



## Seepage metal concentrations beneath long-term operated bioretention systems

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### ABSTRACT

Bioretention systems maintain the natural water cycle and help to mitigate climatic extremes impact on urban areas by retarding, storing, and evaporating stormwater runoff. Although bioretention systems have been operated for more than 25 years, systematic investigations on the hydrological functionality and pollutant retention performance of older systems are rare. We employed laboratory and field experiments to investigate three long-term operated bioretention systems in Germany with the following objectives: (i) physico-chemical substrate characterisation; (ii) an event-based influent and effluent trace metal concentration monitoring covering 22 months and (iii) the calculation of metal retention rates. Regarding the pollution status, we found significantly increased trace metal contents in the soil substrate mainly as a function of the drainage area type and the inflow regime. Nonetheless, all measured metal seepage concentrations fall below the German legislative trigger values. Our current findings demonstrate no risk of groundwater degradation even for old bioretention systems suggesting bioretention as a powerful and sustainable tool for stormwater management. Further research requires the handling of soil substrates modified by stormwater infiltration showing enhanced trace metal contents and a certain amount of technogenic sediments.

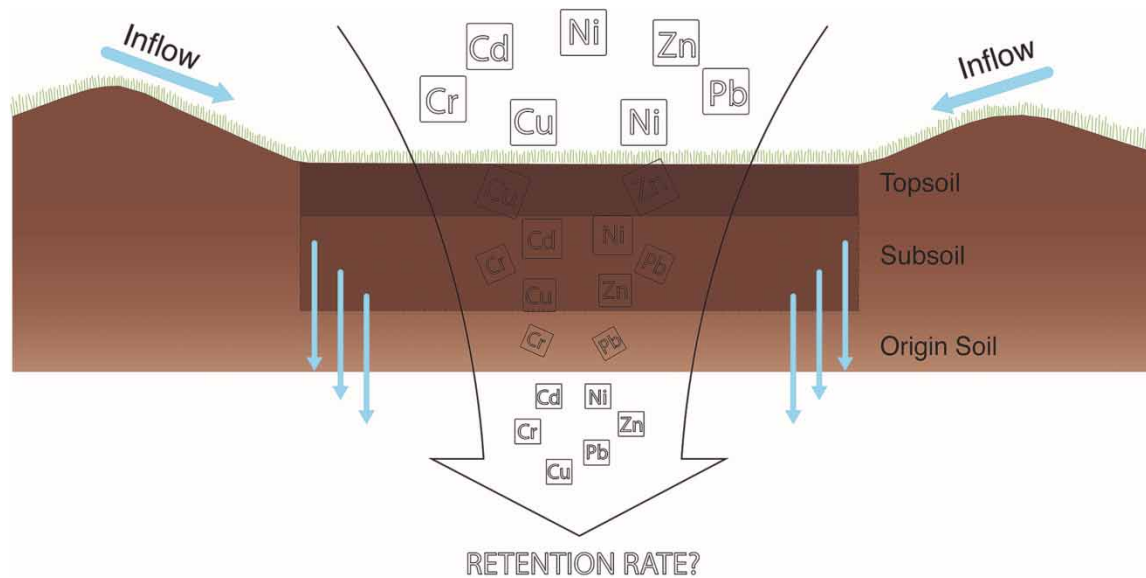
**Key words:** bioretention systems, Blue-Green Systems (BGS), seepage metal concentrations, stormwater management, sustainable urban drainage systems (SUDS), urban runoff

### HIGHLIGHTS

- Systematic investigation of three long-term operated bioretention systems in Germany.
- Synchronised *in situ* monitoring of soil seepage samples and real drainage periods for almost 2 years.
- Fulfilment of German legislative standards considering the average metal seepage concentrations.
- Drainage area types did not reflect seepage qualities.
- High retention rates for all investigated metals after long-term operation.

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## GRAPHICAL ABSTRACT



## INTRODUCTION

Urban areas will continuously grow in future due to the growing world population, even further enforced by the rural exodus (Wong *et al.* 2006; Liu *et al.* 2014). Urbanisation affects soils' natural drainage function by surface sealing and compaction requiring stormwater management to prevent urban flooding. Conventionally, urban stormwater drains by central sewer systems, which discharge directly into surface water bodies during high flow (LeFevre *et al.* 2015). Such conventional overflow systems were developed in the past to rapidly drain urban areas in order to prevent flooding on the one hand and to reduce the runoff volume to be treated in the treatment plant to not exceed its capacity on the other hand. However, runoff water contains a variety of pollutants leading to the degradation of receiving water bodies, hence endangering ecological and human health (LeFevre *et al.* 2015). Over the last decades, alternative management practices for urban drainage have been established, which try to face the multiple problems associated with the conventional urban stormwater management (Fletcher *et al.* 2015). The basic principle of all so-called blue-green infrastructures is to capture and infiltrate runoff water of the surrounding sealed area into soil media, in which containing pollutants are potentially filtered (Ahiablame *et al.* 2012). Subsequently, the infiltrated runoff percolates through the soil media and seepage water is either collected in an underdrain, which discharges into a near-by surface water body, or simply percolates to the groundwater (LeFevre *et al.* 2015). Hence, blue-green infrastructures maintain the natural water cycle and consequently are also referred to as low impact developments (LIDs) or best management practice (BMP) (Winogradoff & Coffman 2002).

One of the most efficient and frequently used types of the LID-BMPs is bioretention, synonymously referred to as rain garden and biofilters, to address the increasing urban stormwater problems (Davis *et al.* 2010; Liu *et al.* 2014). Bioretention systems are applied in residential or commercial settings (Dietz 2007). Usually, they are relatively inexpensive to install and maintain and offer several economic and ecological advantages compared with conventional stormwater management practices. Important benefits of LID-BMPs compared with conventional practices are the reduction of rainwater runoff into the sewer system, stabilisation of the urban water cycle and reduction of the urban heat island effect by promoting evapotranspiration. The infiltrating runoff from urban areas leads to a potential contamination of the bioretention facilities with many different organic and especially inorganic pollutants (Müller *et al.* 2020).

Trace metals, such as cadmium, copper, lead, and zinc, are of major concerns in urban drainage water due to their widespread occurrence, toxicity, and persistence in the environment posing a major threat to receiving surface and groundwater resources (Wong *et al.* 2006; Alloway 2012; Kayhanian *et al.* 2012; Vardhan *et al.* 2019). In general, toxic metals are atmospherically deposited originating from numerous anthropogenic activities, such as traffic, industry, or disposal of municipal waste (Wong *et al.* 2006; Ahiablame *et al.* 2012). However, the primary

source of trace metals in urban stormwater runoff originates from traffic-related depositions and due to the corrosion of construction and buildings with metal coatings (Wong *et al.* 2006; Kayhanian *et al.* 2012; LeFevre *et al.* 2015; Müller *et al.* 2020). Trace metals conveyed by runoff are either present in dissolved form or associated with particles (LeFevre *et al.* 2015). Dissolved metal ions in stormwater are of great concern due to their high mobility while being more bioavailable and toxic than particle-associated metals (LeFevre *et al.* 2015).

During the infiltration of contaminated runoff water into a bioretention system and its percolation through the soil towards the groundwater, the seepage water quality is usually improved by three major removal mechanisms: (i) physical filtering of particle-associated metals (Davis *et al.* 2010; Clark & Pitt 2012; LeFevre *et al.* 2015); (ii) plant-root uptake of dissolved metals from the soil solution (Sun & Davis 2007; LeFevre *et al.* 2015); and (iii) sorption of dissolved metals to the bioretention media (Bradl 2004; Kabir *et al.* 2014; LeFevre *et al.* 2015). Nonetheless, retained metals accumulate in the bioretention media with an increasing operation time and their ultimate sorption behaviour remains uncertain (LeFevre *et al.* 2015). For example, changes of physico-chemical soil conditions or saturated water conditions can remobilize sorbed metals (Tedoldi *et al.* 2016; Costello *et al.* 2020). Moreover, as bioretention cells age, organic matter accumulates, soil texture changes, and preferential flow paths develop, which in turn may alter the metal removal capacity and hydraulic conductivity of bioretention systems (Mitchell Ayers & Kangas 2018, Costello *et al.* 2020).

All these variables make it difficult to anticipate the timespan in which bioretention systems sufficiently retain trace metals from seepage water in order to protect the receiving water bodies or fail to do so. Usually, the risk of metal leaching is assessed by effluent concentrations from underdrain samples or the outflow of soil columns in the laboratory (e.g., Kabir *et al.* 2014). However, especially in Europe, many bioretention systems are bottom-opened and the metal leaching potential cannot be assessed straightforwardly.

This study aims to investigate and describe the pollution status in soil water and soil matrix of three different long-term operated (>20 years) bioretention systems to assess the risk of heavy metal leaching. Therefore, we monitored influent and effluent metal concentrations at each site for nearly 2 years using a rain event-based measurement approach. Field measurements were complemented by physico-chemical soil properties determined in the laboratory. Ultimately, we want to contribute to a better understanding of how bioretention systems develop as they age and whether sufficient performance is ensured in order to mainstream the use of bioretention systems.

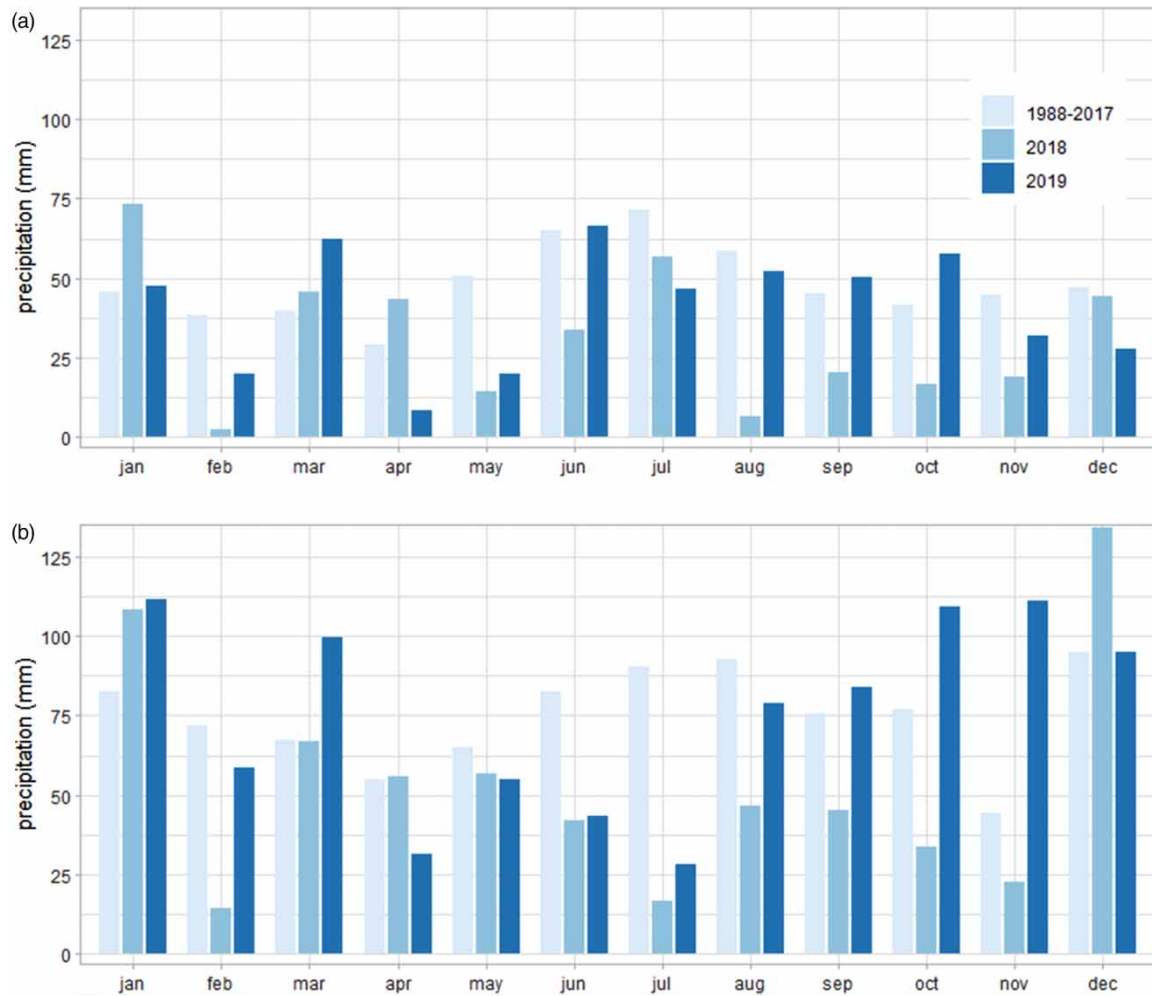
## MATERIALS AND METHODS

### Location and site conditions

All three investigated sites are located in Germany and can be classified as bioretention systems (also known as bioretention cells or areas – raingardens; biofilters or bioswales) according to the nomenclature proposed by Woods-Ballard *et al.* (2007). One site is located in north eastern Germany (Berlin, urban setting, site BS1) and two sites are located in central western Germany (Ruhr valley, urban setting and sub-urban setting, sites BS2 and BS3, respectively; Figure 2). The weather conditions during the measurement period were relatively dry, which is indicated by a pronounced drought from early summer to late autumn 2018 in both regions (Figure 1). In 2018, low annual precipitation rates of 376 mm in Berlin and 643 mm for the Ruhr area were observed (long-term mean: 577 mm Berlin and 899 mm Ruhr area, respectively). In 2019, annual precipitation for Berlin (490 mm) was still relatively low compared with the long-term mean because of low precipitation in February, April, and May. By contrast, annual precipitation in 2019 at the Ruhr area matched the long-term mean, but its monthly distribution was rather unusual with low rates in summer (June and July) and higher rates in autumn (October and November).

### Experimental site description

Site BS1 (42 m<sup>2</sup>) is located in a residential area and receives stormwater areally from the adjacent paved road (average annual daily traffic (AADT) <2,000; Sommer *et al.* 2002), car parking areas, and paved sidewalks. The ratio of the drainage area to the infiltration area accounts for 15.5:1. The system was put to operation in 1997 and consists of a 10 cm topsoil and 20 cm subsoil layer with an organic carbon content between 1.0 and 2.3 mass-%. The topsoil is bottom-lined by a water permeable geotextile and a gravel-filled trench zone with a throttled outflow to the sewage system to maintain surface drainage in the case of runoff supply exceeding the infiltration capacity. In direct vicinity to BS1 exists a test bioretention facility from an earlier research project



**Figure 1** | Monthly precipitation data in 2018 and 2019 in comparison to the long-term average (1988–2017) for Berlin (a) corresponding to BS1 and the Ruhr area (b) corresponding to BS2 and BS3. Data are provided by the German meteorological service (Deutscher Wetterdienst 2020) from the weather station 'Berlin-Tempelhof' (ID 433) and 'Essen-Bredeney' (ID 1303) for (a) and (b), respectively.

with an access shaft to sample seepage water directly from an underdrain (Sommer *et al.* 2002). Samples from this test facility were used as reference samples for our suction cup measurements.

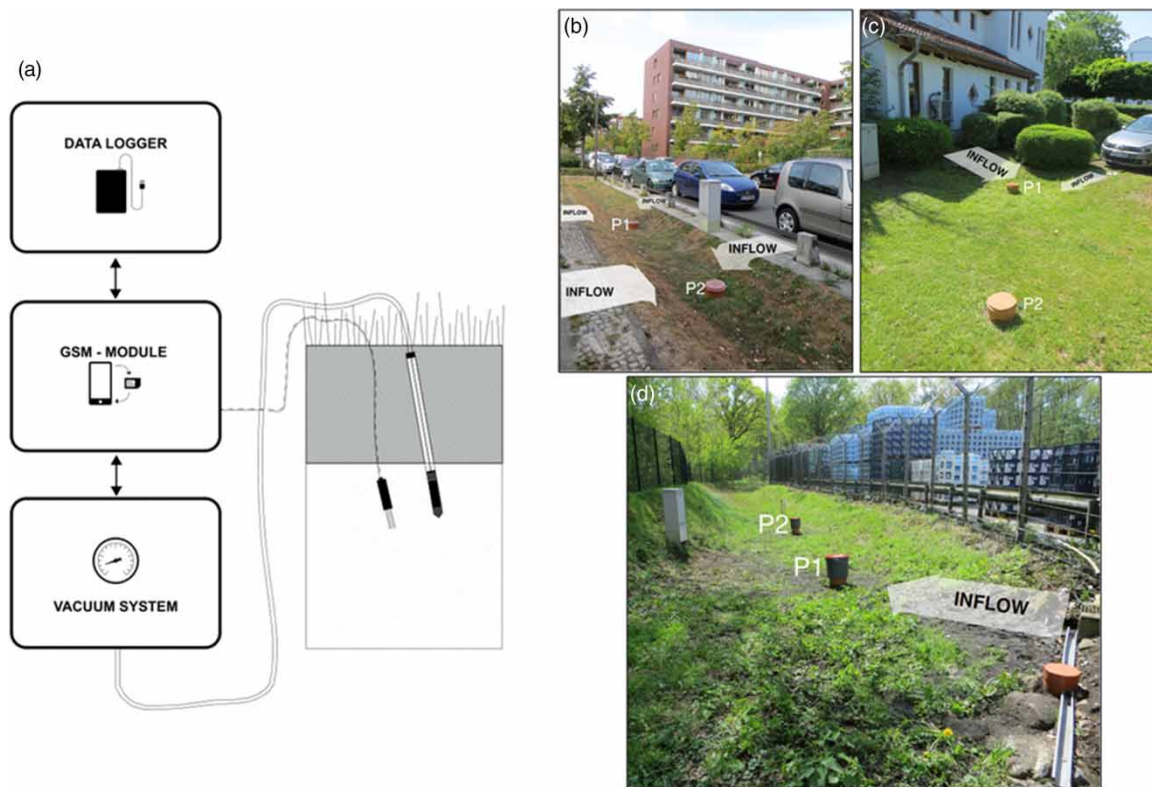
Site BS2 (140 m<sup>2</sup>) was constructed in 1995 and is located in a commercial area. The system receives stormwater runoff centralised by single inlets draining the manoeuvring area of a logistic company. The ratio of bioretention system size to the adjacent drained area is 18.4:1. During the 25 years of operation, a cone consisting of alluvial sediments accumulated around the inlet ( $C_{\text{ORG}} = 7.3\text{--}8.9 \text{ mass-}\%$ ). Underneath the sediment follows the original topsoil layer with an organic carbon content between 2.6 and 4.5 mass-%.

Site BS3 (257 m<sup>2</sup>) is located in a mixed used area (housing and commercial) and receives stormwater runoff from tile-covered roofs having zinc gutters. The ratio of the drained roof area to the infiltration area is 7:1. The system was constructed in 1995, and the 28 cm thick topsoil layer ( $C_{\text{ORG}} = 3.3\text{--}5.5 \text{ mass-}\%$ ) is followed by a gravel-filled trench zone similar to Site BS1. The site is characterised by a coarse texture because it is located on a historical bomb crater filled with ruin rubble. All three infiltration systems are bottom-opened systems, meaning they have a free draining lower boundary with a hydraulic connection to the groundwater body.

### Field measurement

To analyse metal concentrations in the effluent of the infiltration system, it was necessary to install a monitoring system in such a way that it would sample soil seepage water below the lower systems' boundary. We therefore used a novel measurement approach restricting soil water sampling to a given pore size fraction. A





**Figure 2** | (a) Schematic drawing of the measurement approach for seepage water sampling and (b–d) overview of the three experimental sites including measurement points and inflow directions (b = BS1, c = BS3, d = BS2).

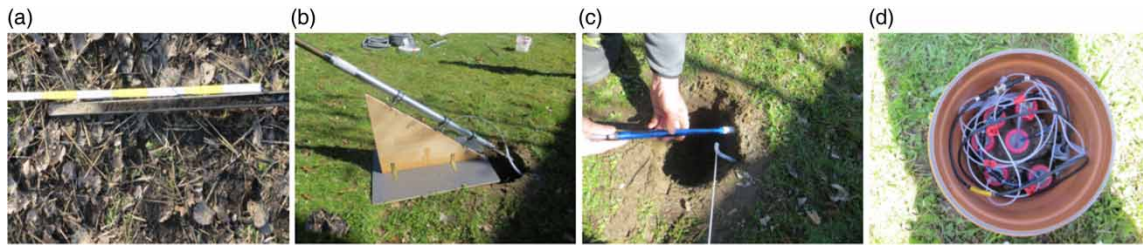
comprehensive description of the measurement approach is given by [Reck \*et al.\* \(2019\)](#) including a thorough technical description and testing. A simplified drawing of the technical approach illustrating the main components is shown in [Figure 2](#). The basic idea of this approach is to carry out operational soil water sampling using suction cups triggered by the actual soil moisture. In the case of an exceedance of the pre-set moisture threshold, a pump evacuates the vacuum system and soil water sampling starts.

In this case, the moisture threshold was set to equal the field capacity (moisture value at soil tension of  $-6.3$  kPa) plus offset of the specific soil to restrict sampling to drainage periods, i.e., the occurrence of gravitational dominated water flow generating deep seepage and potentially contaminant transport.

Each experimental site was equipped with the advanced sampling approach having the same technical equipment and configuration as described in [Reck \*et al.\* \(2019\)](#). Major features per site are: (i) two soil water sampling points, (ii) one to two soil moisture sensors (depth of 25–40 cm; ML2x Decagon Devices), (iii) a tipping bucket rain gauge (Kalyx, Environmental Measurements Ltd) installed 1 m above the soil surface, and (iv) one inflow collector. For areal inflow conditions (site BS1), both sampling points were situated in the swale depression centre, whereas for selective inflow conditions one sampling point was situated close to the stormwater inlet (distance of 2 m) and the other distant to the stormwater inlet (distance of 14 and 5 m at sites BS2 and BS3, respectively).

At each sampling point, we installed five suction cups (SPE20, Meter Group, inner nylon membrane and an outer protective polyethylene membrane; porosity of  $0.2$   $\mu\text{m}$ ; bubble point  $-80$  kPa) radially around the beforehand excavated access shaft with a given angle of  $45^\circ$  ([Figure 3\(b\)](#)). Hence, preferential infiltration along the suction cup is avoided, but installation depth is still controllable. Before cup installation, we prospected the lower boundary of the mollic horizon per measurement point to ensure the porous cup was situated exactly below this horizon (depth of 25–45 cm, [Figure 3\(a\)](#)) to sample percolating water beneath the zone of major trace metal retention. The pre-drilled hole for cup installation was  $0.2$  mm smaller in diameter to ensure a proper hydraulic connection between porous cups without the need of slurring. [Figure 3](#) illustrates the entire sequence of cup installation.

Because of the known variability in the soil water sampling rate between single cups ([Weihermüller \*et al.\* 2007](#)), we restricted the sampling volume to  $150$  ml event<sup>-1</sup> cup<sup>-1</sup> by a hydrophobic membrane valve installed



**Figure 3** | Sequence of suction cup installation with (a) soil prospection for installation depth setup, (b) pre-drilling with given angle and depth, (c) suction cup insertion, and (d) embedded soil water sampling infrastructure after finished cup installation.

upstream of the vacuum tube inside the sampling bottles. Hence, any bias in trace metal concentration due to diluting effects by single cups should be avoided.

Sampling of seepage water was carried out as followed: (i) each measurement station was operated autonomously, meaning sampling was initiated automatically in the case of a drainage period onset defined as a threshold exceedance of the default soil moisture threshold; (ii) the following soil water extraction was conducted the subsequent 4 h by a given vacuum regulated around  $-35$  hPa; and (iii) after sampling, the measurement station was deactivated upon sample collection to avoid any cross-contamination with subsequent precipitation events.

Trace metal concentrations were determined on composite samples per measurement point. Due to their porous membrane and a possible mechanical filtering of particulate substances, suction cups only allow to analyse the dissolved metal fraction in the seepage water. While composite samples were compounded from five collected suction cup samples, pH and EC were measured for each sample (MultiLine P3 Device, WTW Ltd). Additionally, the automatically triggered seepage water sampling was accompanied by a manual inflow sampling (from here on simply referred to as inflow). The inflow collector consisted of a glass bottle (1 l) located in a soil shaft close to the system's inlet in the case of selective inflow conditions or adjacent to the paved surfaces in the case of areal inflow conditions. During the collection of the seepage samples, two aliquots were taken from the runoff collector (30 ml each) and the collector was emptied afterwards. For this reason, inflow concentrations do not necessarily present the exact event concentration because of a possible sample contamination by preceding storm events without deep seepage or storm events after sampling initiation. Consequently, influent samples represent concentration estimates from the seepage sampling period but not the exact event concentration. Subsequently, all field-installed sampling bottles were washed with rinsing acid (10%  $\text{HNO}_3$ ) and ultrapure water ( $\text{EC} < 50 \mu\text{S cm}^{-1}$ ). Each water sample was acidified by one drop of 65% nitric acid and stored at  $4^\circ\text{C}$  until analysis.

### Laboratory measurements

Methods for soil-physical and -chemical property determination applied on the composite and undisturbed soil samples are summarised in Table 1. The pedologic site investigations were performed in September 2018 for sites BS2 and BS3, and for BS1 in May 2014 in a preceding study (Kluge *et al.* 2018). Thereby, the dug soil profiles were differentiated into horizons following the German soil classification scheme ('Bodenkundliche Kartieranleitung. KA5', Eckelmann *et al.* 2006), and soil samples were collected for each horizon for laboratory investigations.

Trace metal concentrations in water samples were analysed by ICP-OES following DIN EN ISO 11885 (2009). Soil seepage samples were analysed without any further pre-treatment. Inflow samples were analysed with following pre-treatment: one aliquot was filtered over a  $0.45 \mu\text{m}$  cellulose acetate filter, and one aliquot was digested with nitric acid (DIN EN ISO 15587-2 2002) before ICP-OES measurements ( $n = 2$ ; 2–6 blanks). Hence, it is possible to distinguish total and dissolved metal concentrations in the inflow. To compare with the existing literature on particulate metal fractions, we calculated a particulate fraction by subtraction of the median dissolved from median total concentration.

### Physical- and chemical-site properties

Table 2 shows relevant physico-chemical soil properties for topsoil layers at each bioretention system (both measurement points at BS2), respectively. The pH values for all sites and depths are found to be in a weakly acidic to neutral milieu, which meet the requirements of the German construction standards for bioretention

**Table 1** | Soil-physical and -chemical laboratory techniques as well as the numbers of analysed samples and replicates

Parameter	Method/Standard	Analysed samples (replicates)
$C_{ORG}$	Dry combustion at 550 °C and differential weighing (DIN ISO 10694 1995)	11 (2)
BD	Drying at 105 °C until constant weight (DIN EN ISO 11274 2014)	10 (3)
pH	Potentiometric in a 0.01 m $Ca_2Cl$ suspension (DIN ISO 10390 2005)	11 (1)
Soil texture	Wet sieving and sedimentation analysis (DIN ISO 11277 2002)	11 (1)
$K_{SAT}$	Water conductivity of saturated undisturbed soil core samples (DIN 19683-9 2012; DIN EN ISO 11274 2014)	8 (6)
Field capacity	Determination of the water retention characteristics – laboratory methods (ISO 11274:2019)	3 (5)
Pseudo total metal content	Aqua regia digestion following DIN ISO 11466 (1997)	66 (1)

**Table 2** | Physical and chemical properties of all investigated bioretention sites

Site	Depth (cm)	BD (g/cm <sup>3</sup> )	Soil texture	$C_{ORG}$ (mass.%)	$K_{SAT}$ (cm/d)	pH (-)	(mg/kg)					
							Cd	Cr	Cu	Ni	Pb	Zn
BS1	0–7	1.45	Sand	2.34	639	6.6	0.40	<5	17	5	23	79
	7–15	1.54	Sand	1.29	588	6.7	0.50	<5	17	5	26	67
	15–27	1.54	Sand	0.99	508	6.8	0.50	<5	16	5	25	57
BS2 P1	0–10	0.77	Sandy loam	8.87	1,911	6.9	0.82	36	80	24	66	1,015
	10–28	0.93	Sandy loam	7.29	1,610	7.0	1.08	43	110	31	91	944
	28–41	1.07	Sand	2.59	1,361	6.8	0.18	8	16	5	11	194
BS2 P2	0–9	0.96	Loamy sand	4.52	1,852	6.6	0.61	23	30	11	45	390
	9–21	1.29	Sand	2.97	1,401	6.5	0.50	18	13	5	30	120
	21–36	1.61	Loamy sand	1.61	469	6.4	–	–	–	–	–	–
BS3	0–6	0.84	Loamy sand	5.50	1,444	6.0	0.14	34	34	42	21	260
	6–28	0.86	Sand	3.29	2,963	6.4	0.04	33	28	41	10	37
Precautionary values for sand according to BBodSchV (1999)							0.4	40	30	20	15	60
Precautionary values for loam/silt according to BBodSchV (1999)							1	70	60	40	50	150

Soil texture classes are according to USDA soil taxonomy. BS2 – P1 represents the closest (3.5 m), and P2 the furthest (14 m) measurement point from the inlet. The upper two soil layers at BS2 – P1 consist of sediments.

facilities (pH between 6 and 8) and indicate optimal chemical soil conditions for the sorption of dissolved metals to the soil matrix (Bradl 2004; DWA A-138 2005; Alloway 2012). pH values in the seepage samples correspond to the solid-phase milieu with values of  $7.5 \pm 0.2$ ,  $6.7 \pm 1.4$ , and  $7.1 \pm 0.5$  for Sites BS1, BS2, and BS3, respectively. The electrical conductivity in the seepage samples was lowest at Site BS1 with  $219 \pm 58 \mu S cm^{-1}$  and twice as high at Sites BS2 and BS3 with  $419 \pm 111$  and  $465 \pm 143 \mu S cm^{-1}$ . The organic carbon content is between 1 and 3 mass-% for the topsoils on average (upper 10 cm). Extraordinary high  $C_{ORG}$  contents between 7.29 and 8.87 mass-% were measured for the upper two soil layers (sediment layers) at BS2 – P1, and for the surface soil layers at BS2 – P2 and BS3 with  $C_{ORG}$  contents of 4.52–5.50 mass-%, respectively. The saturated hydraulic conductivity ( $K_{SAT}$ ) represents a good basis to evaluate the hydrological performance of a bioretention system. The DWA A-138 (2005) specifies an infiltration rate between  $1 \times 10^{-6}$  and  $1 \times 10^{-5}$  m/s (~10 to ~10,000 cm/d, respectively) for bioretention systems in Germany. As shown in Table 2, all measured  $K_{SAT}$  values are within the recommended range. Overall, the bioretention systems show a good hydrological performance and likely maintain their ability to sufficiently infiltrate stormwater which is in line with research by Costello *et al.* (2020) and Spraakman *et al.* (2020).

### Runoff and seepage water sample overview

The collection of water samples (inflow and seepage) was conducted from spring 2018 to the end of 2019 at BS2 and BS3 and to February 2020 at BS1. After a defined initial phase, 149 seepage water samples were collected

and analysed during the sampling period (Table 3). Depending on the climatic conditions at the monitoring sites, this phase refers to the first five events at BS2 and first 2 months after the monitoring station setup of BS1 and BS3 (12 rejected samples in total). It was defined to ensure the development of a proper hydraulic connection between suction cups and soil matrix and to eliminate possible metal contamination of the equipment which may have occurred during installation. While both measurement points at BS1 (areal inflow of runoff water) collected an equal amount of seepage samples, BS2 and BS3, both sites with centralised runoff inflow, collected about twice as many samples at the measurement point close to inlet (P1), respectively. Seepage sampling at BS2 – P2 only occurred at higher precipitation events yielding sufficient runoff to trigger seepage generation at the inlet-distant (14 m) measurement point. At BS1, 14 seepage water samples were additionally collected from an underdrain (referred to as P3, see the section ‘Experimental site description’) and analysed in the same manner. Measured concentrations in these samples serve as reference metal concentrations to water on its passage towards the underlying groundwater body. For 75 individual seepage water measurement events, additional runoff water samples were collected and analysed for dissolved (filtered  $<0.45\ \mu\text{m}$ ) and, in case of adequate sample material, total metal concentrations (nitric acid digestion).

### Statistical data handling

For post-processing of all ICP-OES analyses, we subtracted the mean of blank runs from the respective measurements. Following Hornung & Reed (1990), measurements below the LOQ and LOD were substituted by half of the respective LOQ and LOD values. As indicated by Antweiler & Taylor (2008), the estimation of summary statistics (e.g., mean) for sample sets with  $>70\%$  censoring (values  $<\text{LOD}$ ) gives very imprecise estimates. For that reason, we restrained the calculation of a mean for coherent datasets with  $<70\%$  censored values (sum of values  $<\text{LOQ/LOD}$ ). Subsequently, for datasets with  $>70\%$  censored data, the median corresponds to a specific LOQ or LOD value (marked by ‘<’ sign), and no proceeding calculations or statistics were performed (e.g., the calculation of metal retention capacities or a particulate fraction in inflow water). Statistical parameters, the number of samples below the LOQ and the LOD, and the percentage of censored data are listed in Supplementary Material for inflow samples (Appendix A, Table 5) and seepage samples (Appendix A, Table 6), respectively.

Due to analytical uncertainties, the inflow water metal concentrations of the dissolved fraction exceeded the corresponding total fraction for some events. For these cases, we substituted the lower total value by the higher dissolved value of the same event.

To check for significant shifts in between sample populations, we applied the non-parametric Wilcoxon test because of skewed distributions. In the case of paired populations, the Wilcoxon signed-rank test was used and the Wilcoxon rank-sum test in case of non-paired populations. If the direction of the location shift in between sample sets could be presumed, we performed a one-sided test. In all other cases, we performed a two-sided test. The null hypothesis was rejected for  $p$ -values  $<0.05$ . All statistical tests were performed in R (R Core Team 2021) using the RStudio environment (RStudio Team 2021).

## RESULTS AND DISCUSSION

### Inflow concentrations and soil metal contents

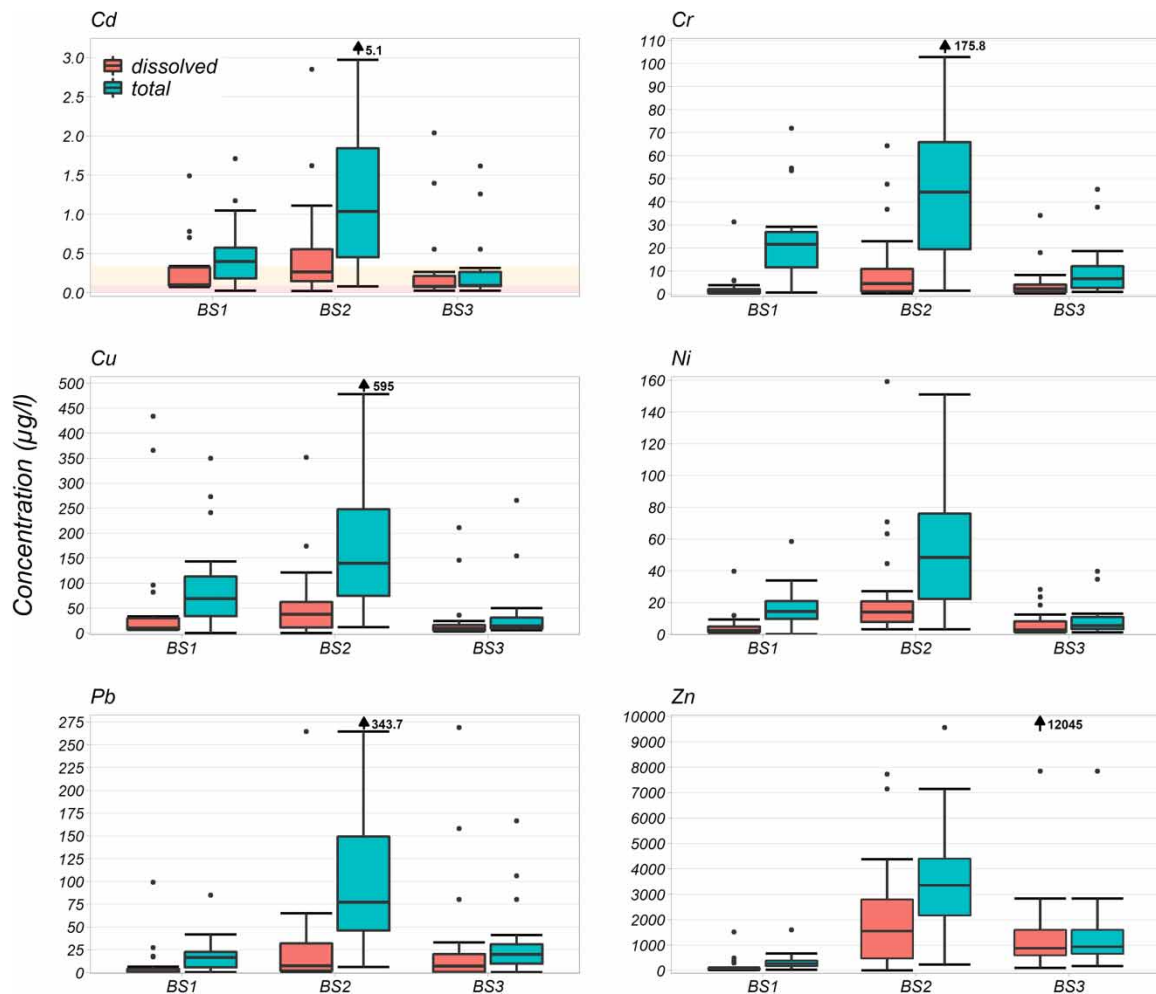
Generally, the drainage area type affects pollution loads in stormwater runoff, thus influencing the performance of a bioretention system (Tedoldi *et al.* 2016) and probably its lifecycle. Figure 4 shows dissolved ( $<0.45\ \mu\text{m}$ ) and

**Table 3** | Overview of collected seepage and runoff water samples for all sites and measurement points

Site	Seepage water						Inflow water	
	Total			Reliable samples <sup>a</sup>			Dissolved	Total
	P1	P2	P3	P1	P2	P3		
BS1	27	27	14	26	26	14	19	18
BS2	39	21		34	19		31	26
BS3	32	15		29	15		25	21

<sup>a</sup>Number of samples after the initial phase.





**Figure 4** | Dissolved and total metal concentrations of the inflow water at the monitoring sites. Arrows and values at top of each subplot mark extreme outliers.

total (dissolved and particulate) metal concentrations measured in the inflow of each site. Measured inflow metal concentrations clearly differ between the investigated sites. In general, highest concentrations were found at BS2 for all metals regarding individual metals as well as the dissolved and total fractions. Lowest inflow concentrations were found at BS1. Concerning the contamination levels, Cu and Zn were identified as the major contaminants. Because dissolved metal concentrations for Cd and Pb at BS1 and Cd at BS3 exceeded 70% of censored data, no particulate fractions were calculated (cf. Supplementary Material, Appendix A, Table 5).

Differences between total and dissolved fractions of all metal inflow concentrations were found especially at BS2 and BS1, indicating a high proportion of particulate metals. Depending on the metal, the particulate fraction equals at least 54% of the total inflow concentrations (Zn at BS2), which corroborates metal fractions in street runoff measured by Lange *et al.* (2020). By contrast, BS3 is mainly charged by roof runoff and hence differences in between dissolved and total fractions were rather minor, indicating that most metals were dissolved. Cu and Zn predominate in the dissolved fraction at site BS3 with 67 and 93%, respectively. Lindfors *et al.* (2017) determined a dissolved fraction of 92% for Zn and a dissolved Cu fraction of 11% in runoff from zinc roofs. Hence, our ratio of dissolved Cu is rather in line with dissolved Cu fractions (73%) reported by Lindfors *et al.* (2017) for copper roofs.

BS1 is located in a residential area and receives runoff from the adjacent sidewalk and road, including a parking lot strip in between the road and bioretention system (<2,000 AADT; Sommer *et al.* 2002). Median metal concentrations of the inflow water for Cd, Cr, Cu, Ni, Pb, and Zn were 0.4, 21.6, 68.9, 14.3, 16.5, and 279.2 µg/l for total and <0.2, 0.9, 10.0, 2.5, <1.3, and 57.7 µg/l for the dissolved fraction, respectively. In comparison to a review by Huber *et al.* (2016), most metal concentration values measured in the inflow comply with runoff concentrations summarised for roads with an AADT of <5,000. Only Zn concentrations rather match those reviewed for parking lots, which are applicable for the runoff area at BS1.

BS2 is charged by runoff from a commercial area, which is characterised by an intense logistic vehicle manoeuvring. Unsurprisingly, metal concentrations at BS2 were significantly higher compared with BS1 and BS3, since trace metals in urban areas mainly originate from traffic activities (Zn, Cu, Cd, Pb, and Ni; Horstmeyer *et al.* 2016; Müller *et al.* 2020), especially at spots with high braking, acceleration, and deceleration activities (Loganathan *et al.* 2013). Median metal concentrations of the inflow water for Cd, Cr, Cu, Ni, Pb, and Zn were 1.0, 44.1, 139.8, 48.5, 77.0, and 3,345.8 µg/l for total and 0.3, 4.5, 37.7, 14.0, 7.3, and 1,553.0 µg/l for dissolved fraction, respectively. These values of dissolved Cr, Cu, Ni, and Pb concentrations correspond to median concentrations of non-urban highways with an AADT of >30,000 (representing the most polluted category for these metals) considering the classification by Huber *et al.* (2016). If considering their total concentrations, Cr, Cu, Ni, and Pb exceed the values of the category non-urban highways double to fourfold and Zn concentrations rather equally reported runoff from zinc roofs (Göbel *et al.* 2007).

The metal concentrations of BS3 at the inflow are comparable to BS1 with some restrictions. Zn is found to be significantly higher (factor 14 in dissolved fraction), which probably relates to the zinc gutters. Overall, Zn is the main contaminant and is found to be mainly dissolved (93% referring to median concentrations). Median metal concentrations of the inflow water for Cd, Cr, Cu, Ni, Pb, and Zn were <0.2, 6.6, 14.3, 5.5, 19.9, and 938.2 µg/l for total and <0.2, 2.2, 9.5, 2.7, 7.1, and 874.0 µg/l for dissolved fraction, respectively. Cu concentrations coincide with respective literature values found for zinc roof runoff (Lindfors *et al.* 2017), whereas Zn is about 14 times lower. At BS3, only the roof gutters are made of zinc and not the roof itself explaining the lower Zn values in our case. However, Cd, Cr, and Ni concentrations match reported concentrations in runoff from roofs with zinc gutters (Göbel *et al.* 2007).

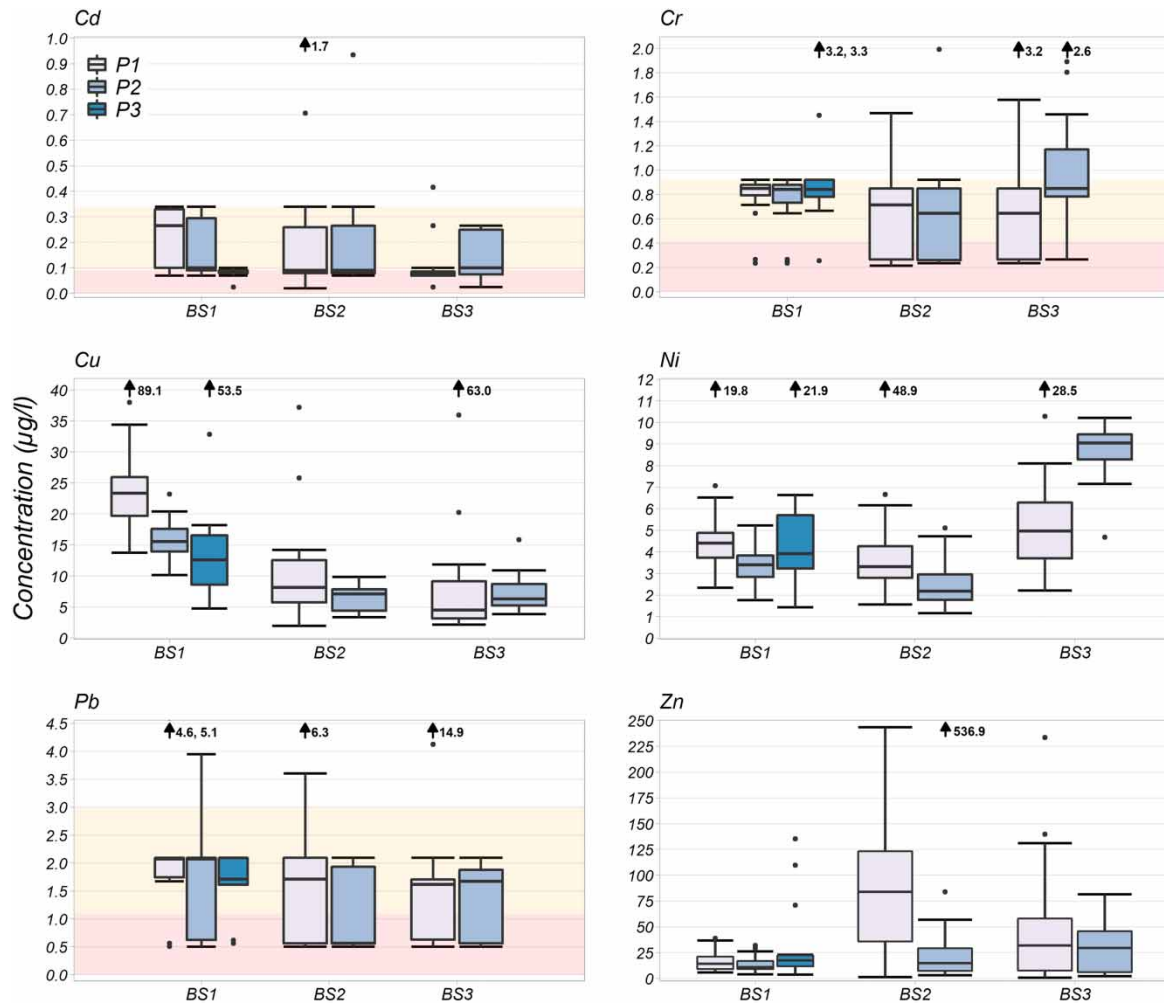
The soil metal contents in between the sites decrease in the order BS2 > BS3 > BS1, which reflects the influence of the presented inflow concentrations (Table 2). At BS2, highest soil metal contents were found close to the inlet point (P1) and significantly decreased at 14 m (Point P2), which was also observed in other studies with a focus on contamination patterns in bioretention systems with selective inflow conditions (Napier *et al.* 2009; Jones & Davis 2013; Tedoldi *et al.* 2017). At BS2 – P1, all trace metals in the sedimented layer (0–28 cm depth) exceeded the respective precautionary values of the German Federal Soil Protection and Contamination Ordinance (Table 2; BBodSchV 1999), except for Ni. Compared with BS2, metal concentrations in the topsoil layers at BS3 and BS1 are significantly lower, and only single elements (Pb, Zn at BS1 and Cu, Ni, Pb, Zn at BS3) exceed the precautionary values for sandy soils. Higher metal contents at BS2 might originate from road-deposited sediments (RDS) in the sedimented cone. RDS show very high metal concentrations (Huber *et al.* 2016; Kim & Ko 2020) but at the same time also high sorption capacities (Robertson & Taylor 2007; Kluge *et al.* 2014). Measured heavy metal concentrations in the sediment cone at BS2 – P1 were 1, 110, 31, 91, and 944 mg/kg for Cd, Cr, Cu, Ni, Pb, and Zn (Table 2). These values agree with recently reported metal concentrations in RDS (Kim & Ko 2020).

Regarding the vertical metal distributions, we observed decreasing metal contents with increasing soil depth (Table 3), which matches observations for bioretention facilities as reviewed by Tedoldi *et al.* (2016). Surprisingly, at the most polluted spot BS2 – P1, the concentration maximum for all metals except for Zn was found in the lower sediment layer (10–28 cm depth). Such an increase of metal contents with depth might relate to: (i) higher physical filtration of particulate metals in this layer and (ii) to variations in inflow metal concentrations during the operation time. Under the assumption of enhanced sorption capacities in RDS, the sediments at BS2 are likely able to additionally bind dissolved metals from the infiltrating water via adsorption. This mechanism might explain the peak of metal contents in the lower sediment layer.

### Soil seepage metal concentration

As a result of site-specific soil conditions, drainage area types, and inflow conditions (centralised or areal), measured seepage water concentrations clearly differ in inter-site comparison (Figure 5). Generally, Cd, Cr, and Pb were determined at low concentration levels for each site and measurement point. As marked by the coloured domains in the subplots of Figure 5, almost the entire datasets of Cd, Cr, and Pb values were found to fall below the LOQ (orange range) or even below the LOD (red range). Therefore, the individual absolute values of Cd, Cr, and Pb in seepage water are at very low concentration levels.

Zn and Cu were found to be the major contaminants in terms of their absolute values reflecting the high inflow concentration at each site. In case of centralised inflow conditions, only Zn at BS2 and Ni at BS3 revealed spatially significantly different seepage concentrations if comparing the paired samples (P1 versus P2) for each event that generated seepage flows (Wilcoxon signed-rank test, one-sided). With a median concentration of



**Figure 5** | Distribution of metal seepage concentration for each bioretention system divided element-wise and grouped by the measurement point. P3 represents water samples of the underdrain at BS1. Arrows and values at the top of subplots mark extreme outliers. For Cd, Cr, and Pb subplots, the red layer marks the average LOD range and yellow marks the average range between the LOD and the LOQ, for which the values are set to half of the receptive limit. For Cu, Ni, and Zn, LOQ and LOD ranges are not illustrated because not a single value was measured below the LOD or LOQ for these elements.

83.9  $\mu\text{g/l}$ , highest Zn concentrations were found close to the inlet (P1) at BS2. Substantially lower median concentrations (14.9  $\mu\text{g/l}$ ) were observed at the inflow-distant measurement point (P2). *Vice versa*, we observed an increase of Ni at BS3 from 5.0 to 9.1  $\mu\text{g/l}$  between P1 and P2. Median Cu concentrations were not significantly shifted between both measurement points at both sites having centralised inflow conditions. Due to the first flush effect in urban runoffs (Li *et al.* 2012), we expected a clear decrease in seepage concentrations with distance for all metals since the high initial loads are more likely to reach the point next to the inflow. However, considering Ni at Site BS3 suggests that local shifts in soil conditions, especially the  $C_{\text{ORG}}$  and solid-phase metal contents, mask the influence of inflow concentrations on seepage qualities. For example, at BS1, Ni and Cu concentrations differed significantly between P1 and P2 regardless of the areal inflow conditions. Such small-scale spatial deviations in seepage qualities instead of homogeneous inflow concentrations confirm the likely influence of local soil properties on metal discharge. Highest Zn seepage concentrations at BS2 – P1 mirror the highest inflow concentrations. *Vice versa* for Cu where inflow concentrations were significantly higher at BS2 compared with BS1. However, we observed highest Cu concentrations at BS1 – P1. High  $C_{\text{ORG}}$  content at the bioretention media of BS2 might increase the sorption of Cu because of its strong affinity to organic matter (Bradl 2004). At the same time, adsorbed metals can be remobilized and transferred downwards by the formation of soluble chelate complexes with dissolved organic matter (DOM) – a mechanism especially affecting Cu and Pb (Bradl 2004; Tedoldi *et al.* 2016). Within the soil, DOM originates mostly from microbial or root activity and during the degradation of organic matter (Pitt *et al.* 1999) causing a seasonal fluctuation in metal mobilisation. Li & Davis (2008)

as well as Trowsdale & Simcock (2011) observed DOM-mediated Cu discharges in summer periods during heavy rain events after droughts. These constellations were rather found at BS1 in both summers and may explain the highest Cu values in seepage water.

Ni concentrations were below 10.0 µg/l on average. For BS1 and BS2, median Ni concentrations are roughly similar in site comparison and between measurement points. By contrast, highest median Ni concentrations were observed for BS3 – P2 with 9.1 µg/l compared with 5.0 µg/l at BS3 – P1. Generally, BS3 showed the highest solid-phase metal contents of all three experimental sites (see Table 2). In combination with the low inflow concentrations, the chemical equilibrium is shifted towards the solid phase at BS3, and Ni concentrations rather reflect re-mobilised Ni from the soil matrix. Thus, relatively high seepage concentrations are rather a result of the high Ni content in the bioretention soil media.

Measurement point BS1 – P3 represents samples of the underdrain. Thus, these samples equal the seepage water quality after a complete percolation through the bioretention media. In comparison to our suction cup measurements (BS1 – P1 and P2) to BS1 – P3, the Wilcoxon test (Wilcoxon signed-ranks test, two-sided) demonstrates no significantly different median between measurement points P1 or P2 to P3. In the case of Cu, P1 deviates significantly from P3 but not from P2. In cases of Cd, Cr, and Pb, >70% of the concentration values of BS1 are <LOD/LOQ, and comparing these values is statistically uncertain. Nonetheless, the good fit of seepage median concentrations (P1 and P2) compared with P3 reveals that our *in situ* sampling approach yields representative seepage quality measurements for the lower boundary of the bioretention systems. Consequently, our measurement approach is applicable to assess the quality of seepage after its percolation through the biologically active soil layer.

Overall, all median heavy metal concentrations are below the threshold values of the path soil to groundwater of the German Federal Soil Protection and Contamination Ordinance (BBodSchV 1999). Consequently, our 2-year monitoring gave no quantitative evidence that long-term operated bioretention systems fail to perform and our sampling raised no demonstrable risk of ecotoxicological impacts considering environmental quality standards.

However, bioretention facilities are characterised by high hydrological loads and massively enhance the groundwater recharge rate. Thus, high loads of metals can be transferred into receiving waters as mentioned, for example, by Werkenthin *et al.* (2016) for roadside swales. For this reason, seepage concentrations need to be judged in combination with underlying soil media and the site-specific groundwater recharge rates to obtain realistic metal loads.

### Metal retention capacities

The metal retention performance of each site was analysed by comparing metal concentrations in the influent and effluent. For centralised inflow conditions (BS2 and BS3), we used the measurement point close to the stormwater inlet (P1) and mean values averaged from both measurement points for areal inflow condition (BS1). Retention calculations were restrained in the dissolved inflow fraction, since suction cups sample only the dissolved fraction. We gradually assessed metal retention capacities, since measured inflow and seepage concentrations do not have the same level of representativeness.

In the first step, we statistically compared the distributions applying the Wilcoxon test to check for significant shifts between both distributions (Table 4). Except for Cu at Site BS1, all distributions show a significant shift in the location between the influent and effluent concentrations. All shifts are negative, except for Ni, at Sites BS1 and BS3. Transferred to our investigated bioretention sites, this difference in distributions implies a highly significant reduction in Zn concentrations during the soil transfer at all sites, and the significance of reduction decreases in the order Zn > Cu > Ni. In inter-site comparison, the concentration shift between inflow and seepage decreases in the order BS2 > BS3 > BS1.

In the second step, we plotted the pairs of dissolved inflow and seepage concentrations per event to investigate the retention performance in more detail (Figure 6).

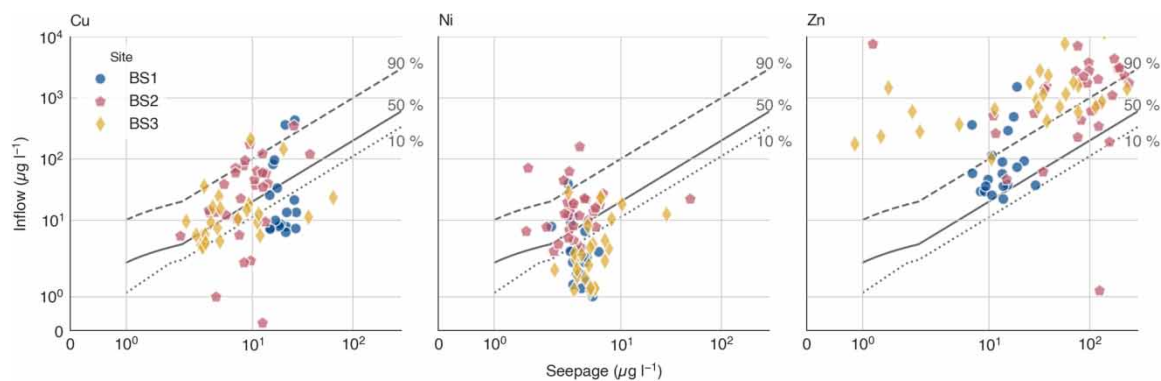
The retention capacities show a wide scatter ranging from 0 to >90% for all metals and all sites. In line with the comparison of the distributions, the event-wise inflow and seepage concentrations mirror the shift in location and its significance as proposed by the Wilcoxon test. In the overall site comparison, retention capacities decrease in the order BS2 > BS3 > BS1. Minor retention capacities were found for Ni and might be a result of re-mobilised metals from the soil matrix as discussed before. All sites retain Zn efficiently with 75% of all events (56 of 74) demonstrating retention rates >80%. With 50% of all events (37 of 74) showing a reduction >50%, Cu was retained to a high extent as well. Such a very high retention of Zn and Cu agrees with findings of Davis *et al.*



**Table 4** | Results of the statistical comparison between the distributions of seepage and dissolved inflow concentrations applying the Wilcoxon rank-sum test

Site	Element	Difference in location
BS1	Cu	7.76'
	Ni	1.40*
	Zn	-39.73***
BS2	Cu	-26.94***
	Ni	-9.99***
	Zn	-1,510.81***
BS3	Cu	-4.01**
	Ni	1.91*
	Zn	-831.56***

The difference in location denotes the median of the difference between the sample from the seepage and inflow concentrations. Negative values indicate a concentration reduction from the influent to the effluent and *vice versa* for positive values. Levels of significance are \*\*\* $p$ -value <0.001, \*\* $p$ -value <0.01, \* $p$ -value <0.05, '  $p$ -value <0.1.

**Figure 6** | Event-pairs of dissolved inflow and seepage concentrations constrained by the element and the experimental site. The dotted, solid, and dashed lines equal a retention of 10, 50, and 90%, respectively. Note the logarithmically scaled axes.

(2003), Kabir *et al.* (2014) and Lange *et al.* (2020). Transposing the scatter of retention rates to a mean metal reduction, these values suggest an averaged reduction of Ni by 23, 78, and 25%, of Cu by 66, 82, and 82%, as well as Zn by 92, 96, and 98% at Sites BS1, BS2, and BS3, respectively. Considering the long-term operation time (>20 years) of the three facilities, it can be concluded that Cu, Ni, and Zn are still retained efficiently by the soil media.

Surprisingly, the highest retention capacities of all three experimental sites coincide with the highest solid-phase content (BS2). This point is of major importance because it demonstrates the necessity to integrate effluent measurements at the lower system boundary (e.g., seepage sampling) to reliably assess the risk of groundwater pollution. The environmental legislations in many European countries only use total metal soil contents to derive mitigation strategies for environmental and human health exposure (Kumpiene *et al.* 2017). For the groundwater pollution assessment, a greater risk may be deduced from higher metal contents or *vice versa*. However, such conclusions might be misled as our findings suggest. Moreover, findings at BS2 indicate that the accumulation of RDS around the inlet improved the metal retention performance due to a continuous supply of new material with high  $C_{TOT}$  contents in conjunction with numerous sorption sites. These findings agree with Robertson & Taylor (2007) and Kluge *et al.* (2014), who found high adsorption capacities in RDS. Altogether, it may suggest that some kind of 'dirt-cleans-dirt' effect exists, where the inflow of polluted RDS and their sedimentation around the inlet of a bioretention system improve the local retention capacity. Ultimately, it seems to be an ambivalent situation, because on the one hand the fate of accumulated heavy metals remains unclear (Tedoldi *et al.* 2017) and high metal concentration in RDS (Loganathan *et al.* 2013; Huber *et al.* 2016) may pose a growing threat for the underlying groundwater.

Admittedly, our mode of sampling inflow water must be critically reflected. Metal concentrations in stormwater runoff clearly vary during a rain event. Large loads of metals usually occur during the initial runoff hydrograph

referring to the so-called first flush effect (Li *et al.* 2012). Based on our simplified runoff sampling (single bottle sampling per event), it is unknown whether the measured inflow concentrations exactly represent the measured seepage concentrations. For that reason, our deduced retention performance might, to some extent, change in the case of more precise inflow measurements. Another question regarding the retention capacity exists in terms of the particulate metal fraction in the inflow. If considering the total inflow concentration in calculating the retention fraction yields an averaged reduction of Ni by 75, 91, and 45%, of Cu by 78, 94, and 88%, as well as Zn by 96, 98, and 98% for the Sites BS1, BS2, and BS3, respectively. Compared with the dissolved fraction, these values suggest that especially the elements Ni and Cu might be retained to a higher degree. However, the calculation of the total retention includes the assumption that particulate metals in the inflow are removed totally via mechanical filtration, because seepage samples driven by suction cups can only sample the dissolved metal fraction ( $<0.45 \mu\text{m}$ ).

## CONCLUSIONS

This study seeks to improve our understanding of the behaviour of bioretention systems with increasing operation time regarding heavy metal transfer processes. To investigate the metal retention of three long-term operated bioretention systems, we installed an automated and event-driven seepage water monitoring system to investigate the dissolved and thus mobile metal fraction in the leachate. The monitoring was conducted for nearly 2 years and included additional inflow sampling for each precipitation event-generating seepage. The study was further complemented by investigations of physico-chemical soil properties of the bioretention media. Regarding the hydraulic performance, all sites are in a good condition and meet the requirements by the German standards for bioretention facilities (DWA A-138 2005).

Results of this study reveal that the inflow water concentration and subsequently the soil media metal content are both mainly determined by the drainage area type (commercial versus residential area). Moreover, stormwater from highly trafficked areas conveys many RDS, which accumulate around the inlet in facilities with selective inflow conditions and build-up a local sediment cone with high  $C_{\text{TOT}}$  and metal contents. Regardless of the site, Cu and Zn were identified as the major pollutants among the investigated metals. The metal content in the solid phase generally decreased with increasing depth, which may indicate the high retention of dissolved metals within the topsoil layers or in case of particulate metals a sufficient physical filtration. The legislative trigger values for the pathway soil to groundwater by the German Federal Soil Protection and Contamination Ordinance (BBodSchV 1999) were exceeded very rarely and only for single events and elements during the measurement period. In contrast to the inflow concentrations and the metal media contents, the soil seepage concentrations did not reflect the drainage area type and the magnitude of measured concentrations was similar between the sites. Consequently, the calculated retention capacities were found to be highest in the bioretention system installed at the commercial area with very high inflow concentrations, which we attributed to the sedimentation of RDS that deliver many sorption sites.

Overall, the major pollutants of Cu and Zn were retained efficiently at all investigated sites. In conclusion, all three investigated bioretention systems fulfil their expected functionality regarding the infiltration of runoff water and their retention of metals after more than 20 years of operation. However, to better understand which factors might change the high retention performance, these investigations should be expanded to more bioretention systems of different designs and drainage area types. Additionally, systematic measurements of inflowing metal concentrations are required to better assess the retention performance of nature-based solutions for stormwater management.

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## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

## REFERENCES

- Ahiablame, L. M., Engel, B. A. & Chaubey, I. 2012 Effectiveness of low impact development practices: literature review and suggestions for future research. *Water, Air, & Soil Pollution* **223**, 4253–4273. doi:10.1007/s11270-012-1189-2.
- Alloway, B. J. 2012 *Heavy Metals in Soils: Trace Metals and Metalloids in Soils and Their Bioavailability Bd. 22*. Springer Science & Business Media. doi:10.1007/978-94-007-4470-7.
- Antweiler, R. C. & Taylor, H. E. 2008 Evaluation of statistical treatments of left-censored environmental data using coincident uncensored data sets: I. Summary statistics. *Environmental Science & Technology* **42** (10), 3732–3738. doi: 10.1021/es071301c.
- BBodSchV. 1999 *Bundes-Bodenschutz- und Altlastenverordnung vom 12. Juli 1999 BGBl. I S. 1554, die Zuletzt durch Artikel 3 Absatz 4 der Verordnung vom 27. September 2017 BGBl. I S. 3465 geändert worden ist. (Federal Soil Protection and Contaminated Sites Ordinance)*. Bundesministerium für Umwelt, Naturschutz und nukleare Sicherheit BMU, Berlin, Germany.
- Bradl, H. B. 2004 Adsorption of heavy metal ions on soils and soils constituents. *Journal of Colloid and Interface Science* **277**, 1–18. doi:10.1016/j.jcis.2004.04.005.
- Clark, S. E. & Pitt, R. 2012 Targeting treatment technologies to address specific stormwater pollutants and numeric discharge limits. *Water Research* **46**, 6715–6730.
- Costello, D. M., Hartung, E. W., Stoll, J. T. & Jefferson, A. J. 2020 Bioretention cell age and construction style influence stormwater pollutant dynamics. *Science of the Total Environment* **712**, 135597. doi:10.1016/j.scitotenv.2019.135597.
- Davis, A. P., Shokouhian, M., Sharma, H., Minami, C. & Winogradoff, D. 2003 Water quality improvement through bioretention: lead, copper, and zinc removal. *Water Environment Research* **75**, 73–82. doi:10.2175/106143003(140854).
- Davis, A. P., Traver, R. G. & Hunt, W. F. 2010 Improving urban stormwater quality: applying fundamental principles. *Journal of Contemporary Water Research & Education* **146**, 3–10.
- Deutscher Wetterdienst. 2020 Historical and recent daily climate data of the measurement stations ‘Berlin-Tempelhof’ ID 433, and ‘Essen-Bredeneu’ ID 1303. Recalled on 28th March 2020. Available from: [https://opendata.dwd.de/climate\\_environment/CDC/observations\\_germany/climate/daily/more\\_precip/](https://opendata.dwd.de/climate_environment/CDC/observations_germany/climate/daily/more_precip/).
- Dietz, M. E. 2007 Low impact development practices: a review of current research and recommendations for future directions. *Water, Air, and Soil Pollution* **186**, 351–363. doi:10.1007/s11270-007-9484-z.
- DIN 19683-9. 2012 *Soil Quality – Physical Laboratory Tests – Part 9: Determination of the Saturated Hydraulic Water Conductivity in the Cylindrical Core-Cutter*. Beuth Verlag, Berlin, Germany.
- DIN EN ISO 11274. 2014 *Soil Quality – Determination of the Water-Retention Characteristic – Laboratory Methods ISO 11274:1998+Cor 1:2009; German Version EN ISO 11274:2014*. Beuth Verlag, Berlin, Germany.
- DIN EN ISO 11885. 2009 *Water Quality – Determination of Selected Elements by Inductively Coupled Plasma Optical Emission Spectrometry ICP-OES; German Version EN ISO 11885:2009*. Beuth Verlag, Berlin, Germany.
- DIN EN ISO 15587-2. 2002 *Water Quality – Digestion for the Determination of Selected Elements in Water – Part 2: Nitric Acid Digestion ISO 15587-2:2002; German Version EN ISO 15587-2:2002*. Beuth Verlag, Berlin, Germany.
- DIN ISO 10390. 2005 *Soil Quality – Determination of pH ISO 10390:2005*. Beuth Verlag, Berlin, Germany.
- DIN ISO 10694. 1995 *Soil Quality – Determination of Organic and Total Carbon After Dry Combustion Elementary Analysis ISO 10694:1995*. Beuth Verlag, Berlin, Germany.
- DIN ISO 11277. 2002 *Soil Quality – Determination of Particle Size Distribution in Mineral Soil Material – Method by Sieving and Sedimentation ISO 11277:1988+ISO 11277:1998/Cor.1:2002*. Beuth Verlag, Berlin, Germany.
- DIN ISO 11466. 1997 *Soil Quality – Extraction of Trace Elements Soluble in Aqua Regia ISO 11466:1995*. Beuth Verlag, Berlin, Germany.
- DWA-A 138. 2005 *Planung, Bau und Betrieb von Anlagen zur Versickerung von Niederschlagsabwasser (Design, Construction and Operation of Rainwater Drainage Systems)*. Deutsche Vereinigung für Wasserwirtschaft, Abwasser und Abfall e.V., Hennef, Germany.
- Eckelmann, W., Sponagel, H., Grotenthaler, W., Hartmann, K.-J., Hartwich, R., Janetzko, P., Joisten, H., Kühn, D., Sabel, K.-J. & Traidl, R. 2006 *Bodenkundliche Kartieranleitung. KA5. (Manual of Soil Mapping. 5th Edition (KA5))*. Schweizerbart Science Publishers, Stuttgart, Germany.
- Fletcher, T. D., Shuster, W., Hunt, W. F., Ashley, R., Butler, D., Arthur, S., Trowsdale, S., Barraud, S., Semadeni-Davies, A., Bertrand-Krajewski, J.-L., Mikkelsen, P. S., Rivard, G., Uhl, M., Dagenais, D. & Viklander, M. 2015 *SUDS, LID, BMPs, WSUD and more – the evolution and application of terminology surrounding urban drainage*. *Urban Water Journal* **12**, 525–542. doi:10.1080/1573062X.2014.916314.
- Göbel, P., Dierkes, C. & Coldewey, W. G. 2007 Storm water runoff concentration matrix for urban areas. *Journal of Contaminant Hydrology* **91**, 26–42. doi:10.1016/j.jconhyd.2006.08.008.
- Hornung, R. W. & Reed, L. D. 1990 Estimation of average concentration in the presence of nondetectable values. *Applied Occupational and Environmental Hygiene* **5** (1), 46–51. doi:10.1080/1047322X.1990.10389587.

- Horstmeyer, N., Huber, M., Drewes, J. E. & Helmreich, B. 2016 Evaluation of site-specific factors influencing heavy metal contents in the topsoil of vegetated infiltration swales. *Science of the Total Environment* **560–561**, 19–28. <https://doi.org/10.1016/j.scitotenv.2016.04.051>.
- Huber, M., Welker, A. & Helmreich, B. 2016 Critical review of heavy metal pollution of traffic area runoff: occurrence, influencing factors, and partitioning. *Science of the Total Environment* **541**, 895–919. doi:10.1016/j.scitotenv.2015.09.033.
- Jones, P. S. & Davis, A. P. 2013 Spatial accumulation and strength of affiliation of heavy metals in bioretention media. *Journal of Environmental Engineering* **139**, 479–487. doi:10.1061/asceee.1943-7870.0000624.
- Kabir, M. I., Daly, E. & Maggi, F. 2014 A review of ion and metal pollutants in urban Green water infrastructures. *Science of the Total Environment* **470**, 695–706. doi:10.1016/j.scitotenv.2013.10.010.
- Kayhanian, M., Fruchtmann, B. D., Gulliver, J. S., Montanaro, C., Ranieri, E. & Wuertz, S. 2012 Review of highway runoff characteristics: comparative analysis and universal implications. *Water Research* **46**, 6609–6624. doi:10.1016/j.watres.2012.07.026.
- Kim, D.-G. & Ko, S.-O. 2020 Road-deposited sediments mediating the transfer of anthropogenic organic matter to stormwater runoff. *Environmental Geochemistry and Health*, 1–15. doi:10.1007/s10653-020-00703-2.
- Kluge, B., Werkenthin, M. & Wessolek, G. 2014 Metal leaching in a highway embankment on field and laboratory scale. *Science of the Total Environment* **493**, 495–504. doi:10.1016/j.scitotenv.2014.05.120.
- Kluge, B., Markert, A., Facklam, M., Sommer, H., Kaiser, M., Pallasch, M. & Wessolek, G. 2018 Metal accumulation and hydraulic performance of bioretention systems after long-term operation. *Journal of Soils and Sediments* **18**, 431–441. <https://doi.org/10.1007/s11368-016-1533-z>.
- Kumpiene, J., Giagoni, L., Marschner, B., Denys, S., Mench, M., Adriaensen, K., Vangronsfeld, J., Puschnreiter, M. & Renella, G. 2017 Assessment of methods for determining bioavailability of trace elements in soils: a review. *Pedosphere* **27**, 389–406. <https://doi.org/10.1016/S1002-01601760337-0>.
- Lange, K., Österlund, H., Viklander, M. & Blecken, G.-T. 2020 Metal speciation in stormwater bioretention: removal of particulate, colloidal and truly dissolved metals. *Science of The Total Environment* **724**, 138121. doi:10.1016/j.scitotenv.2020.138121.
- LeFevre, G. H., Paus, K. H., Natarajan, P., Gulliver, J. S., Novak, P. J. & Hozalski, R. M. 2015 Review of dissolved pollutants in urban storm water and their removal and fate in bioretention cells. *Journal of Environmental Engineering* **141**. doi:10.1061/asceee.1943-7870.0000876.
- Li, H. & Davis, A. P. 2008 Heavy metal capture and accumulation in bioretention media. *Environmental Science & Technology* **42**, 5247–5253. <https://doi.org/10.1021/es702681j>.
- Li, W., Shen, Z., Tian, T., Liu, R. & Qiu, J. 2012 Temporal variation of heavy metal pollution in urban stormwater runoff. *Frontiers of Environmental Science & Engineering* **6**, 692–700. <https://doi.org/10.1007/s11783-012-0444-5>.
- Lindfors, S., Österlund, H. & Viklander, M. 2017 Truly dissolved and labile Cu and Zn in urban runoff from a parking lot, an industrial area and copper and zinc roofs. In: *14th IWA/IAHR International Conference on Urban Drainage, ICUD 2017*, September 10–15, Prague.
- Liu, J., Sample, D., Bell, C. & Guan, Y. 2014 Review and research needs of bioretention used for the treatment of urban stormwater. *Water* **6**, 1069–1099. doi:10.3390/w6041069.
- Loganathan, P., Vigneswaran, S. & Kandasamy, J. 2013 Road-deposited sediment pollutants: a critical review of their characteristics, source apportionment, and management. *Critical Reviews in Environmental Science and Technology* **43**, 1315–1348. doi:10.1080/10643389.2011.644222.
- Mitchell Ayers, E. & Kangas, P. 2018 Soil layer development and biota in bioretention. *Water* **10** (11), 1587.
- Müller, A., Österlund, H., Marsalek, J. & Viklander, M. 2020 The pollution conveyed by urban runoff: a review of sources. *Science of the Total Environment* **709**, 136125. doi:10.1016/j.scitotenv.2019.136125.
- Napier, F., Jefferies, C., Heal, K. V., Fogg, P., Arcy, B. J. & Clarke, R. 2009 Evidence of traffic-related pollutant control in soil-based sustainable urban drainage systems SUDS. *Water Science and Technology* **60**, 221–230. doi:10.2166/wst.2009.326.
- Pitt, R., Clark, S. & Field, R. 1999 Groundwater contamination potential from stormwater infiltration practices. *Urban Water* **1**, 217–236. doi:10.1016/s1462-07589900014-x.
- R Core Team. 2021 *R: A Language and Environment for Statistical Computing*. R Foundation for Statistical Computing, Vienna, Austria. Available from: <https://www.R-project.org/>.
- Reck, A., Paton, E. & Kluge, B. 2019 Advanced in situ soil water sampling system for monitoring solute fluxes in the vadose zone. *Vadose Zone Journal* **18**, 1–8. doi:10.2136/vzj2019.01.0008.
- Robertson, D. J. & Taylor, K. G. 2007 Temporal variability of metal contamination in urban road-deposited sediment in Manchester, UK: implications for urban pollution monitoring. *Water, Air and Soil Pollution* **186**, 209–220.
- RStudio Team 2021 *RStudio: Integrated Development Environment for R*. RStudio, PBC, Boston, MA. Available from: <http://www.rstudio.com/>.
- Sommer, H., Engel, N., Rehfeld-Klein, M. & Sieker, F. 2002 Messtechnische Untersuchung zur Funktion eines Mulden-Rigolen-Systems (Measurement investigation of the function of a swale-trench system). *GWF-Wasser/Abwasser* **143**, 864–871.
- Spraakman, S., Setters, T. V., Drake, J. & Passeport, E. 2020 How has it changed? A comparative field evaluation of bioretention infiltration and treatment performance post-construction and at maturity. *Ecological Engineering* **158**, 106036. <https://doi.org/10.1016/j.ecoleng.2020.106036>.
- Sun, X. & Davis, A. P. 2007 Heavy metal fates in laboratory bioretention systems. *Chemosphere* **66**, 1601–1609. doi:10.1016/j.chemosphere.2006.08.013.



- Tedoldi, D., Chebbo, G., Pierlot, D., Kovacs, Y. & Gromaire, M.-C. 2016 Impact of runoff infiltration on contaminant accumulation and transport in the soil/filter media of sustainable urban drainage systems: a literature review. *Science of The Total Environment* **569**, 904–926. doi:10.1016/j.scitotenv.2016.04.215.
- Tedoldi, D., Chebbo, G., Pierlot, D., Branchu, P., Kovacs, Y. & Gromaire, M.-C. 2017 Spatial distribution of heavy metals in the surface soil of source-control stormwater infiltration devices – inter-site comparison. *Science of the Total Environment* **579**, 881–892. doi:10.1016/j.scitotenv.2016.10.226.
- Trowsdale, S. A. & Simcock, R. 2011 Urban stormwater treatment using bioretention. *Journal of Hydrology* **397**, 167–174. doi:10.1016/j.jhydrol.2010.11.023.
- Vardhan, K. H., Kumar, P. S. & Panda, R. C. 2019 A review on heavy metal pollution, toxicity and remedial measures: current trends and future perspectives. *Journal of Molecular Liquids* **290**, 111197. doi:10.1016/j.molliq.2019.111197.
- Weihermüller, L., Siemens, J., Deurer, M., Knoblauch, S., Rupp, H., Göttlein, A. & Pütz, T. 2007 In situ soil water extraction: a review. *Journal of Environmental Quality* **36**, 1735–1748. doi:10.2134/jeq2007.0218.
- Werkenthin, M., Kluge, B. & Wessolek, G. 2016 Assessment of metal retention in newly constructed highway embankments. *Environmental Science and Pollution Research* **23**, 23619–23629. doi:10.1007/s11356-016-7526-z.
- Winogradoff, D. & Coffman, L. 2002 *Prince George's County Bioretention Manual*. Programs and Planning Division of the Department of Environmental Resources, Prince George's County, Maryland.
- Wong, C. S., Li, X. & Thornton, I. 2006 Urban environmental geochemistry of trace metals. *Environmental Pollution* **142**, 1–16. doi:10.1016/j.envpol.2005.09.004.
- Woods-Ballard, B., Kellagher, R., Martin, P., Jefferies, C., Bray, R. & Shaffer, P. 2007 *The SUDS Manual C697*. CIRIA, London.

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