

Metal accumulation and hydraulic performance of bioretention systems after long-term operation

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Abstract

Purpose Stormwater bioretention systems are widely used to treat diffuse infiltration of runoff from paved surfaces and roofs. Substantial questions remain about the hydraulic performance and the accumulation of pollutants in systems over the long term. Data of metal accumulation of systems with operational times >10 years currently is limited. This study deals with the accumulation of metals in a variety of long-term operational bioretention systems (11–22 years) to derive further operation recommendations for the water authorities. **Materials and methods** The hydraulic conductivity of the bioretention systems in field was measured using a double ring infiltrometer. Media soil samples from 22 diverse designed systems were collected across the surface and at intervals up to a depth of 65 cm to determine the spatial accumulation of Zn, Cu, Pb and Cd. Leaching experiments of selected bioretention media soils were derived to assess the metal leachability by water. **Results and discussion** The hydraulic performance of most bioretention systems still met the technical guidelines of Germany even after long-term operation. Considerable metal accumulation occurred in the topsoil (0–20 cm). Median concentrations of all metals are highest at the soil surface (0–10 cm), decreasing with increasing depth. High concentrations

were determined at the inflow points of the runoff waters, whereas concentrations at more than 1.5 m distance from the inflow were only slightly increased compared to the initial soil concentrations. Leachability tests have shown that most of the metals deposited in bioretention soils are only slightly water soluble. No concentrations exceeding the threshold values of the German Soil Contamination Ordinance for the pathway soil to groundwater could be determined.

Conclusions The hydraulic conductivity of the bioretention systems is given even well after long-term operation. Most of the metal accumulation is concentrated in the top 20 cm; concentrations decrease rapidly and mostly reach background/initial concentrations after depths of 30 cm. The water-soluble metals are all below the trigger values of the German Soil Act. This underlines the strong retention capacity of long-term bioretention systems after long-term operational times.

Keywords 1:10 elution · Bioretention · Hydraulic performance · Leaching potential · Long-term operation time · Metal accumulation

1 Introduction

Due to rapid urbanization, sealed areas for infrastructural, residential and commercial purposes are considered to increase worldwide (García et al. 2014; Liu et al. 2014). This in turn leads to reduced rainwater infiltration and evaporation and as a consequence, to larger storm water volumes and runoff peaks in urban areas (Davis et al. 2003; Fletcher et al. 2013). Stormwater runoff often contains high levels of contaminants from road and building materials, vehicle components and traffic activity or dog faeces (Folkesson et al. 2009; Parker et al. 2010). The most recognized and examined contaminants in stormwater runoff are suspended solids, heavy metals,

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PAHs, microbial contaminants and nutrients such as nitrogen and phosphorus (Blanchard et al. 2001; Göbel et al. 2007; Ingvertsen et al. 2012a; Kayhanian et al. 2012; Li et al. 2014).

To reduce the increasing amount of runoff water and minimize the contamination of receiving water, water authorities in many countries all over the world encourage structural stormwater treatment techniques and processes, often as part of their “best management practices (BMPs)” (Hunt et al. 2006; Marsalek et al. 2006; Prince George’s County 2007; Lim et al. 2015). The diffuse infiltration of runoff from sealed surfaces or roofs into bioretention systems (also known as bioretention cells, areas; raingardens; biofilters; bioswales) is an important component of the BMPs practice and is common in commercial and residential areas (Davis et al. 2006; Dietz 2007; Boivin et al. 2008; Piguet et al. 2008).

In all these systems, the soil serves as a natural filter to reduce runoff quantity via water retention and evapotranspiration and to promote pollutant retention through physical and chemical retardation. They are often constructed as shallow depressed sites to pool and infiltrate stormwater. The design and construction differs widely; however, they are generally constructed using several layers of soil media (Lundy et al. 2012; LeFevre et al. 2014). The design often includes a top soil filter layer of 10–20 cm thickness with an adequate organic matter content that provides precipitation water, root space and nutrients for the vegetation and soil microorganisms which positively supports mechanical, chemical and biological pollutant retardation and removal (Davis et al. 2001; Roy-Poirier et al. 2010; LeFevre et al. 2014). At the bottom of the system, a high permeability layer improves the drainage of the infiltrated runoff water. Between these two layers, a layer with intermediate grain sizes is commonly found which limits the migration of fine soil particles to the bottom layer (Lundy et al. 2012).

In some cases, underdrain structures are installed to meliorate underlying native soil with low permeability (Roy-Poirier et al. 2010). To avoid erosion processes of the systems, they are often covered with grass, small plants, shrubs, or trees.

It is essential for the performance of the bioretention systems that the soils offer high infiltration rates while at the same time providing sufficient retardation time for the runoff waters. International guideline recommendations for the hydraulic conductivity (K) of the soil vary considerably, ranging from $3.5e-06$ m/s in the USA/New Zealand, to $1.4e-05$ and $5.5e-05$ m/s in Australia to $e-04$ and $e-06$ m/s in Austria/Germany (Le Coustumer et al. 2009; DWA-A 138 2005). Lindsey et al. (1992) and Le Coustumer et al. (2009, 2012) who investigated the hydraulic performance of biofiltration systems after long-term use, reported that after a few years of operation, more than 40 % had a lower hydraulic conductivity than that recommend in the guidelines. Li and Davis (2008a) determined a decrease of hydraulic conductivity of two soil filter media in column experiments from $1.5e-02$ and $2.e-02$ to less than $2.8e-05$ m/s due to particle clogging.

Several studies were performed to analyse the efficiency of pollutant reduction of bioretention systems under field and laboratory conditions (e.g. Davis et al. 2006; Ingvertsen et al. 2012a, 2012b; Søberg et al. 2014). In most cases, bioretention systems reduced the mass of a variety of pollutants in the outflow by a very high degree (Kabir et al. 2014). Hunt et al. (2006) found that removal of the incoming metals Zn and Cu was more than 98 % and for Pb approximately 80 %. Li and Davis (2009) determined median reduction rates of between 92–99, 65–96 and 83–100 % for Zn, Cu and Pb respectively, in two bioretention cells over a period of 15 storm events. Zhang et al. (2014) determined high removal rates for TPHs, glyphosate, DBP, DEHP, pyrene and naphthalene (>80 % load reduction) at a biofilter system in Australia.

For more than 20 years, bioretention systems (areas or swales) have represented the state-of-the-art for the management of surface and roof runoff for urban areas in Germany. Nevertheless, the planning guidelines in Germany only deal with these systems during the construction and installation phase; substantial questions remain about the long-term performance and contamination status of these systems.

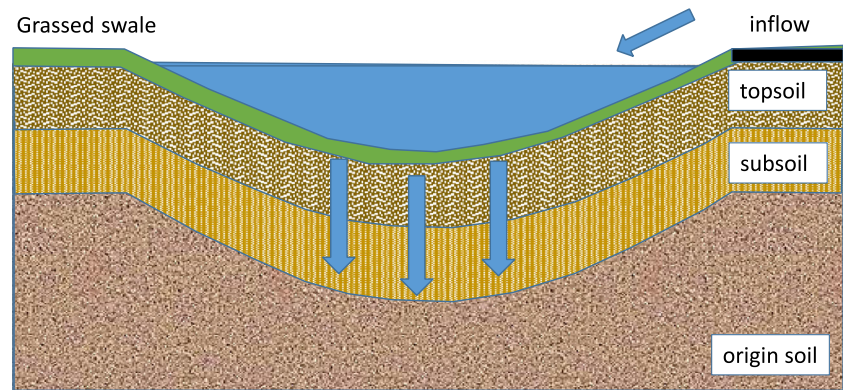
The issues of particular importance are the potential for clogging and their long-term pollutant removal capabilities (Le Coustumer et al. 2012). Some studies can be found concerning the accumulation of metals over operational periods from 2 to 10 years (Dietz and Clausen 2006; Achleitner et al. 2007; Li and Davis 2008b; Jones and Davis 2013; Lucke and Nichols 2015). To our knowledge, bioretention metal accumulation data are currently limited and only two studies deal with the pollutant status of bioretention systems over operational times >10 years (Ingvertsen et al. 2012a; Paus et al. 2014). Thus, the aim of this study was to determine the spatial accumulation and leaching potential of metals (Zn, Cu, Pb, Cd) in a variety of long-term operational bioretention systems (11–22 years) to derive further operation recommendations for the water authorities.

2 Material and methods

2.1 Study sites

The investigated bioretention systems are located in urban areas of different cities in Germany. The systems are bioswales with or without underlying underdrains (Fig. 1). The systems treat stormwater from parking lots, sidewalks, residential roads, or roofs and are constructed according to the technical guidelines of the German Association for Water, Wastewater and Waste (DWA-A 138 2005). Table 1 summarizes the requirements for the construction of bioretention swales in Germany, whereas Table 2 shows a classified register of the systems recording the use of the surrounding area, drainage area type and years of operation.

Fig. 1 Schematic diagram of a typical German bioretention swale without an underdrain according to DWA-A 138 (2005)



2.2 Sampling and data analysis

Soil samples were collected from low points at the inflow zones of each site and from a second point at 1.5 m distance from the inflow. A stainless steel shovel was used to dig up a profile (70 × 70 cm). The depth of the profile depended on the bioswale base, limited by the installation of a geotextile, an underdrain or the origin soil. Therefore, the sampling depth varied as well as the bioretention soil thicknesses across the sampled sites. In general, samples of depths 0–10, 10–20, 20–30 cm were collected; for some systems as well, the soil depth was 30–65 cm. Composite soil samples of the prescribed depths were taken to a laboratory for chemical analysis. Before analysis, all soil samples were air dried and sieved through <2 mm.

In order to verify the influence of land use and drainage area type on metal accumulation and distribution of the sampled bioretention systems, the data were classified according to the following runoff categories: (a) commercial, (b) main roads, (c) roofs (=roofs with zinc gutters and downpipes), (d) parking lots and (e) residential roads.

2.3 Measurements of hydraulic conductivities

The hydraulic conductivity of the bioretention systems in field was determined using a double ring infiltrometer according to DIN 19682-7 (2015)-08. In total, 48 measurements were conducted.

Table 1 Requirements for the construction of bioretention swales in Germany (DWA-A 138 2005)

Construction height	10 cm topsoil/20 cm subsoil
pH	6–8
Hydraulic conductivity (Kf)	10^{-4} to 10^{-5} m/s
Soil texture	fine and medium sand (<10 M.% clay/silt)
Organic matter (top soil)	1–3 M.%
Max. initial pollutant loads of the built-in soils	≤Z0 soil according to LAGA (2003)

2.4 S4 elution (batch S4) of system no. 9

The metal leachability by water was determined according to DIN 38414-4 (1984). This method is well established and widely used in Germany and other countries (Delay et al. 2007; Alvarenga et al. 2012; Kluge et al. 2014). Deionised water was added to the soil samples with a ratio of soil to water by 1:10. Afterward, the samples were treated in an overhead shaker for 24 h and then filtered through 0.45 μm.

2.5 Elemental analyses

The pseudo total metal concentrations of soil samples was determined by aqua regia digestion in closed vessels in a microwave oven (Perkin Elmer) according to DIN ISO 11466 (1997). The concentrations of Cd, Cu, Pb and Zn in the digests were measured by ICP-MS (Varian 810/820-MS) and by graphite tube atomic absorption spectrophotometer (Varian SpectrAA 800Z) according to ISO 11885 (2007). All samples were analysed in triplicates. Quality assurance procedures were carried out to ensure reliability of the results. Analytical grade reagents and bi-distilled water were used during all procedures. A certified reference material was used as the quality control sample. Limits of quantification (LOQ) for measured metals were as follows: Pb 0.5 ppb, Cu 1 ppb, Cd 0.2 ppb and Zn 10 ppb. The concentrations of Cd, Cu, Pb and Zn in the soil solutions of S4 elution were measured by ICP MS (Varian 810/820-MS). The LOQ were as follows: Pb 10 ppb, Cu 1 ppb, Cd 0.5 ppb and Zn 1 ppb.

2.6 Comparison of metal concentrations with concentrations at the construction phase

Only limited information on the initial metal concentrations of the soils used in construction of the bioretention systems was available. Therefore, it is difficult to determine pollutant accumulation due to runoff infiltration. Nevertheless, in Germany, it is required to use clean soils to build bioretention systems.

Table 2 Inventory of the investigated bioretention swales

No.	Use of surrounding area	Drainage area type	Location	Infiltration system	Years of operation
1	Residential	Residential road; sidewalks	Berlin	Bioswale	19
2	Residential	Residential road	Berlin	Bioswale	19
3	Commercial	Main road	Berlin	Bioswale/underdrain	19
4	Residential	Residential road	Berlin	Bioswale/underdrain	19
5	Residential	Residential road; sidewalks	Berlin	Bioswale	19
6	Commercial	Road; sidewalk	Berlin	Bioswale	17
7	Commercial	Road; sidewalks	Berlin	Bioswale	15
8	Residential	Main road	Berlin	Bioswale	18
9	Residential	Roofs	Dortmund	Bioswale/underdrain	21
10	Residential	Residential road; parking lots	Dortmund	Bioswale/underdrain	14
11	Commercial	Roofs; parking lots; truck manoeuvring areas	Dortmund	Bioswale	22
12	Commercial	Parking lots; truck manoeuvring areas	Dortmund	Bioswale	21
13	Residential	Parking lots; residential road	Dorsten	Bioswale/underdrain	16
14	Residential	Parking lots; sidewalks; roofs residential road	Dortmund	Bioswale/underdrain	16
15	Residential	Parking lots	Dortmund	Bioswale/underdrain	19
16	Commercial	Parking lots	Dinslaken	Bioswale/underdrain	18
17	Commercial	Parking lots	Dortmund	Bioswale	18
18	Residential	Roofs	Bottrop	Bioswale	18
19	Residential	Roofs	Bochum	Bioswale	16
20	Residential	Roofs; residential road	Bochum	Bioswale	16
21	Commercial	Parking lots	Leverkusen	Bioswale	16
22	Commercial	Roofs	Leverkusen	Bioswale	11

This point is formalized in a technical regulation put forward by the German Association for Water, Wastewater and Waste (DWA-A 138 2005). The DWA requires that soils used in the construction of bioretention systems must have lower initial concentrations than the Z0 soil as defined by the Working Group for Waste (LAGA 2003) as a recycling (construction) standard.

The concentrations defined for the Z0 soil are equal to the precautionary values for the soil texture sand prescribed by the German Federal Soil Protection and Contamination Ordinance (BBodSchV 1999) (see Table 3). We compared our results to these Z0 concentrations and defined them as maximum initial concentrations in order to have an indication of metal accumulation by infiltrated water. Table 3 presents the assignment criteria and the precautionary values of the German Soil Act (BBodSchV 1999) for different soil textures.

3 Results and discussion

3.1 Hydraulic performance

The hydraulic conductivity represents a good basis for evaluating the hydraulic performance of bioretention systems. The

DWA A-138 (2005) defines a technically infiltration rate of between $1e-03$ and $1e-06$ m/s. The results were differentiated by the texture into two categories: (i) sand/loamy sand and (ii) sandy loam/silty loam (Fig. 2). They express the main texture classes (sand and loam/silt), which are dominant in the two regions studied (Berlin/Brandenburg and Nord-Rhine Westfalia).

The median of the hydraulic conductivity in predominantly sandy systems is $1.3e-04$ m/s, ranging from $9.2e-04$ to $5.6e-07$ m/s, at the sandy loam/silty loam systems the median hydraulic conductivity is $1.4e-05$, with a range between $7.5e-05$ and $6.9e-07$ m/s (Fig. 2). Thus, most of the sampled systems correspond well with the recommended hydraulic conductivity of the technical guidelines in Germany. Only 4 topsoils of in total 48 show lower hydraulic conductivity than $1e-6$. Lindsey et al. (1992) and Le Coustumer et al. (2009, 2012), who also investigated the hydraulic performance of biofiltration systems after long-term use, reported that more than 40 % of the investigated systems had a lower hydraulic conductivity than that recommend in the guidelines.

3.2 Vertical distribution of metals in bioretention soils

Metal concentrations in bioretention soils differ widely from system to system. Figure 3 shows the total concentrations of

Table 3 Z0 soil concentrations (LAGA 2003) and precautionary values of the German Soil Act (BBodSchV 1999)

Metal	Assignment criteria			Pathway soil to groundwater
	For construction use ^a Z0 soil mg/kg	Precautionary value ^b Sand mg/kg	Precautionary value ^b Silt/loam mg/kg	Trigger value ^b µg/L
Cd	0.4	0.4	1	5
Pb	40	40	70	25
Cu	20	20	40	50
Zn	60	60	150	500

Trigger values are valid for the transition area of the unsaturated to the water-saturated zone

^aLAGA (2003)

^bBBodSchV (1999)—Federal Soil Act and Federal Soil Protection and Contaminated Sites Ordinance

Cd, Pb, Cu and Zn at different bioretention soil depths. The median concentrations of all metals are highest at the soil surface (0–10 cm) and decrease with increasing depth. Zn concentrations in the first soil layer (0–10 cm) of the bioretention systems are the highest, with levels ranging from 2 to 1800 mg/kg, followed by Cu (5.7 to 210 mg/kg), Pb (12 to 160 mg/kg) and Cd (0.07 to 1.2 mg/kg). Highest metal concentrations often were found at the inflow points of the systems. This might be a consequence of heterogeneous infiltration pattern caused by rainfall events with low to medium intensity. At these events, only small inlet parts of the bioretention systems are flooded and led to a uniform runoff and consequently metal distribution. Jones and Davis (2013) also found a decrease of metal concentrations with distance from the inflow point at a bioretention cell after a 4-year operational time.

When we compare the median concentrations with the Z0 values (maximum initial concentrations; see Table 3), the median concentrations at the topsoil increased by factor of 2.3 for Zn, 1.3 for Cu, 1.2 for Pb and 1.1 for Cd. Higher median concentrations than Z0 are also determined at the soil depth 10–20 cm for Cd and Zn. Relating all metal concentrations to the precautionary values of the BBodSchV (1999), out of a total of 119 soil samples, we determined 72 exceeding the

threshold values for Zn, 40 for Cu, 33 for Pb and 22 for Cd. At the soil depth 20–35 cm, a sharp decrease of the median concentrations of all metals was observed, which is to be expected as most topsoils used as bioretention media usually have adequate retention capacities due to filtration, precipitation and sorption processes.

The determined metal capture in the top soil layers of the bioretention systems agrees with the findings of previous laboratory and field studies related to stormwater bioretention (e.g. Davis et al. 2003; Li and Davis 2008a; Hatt et al. 2008; Blecken et al. 2011; Ingvertsen et al. 2012b; Jones and Davis 2013; Paus et al. 2014). The finding that most of the metals are concentrated at the top layer is also in line with other studies related to the long-term accumulation of metals by infiltrating runoff water, among other sources.

In a review of metal concentrations in roadside soils, Werkenthin et al. (2014) pointed out that the concentrations decreased between 57 and 75 % at soil depths between 15 and 75 cm compared to topsoil concentrations. At soil depths between 10 and 30 cm, Dierkes and Geiger (1999) determined a reduction of Cu, Pb and Zn concentrations ranging from 75 to 93 %; for Cd, the reduction was much less (30–40 %). The sample data suggest that considerable metal accumulation has occurred at the soil depth 0–20 cm during the long-term operation of the systems.

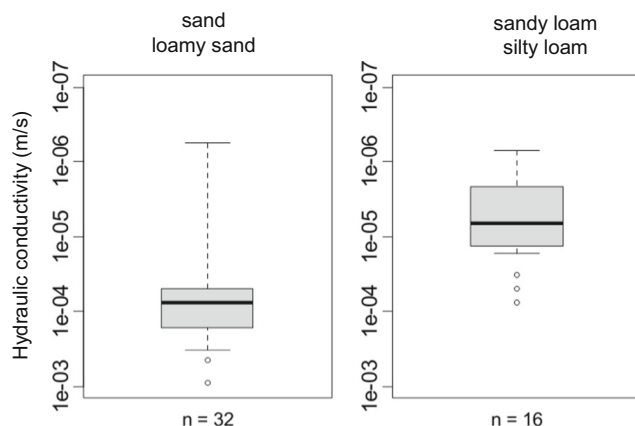
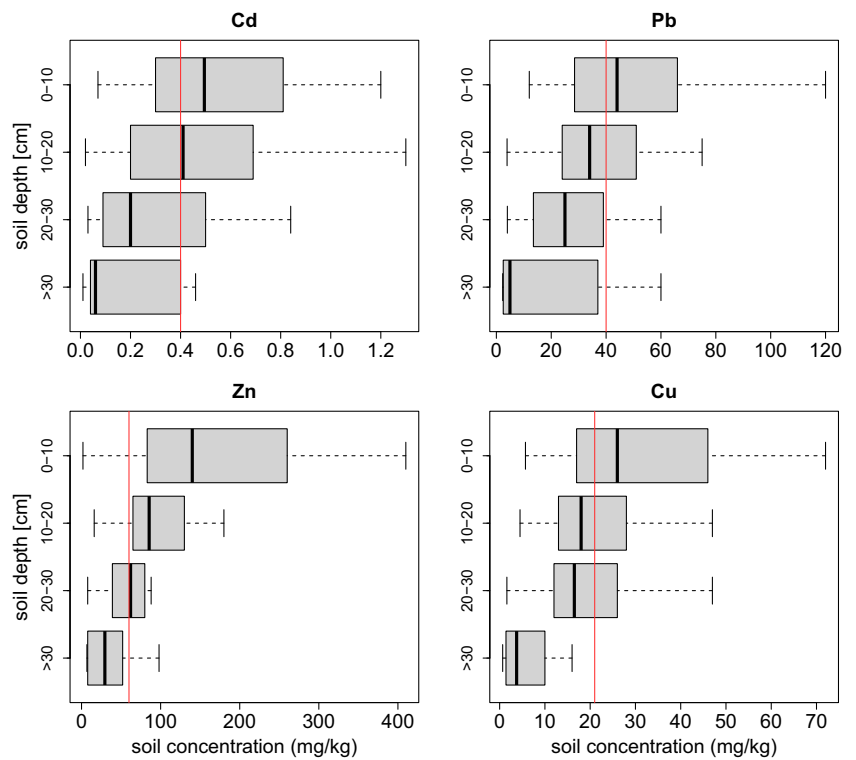


Fig. 2 Hydraulic conductivity (m/s) of 48 bioretention systems measured by double ring infiltrometer

3.3 Influence of land use type on metal accumulation

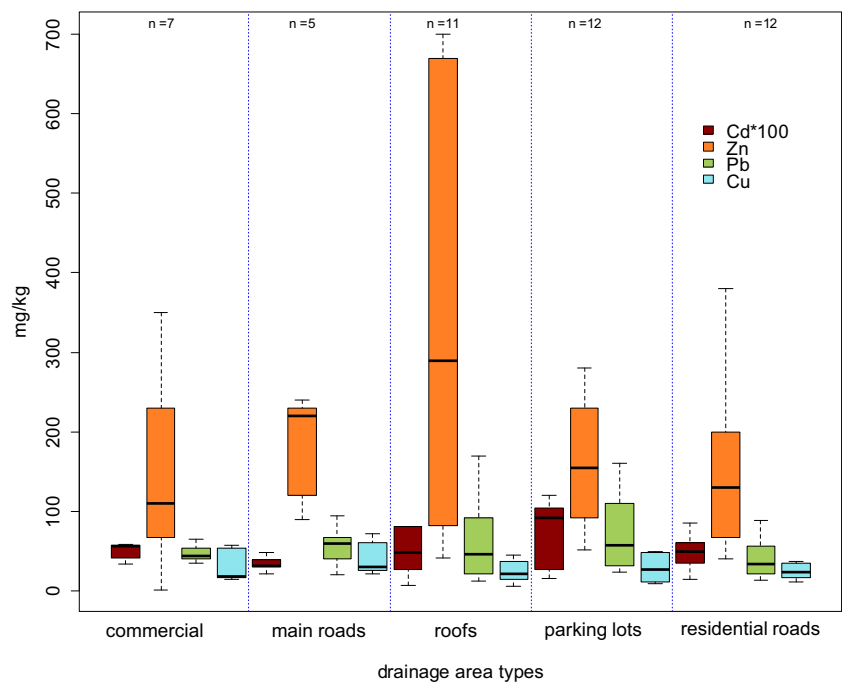
In order to compare various drainage area types on metal accumulation and distribution of the sampled bioretention systems, Fig. 4 shows the metal concentration in the topsoil (0–10 cm) for the following runoff categories: (a) commercial, (b) main roads, (c) parking lots, (d) roofs with zinc gutters and (e) residential roads. Median Zn concentrations are significantly higher in topsoils at systems with runoff drainage from roofs with zinc gutters, followed by systems with runoff drainage from main roads. Higher median concentrations of Cd and Pb were found in systems with runoff drainage from parking lots.

Fig. 3 Heavy metal concentrations in different soil depths of all bioretention systems year 2014 ($n = 22$). Note: n in the diagram refers to the sample size of the sampled and analysed horizons; the red line marks the maximum allowed concentration of the built-in soils (Z0) as defined by the Working Group for Waste (LAGA 2003). Outliners are not shown



Median Cu soil concentrations are more or less in the same order of magnitude for all drainage area types. Lowest metal concentrations are determined for the drainage area types “commercial” and “residential roads”. The distribution of the metals in topsoil corresponds well with the distribution of average metal concentrations from different runoff surface types reported by Göbel et al. (2007).

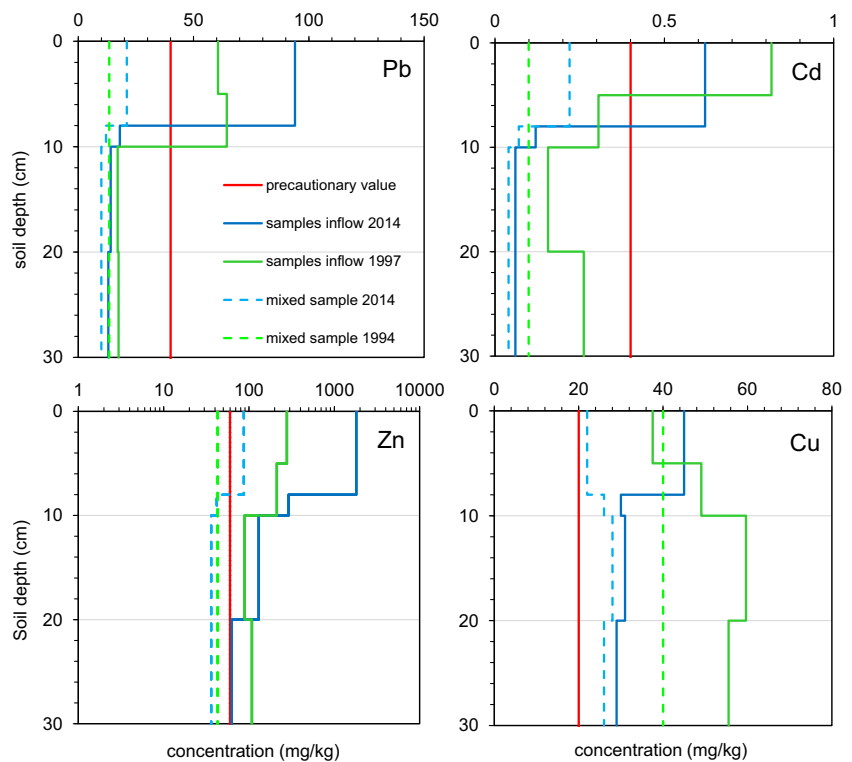
Fig. 4 Heavy metal concentrations in topsoil (0–10 cm) of all bioretention systems classified by different drainage area types: commercial, main roads, parking lots, roofs with zinc gutters and residential roads



3.4 Long-term accumulation and comparison of metal concentrations at different time steps of two bioretention systems

Figure 5 shows the heavy metal concentrations in soil matrix of system no. 9 at different time steps. The coordinates, metal soil concentrations and S4 elution concentrations of different

Fig. 5 Heavy metal concentrations in different soil depths and sampling years (1994, 1997, 2014) of system no. 9. Mixed samples = 1.5 m distance from the inflow. Red line represents the precautionary value of the BBodSchV (1999) for the soil texture sand



soil depths were obtained from the data of a technical report of Schöttler and Remmler (1999), for system no. 11 from a technical report by Hiller et al. (2001).

The system is located in a residential area; the catchment area of the infiltration water is a roof; and the water passes through a galvanized roof gutter and infiltrates directly into the adjacent soil. The soil texture of the soil medium is silty sand.

Metal concentrations determined in the year 2014 mostly accumulated at the upper 10 cm, rapidly decreasing to initial concentrations with increasing depth. Compared to the initial concentrations of the topsoil (0–10 cm) of the year 1994 at the inflow, Pb, Cd and Zn concentrations increased by a factor of 7, 6 and 42. In contrast, the metal concentrations of the topsoil (0–10 cm) at 1 m distance to the inflow (mixed sample 2014) are, compared to the initial concentrations of 1994, only slightly increased. The high Zn concentration in the inflow point (0–10 cm) could be explained due to the corrosion of Zn from the galvanized rain gutter. Göbel et al. (2007) pointed out that runoff from roofs constructed with Zn gutters and downpipes often release higher metal concentrations than runoff from high trafficked areas. Due to corrosion by rain, not only Zn in particular but also Cd and Cu are released to the environment. He et al. (2001) investigated corrosion rates of zinc roofing materials and measured decreasing metal concentrations in roof runoff after initially high concentrations, reaching more or less constant concentrations with time.

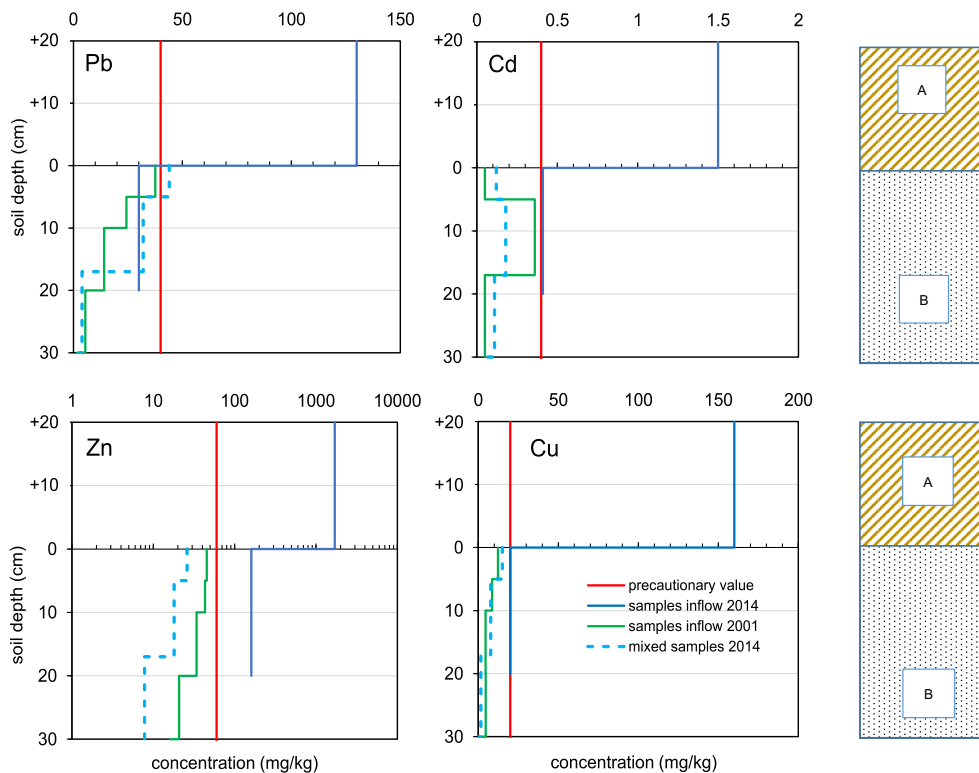
This trend is also reflected by the rapid increase of all soil metal concentrations in the topsoil within the first 3 years of operation, except Cu (comparison of the mixed sample 1994

to the inflow sample 1997; Fig. 4). The Cu concentrations at the topsoil of both mixed and inflow samples of the year 2014 show a different distribution with lower concentrations compared to 1994, similarly for 1997. One explanation for these finding could be the preferred association of Cu with organic matter, which significantly increases the leachability compared to Pb and Zn and thus leads to a stronger leaching to the subsoil. Increased Cu leaching from bioretention media (soils) was also found by Sauvé et al. (2000), Li and Davis (2008a), Trowsdale and Simcock (2011) and Mullane et al. (2015). Li and Davis (2008a) observed a strong release of Cu from their investigated bioretention media and also put this down to a stronger association with organic matter and a high solubility by water.

Increased leaching and displacement of Cu was also detected in roadside soils, which is similarly characterized by high runoff infiltration (Bäckström et al. 2004 and Kocher et al. 2005).

The Cd concentrations at the inflow of the year 1997 were higher than those of the year 2014 at all investigated depths. A rapid leaching and a transfer to lower soil layers might be a reason for this, since Cd is the most mobile of the elements analysed (Alloway 2006) and mainly occurs in a dissolved fraction in runoff (e.g. Dierkes and Geiger 1999; Preciado and Li 2006). Concentrations exceeding the precautionary values of the BBodSchV were determined at the topsoil layer (0–10 cm) for all investigated metals at the inflow, whereas the concentrations at 1.5 m distance are well below the values with the exception of Zn in the mixed sample 2014 and Cu at all sample depths.

Fig. 6 Heavy metal concentrations in different soil depths and sampling years (2001 and 2014) of system no. 11. Note: Sample inflow 2001 describes the initial concentration of the built-in soils year 2001. A sediment accumulation zone; B original bioretention soil layer



Cd, Pb, Cu and Zn soil concentrations of system no. 11 are shown in Fig. 6. The coordinates and metal soil concentrations were obtained from a technical report by Hiller et al. (2001). The system is located in a commercial area; the catchment area of the infiltration water is an asphalt parking and truck-manoeuvring area at a logistics center; and the water passes through a channel of concrete and infiltrates into the adjacent soil. The soil texture of the soil media ranges from silty sand at the top layer (0–10 cm) to medium sand at the bottom layer (10–65 cm).

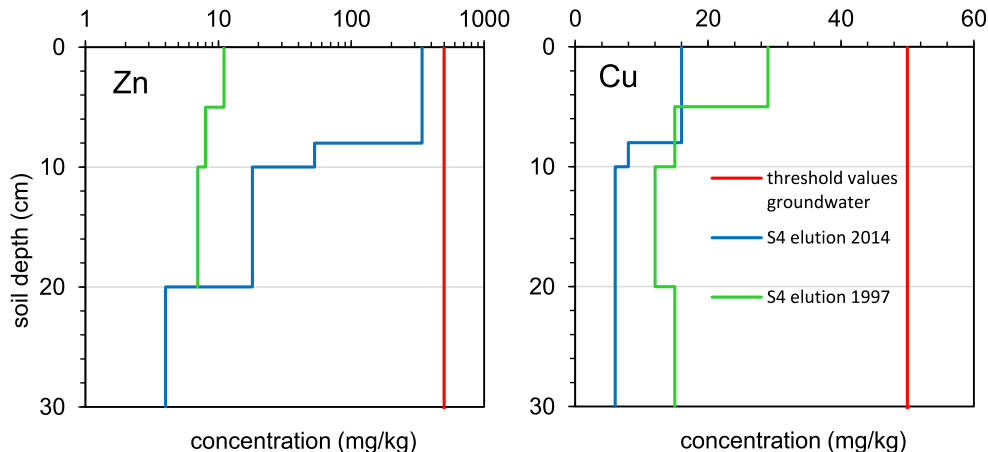
Compared to the raised portion of the system, at 1.5 m distance from the inflow, infiltrating water led to a sediment deposition of about 20 cm due to washed up and accumulated particles. The concentrations of all metals are strongly

increased at the accumulated sediment (0 ± 20 cm) compared to the soil metal concentration of the years 2001 and 2014.

In contrast, data of mixed samples at 1 m distance from the inflow for the year 2014 show only a slight increase in metal concentrations compared to the inflow samples of the year 2001, except Zn and to a certain extent Cd. For these metals, decreasing metal concentrations at the top soil (0–10 cm) could be observed.

One explanation for this finding is a slight leaching of the initial soil metal concentrations due to long-term infiltration over a timespan of 13 years. Dietz and Clausen (2006) also found a decrease of metals in a bioretention system after 2 years of operation and pointed out that the investigated bioretention soils serve as a source of metals. Our data suggest

Fig. 7 S4 elution concentrations of Zn and Cu of the inflow soil samples at different depths from system no. 9, sampling years 1997 and 2014. Red line represents the threshold values of the German Soil Act for the soil to groundwater path. Note: S4 elution data of Pb and Cd of the year 1997 are below the detection limit; therefore, no comparison is shown



that the accumulated sediment is a likely sink for the incoming metals. The concentrations of Zn and Pb at the inflow are at 1770 and 130 mg/kg respectively, much higher than those determined by other authors (Jones and Davis 2013; Paus et al. 2014). Jones and Davis (2013) reported concentrations of Pb = 64 mg/kg, Cu = 49 mg/kg and Zn = 257 mg/kg near the inflow at a bioretention system after 4 years of operation. Observed mean Zn and Cu concentrations of Paus et al. (2014) at the topsoil layer of different bioretention systems after periods of operation ranging from 7 to 13 years are 2–6 times lower than those reported by Jones and Davis (2013). The higher concentrations could be explained by the longer operation time of the investigated bioretention systems and by the

different runoff origin. All metal concentrations at the sediment are at or above the precautionary values of the BBodSchV (1999); all metals at the origin top layer (0–10 cm) of the year 2014 (inflow) and at 1.5 m distance are below the precautionary values of the BBodschV (1999), except Pb, for which we observed a marginal exceeding of the precautionary values at the soil depth 0–5 cm.

3.5 Batch leaching tests of the soils in long-term operating bioretention systems

The concentrations of water-soluble heavy metals determined by S4 elution of system no. 9 are presented exemplarily in

Table 4 Soil texture, pH, C_{org} and (pseudo-) total heavy metal concentrations in soil and soil solution (S4 elution) in different soil depths of different bioretention systems, year 2014

No.	depth cm	Soil texture USDA	pH –	C _{org} M.%	Cd SP mg/kg	Cd S4 µg/L	Cu SP mg/kg	Cu S4 µg/L	Pb SP mg/kg	Pb S4 µg/L	Zn SP mg/kg	Zn S4 µg/L
1	0–12	Sandy loam	7.2	3.6	0.4	<0.5	72	35.3	41	<10	240	7.3
	12–26	Sandy loam	6.9	1.2	0.2	<0.5	19	24.1	24	<10	65	17.8
	26–44	Sandy loam	6.8	1.0	0.4	<0.5	23	18.6	39	<10	98	6.3
9	0–8	Sandy loam	6.8	5.7	0.62	<0.5	45	16.3	94	<10	1800	330
	8–10	Loamy sand	6.6	4.0	0.12	<0.5	30	7.7	18	<10	290	50.7
	10–20	Sand	6.9	3.5	0.06	<0.5	31	5.9	14	<10	130	16.4
	20–30	Loamy sand	7.1	3.2	0.06	<0.5	29	4.6	13	<10	63	2.3
11	0–15	Silty loam	6.0	6.2	0.91	<0.5	50	25	62	<10	660	72.3
	15–25	Silty loam	6.1	2.3	0.73	<0.5	21	12.7	48	<10	160	8.2
6	0–15	Sand	6.7	1.5	0.16	<0.5	21	4	35	<10	68	9
	15–39	Sand	7.3	1.0	0.09	<0.5	5.4	2	11	<10	35	5
	39–90	Sand	7.2	0.1	0.05	<0.5	1.4	2	2.5	<10	9.1	11
7	0–18	Loamy sand	6.2	1.5	0.15	<0.5	11	4	20	18	41	11
	18–23	Sand	6.5	1.2	0.25	<0.5	13	3	47	<10	67	4
	23–37	Sand	6.4	1.0	0.2	<0.5	9.8	4	36	<10	44	7
	37–65	Sand	6.4	0.1	0.02	<0.5	1.6	6	2.5	<10	6.9	4
8	0–8	Sandy loam	7.0	3.1	0.21	<0.5	30	8	67	<10	120	30
	8–20	Sandy loam	7.8	0.9	0.2	<0.5	29	6	75	<10	87	12
	20–40	Sandy loam	7.8	0.8	0.2	<0.5	27	5	60	<10	88	5
16	0–10	Sandy loam	5.7	2.1	0.58	<0.5	9	1.8	50	<10	120	30.6
	0–23	Sandy clay loam	5.6	1.3	0.59	<0.5	8.3	2.5	47	<10	69	21
	23–45	Sand	5.5	1.1	0.46	<0.5	6.8	2.2	37	<10	52	9
18	0–14	Sandy loam	5.5	3.0	0.8	<0.5	19	1.8	90	<10	690	460.2
	14–27	Sandy loam	6.0	2.7	1.3	<0.5	16	1.4	59	<10	290	110.8
	27–44	Sandy loam	5.7	0.9	0.28	<0.5	6.9	<1	20	<10	44	9.2
	90–100	Sand	6.0	0.7	0.03	<0.5	4	<1	6.2	<10	9.7	2
21	0–5	Sandy loam	5.9	4.3	0.34	<0.5	57	6.8	35	<10	350	22.3
	5–30	Sandy loam	6.2	1.1	0.39	<0.5	9	1	32	<10	62	3.7
	30–40	Sand	6.3	0.2	0.05	<0.5	10	<1	11	<10	34	1
20	0–9	Sandy loam	6.1	3.5	0.39	<0.5	33	5.1	34	<10	160	14.8
	9–19	Sand	6.3	0.9	0.3	<0.5	23	2.9	32	<10	140	10.9
19	0–14	Silty loam	5.9	3.1	0.48	<0.5	15	2.4	46	<10	650	209.4

SP solid phase extraction with aqua regia digestion according to DIN ISO 11466 (1997), S4 soil solution concentration according to DIN 38414-4 (1984)

Fig. 7. The results show that most of the metals deposited in bioretention soils are only slightly water soluble. According to the threshold values of the BBodSchV for the soil to groundwater path, no concentrations measured exceeded the precautionary value. Pb and Cd concentrations of the S4 elution were below the detection limit; therefore, no data comparison was possible. A comparison of the water-soluble concentrations of the years 1997/2014 shows a strong increase in Zn and a moderate decrease of Cu. This is in line with the finding of increasing Zn concentrations and decreasing Cu concentrations of the topsoil of system no. 9 (Fig. 4). Overall, we eluted 49 samples of 11 sample points of six different bioretention systems in different depths with the method of 1:10 elution (Table 4). When we compared the trigger values of the BBodSchV (1999)—pathway soil to groundwater—for all investigated metals, no exceeding values were detectable (Tables 3 and 4).

4 Conclusions

After long-term operation, the hydraulic performance (k_{sat}) of the bioretention systems is mostly given and corresponds well with the recommended hydraulic conductivity of the technical guidelines of Germany. Mostly, metal accumulation is concentrated in the top 20 cm, which is in agreement with findings of other authors. In the underlying soil layers, all concentrations decrease rapidly and mostly reach background, i.e. initial concentrations after depths >30 cm. However, in some cases, we found very high metal concentration even in deeper soil layers. Obviously, this is a result of pre-polluted built-in substrates (anthropogenic and technical materials, diagnosed by colour, texture, smell), which were used as construction substrate for the bioretention systems.

However, very high metal concentrations were found at the inflow points with runoff drainage from roofs and parking lots. The same is true for drainage runoff from commercial areas. This might be a consequence of heterogeneous infiltration pattern caused by rainfall events with low to medium intensity. Then, only small inlet parts of the bioretention systems are flooded. Consequently, the following reasons for the heterogeneous metal distributions can be pointed out: (a) first flush effects, (b) system geometry of the bioretention system and (c) heterogeneity of the soil hydraulics.

In order to improve bioretention systems, we suggest regularly a replacement of the accumulated sediments or soils at the inflow points after 20–25 years as a routine maintenance. Another possibility could be a technical optimization of the bioretention systems leading to a more homogeneous infiltration of runoff water. Both might be an adequate solution to avoid critical violation of thresholds in order to protect groundwater resources.

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References

- Achleitner S, Engelhard C, Stegner U, Rauch W (2007) Local infiltration devices at parking sites—experimental assessment of temporal changes in hydraulic and contaminant removal capacity. *Water Sci Technol* 55(4):193–200
- Alloway BJ (2006) Heavy metals in soils. Environmental pollution. Springer, Dordrecht
- Alvarenga P, Palma P, De Varennes A, Cunha-Queda AC (2012) A contribution towards the risk assessment of soils from the São Domingos mine (Portugal): chemical, microbial and ecotoxicological indicators. *Environ Pollut* 161:50–56
- Bäckström M, Karlsson S, Backman L, Folkesson L, Lind B (2004) Mobilisation of heavy metals by deicing salts in a roadside environment. *Water Res* 38:720–732
- BBodSchV (1999) Bundes-Bodenschutz- und Altlastenverordnung vom 12. Juli 1999 (BGBl. I S. 1554)
- Blanchard M, Teil MJ, Ollivon D, Garban B, Chesterikoff C, Chevreuil M (2001) Origin and distribution of polyaromatic hydrocarbons and polychlorobiphenyls in urban effluents to wastewater treatment plants of the Paris area (France). *Water Res* 35:3679–3687
- Blecken GT, Marsalek J, Viklander M (2011) Laboratory study of stormwater biofiltration in low temperatures: total and dissolved metal removals and fates. *Water Air Soil Pollut* 219(1–4):303–317
- Boivin P, Saade M, Pfeiffer HR, Hammecker C, Degoumois Y (2008) Depuration of highway runoff water into grass-covered embankments. *Environ Technol* 29:709–720
- Davis AP, Shokouhian M, Sharma H, Minami C (2001) Laboratory study of biological retention for urban stormwater management. *Water Environ Res* 73(1):5–14
- Davis AP, Shokouhian M, Sharma H, Minami C, Winogradoff D (2003) Water quality improvement through bioretention: lead, copper, and zinc removal. *Water Environ Res* 75(1):73–82
- Davis AP, Shokouhian M, Sharma H, Minami C (2006) Water quality improvement through bioretention media: nitrogen and phosphorus removal. *Water Environ Res* 78(3):284–293
- Delay M, Lager T, Schulz HD, Frimmel FH (2007) Comparison of leaching tests to determine and quantify the release of inorganic contaminants in demolition waste. *Waste Manag* 27(2):248–255
- Dierkes C, Geiger WF (1999) Pollution retention capabilities of roadside soils. *Water Sci Technol* 39:201–208
- Dietz ME (2007) Low impact development practices: a review of current research and recommendations for future directions. *Water Air Soil Pollut* 186(1–4):351–363
- Dietz ME, Clausen JC (2006) Saturation to improve pollutant retention in a rain garden. *Environ Sci Technol* 40(4):1335–1340
- DIN 19682-7 (2015) Soil quality—field tests—part 7: determination of infiltration rate by double ring infiltrometer. German Institute for Standardization, Berlin
- DIN 38414-4 (1984) German standard methods for the examination of water, waste water and sludge; sludge and sediments (group S); determination of leachability by water (S4). German Institute for Standardization, Berlin
- DWA-A 138 - German Association for Water, Wastewater and Waste (2005) Planung, Bau und Betrieb von Anlagen zur Versickerung von Niederschlagswasser, 2005 [Design, construction and operation of rainwater drainage systems] (in German)

- Fletcher TD, Andrieu H, Hamel P (2013) Understanding, management and modelling of urban hydrology and its consequences for receiving waters: a state of the art. *Adv Water Resour* 51:261–279
- Folkeson L, Bækken T, Brenčić M, Dawson A, François D, Kuřimská P, et al. (2009) Sources and fate of water contaminants in roads. In *Water in Road Structures*. Springer, Netherlands, pp 107–146
- García P, Pérez ME, Guerra A (2014) Using TM images to detect soil sealing change in Madrid (Spain). *Geoderma* 214:135–140
- Göbel P, Dierkes C, Coldewey WG (2007) Storm water runoff concentration matrix for urban areas. *J Cont Hydrol* 91(1):26–42
- Hatt BE, Fletcher TD, Deletic A (2008) Hydraulic and pollutant removal performance of fine media stormwater filtration systems. *Environ Sci Technol* 42(7):2535–2541
- He W, Wallinder IO, Leygraf C (2001) A comparison between corrosion rates and runoff rates from new and aged copper and zinc as roofing material. *Water Air Soil Pollut Focus* 1(3–4):67–82
- Hiller D, Winzig G, Dornauf C (2001) Bodenchemische Untersuchungen von Versickerungsanlagen als Grundlage für eine nachhaltige Niederschlagswasserbewirtschaftung im Sinne des Boden- und Grundwasserschutzes. Technical report, Essen (in German)
- Hunt WF, Jarrett AR, Smith JT, Sharkey LJ (2006) Evaluating bioretention hydrology and nutrient removal at three field sites in North Carolina. *J Irrig Drain E Asce* 132(6):600–608
- Ingversen ST, Cederkvist K, Régent Y, Sommer H, Magid J, Jensen MB (2012a) Assessment of existing roadside swales with engineered filter soil: I. Characterization and lifetime expectancy. *J Environ Qual* 41(6):1960–1969
- Ingversen ST, Cederkvist K, Jensen MB, Magid J (2012b) Assessment of existing roadside swales with engineered filter soil: II. Treatment efficiency and in situ mobilization in soil columns. *J Environ Qual* 41(6):1970–1981
- ISO 11466 (1997) Soil quality—extraction of trace elements soluble in aqua regia. German Institute for Standardization, Berlin
- ISO 11885 (2007) Water quality—determination of selected elements by inductively coupled plasma optical emission spectrometry (ICP-OES). German Institute for Standardization, Berlin
- Jones PS, Davis AP (2013) Spatial accumulation and strength of affiliation of heavy metals in bioretention media. *J Environ Eng* 139(4):479–487
- Kabir MI, Daly E, Maggi F (2014) A review of ion and metal pollutants in urban green water infrastructures. *Sci Total Environ* 470:695–706
- Kayhanian M, Fruchtmann BD, Gulliver JS, Montanaro C, Ranieri E, Wuertz S (2012) Review of highway runoff characteristics: comparative analysis and universal implications. *Water Res* 46:6609–6624
- Kluge B, Werkenthin M, Wessolek G (2014) Metal leaching in a highway embankment on field and laboratory scale. *Sci Total Environ* 493:495–504
- Kocher B, Wessolek G, Stoffregen H (2005) Water and heavy metal transport in roadside soils. *Pedosphere* 15:746–753
- LAGA-Bund/Länder-Arbeitsgemeinschaft Abfall (2003) Anforderungen an die stoffliche Verwertung von mineralischen Reststoffen/Abfällen. Technical regulations. (in German)
- Le Coustumer S, Fletcher TD, Deletic A, Barraud S, Lewis JF (2009) Hydraulic performance of biofilter systems for stormwater management: influences of design and operation. *J Hydrol* 376(1):16–23
- Le Coustumer S, Fletcher TD, Deletic A, Barraud S, Poelsma P (2012) The influence of design parameters on clogging of stormwater biofilters: a large-scale column study. *Water Res* 46(20):6743–6752
- LeFevre GH, Paus KH, Natarajan P, Gulliver JS, Novak PJ, Hozalski RM (2014) Review of dissolved pollutants in urban storm water and their removal and fate in bioretention cells. *J Environ Eng* 141(1):04014050
- Li H, Davis A (2008a) Urban particle capture in bioretention media. I: laboratory and field studies. *J Environ Eng* 134(6):409–418
- Li H, Davis AP (2008b) Heavy metal capture and accumulation in bioretention media. *Environ Sci Technol* 42(14):5247–5253
- Li H, Davis AP (2009) Water quality improvement through reductions of pollutant loads using bioretention. *J Environ Eng* 135(8):567–576
- Li YL, Deletic A, McCarthy DT (2014) Removal of *E. coli* from urban stormwater using antimicrobial-modified filter media. *J Hazard Mater* 271:73–81
- Lim HS, Lim W, Hu JY, Ziegler A, Ong SL (2015) Comparison of filter media materials for heavy metal removal from urban stormwater runoff using biofiltration systems. *J Environ Manag* 147:24–33
- Lindsey G, Roberts L, Page W (1992) Inspection and maintenance of infiltration facilities. *J Soil and Water Cons* 47(6):481–486
- Liu J, Sample DJ, Bell C, Guan Y (2014) Review and research needs of bioretention used for the treatment of urban stormwater. *Water* 6(4):1069–1099
- Lucke T, Nichols PW (2015) The pollution removal and stormwater reduction performance of street-side bioretention basins after ten years in operation. *Sci Total Environ* 536:784–792
- Lundy L, Ellis JB, Revitt DM (2012) Risk prioritisation of stormwater pollutant sources. *Water Res* 46(20):6589–6600
- Marsalek J, Watt WE, Anderson BC (2006) Trace metal levels in sediments deposited in urban stormwater management facilities. *Water Sci Technol* 3(2):175–183
- Mullane JM, Flury M, Iqbal H, Freeze PM, Hinman C, Cogger CG, Shi Z (2015) Intermittent rainstorms cause pulses of nitrogen, phosphorus, and copper in leachate from compost in bioretention systems. *Sci Total Environ* 537:294–303
- Parker JK, McIntyre D, Noble RT (2010) Characterizing fecal contamination in stormwater runoff in coastal North Carolina, USA. *Water Res* 44(14):4186–4194
- Paus KH, Morgan J, Gulliver JS, Leiknes T, Hozalski RM (2014) Assessment of the hydraulic and toxic removal capacities of bioretention cells after 2 to 8 years of service. *Water Air Soil Pollut* 225:1803
- Piguet P, Parriaux A, Bensimon M (2008) The diffuse infiltration of road runoff: an environmental improvement. *Sci Total Environ* 397:13–23
- Preciado HF, Li LY (2006) Evaluation of metal loadings and bioavailability in air, water and soil along two highways of British Columbia, Canada. *Water Air Soil Pollut* 172:81–108
- Prince George's County PGC (2007) Bioretention manual, PGC, Maryland, Dept. of Environmental Resources, Environmental Services Div., Landover, Md
- Roy-Poirier A, Champagne P, Filion Y (2010) Review of bioretention system research and design: past, present, and future. *J Environ Eng* 136(9):878–889
- Sauvé S, Hendershot H, Allen HE (2000) Solid-solution partitioning of metals in contaminated soil: dependence on pH, total metal burden, and organic matter. *Environ Sci Technol* 34:1125–1131
- Schöttler U, Remmler F (1999) Abschlußbericht zum BMBF-Vohaben Naturnahe Regenwasserbewirtschaftung aus der Sicht des Boden- und Grundwasserschutzes. Technical report. Institut für Wasserforschung GmbH, Dortmund. (in German)
- Søberg L, Blecken GT, Viklander M (2014) The influence of temperature and salt on metal and sediment removal in stormwater biofilters. *Water Sci Technol* 69(11):2295–2223
- Trowsdale SA, Simcock R (2011) Urban stormwater treatment using bioretention. *J Hydrol* 397(3):167–174
- Werkenthin M, Kluge B, Wessolek G (2014) Metals in European roadside soils and soil solution—a review. *Environ Pollut* 189:98–110
- Zhang K, Randelovic A, Page D, McCarthy DT, Deletic A (2014) The validation of stormwater biofilters for micropollutant removal using in situ challenge tests. *Ecol Eng* 67:1–10