofilter inflow and outflow concentrations of metals (μ g/L) from selected studies. (diss. dissolved concentrations; non-veg. non-vegetated; In. inflow concentrations; Out.: outflow concentrations) (partly based on Søberg 2014)

Filter type	Value(s)	Cu		Pb		Zn		Reference
		In	Out	In	Out	In	Out	
Field scale	Total, mean	56.8	1.9	41.4	10.2	98.3	20.6	Glass and Bissouma (2005)
Field scale	Total, mean/range	10	3-4	58	<2-4	107	44-48	Davis (2007)
Field scale	Total, mean	-	-	-	-	72	17	Hunt et al. (2008)
Field scale	Total, mean/range	10	46	6	2–3	100	13–30	Hatt et al. (2009)
Field scale	Total, mean	60	5	110	7	330	13	
Field scale	Total, mean	19	16	6	3	71	12	Li and Davis (2009)
Field scale	Total, mean	13	9	<2	<2	15	3	
Field scale	Total, mean	16	6.3	17	4.5	120	47	Chapman and Horner
Field scale	Dissolved, mean	3.6	2.9	<1	<1	49	26	(2010)
Field, with submerged zone highway	Total, mean	20	52	80	22	130	280	Li et al. (2014)
Field highway	Total, mean	20	62	80	5	130	310	
Field, residential	Total, mean	60	5	110	7	330	13	Hatt et al. (2009)
Carpark, 3 filter cells	Total, mean	10	6	6	2	100	13/15/30	
Sludge as filter medium	Total, mean	241	4.5	90.3	0.2	1127	2.1	

The total metal removed by bioretention filters often exceeds 80–90% (Hatt et al. 2009; Muthanna et al. 2007b; Read et al. 2008; Sun and Davis 2007).

As for most compounds removed by bioretention filters (see, e.g. Chap. 2) the processes and properties are, to varying degrees, affected by ambient conditions, e.g. the drying/wetting pattern, ambient temperatures, road salt in the runoff, the pollutant concentrations in the runoff and the runoff intensity (Hatt et al. 2007b; Blecken et al. 2009; Søberg et al. 2014b; Muthanna et al. 2007a; Denich et al. 2013; Bratieres et al. 2008), the filter design (e.g. water saturated zone, different filter materials) (Dietz and Clausen 2006; Davis et al. 2009; Hatt et al. 2008).

Although dissolved metal removal has been shown to vary far more than the quite stable total metal removal, only the total metal removal has been investigated in most biofilter studies (see Table 5.1). Dissolved metal removal has been considered in fewer of the investigations (Muthanna et al. 2007b; Read et al. 2008; Hatt et al. 2007a; Sun and Davis 2007; Søberg et al. 2014b). In pilot-scale stormwater biofilters, Muthanna et al. (2007b) found removal rates of dissolved Zn up to 70%, whereas leaching was observed for both dissolved Cu and Pb. In a laboratory study investigating biofilter columns at three different temperatures, Blecken et al. (2011) found lower removal efficiencies (24–66%) for dissolved Cu and Pb compared to Zn and Cd (99%), and a negative correlation between temperature increase and removal of dissolved Cu and Pb. In a study about temperature and salt influence on metal removal in laboratory pilot-scale bioretention filters, Søberg et al. (2014a) found high removal of dissolved Zn and Cd (>90%), whereas removal of dissolved Cu and Pb was less efficient, ranging from -1345 to 71% being deteriorated by the presence of salt, particularly in connection with high temperature.

Although some findings indicate that dissolved metal removal is significantly lower than total metal removal and, in particular, Cu leaching was observed (Hatt et al. 2007a; Chapman and Horner 2010; Muthanna et al. 2007b), biofilters seem to have potential to provide adequate dissolved metal treatment if filter material with specific sorption properties is used (Sun and Davis 2007; Hsieh and Davis 2005). An efficient removal of dissolved metals has also been reported for bioretention filters where sandy soils with only little organic content are used as filter material (e.g. Blecken et al. (2011) reported removal rates of >99% for dissolved Zn and Cd and >60% for dissolved Cu when using filter material with 90% sand). Numerous studies have further tested various filter materials to enhance metal treatment. Examples are zeolites and peat (Färm 2003), blast furnace slag, chitosan, crab shell, peat, sawdust and sugar cane (Vijayaraghavan et al. 2010), limestone, shell sand, zeolite, and olivine (Wium-Andersen et al. 2012). Many of these results are derived from short-term laboratory studies; when these results are transferred to praxis, it is important to consider long-term behaviour of the material (e.g. breakdown and release of associated pollutants over time). When choosing filter materials for bioretention systems, one thus has to compromise between infiltration rate, adsorption capacity and support of plant growth.

Typically, metals do not ingress far into the filter material, but are trapped on or near the top of the filter due to both mechanical removal and sorption processes (e.g. Davis et al. 2001; Grotehusmann et al. 2017). Grotehusmann et al. (2017) found that metals accumulate on the filter surface and in the first 10–15 cm of the filter layer in correlation with how much calcium carbonate (CaCO₃) is available, which is often added as additional layer on top of the filter surface at the large-scale sites investigated in Germany. Although in general, high inflow values of CaCO₃ onto the filter could also lead to building up a carbonate layer, due to the hydraulic conditions on the filter surfaces, it is usually limited to areas close to the inflow and did not result in overall clogging of the filter surface. However, when CaCO₃ is added as additional surface layer or mixed into the filter material, the additive itself may not be contaminated with heavy metals, e.g. lead (Grotehusmann et al. 2017). The high metal removal in the upper layer facilitates filter maintenance since merely scraping off the top layer may remove a high proportion of accumulated metals from the system, and thus postpone the need to replace the whole filter media (Hatt et al. 2008).

Some field investigations predicted that the accumulation of fine stormwater sediment on top of the filter material and in the upper layers reduces the hydraulic conductivity relatively quickly, sometimes even within several months, and leads to clogging (Li and Davis 2008). However, Grotehusmann et al. (2017) could not confirm this in large-scale investigations on filters designed according to German standards. The main reason for this finding was oversized filter layers which led to low long-term loads of fine sediments.

During winter in cold or temperate climates, pollutant concentrations are particularly high, and de-icing salt often affects metal partitioning towards the dissolved phase (Marsalek et al. 2003; Oberts 2003). The presence of salt has been shown to substantially influence the ability of stormwater biofilters to remove metals. The latter is particularly pronounced for dissolved metals (Søberg et al. 2014b). Søberg et al. (2014b) found that ion exchange by Na⁺ was probably entirely responsible for the leaching of dissolved Pb from the filter material.

In winter, plant metal uptake is generally inhibited by salt in stormwater runoff (Fritioff et al. 2004) and low temperatures generally reduce biological activities. Søberg et al. (2014b) examined the impact of temperature, salt and a submerged zone on metal uptake in three native (Northern Sweden) wet/drought tolerant plant types: Juncus conglomeratus, Phalaris arundinacea and Carex panacea. They found a generally higher metal uptake at low temperature. Their results suggested that the three plant species were not particularly affected by different temperatures and/or the presence/absence of a submerged zone in the filter and/or salt in stormwater. This indicates the potential to use the investigated plant species for targeted cold climate biofilter design. Additionally, Denich et al. (2013) found that biofilter vegetation was capable of withstanding high salt exposure. Despite the reduced biological activity in cold seasons as described in Chap. 2, metal retention was good for both seasons with mass reductions of 90, 82 and 72% of Zn, Pb and Cu, respectively (Muthanna et al. 2007b). The latter is supported by findings of a study evaluating seasonal performance variations (Roseen et al. 2009), where seasonal contaminant removal performance was found to vary little for stormwater biofilters.

Swales and Buffer Strips

Grotehusmann et al. (2017) found out that a major part of the metals is already captured within the first 10 cm of the buffer strip leading to the swale. Since the buffer strips contain rather coarse media, the metal accumulation was also found in deeper layers (25–30 cm). The authors revealed that a major part of the retention was, thus, already provided by the shoulder, and concluded that the treatment of swale effluent, as often practiced in Germany, is not necessary.

Reported removal percentages of metals in swales vary as follows: Bäckström et al. (2006) report about 20% metal removal while Stagge et al. (2012) and Knight et al. (2013) report very efficient metal removal rates. Bäckström et al. (2006) found that the particle size distribution influences the removal efficiency: only large particles >250 μ m settle in swales. In general, the pollution removal capacities for dissolved pollutants and small particles are low. Thus, Bäckström et al. (2006) conclude that, while efficient for flow retention, swales cannot produce consistently high pollutant removal.

Although swales commonly tend to be comprised of grass, they can have particular design modifications (such as wetland planting) to improve nutrient reduction (Winston et al. 2012). Metal uptake by plants can be significant. This uptake is specific to metal and plant species (Zhang et al. 2012). Most plants accumulate the metals in their roots, but also transport to the leaves occurs (Weis and Weis 2004). It is, thus, important that swales and buffer strips be harvested regularly.

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Chapter 6 Emerging Contaminants: Occurrence, Treatment Efficiency and Accumulation Under Varying Flows

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Abstract Emerging contaminants became a major topic in water treatment when laboratory detection methods for concentrations at a nanogram-scale improved approximately two decades ago. Research on using ecotechnologies to remove emerging contaminants in variable stormwater and wastewater flows has been conducted for more than a decade, but so far, not all removal mechanisms are well understood and only few setups have been investigated. This chapter summarises the current knowledge, focussing on pesticides and emerging contaminants listed on the watch list of the European Union. However, large-scale investigations are still rare and further research will have to be conducted in this field to enable practitioners to provide recommendations for design and maintenance of treatment facilities in the field of ecotechnologies.

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Introduction

Emerging contaminants are substances that have been detected in the environment, but which are currently not included in routine monitoring programmes and whose fate, behaviour and toxicological effects are not well understood.¹ Originating from manifold products, including human and veterinary medicines, nanomaterials, personal care products, paints and coatings, emerging contaminants have caused concern over the last decade as they may have harmful effects on human and ecological health. Because these contaminants often make their way into the environment via wastewater discharges, urban stormwater and runoff from agricultural land, research has investigated or is still investigating how to remove a wide range of them in treatment systems, including both conventional ones and ecotechnologies. Depending on their context, these studies focus on substances mentioned in regulations such as the EU Water Framework Directive (EU 2000), ecotoxicological issues in receiving waters (Strobel et al. 2016) or human health risks in urban water cycles (Pal et al. 2014).

Because chemicals released into the water cycle are becoming more diverse, the analytical effort required to detect all relevant contaminants in the environment has increased significantly.

Emerging contaminants can be divided into inorganic and organic compounds, e.g. metals, pesticides or phenols. Their removal mechanisms rely on their chemical composition and involve physical processes (sedimentation, filtration), biological processes (uptake by plants and microorganisms) and chemical processes (sorption, precipitation and co-precipitation, oxidation and hydrolysis, metal carbonisation and sulfidation, etc.) (Gasperi et al. 2012a; Zhang et al. 2012).

Since the sheer number of industrially manufactured chemical compounds makes it impossible to monitor all of them, indicator substances are being sought. **Source indicators** require a comparably high polarity, a low sorption tendency and a high persistency towards chemical and biological attenuation processes, while **process indicators** should exhibit a defined reactivity and behaviour towards a respective process.

Jekel et al. (2015) suggest monitoring a representative subset of source or process indicator substances 'with similar characteristics with respect to application, source, physicochemical properties or reactivity'. These indicators should

- have known sources, and be common, distinct and continuously released,
- have a well-known fate (e.g. photolysis, biodegradation, adsorption and others)
- occur in all natural compartments of the water cycle and
- be detectable at low concentrations with comparably low effort by widely available methods.

¹EU Network of Reference Laboratories, Research Centres and Related Organisations for Monitoring of Emerging Environmental Substances (NORMAN) <www.norman-network.net>.

For stormwater-driven urban runoff, Jekel et al. (2015) propose Mecoprop or Diuron as indicator substances, which are common herbicides used to protect building facades. For agricultural runoff as a non-point source (Babut et al. 2013), many studies focus on pesticides whose movement through ecosystems depends on (Poissant et al. 2008)

- their chemical properties such as soil organic carbon/water distribution coefficient (K_{oc}),
- half-life $(t_{1/2})$,
- vapour pressure, etc.,
- the environmental conditions and
- how they are applied.

The local and regional context—including hydrological regimes, soil properties and type of farming system—contribute to the global distribution of pesticides released to the environment. In this context, assuming widespread application, constructed wetlands (CWs) appear to mitigate the pesticide transfer at a catchment scale (Gregoire et al. 2009; Tournebize et al. 2017).

It is not always easy, however, to assess the contribution of different pollutants from specific sources, since stormwater at appropriate sampling points often involves runoff from different types of surfaces. It may, therefore, be useful to investigate potential sources in the lab and on a pilot-scale in order to identify the sources as well as estimate the concentrations of the relevant substances that different materials emit (Wangler et al. 2012; Winters et al. 2015; Andersson Wikström et al. 2015). As for all pollutants detected in stormwater, emerging pollutants can also vary within and between events, and between catchments. For instance, Gasperi et al. (2014) analysed micropollutants in stormwater from three different French catchments with different land use. They report differences of PAH concentrations between the three areas while they did not detect any differences in alkylphenol concentrations. However, given the limited number of studies dealing with emerging compounds (compared to those on e.g. metals and nutrients), the extensive number of such compounds and relatively high analysis costs, data is still lacking regarding concentration variations. Many of the studies conducted so far are rather screening tests to understand which compounds can be present in the runoff instead of being detailed investigations evaluating variations and processes it. However, such compounds have gained increasing interest and further in-depth analyses are likely to be published in near future.

Since there is such a broad spectrum of possible compounds, removal processes are not yet well understood and are currently being examined in different research projects, which is also the case for assessing the purification potential of CWs to dissipate pesticide pollution. Since pesticides have a great diversity of uses and properties as to how they transfer and dissipate, these investigations are quite complicated. Nonetheless, in addition to sedimentation for pesticides attached to settleable solids, pesticides can be eliminated from the water column through transfer (corresponds to sorption phenomena) and transformation processes, which produce new molecules (metabolites or degradation byproducts) from the so-called parent pesticide molecule.

Concentrations of Selected Emerging Contaminants

Since there are such a large number of organic and inorganic compounds worldwide and different definitions of 'emerging contaminants' in literature and legislation, comparing studies on these substances is quite difficult.

One way of structuring them is to follow legislative approaches. As one example, the Water Framework Directive of the European Union (EU WFD) defines 'priority substances' as substances presenting 'a significant risk to or via the aquatic environment' (EU 2000). It also gives a list of 'indicative main pollutants', containing substances that are commonly seen as emerging contaminants. These are

- organohalogen compounds and substances that create such compounds in water,
- organophosphorus compounds,
- organotin compounds and
- cyanides, metals, arsenic and their compounds.

Another group of contaminants is defined by substances that have a negative effect on organisms, for example, carcinogenic or mutagenic as well as endocrine disrupters or organic toxic substances that are persistent and bio-accumulative. Biocides and plant protection products can also be placed in this group since they are predominantly designed to have a special effect on organisms.

Later, a list of substances classified as priority substances was released and 19 of these marked as 'priority hazardous substances'. A proposal of the European Commission (European Commission 2012) to add threshold values for several priority substances to the Water Framework Directive led to the first watch list for emerging contaminants in surface waters (European Commission 2015), containing 11 substances or group of substances, of which three (17 β -estradiol, 17 α -ethinyl estradiol and erythromycin) are also listed in Drinking Water Contaminant Candidate List 3 of the US Environmental Protection Agency (US EPA 2009).

The substances included in this chapter are listed on the EU watch list; additionally, it contains a specific group of components identified in the relevant studies to have a strong impact on the different flows.

Pesticides are regularly detected in stormwater (e.g. Zgheib et al. 2011b). These include glyphosate, 3-(3,4-dichlorophenyl)-1,1-dimethylurea, better known under its trade name Diuron, and isoproturon. In urban areas, pesticides, often also referred to as biocides, are used for gardening activities in addition to their use as additives in or on building materials. The latter has been shown to be one of the main sources of pesticides in urban waters (Burkhardt et al. 2007). Bollmann et al. (2014) conducted a study in Denmark and showed that pesticides enter stormwater from varying types of urban areas including both central areas, but also suburbs.

Today, many substances are still found in stormwater although they have been banned from general use or are limited to selective purposes because they are harmful towards humans or the environment. For instance, in the European Union, **terbutryn** may be used to protect building surfaces from mould and also mecoprop and carbendazim are part of building materials, but they may not be used as pesticides in Germany, Switzerland and Austria. Consequently, mecoprop was found in stormwater in a Swiss study (Burkhardt et al. 2007). Another example is **organotin compounds** such as tributyltin (TBT), which was detected together with dibenzothiophene (DBT) in stormwater collected from three storm drains (n = 20) in Paris (Zgheib et al. 2011a, b). In Sweden, organotin compounds have been measured in stormwater from waste sorting areas and a deicing area at Stockholm-Arlanda Airport (Junestedt et al. 2004). The major sources of organotin compounds are industrial point sources, diffuse urban effluents and industry/household wastewater via wastewater treatment plants (Swedish EPA 2007).

Another major source of organic compounds is again road runoff. **Hydrocarbons** and **alkylphenol** sources in road runoff are accidental oil spills or deliberate dumping of oil or fuel, vehicle emissions, atmospheric deposition, leaching and/or erosion from road pavements.

Hydrocarbons are one of the most prevalent pollutants in both soil and water in urban areas. One important group found regularly in stormwater which poses toxic hazards is PAHs (Polycyclic aromatic hydrocarbons). PAHs are formed by incomplete combustion of organic matter (oil, coal, waste, etc.) and are thus found in traffic emissions and in bitumen, which is used in asphalt paving and roofing materials. Other relevant hydrocarbons in the stormwater context are benzene and alkenes, both of which are present in gasoline and the exhaust from its combustion.

Alkylphenols consist of a phenol group with one or more alkyl chains. They are a group of industrial chemicals used primarily for the preparation of surfactants, such as ethoxylates, in various types of detergents (KemI 2015). The most common alkylphenols for industrial applications are nonylphenol and octylphenol (Björklund 2011). Besides from road runoff, they can also occur in wash-off from building materials, e.g. in joint sealers and as pore builders in concrete (Björklund et al. 2007). For alkylphenols, the dissolved fraction dominates with a proportion of 65–85% (Bressy et al. 2011) which may affect treatment. Bressy et al. (2011) reported a high variability of the measured concentrations, up to a factor of ten, from one rainfall event to another.

Phhalates originate from a variety of products such as binding materials and pigments in varnishes and paints, but traffic is again a major source of phthalates in stormwater (Björklund et al. 2007). Björklund (2011) studied the presence of phthalates in water and showed that di-isononyl phthalate (DINP) was the phthalate which occurred in highest concentrations in water, snow and sediment. Also, Bis (2-ethylhexyl) phthalate (di-2-ethylhexyl phthalate, diethylhexyl phthalate (DEHP), which historically is the most widely used phthalate, was detected in surface water and exceeded the limits set in guidelines of the European Union in several cases. Phthalates are a group of industrial chemicals that are mainly used as plasticizers in PVC and other plastics (Swedish EPA 2009).

Another critical group of industrial chemicals is **per- and polyfluorinated alkyl substances (PFAS)**, which are used in many products due to their water and fat-repellent properties (Swedish EPA 2016). Their use in products such as fire-fighting foam is especially problematic in the stormwater context since the nearby environment—groundwater and surface water bodies—may be contaminated because of a direct discharge with surface runoff. PFAS, including perfluorooctane sulfonate (PFOS), and perfluorooctanoic acid (PFOA) has been detected in stormwater from different types of sites and drainage from a roof (Pirzadeh et al. 2015).

Polychlorinated Biphenyls (PCBs) are found in lubricants and hydraulic oils; they often enter the environment through landfills. Large sources of PCBs are created from combustion byproducts and the chemical industry (Wiberg et al. 2009). Two Norwegian studies have shown that facade paints and plaster from the period 1950–1970 are major sources of PCB in the urban environment (Andersson et al. 2004; Jartun et al. 2009). Thus, PCBs have also been detected in stormwater (Zgheib et al. 2011a, b). Table 6.1 summarises concentrations in stormwater and gives representative for measurements, which are based on different studies.

Pollutant group and analysed pollutant		Value(s)	Concentration (µg/L)	References	
Hydrocarbons	PAH (16)	Representative interval	0.2–5		
		Housing area	1.4	Gasperi et al.	
		Industrial area	1.1	(2014)	
		Commercial area	2.9	Zgheib et al. (2011b)	
Alkylphenols	Nonylphenol	Representative interval	0.1–4		
		Min-max in same catchment	1.6–9.2	Zgheib et al. (2011b)	
	Octylphenol	Housing area	0.06		
		Commercial area	0.1		
		Highway	0.32	Stachel et al. (2010)	
Phtalates	DEHP	Representative interval	1–30	Zgheib et al. (2011b)	
		Housing area	13		
		Commercial area	27		
		Highway	8.6	Stachel et al. (2010)	
Polychlorinated biphenyls	PCB (7)	Representative interval	0.2–0.5		
Organotin compounds	DBT	Representative	0.01–0.8		

Table 6.1 Concentrations of organic compounds observed in stormwater

The estimations for 'representative intervals' are based on measured mean and median concentrations from various studies, storm events and catchments

Studies on combined sewer overflows (CSOs) often focus on the emerging contaminants that derive from the wastewater component. These often include pharmaceuticals and industrial chemicals, which can also be found from very specific sources. A literature review on substances from the EU watch list, limited to those which have been directly tested in stormwater, agricultural diffuse runoff or combined sewage before discharge into the surface waters, revealed very few references. Substances from this list were found by Deffontis et al. (2013): estrone and 17 β -estradiol occurred both in dry and wet weather flow in two stormwater outlet. Diclofenac, however, was detected in three studies on CSOs in Germany in concentrations ranging from approx. 390–1500 ng/l (Tondera et al. 2013; Christoffels et al. 2014; Scheurer et al. 2015).

Gasperi et al. (2012b), Madoux-Humery et al. (2013, 2015), Launay et al. (2016) have also conducted studies specifically on CSOs with a focus on other substances. However, most studies concentrate on taking samples in the surface waters (Pailler et al. 2009; Weston et al. 2015), often upstream and downstream of the discharge location.

Pesticide transfer via agricultural runoff or drainage is often less than 0.5% of the dose applied and rarely exceeds 3% (Kladivko et al. 2001; Boithias et al. 2014). The quantities transferred are on the order of several grams of all pesticides combined per hectare and year. Generally, the first high-flow events after the substances are applied contain concentrated pesticides (Kladivko et al. 2001; Branger et al. 2009) and therefore present the highest risk of pesticide transfer. The general behaviour of pesticide transfer is restricted to the period after uses, meaning that certain flows present no risk of transfer (except for remnant persistent pesticides such as Atrazine). Pesticide concentrations clearly depend on the spatial scale. In the upstream part of the watershed, the concentrations are significantly higher (>1 μ g/L) than in the lower course area (Fig. 6.1). Since farmers use pesticides in different amounts and at different intervals, the concentrations found to vary widely, creating this large-scale dilution. Dissipating in the watershed, the concentrations decrease even if the flows remain conservative.

Selecting indicator substances as described before can also be used to select inorganic indicator parameters, e.g. metals from roof installations for runoff from separate sewer system or cadmium for road runoff.

Weyrauch et al. (2010) developed another approach for CSOs and chose substances as indicator parameters that are eliminated to a high extent in WWTPs; thus, their occurrence in surface waters can be traced back to the CSO as a source. This could also be transferred to stormwater in separate sewer systems or diffuse agricultural runoff.

Regardless of the approach chosen, the occurrence of emerging contaminants strongly depends on the rainfall patterns, the structure of the catchment area and its use, even more than for other substance groups. Most treatment facilities described in this book rely on different mechanisms although the complete removal processes are not yet well understood, and still the subject of ongoing investigations across a range of laboratories.



Fig. 6.1 Scale effect of 26 pesticides concentrations in three Irstea experimental sites in the Seine-et-Marne department, considering equivalent soil use (>80% agricultural use) (adapted from Tournebize et al. 2017)

Treatment Systems

Very little data is available on the treatment performance of the aforementioned emerging contaminants, but is summarised for each technology in this section. However, it has to be noted that these pollutants have only been investigated in relatively few studies and that their chemical characteristics cannot be easily grouped due to the sheer amount of different organic compounds. Therefore, it is more difficult to draw general conclusions, in contrast to the treatment of suspended solids, metals and nutrients.

Stormwater Ponds and Basins

Van Buren et al. (1997) evaluated the treatment performance of a stormwater pond in Canada and reported a mean removal of 24% for oils and 12% for phenols. Roseen et al. (2009) observed a high removal of total petroleum and hydrocarbons around 90% for stormwater ponds. In contrast, Andersson et al. (2012) did not observe any significant treatment of PAHs when evaluating the performance of five stormwater ponds. However, the results from the different ponds included in the study were partly contradictory; some ponds reduced PAH concentrations while for others outflow concentrations exceeded those in the inflow (DeLorenzo et al. 2012).

Tournebize (2016) explored the behaviour of pesticides in ponds and basins. In addition to their dilution effect, such water reservoirs stimulate the dissipation process of pesticides. Concentrations in dissolved and particulate phases are similar to those measured in upstream waterbodies and much higher than in downstream


Fig. 6.2 Pesticides concentration in sediment from pond and basins according to KOC values (from Tournebize 2016, based on 45 references)

waterbodies. Tournebize's review highlighted the concentration gradient between upstream and downstream sediments of the pond system and concludes that, due to their water storage function, ponds and basins contribute to downstream water quality of waterbodies.

There is no obvious relationship between pesticide persistency and the properties K_{OC} and DT_{50} measured in ponds and basins. Nevertheless, the pesticides in the sediment showed the most explicit relationship between their chemical properties and accumulation in pond sediments: a statistical evaluation showed that pesticides with a K_{OC} above 4000 ml/g were strongly bound in the pond sediments, as shown in Fig. 6.2.

Constructed Wetlands

The removal of pesticides in constructed stormwater wetlands (CSWs) has proved to be effective (Maillard and Imfeld 2014). However, the removal efficiencies vary between different storm events and during different seasons, e.g. due to temperature dependent (bio)chemical processes (Maillard et al. 2011). Terzakis et al. (2008) observed a mean removal of PAHs of 59% when evaluating a CSW in Greece.



Fig. 6.3 Processes involved in pesticide retention in a shallow artificial wetland (Passeport et al. 2011). The numbers indicate results from the Irstea experiments on S-metolachlor (Hoyos-Hernandez 2010) and exposiconazole

Similarly, Schmitt et al. (2015) and Tromp et al. (2012) reported a significant treatment of various PAHs in a combined pond-wetland system. However, a large proportion of these PAHs were particle-bound and, thus, already removed in the pond (Fig. 6.3).

The biodegradation of molecules is a slow process which profits from long retention times. The hydraulic functioning of a CW, most particularly the residence time, is a key factor in optimising biological processes. In controlled experiments, drained waters were hydraulically controlled as the inflow of an experimental CW insuring about 8 days of retention time (Blankenberg et al. 2007; Hunt et al. 2008; Braskerud and Haarsad 2003). However, other experiments in natural systems showed a stronger dependency on the background conditions such as rainfall (Passeport et al. 2013; Tournebize et al. 2013; Vallée et al. 2015). Both approaches lead to similar retention average efficiency of 32% (SD = 22%) and 39% (SD = 40%) for controlled and uncontrolled conditions, respectively.

Similarly, vegetation has direct and indirect effects on the dissipation of pesticides. By airing sediments, vegetation increases microbial activity; by creating roughness, it decreases the flows, thus increasing the hydraulic retention time of pesticides (Brix 1997), and favours particle sedimentation. Decomposing vegetation provides organic carbon to microorganisms (Moore et al. 2002). It also serves as an adsorbing surface for pesticides and can at times remove some. Developing biofilms can promote biodegradation of pesticides and help stabilise sediments (Brix 1997).

The pesticides' properties influence the removal efficiency, whereof adsorption coefficient is one of the most important factors. Splitting the removal efficiency according to K_{OC} , as shown in Fig. 6.4, revealed an average removal efficiency



Fig. 6.4 Removal efficiency of pesticides in surface-flow wetlands based on a comparison of input/output mass removal. Three categories of Koc were selected: low (<400 ml/g), moderate (>400 ml/g and <1000 ml/g) and strong (>1000 ml/g). N indicated the number of data (adapted from Tournebize et al. 2017)

of 25% (SD = 32%) for pesticides such as MCPA, bentazone, metalaxyl, isoproturon, chlortoluron, metamitrone, s-metolachlor, ethofumesate, atrazine and metazachlor. Their K_{OC} values are low. Moderate and strong K_{OC} values show higher removal potential with 49% (SD = 30%) and 51% (SD = 29%) respectively. These groups include pesticides such as boscalid, chlorothalonil, napropamide, tebuconazole, azoxystrobin, propyzamide and propiconazole, fenpropimorph, epoxiconazole, chlorpyrifos, prosulfocarbe, difflufenilcanile, aclonife and pendimethalin.

Surface-flow constructed wetlands have real potential to reduce the concentrations and flows of agricultural pollutants. However, their performance depends on hydrological conditions and seasonality. CWs should, therefore, not be considered as a permit to pollute, but rather as a complementary tool to actions implemented at the agricultural plot scale to reduce the pressure of pollution (reduction of inputs).

The vegetation in subsurface-flow CWs may also be a potential compartment for sorption processes. Sorption phenomena should be considered as a temporary phenomenon that delays the transfer of peaks of pesticide concentrations through the CWs and attenuates peaks in these concentrations. As in the soil compartment, the degradation of pesticides has multiple sources in CWs: due to the effect of light (photodegradation), water molecules (hydrolysis) and particularly microorganisms (biodegradation).

Although using CWs to remove pesticide pollution has shown very promising results, the removal rate of different pesticides varies extremely between negative values (higher output than input concentration) and a complete removal (Blankenberg et al. 2007; Maillard et al. 2011; Stehle et al. 2011; Vymazal and Březinová 2015).

The removal of other emerging contaminants in constructed wetlands is described in the section Bioretention filters for vertical-flow systems.

Bioretention Filters

The processes for the removal of emerging contaminants in bioretention filters include filtration of particle-bound substances and diverse chemical, biochemical and physical processes. Randelovic et al. (2016) describe in detail several of these processes and include modelling approaches.

The few studies investigating the treatment of pesticides in bioretention filters indicate a good potential for purification of glyphosate (removal efficiency >80%; Zhang et al. 2014). However, the purification of triazines (atrazine, prometryn, simazine) was lower ($\sim 35\%$; Zhang et al. 2015). Lefevre et al. (2012) showed a 93% purification rate for naphthalene for vegetated biofilters and 78% for non-vegetated control filters. Removal mechanisms were adsorption (56–73%), mineralisation (12–18%) and plant uptake. Even Diblasi et al. (2009), Zhang et al. (2014) observed decreased levels of PAHs (removal efficiency >80%) in biofilters.

Some results on the treatment of emerging contaminants from CSOs were published by Tondera et al. (2013), Christoffels et al. (2014), Scheurer et al. (2015) on large-scale treatment facilities in Germany. Diclofenac was removed by $73 \pm 3\%$ (Tondera et al. 2013, n = 8) and $81 \pm 21\%$ (Scheurer et al. 2015, n = 5). Christoffels et al. (2014) chose a different way to evaluate the events since diclofenac could only be quantified in 68% of 343 single samples from 33 events, but in only 9% of the outflow samples (quantification limit: 100 ng/L). Hence, the maximum concentration of diclofenac in the outflow was 65% lower than that in the inflow. Other substances investigated in these studies were other pharmaceuticals, e.g. carbamazepine which could not be removed sufficiently, and industrial chemicals such as bisphenol A.

In a laboratory scale study, Janzen et al. (2009) simulated the CSO matrix with spiked artificial wastewater. The filter consisted of three layers, each with a height of 0.185 m: a reed planted peat layer on top, a sand layer (0/2) and a gravel layer (2/8). The micropollutants tested, e.g. antioxidants, plasticizers as well as chemical UV filters, showed reduction rates from 80 to 90%, with the exception of N-butylbenzene-sulfonamide (NBBS), which had an elimination rate of only 20%. When identifying the most relevant elimination mechanism for four of the seven investigated compounds, the authors assumed them to be biological as well as chemical degradation processes. In both the sand and drainage layer, Janzen et al. (2009) evaluated additional elimination processes, most probably sorption on the biomass in the layers. In total, sand performed better than gravel.

The ecological benefit of using bioretention filters for the removal of emerging contaminants was investigated by McIntyre et al. (2015): the toxicity of highway runoff on several aquatic species including juvenile salmon was tested before and after treatment with mesocosm bioretention filters. The treated stormwater had a

less harmful effect on the aquatic species than the untreated one. However, long-term experience with the removal of pollutants in urban runoff through biofilters is still lacking and the removal will be influenced by various factors as described above.

Swales and Buffer Strips

There are studies on pesticide and PAH removal in swales and buffer strips (e.g. Andersson Wikström et al. 2015). However, the results vary extremely from depending on whether single compounds or groups of pesticides were investigated. The removal of PAHs also fluctuated between different treatment sites. As for nutrient concentrations (see Chap. 3), common swales for stormwater management cannot be regarded as a complete treatment system for emerging contaminants and have to be complemented with other facilities if quality treatment is targeted.

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Chapter 7 Modelling Under Varying Flows

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Abstract Constructed wetlands (CWs) subjected to variable loads present a series of challenges for designers and researchers. Classical design approaches (e.g. rule of thumbs or first-order kinetic model) are not suited to properly estimate the removal efficiencies of CWs under varying flows. The internal removal processes of CWs are expected to be influenced by the variation of influent pollutant concentrations and hydraulic loads for particular CW applications (e.g. stormwater or combined sewer overflow treatment). A powerful tool to properly study and design CWs under varying flows is given by mathematical model. Either for design or research purposes, mathematical models have been developed to simulate CWs subjected to varying flow and are revised in this chapter. Models used to simulate the hydraulic behaviour as well as the treatment performances of variable-flow CWs are reviewed. Moreover, future perspectives of mathematical models in this field are analysed in terms of design-support tools, process-based model for design purposes, and limitation for a wider application.

Modelling of Treatment Systems

Variable flows pose one of the most complex challenges for modelling of constructed wetlands (CWs). The treatment of stormwater and combined sewer overflow (CSO) is highly dependent on the stochastic variability of rain events, thus compromising the reliability of conventional design approaches, such as rules of thumb or first-order kinetics. Moreover, the internal removal processes suffer from varying influent pollutant loads, which can lead to sporadic failures in treatment performance (Galvão and Matos 2012). For these reasons, wetland scientists

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developed specialised mathematical models that provide novel tools to assess CWs performance under varying flows and support design for such conditions. The hydraulic functioning of variable-flow systems differs fundamentally from that of CWs receiving consistent flows. Therefore, we describe the specific hydraulic modelling tools developed for them separately from the treatment models. Finally, we provide a critical review of available modelling tools for variable-flow CWs and suggest future research directions.

Modelling Hydraulics of Variable-Flow Constructed Wetlands

To treat water from stochastic rain and flow events, treatment systems have been developed with high accumulation volumes (either in a single stage above the CW bed or in separate free water surface/accumulation pond wetlands) and with restricted orifices to throttle the outflow. This way, extended residence times are achieved, thus promoting effective wastewater treatment. Consequently, these systems saturate during rain events and face unsaturated conditions in the subsequent emptying phase, showing a complex hydraulic functioning. Mathematical models of the hydraulics of these systems have been developed to both support design and investigate their internal hydraulic functioning.

Meyer and Dittmer (2015) developed the numerical model RSF_Sim to support the design of retention soil filters (RSFs), which are widespread in Germany for CSO treatment. Inspired by RSF_Sim, Pálfy et al. (2015a) provided a modelling toolkit called Orage, to simulate and scale French CSO-CWs, which receive non-settled inflows. The hydraulics of these models are discussed together below, highlighting their similarities and differences. Both models use continuously stirred tank reactors in series (TIS) to simulate flow dynamics. In RSF_Sim, the wetland is divided into three conceptual tanks in a single series (TIS) (Fig. 7.1—left). These are from top to bottom:

- retention tank (surface water).
- process layer (subsurface water) and
- drainage layer (subsurface water).

Hydraulics can be simplified to such a degree because an outflow-limiting orifice is used, which is represented in the model by an outflow rate constant. As such, both tools assume a uniform vertical flow through the reactive media.

Orage can model both the German and the French CSO systems since it can simulate a single operating filter (analogous to the German approach) and twin filters (French approach) as well. To simulate twin-sided filters, model space is divided into seven discrete tanks, as the three compartments (TIS) of RSF_Sim are doubled and a storage basin (surface water) common to both sides is added (Fig. 7.1—right). In reality, there is an impermeable wall with a single



Fig. 7.1 TIS in RSF_Sim [left, based on Meyer and Dittmer (2015)] and twin TIS with common basin and fixed level cross-connection between the retention tanks in Orage [right, based on Pálfy et al. (2015a)]. The drainage layer in Orage is permanently saturated

cross-connection point so that the two filters are separated in the model accordingly. Calibration with long-term time series of inflow and outflow records showed that fitting the hydraulic model with tracer tests was unnecessary, enabling us to simplify the modelling significantly and to make practical applications easy.

Fournel et al. (2013) used an existing state-of-the-art finite element model (Hydrus-1D) and introduced new features to deal with specificities of CWs for stormwater management. An additional conceptual layer at the outlet of the wetland was added to mimic the local head loss resulting from the flow-limiting orifice. This layer always remains saturated and only its saturated conductivity needs to be calibrated. Through calibration against the outflow data, the hydrodynamic parameters for the filter materials can be obtained by inverse modelling. However, since the outflow is not linearly proportional to the hydraulic head above (because it is simulated by the saturated layer), when large variations are observed in the pressure head (Fig. 7.2), the model has to be recalibrated. Another conceptual layer with high permeability and a 100% porosity was set at the top of the model space to account for water storage above the filter. This provides a simple means to simulate 1D ponding.

Preferential flows which are rapid and gravity-driven in the largest pores of the media are difficult to model. Maier et al. (2009) suggested using a bimodal hydraulic conductivity function to account for flow in large pores at a high saturation level. Another method to account for these preferential flows is suggested by Morvannou et al. (2013) and uses a dual-porosity model. The authors successfully applied it to model premature tracer breakthrough in CW using HYDRUS-2D (Fig. 7.3).

Rizzo et al. (2015) developed a mathematical model to highlight the function of CSO-CWs as bioretention systems capable of re-naturalising the hydrograph of receiving streams (Fletcher et al. 2013; Walsh et al. 2005). This model shows that a CSO-CW not only treats polluted CSO, but also flattens the hydrograph at the outlet towards a pre-development state (low peak and extended duration). The



Fig. 7.2 Calibration and validation of the model from Fournel et al. (2013). (i) and (ii) depict the results obtained and experimental values of the calibration event. (iii) and (iv) show the results obtained on the validation event with and without adjusting the virtual layer K_s value



Fig. 7.3 Outflow concentration and cumulative mass recovery of a tracer test on a French first-stage VFCW simulation, using both equilibrium (default) and a dual-porosity model (Morvannou et al. 2013)



mathematical model was used to simulate a full-scale CSO-CW (Masi et al. 2016) situated in Gorla Maggiore, Italy (45° 40' N, 8° 53'E). This CSO-CW had two stages: a vertical-flow constructed wetland (VFCW) followed by a free water surface (FWS) system. The VFCW and FWS are both equipped with throttling orifices to increase residence times. CSO volumes can be accumulated both above the filter bed of the VFCW and within the FWS. The mathematical model is mono-dimensional and simulates the unsaturated water flow in VF beds (Richard's equation), the accumulated water heights of the ponding layer above the VFCW and within the FWS (mass balance equations). Proper hydraulic equations are included to simulate the VFCW effluent regulated by throttle orifices and the VFCW and FWS overflows. This enables simulation of outflows from each stage of the CW through consecutive events (e.g. multiple average CSO events, Fig. 7.4).

Modelling Treatment Performance of Variable-Flow Constructed Wetlands

Estimating treatment performance of variable-flow wetlands is a challenging task due to the effect inflow stochasticity has on removal efficiencies. Different modelling tools have been developed with the following aims: (i) design-oriented models, (ii) first-order kinetic models to investigate temporal variability in removal efficiencies with a tool of limited complexity and (iii) process-based models to gain deeper insights into internal and variable treatment processes. In the following, we give an overview of some available tools.

Design-Oriented Models for CSO-CWs

RSF_Sim and Orage share similar core models and have been developed to provide a support tool for designers dealing with variable-flow CWs. As described earlier, hydraulic is represented in both models by means of continuously stirred tank reactors. Removal processes are modelled in a single tank which represents the so-called process layer, while the other tanks are used only to simulate hydraulic flows and storage. The modelled pollutants are total suspended solids (TSS), chemical oxygen demand (COD) and NH₄–N. RSF_Sim simulates the dissolved and particulate COD fractions separately, while Orage simulates total COD without fractionation.

Both models assume a constant background concentration of TSS independent of the inflow. COD removal depends on the length of the previous dry period in both models. Additionally, Orage accounts for climatic and seasonal influences by adjusting the dry period's length to a relative value via predefined multipliers (e.g. one month drought will have a stronger impact under Mediterranean summer than under Oceanic summer and winter, and so on). Sorption processes simulated by Orage are based on the same principles as in RSF_Sim. However, Orage accounts for hydraulic shortcuts at the beginning of events, when the influent percolates through a limited zone near the inlet (Pálfy et al. 2015a). As such, in Orage, performance parameters of adsorption and COD removal are selected from internal tables according to the predicted operation mode (shortcutting or plug-flow) at the actual time step; furthermore, in the case of shortcutting, the dynamic mass of water-contacted media is estimated based on the infiltrating volume and Darcy's law. This approach permits modelling shortcutting effects within the framework of a TIS hydraulic model.

Both RSF_Sim and the core model of Orage have proved to be robust and able to predict outflow concentrations that compare well with measured data (Meyer and Dittmer 2015; Pálfy et al. 2015a; Tondera et al. 2013). The intra-event adsorption and inter-event nitrification equations closely reproduce the natural dynamics of NH_4 –N, even at the event level, with concentration breakthrough well-predicted by the two-stage isotherm (Fig. 7.5).



Fig. 7.5 Measurements and simulated series of two consecutive events with extreme NH_4-N loads in RSF_Sim (based on Meyer and Dittmer 2015). Note how the saturation of adsorption sites led to breakthrough and how capacities were regenerated both in reality and in the model during the inter-event (not charted) which separated the two events

First-Order Kinetic Model

Precipitation events generate not only urban runoff, but can also set other non-point sources of pollution in motion. Kadlec (2010) developed a model of pulse-fed FWS systems, targeting NO_3 –N. Non-point source agricultural runoff tends to carry considerable concentrations of this pollutant, which is readily leached from soils by infiltrating rainfall. The stochasticity of loadings impairs basing the analysis and prediction of system behaviour on detention time, hydraulic load or areal pollutant load, as usually done for CWs fed by steady influent loads. For example, event mass removal ranged from 17 to 100% at the full-scale sites of experimentation. However, a dynamic mass balance model was developed which considers the stochasticity of influent load. The model processed and returned time series of flows and concentrations from which mass exports were able to be computed accurately. This approach was applied by Kadlec (2012) and Tanner and Kadlec (2013).

Hydraulics was simulated by TIS reactors and fitted to tracer curves. Each tank had fixed and equal stages. Depending on the residence time, the ideal number of TIS was found between 1.7 and 9 (Tanner and Kadlec 2013). The mass transport was calculated between tanks, and nitrate losses were modelled using first-order areal removal.

Parameter calibration was based on mass-flow fitting instead of concentrations in order to omit biasing towards inter-event periods, which do not have flows. Removal rate was adjusted to seasonal temperatures assuming a sinusoidal pattern, and a modified Arrhenius equation.

The model provides insight into why intra-event treatment effects might sometimes be negligible. Displacing antecedent water is the key factor for events involving low-flow volumes compared to the available storage. In these cases, the effluent nitrate–N concentrations are reduced to low levels due to the extended detention during the preceding inter-event and not because of high removal rate coefficients. The bias is revealed by the dynamic mass balances. Changes in the stored mass and the reacted mass can be quantified because the hydraulic model is fitted to tracer tests. The degradation of stored nitrate in the batch-like inter-event is accounted for separately, following the loadings.

Tanner and Kadlec (2013) used the same approach, considering rain, evapotranspiration and infiltration losses. The model was successfully calibrated to simulate nitrate removal in an FWS wetland (Fig. 7.6) at Toenepi Stream, Waikato, New Zealand. Simulations predicted a better areal removal rate if wetlands are installed at locations where the flow variability is less pronounced. The predicted rate declined markedly when the area of the wetland was enlarged by 0.5-5% of the 2.6 ha catchment area. Furthermore, reversing the seasonal trend of temperatures to cause the high variability flows to coincide with warmer temperatures led to a moderate improvement in NO₃–N load reduction.



Fig. 7.6 Modelled and measured NO₃–N concentrations in the inflow (black) and the outflow (grey) of a wetland receiving subsurface tile drainage (area 293 m², catchment 2.6 ha): **a** concentrations and **b** mass. (based on Tanner and Kadlec 2013)

Carbon availability and oxygen inhibition were not found to be limiting NO_3 –N removal in the studied wetlands and the influence of other N transformations was minor. However, for other scenarios, the inclusion of these factors in the model might become necessary.

Analogous first-order dynamic models have also been used to predict phosphorus removal rate in event-driven wetlands in the USA treating pumped inflows of water from the Des Plaines River (Kadlec and Reddy 2001) and stormwater treatment areas protecting the Everglades (Walker and Kadlec 2011). Fig. 7.7 Measured and simulated COD (above) and NH₄–N (below) outflow in CSO-CW columns, using HYDRUS/CW2D (based on Pálfy et al. 2015b). The five consecutive loadings (E1-E5) were fed on simulation domains which had a quasi-stable biomass at the beginning of the simulation (simulations covered inter-events, but these are concentrations with no actual outflow)



Process-Based Model

HYDRUS is a model package that simulates variably saturated flow, heat and solute transport in porous media. The software numerically solves Richard's equation and the convection-dispersion equation for heat and solute transport (Šimůnek et al. 2011). Thanks to the wetland module extension (Langergraber and Šimůnek 2012), it can perform process-based modelling of CWs via two different biokinetic models. Of these, CW2D (Langergraber and Šimůnek 2005) was applied on event-driven VFCWs treating combined sewer overflow (CSO-CWs). Although CW2D simulates 12 different components (C, N and P cycles), the key water quality constituents targeted by CSO-CWs and represented in the biokinetic model are COD and NH_4 –N. Process kinetics and stoichiometry are structured much like in activated sludge models (ASM, Henze et al. 2000).

HYDRUS/CW2D was originally designated to simulate continuous or frequent feeding patterns, so issues may arrive when modelling a long dry period followed by a sudden strong inflow. Nonetheless, the tool has all the sub-models necessary to describe internal processes except particulate transport (filtration). Modelling research has progressively described and extended the limits of CSO-CW application (Henrichs et al. 2007, 2009; Meyer 2011; Meyer et al. 2013). The latest work of Pálfy et al. (2015b) achieved a quasi-stable biomass after self-inoculating runs, and a good fit to a series of lab-scale column experiments for both COD and NH_4 –N (Fig. 7.7).

Future Perspectives

Novel Approaches for Design of Variable-Flow Constructed Wetlands

As well as modelling the hydraulic and treatment performance of variable-flow CWs, the Orage toolkit (Pálfy et al. 2015a) provides a range of additional user-friendly tools to support design and implementation. A simplified user-friendly interface has been developed, thus broadening the number of potential users (e.g. designers, stakeholders). There are fewer user-defined parameters, therefore hand-ling is less complex. An autonomous iterative optimisation tool enables the user to identify the smallest filter area able to satisfy the appropriate legislative limits (entered by the user). In the future, these tools could be extended by including the relevant costs of land, filter media etc. to enable economic optimization.

Process-Based Models as Design Tools

Current process-based models are able to simulate the treatment performance of variable-flow CWs. HYDRUS/CWM1 and BIO PORE have been validated for horizontal-flow constructed wetlands (HFCWs) subjected to time-variable loads on experimental pilot data (Rizzo et al. 2014; Samsó and Garcia 2013) and have been used to gain more insights on HFCW subjected to sudden loads (e.g. Rizzo and Langergraber 2016). As shown in the previous subsection, attempts have been made to model CSOs with HYDRUS/CW2D (Henrichs et al. 2007, 2009; Meyer 2011; Meyer et al. 2013), with recent work by Pálfy et al. (2015a) providing marked improvements. However, the time-consuming calibration process of flow, transport and biokinetic parameters demand an in-depth understanding of the model formulation, which can be a drawback for the application of such process-based models for CW design. This has led to claims that process-based models are ill-suited for design-support purposes (Meyer et al. 2015). Another barrier is that model accuracy needs to be proven at full-scale and related to the design standards of each country. For example, systems in France receive unsettled CSO flows and use coarse media, whilst systems in Germany receive pre-settled CSO flows and use fine media. In order to open the way for design-support modelling of CSO-CWs using process-based models, future efforts should be dedicated to (i) combining simulation experience from a representative range of CSO-CW sites and (ii) fully calibrating and validating the process-based models at full-scale, including in-depth sensitivity analysis of each of the parameters currently required in the models.

Despite these issues, process-based models remain highly attractive for future designers, because they provide a comprehensive understanding of system functionality. Recently, two CSO-CW treatment plants have been designed exploiting the potential of the hydraulic model proposed by Rizzo et al. (2015), which simulates both the unsaturated (Richard's equation) and the ponded water accumulation (mass balance equation) of the VF-FWS scheme adopted by the designers. These two examples, based on systems designed by the Italian engineering company IRIDRA, are briefly analysed below to highlight the utility of process-based models (modelling only hydraulics in this case) during the design phases.

Example 1: CSO-CW proposed for Capiago Intimiano (Italy)

This WWTP (45° 46'N, 9° 07'E) treats the first flush of CSO from a 5.2 ha urban area (450 person equivalent) in accordance with regulations from Lombardy, Italy. It comprises a two-stage gravity-flow system (see Fig. 7.8), where the first stage is a VFCW bed (300 m²). Following the French approach (Morvannou et al. 2013), the VF bed is filled with a highly permeable media (gravel) to treat the first flush. The second stage is a surface-flow (FWS—350 m²) system that provides tertiary treatment for the first flush and secondary treatment for the second flush. A throttled orifice has been installed in both stages to regulate the discharge, providing sufficient retention time to achieve adequate treatment.



Fig. 7.8 CSO-CW system designed for Capiago Intimiano (Italy), with inflows entering from the top and outflows from the bottom left of the diagram

The FWS CW works as a detention basin with a maximum surface area of 650 m^2 , reducing the hydraulic peaks in the discharge to the receiving stream and the risks of downstream flood events. This promotes sustainable urban drainage by switching back the post-development (high peak and short duration) release pattern to a pre-development (low peak and high duration) hydrograph (Fletcher et al. 2013). Although the FWS stage is the main contributor of the flood reduction, the VF bed provides additional detention volume both in the porous media and in the available ponded volume above the surface. The hydraulic functioning of the system is complex due to the multifaceted flow and storage state in operation. The detention effect due to unsaturated flows in the VFCW is relevant only at the beginning of the load and during the last emptying phase. Saturated flows from ponding above the VFCW and in the FWS are dominant, and this is when the limiting orifice provides an important function. Because VFCW could overflow into the FWS storage area, accurate estimation of detention and peak reduction capabilities was critical during the design phase. The mathematical hydraulic model proposed by Rizzo et al. (2015) (described previously) was used to simulate the potential detention capacity of the system over a range of CSO hydrographs, from rain events of 5 and 10 mm per h and with return time of 10 years (estimated with a hydrological runoff model). The results illustrated in Fig. 7.9 show the strong capacity of the CSO-CW system to attenuate the influent peaks for all influent CSO hydrographs. Furthermore, the effluent discharges are three to four times longer than those of the untreated CSOs; the volumes held in the different stages of the system during the CSO event are equal to 87, 75 and 57% of the total CSO volume, while the peaks are reduced by 84, 60 and 36% (results for rain events of 5, 10 mm per h and with return time of 10 years, respectively). Moreover, the contribution of the VF stage is substantial for 5 mm per h rain event (47.4% of the total held volume), relevant for 10 mm per h rain event (20.8%) and only negligible for high return times (5.6% for rain event with return time of 10 years).

These modelling results have been used to quantitatively demonstrate the hydraulic detention capacity of the proposed system to the public authorities and stakeholders, helping to convince them that the CSO treatment with CWs is an adequate solution to also provide the additional ecosystem service of flood risk mitigation.

Example 2: CSO-CW upstream of the Carimate WWTP (Italy)

The proposed treatment plant to be built in Carimate ($45^{\circ} 42'$ N, $9^{\circ} 07'$ E) has been designed to treat the first flush and part of the second flush of a CSO upstream of a centralised WWTP that treats the wastewater of 11 towns in Como province (70,040 inhabitants). Again, the CSO-CW is a two-stage system where the first stage comprises two VF beds [8500 m² of total area) designed according to the French approach (highly permeable filter media with gravel (Morvannou et al. 2013)]. The second stage is an FWS CW (4500 m²). The system is fed by a pumping system, with throttle valves installed in both VFCW and FWS stages to regulate the effluent discharge and provide sufficient retention time to promote CSO treatment. The VFCW overflow is discharged to the FWS.



Fig. 7.9 Modelling results showing the detention effect of sequential stages of the CSO-CW system designed for Capiago Intimiano (Italy) for different influent CSO hydrographs

CSOs occur frequently upstream of the WWTP, with an almost constant discharge rate, sometimes lasting for several days even during dry weather (on average 75 CSO events per year with up to 10 consecutive CSO days, data from 2012 to 2014). This is due to infiltration into the sewer network (e.g. excess water infiltrating at joints, ruptures etc.). In order to reduce the impact of CSO pollutant loads to the receiving water body (Seveso River), the CW has been designed to treat the first flush of the CSO event (defined as up to 1300 m³/h for a maximum of 7 hours per day, i.e. a maximum of 9000 m³/d). A modelling approach has been adopted to design the effluent discharge regulated by the throttle valve to (i) verify the absence of VFCW overflow for the design CSO event and (ii) to illustrate the internal hydraulic mechanism of the system to the public authorities and to the stakeholders.

The mathematical hydraulic model proposed by Rizzo et al. (2015) (described previously) was used to simulate the response of the initial VFCW stage to the designed CSO event. The results are shown in Fig. 7.10. The simulation confirms that the VFCW overflow is not activated for the chosen throttle valve design. Thus, the entire designed CSO volume is treated by the VF stage, prolonging the CSO effluent hydrograph (Fig. 7.10a). The VF beds fill quickly in almost two hours (Fig. 7.10c), with first top-down then bottom-up filling of the porous media. Subsequently, the CSO volumes start to accumulate on the top of the VF beds, up to the maximum level of 40 cm (Fig. 7.10b). At the end of the design CSO event, the VF bed is slowly emptied, moderated by the throttle valve (first ponded water and subsequently the VFCW porous media: Fig. 7.10c, d, respectively), providing



Fig. 7.10 Modelling results describing the hydraulic functioning of the CSO-CW system designed for Carimate WWTP (Italy): influent versus effluent discharges from the VFCW (**a**), ponded water height above the VFCW (**b**), VFCW filling (**c**) and emptying (**d**) phases

sufficient residence times for effective pollutant removal. Note that the average effluent discharge during the design CSO event is only 29 l/s, significantly lower than the maximum allowed effluent discharge, which is reached only when the VF beds become completely ponded.

These results show that a modelling approach can enable more precise design of CSO-CWs. For instance, in case a more simplified assumption is made of the effluent VFCW discharge—that it is equal to the maximum effluent discharge driven by the throttle valve—the proposed VFCW area would need to be substantially overestimated to guarantee the same residence times. Moreover, as for Example 1, the results of the simulation model have helped explain how the system works to the public authorities and stakeholders, fostering trust in the efficacy of the CSO-CW solutions being proposed.

Limitations of the Wider Application of Models for Variable-Flow Constructed Wetlands

Although mathematical CW models of variable wastewater flow treatment have reached a high complexity for both hydraulic and treatment processes (see previous subsections), further efforts are still needed to provide more complete tools with wider application for research, development and design. The coupled modelling of surface and subsurface flows is particularly important. Water often arrives at one side of VFCWs and progressively floods them until ponding occurs. These transition periods are essential for determining potential adsorption/leaching and identifying hydraulic shortcuts. There is currently no model in the field of CW capable of such coupling.

Clogging is the main process compromising CW durability due to the accumulation of both inert organic matter and biofilm (Kadlec and Wallace 2009). Recent mathematical models for the simulation of clogging processes have been proposed in the literature (Samsó and Garcia 2013, 2014; Rajabzadeh et al. 2015; Samsó et al. 2016). These models require validation and calibration for variable-flow CWs. Moreover, the long-term resilience of CW to variable flows also needs to be examined in more detail. From a design point of view, mathematical models that are able to accurately simulate clogging processes would allow designers to predict the durability of CWs treating variable flows, to optimise these aspects of design and account for them in economic assessments and infrastructure planning. The conceptual models and empirical equations proposed so far-to account for hydraulic permeability reduction due to clogging-focus primarily on biomass growth. Clogging in CW systems receiving stormwaters is more likely to be the consequence of suspended solids settling and filtration. More knowledge is required on the mechanisms of suspended solids deposition and degradation and its effect on hydrodynamic properties.

The so-called emerging pollutants (e.g. metals, pesticides) are also starting to be monitored for variable flows such as CSO and agricultural runoff because they have a high potential impact on receiving water bodies. Some mathematical models have been proposed in the literature for the simulation of emerging pollutants in wetlands or WWTPs, for example, associated with the iron cycle in paddy fields (e.g. Rizzo et al. 2013) or micropollutants in biological wastewater treatments (e.g. Pomiès et al. 2013). However, to our knowledge, these mathematical models have still neither been included nor validated/calibrated for application in systems receiving highly variable inflows. The coupling of current CW models (such as HYDRUS or BIO PORE) with emerging pollutant models would provide a potentially powerful tool for future CW research, development and design. From a research and development point of view, such models would allow the researcher to better investigate the relative contribution of different removal processes (e.g. adsorption, biokinetic removal, plant uptake), allowing development of new, optimized solutions. A modelling approach during the design phase would make it possible to estimate emerging pollutant removal.

Intensifying CWs by active aeration is a promising option to treat variable-flow wastewaters (Wu et al. 2014). An example for these systems is the 4800 m² CSO-CW treatment plant proposed by IRIDRA for Merone, Italy ($45^{\circ} 47'N, 9^{\circ} 15'E$). This system would treat the first flush and part of the second flush of a CSO upstream of a centralised WWTP servicing 38 towns in the Como province (120,000 inhabitants—4800 m² requested area). The advances of this active aeration approach are the reduced bed area and the ability to actively manage removal mechanisms through control of air inflow rates. Aerated CWs are currently

designed with oxygen mass balance calculations empirically adapted to this particular application. To our knowledge, process-based models for aerated CWs have not yet been developed. This is a significant lack within the scientific community and also within the field of variable-flow wastewater treatment with CWs. Indeed, a process-based model simulating the internal processes occurring in aerated wetland removal processes would allow improved investigation, optimization and design of aerated systems treating variable flows.

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