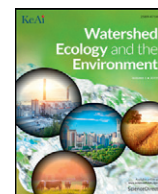


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Using biochar to purify runoff in road verges of urbanised watersheds: A large-scale field lysimeter study

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ABSTRACT

Urban runoff from traffic areas is a major source of pollution that degrades the quality of adjacent surface waters. Green infrastructure provided by the substantial amount of roadside land at urban fringe areas can be used to better manage and infiltrate this urban runoff. When establishing urban green areas, recycled materials should be preferred in order to achieve economically feasible and environmentally responsible solutions. Wastewater treatment plants within e.g. the EU yield substantial amounts of solids containing nutrients and metals that need to be utilised in a sustainable way. However, soil composted from such sewage sludge is being used widely in constructing green infrastructure, such as parks and road verges, which may jeopardise their use for stormwater management even though the effects of sewage sludge at road verges on the quality of runoff waters have not been subjected to scientific examination. Biochar has been suggested to retain pollutants and may also meet the criteria of being recycled material. We established artificial biofilter structures, mimicking road verges, in large-scale field lysimeters under cold climatic conditions in southern Finland to study the ability of biochar to retain pollutants leaching from composted sewage sludge and from infiltrating artificial stormwater. The topmost 15 cm consisted of an organic layer of either natural peat (Peat) or soil composted from sewage sludge either mixed with birch-derived biochar (3% by volume) or without this biochar (Comp+bc and Comp, respectively). At the end of the 1st growing period grasses growing in the lysimeters had taken up to 32% of phosphorus in the top soil containing compost. Leaching of phosphorus did not differ between the treatments, while nitrogen (N) leaching was ten times larger from Comp than Peat. Leaching of heavy metals, such as nickel and copper, was also significantly higher from compost soils than peat, but biochar significantly reduced metal leaching from compost (up to 50%). Two years after establishment, lysimeters were irrigated with artificial stormwater mimicking runoff from roads with heavy traffic. Comp leached more N compared to Peat. However, biochar significantly reduced N load infiltrated through the compost by 44%. Mixing sewage sludge-originated compost with biochar, and adding a 5 cm thick layer of biochar underneath the organic soil layer can substantially reduce leaching of N and heavy metals. However, given the substantial amount of roadsides in urban fringe areas, the extensive use of sewage sludge and other N-rich materials in such areas should be considered carefully.

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1. Introduction

Cities are disproportionately located along aquatic ecosystems and have been identified as hotspots for the accumulation of nutrients and metals and the subsequent leakage of these contaminants into adjacent water bodies (Grimm et al., 2008; Pouyat et al., 2007). Urban runoff, also known as stormwater, is a notable carrier of pollutants in urban areas that can degrade the quality and health of adjacent surface waters (Fletcher et al., 2013). Elevated runoff volumes due to the high proportion of impervious surfaces increase pollutant loads (Carey et al., 2013),

resulting in a significant relationship between the percentage of impervious surfaces and the loading of nutrients and metals (Valtunen et al., 2014, 2015). As conventional urban water management is unlikely to meet challenges brought about by an increasingly urbanising world, there is an urgent need to develop innovative solutions to sustainably manage stormwater in cities (Fitzhugh and Richter, 2004; Larsen et al., 2016). For example, various biofiltration structures have been developed to diminish and treat urban runoff that would be otherwise conveyed - unpurified - directly to adjacent water bodies via underground sewer systems (Ahiablame et al., 2012; Carey et al., 2013; Driscoll et al., 2015; Hatt et al., 2009). Biofiltration systems and/or other green infrastructure have been shown to reduce nutrient export from cities at the watershed-scale (Pennino et al., 2016), while in other cases

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they have been less effective (Liu et al., 2017). Given the limited and inconsistent knowledge of the efficiencies of various green infrastructure solutions, empirical studies are needed to better understand the factors, such as design (including soil materials, vegetation type), pollutant loads, and climate and local conditions that influence the success of stormwater management (Ahiablame et al., 2012; Liu et al., 2017). This is especially the case in northern latitudes where the functioning of bio-based infiltration treatments is supposed to be inefficient during cold months (Blecken et al., 2011; Muthanna et al., 2007).

Traffic is a main source of pollutants in cities, with traffic volume correlating strongly with pollutant loads in roadside environments (Carey et al., 2013; Kuoppamäki et al., 2014; Westerlund and Viklander, 2006). Consequently, contaminants at road edges can pose important but still poorly known risk to both surface and ground waters (Kuoppamäki et al., 2014; Valtanen et al., 2014; Westerlund and Viklander, 2006). A common practice to mitigate the adverse effects of road-derived runoff is grass swales that replace or supplement traditional curbs and gutters. Adsorption/absorption processes by such low-cost bioretention features can efficiently control phosphorus and metals bound in suspended solids but not pollutants that are in dissolved form (reviewed by Ahiablame et al., 2012).

The composition of bioretention media is typically mostly sand and vegetation is planted on a variety of mixtures containing soil, sand, mulch, and organic matter (Ahiablame et al., 2012). In Finland, peat is usually mixed in soil products, but the use of peat is not sustainable due to its extremely slow regeneration rate. Biofilter structures should be environmentally sustainable with a low environmental footprint, minimal economic cost (Hatt et al., 2009), readily available and materials preferably recycled rather than virgin to promote circular economy (Mohanty et al., 2018). For instance, the over 50,000 wastewater treatment plants within the European Union yield over 10 million tons of dry solids per year (Eurostat, 2018). This sewage sludge contains high amount of nutrients and organic matter to be used as fertilizer or a soil improving agent. Within EU countries, 37% of the total annual production of sewage sludge is used in agriculture, while the rest is used for other land applications (Olofsson et al., 2012). In cities worldwide, 10–30% of the city area is covered with roads and streets, with an equal proportion being road verges. Due to the substantial amount of roadside land at urban fringe areas, the use of sewage sludge to support vegetation growth has become a common practice there. However, as sewage sludge can exceed the threshold values for metal concentrations (The Sewage Sludge Directive 86/278/EEC) and also contain organic pollutants (Zennegg et al., 2013), microplastics (Lusher et al., 2017) and unwanted pathogens (Bagge et al., 2005), its use in green infrastructure has been questioned (Di Bonito, 2008). Furthermore, the effects of sewage sludge at road verges on the quality of runoff waters have not been subjected to scientific examination.

To improve their capacity to retain stormwater and contaminants therein, various types of materials have been added in biofilter systems. For example, when added to the soil, biochar has been shown to reduce the leaching of nutrients and metals (Wei et al., 2018) in agriculture (Konczak and Oleszczuk, 2018) and when applied to green roofs (Kuoppamäki et al., 2016). Furthermore, biochar appears to be a promising material for stormwater treatment and meets the criteria for having a low environmental footprint and being recycled material (Mohanty et al., 2018). The manufacturing process of biochar does not allow CO₂ to be released from the feed material, making biochar a carbon sink (Lehmann, 2007). However, results from large-scale field applications of biochar to treat stormwater are lacking.

We established large-scale biofilter structures in field lysimeters (2 m³) to mimic road verges under cold climatic zone in southern Finland. The lysimeters were composed of three soil types, and the efficacy of these soils to retain or the risk of these soils to leach nutrients and metals were monitored for two years. After this the lysimeters were added with artificial stormwater and they were monitored for one month to study the retention of nutrients and metals. We

hypothesized that: 1) the leaching of nutrients and metals from soil composted from sewage sludge is higher than that from nutrient-poor peat but 2) this leaching is reduced by amending sludge based compost with biochar.

2. Materials and methods

2.1. Treatment setup

To simulate road verges, we established an experiment in nine large-scale field lysimeters (depth 2 m, diameter ca. 1 m, stainless steel), situated in the City of Lahti, southern Finland (60°59'00"N, 25°39'20"E) in June 2014. As illustrated in Fig. 1, on the bottom of each lysimeter, a 0.5 m thick layer of coarse gravel (8–16 mm) was placed as a drainage layer. Above this layer, separated with a filter fabric, another 0.5 m of filter sand (0.02–31.5 mm) was added. Above the sand layer, a 0.4 m thick layer of coarsely crushed stones (0–90 mm) and then 0.3 m finely crushed stones (0–32 mm) were placed. Finally, a top layer, 0.15 m thick, of growing substrate was added consisting either of i) soil with composted sewage sludge (50%, v/v) mixed with fine sand (50%, v/v); hereafter referred to as “Comp”), ii) the same compost soil amended (5% by volume) with slow pyrolysis (380–420 °C for 2 h; see Kuoppamäki et al., 2016) biochar made of birch; hereafter “Comp+bc”) or iii) commercial peat (75%, v/v) mixed with sand (25%, v/v) soil for gardening purposes (hereafter “Peat”). In the Comp+bc treatment a 0.05 m layer of biochar was also added below the growing substrate. Thus, the uppermost 0.2 m layer contained 30% biochar and the total lysimeter 3% (v/v). Each treatment had three replicates. The structure and layers of soil in the lysimeters followed what is commonly used in road structures in southern Finland. All lysimeters were planted with seeds of a grass mixture (*Festuca ovina*, *F. rubra*, *Phleum pratense*) typical to road verges in Finland. The soils were derived from commercial producers: sewage sludge from the Helsinki Region Environmental Services Authority HSY, and peat (Belinda, enriched with NKP fertilizer) from Europeat Ltd. Properties of the compost, biochar and peat used are listed in Table 1.

2.2. Measurements

Outflow rate of the infiltrated water was measured at 10 min resolution using pressure sensors set in outflow water tanks of 20 l. The mass of each lysimeter was continuously measured with scales and was used to calculate outflow rate when water tanks overflowed. However, in February and March 2016, technical problems caused by accumulating snow on top of the lysimeters prevented us from calculating runoff volumes. Soil temperature and moisture were measured continuously throughout the study with sensors placed at 0.2 m and 1.6 m depths from the surface of each lysimeter (Fig. 1). A local Vaisala WXT520 Micro Weather Station collected data on rainfall, air moisture and wind velocity at 10 min intervals.

Grass growing in the lysimeters were cut at the base of the shoots at the end of the growing season in 2014. Aboveground biomass of the vegetation was room dried for 6 weeks, and dry mass (g) calculated.

2.3. Sampling during the stabilising period

Before irrigating the lysimeters with artificial stormwater (see below), they were left to stabilise under ambient precipitation for 23 months. During this period, potential leaching of nutrients and metals from the lysimeter soil was monitored by taking samples from infiltrating water at 2–4 month intervals, when enough, i.e. >10 l (hereafter litres are abbreviated as l) had infiltrated through each lysimeter. The first such runoff event was 5 months after establishing the experiment on the 26th November 2014, when the first set of runoff samples for nutrient and metal analyses was taken. Thereafter, in 2015 samples were taken on 22nd January, 27th May, 17th and 10th December, while

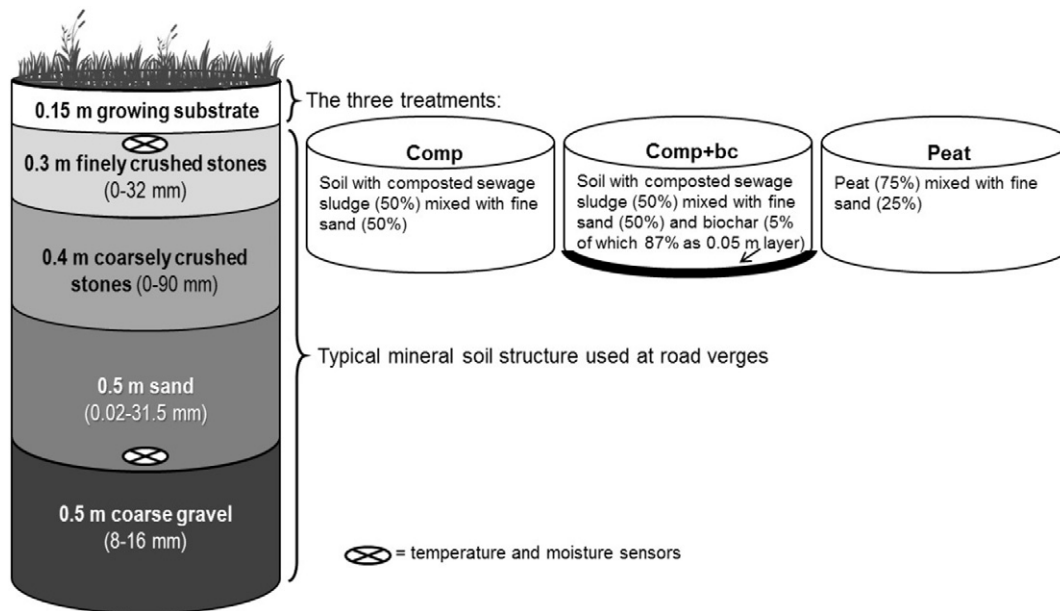


Fig. 1. Schematic presentation of the soil layers in a biofiltration system constructed in a lysimeter and the three treatments in the topmost growing substrate. Percentages of different soil materials are all by volume. The location of the temperature and moisture sensors placed on the top and bottom of the lysimeters is also shown.

in 2016 samples were taken on 17th February and 22nd March. Thus, during the stabilisation period sampling was carried out following a total of 7 runoff events.

2.4. Sampling during the irrigations

In mid-May 2016 (mean monthly temperature + 8 °C), when the growing season had already started and spring snowmelt had passed, the lysimeters were irrigated with artificial stormwater, simulating runoff from an urban core area with 89% impervious surface (see [Valtanen et al., 2014](#)). The content of the artificial stormwater was as follows: soluble phosphorus (K_2HPO_4) 1 mg/l, nitrate ($NaNO_3$) 10 mg/l, zinc ($ZnSO_4 + 7H_2O$) 0.5 mg/l, copper ($CuSO_4 + 5H_2O$) 0.3 mg/l, lead ($PbNO_3$) 0.1 mg/l, aluminium ($Al_2(SO_4)_3 + 18H_2O$) 5 mg/l and chromium (CrCl) 0.1 mg/l. Irrigation occurred during two consecutive days. The first irrigation on 11th May represented a 5 mm precipitation event of 1 h duration and the second on 15th May represented a 10 mm event of 2 h duration so that the volume of stormwater per lysimeter was 40 l and 80 l, respectively.

It was assumed that the area of the lysimeter biofilters represents 10% of their catchment area, with a 0.9 runoff coefficient. 30% of

precipitation events at the locality during a 4 year measuring campaign are between 5 mm and 10 mm (K. Kuoppamäki, unpublished data); such rain events were considered representative and, importantly, large enough to generate runoff from the lysimeters. Samples from infiltrating water were taken on two consecutive days following the irrigation event as well as once a month later, assuming a delayed outflow of stormwater.

Before terminating the experiment, final discharge samples were taken on the 17th June 2016.

2.5. Laboratory analyses

Water samples that infiltrated through the lysimeters as well as a composite sample of the stormwater used for irrigation were first measured for pH by using a WTW Inolab pH 720 and electric conductivity by using a WTW Cond 330i meter. All water samples were stored at -20 °C until the analysis of nutrients and metals.

Samples for total nutrient analyses were first oxidised in an autoclave at 120 °C for 30 min. Total phosphorus (TP) was measured spectrophotometrically after the addition of a molybdate reagent (SFS 3026). A high Performance Liquid Chromatography (Shimadzu Prominence HPLC) instrument was used to determine total nitrogen (TN) with 0.04 M sodium chloride (NaCl) as the eluent and using an ion-exchange column (Waters IC-Pak Anion HC). The chromatogram was recorded at 225 nm. For the analysis of total dissolved metal content in the artificial stormwater and in the water leachates, concentrated HNO_3 (50 μ l) and 50 μ l of 1 mg/l indium solution was added as an internal standard and the sample was mixed well with vortex. For total dissolved metals, samples were filtered through a 0.45 μ m syringe filter (Whatman). One ml of the sample was then diluted with 4 ml of water and 50 μ l of indium (internal standard 1 mg/l) was added to the diluted sample. Five ml of the filtered sample was transferred into a test tube. Finally, metal concentrations were determined using a Perkin-Elmer Elan 6000 ICP-MS according to SFS-EN ISO 17294-2 (2005).

Biochar and grass samples (above ground biomass only) were analysed for total metals (Cd, Cr, Cu, Ni, Zn and Pb) and TP concentration using Elan 6000 ICP-MS. As a pre-treatment, the samples were digested with a MARS 6 Microwave Digester. In addition, total carbon (TC) and

Table 1
Characteristics of materials used in the growing substrate layer of the lysimeters.

	Peat	Comp	Biochar
N mg/kg	530*1	710*1	0.0*1, 4300*2
P mg/kg	90*1	80*1	330*2
K mg/kg	690*1	150*1	1500
Organic matter %	-	16	95
Cu mg/kg	-	-	<LOQ (Carey et al., 2013)*2
Cr mg/kg	-	25	<LOQ (Driscoll et al., 2015)*2
Zn mg/kg	-	200	<LOQ (100)*2
Pb mg/kg	-	12	<LOQ (Ahmad et al., 2014)*2
Mn mg/kg	-	-	170*2
Fe mg/kg	-	-	3100*2
Bulk density g/l	340	490	270
BET-area m2/g	-	-	7.0
pH	6.2	6.0	6.6

*1 soluble, *2 total, Sewage sludge metal analysis: SFS-EN 13650.

TN concentrations of the samples were measured using a LECO CSN – analyser (Leco Incorporation Inc.)

2.6. Data analyses

The effects of treatment (Comp, Comp+bc or Peat) on the event mean concentrations (total load divided by total runoff volume; see supplementary material) and loads of nutrients (TP, TN) and metals (Cd, Cr, Cu, Ni, Pb, Zn) in leachate water during the stabilisation period were tested using repeated measures ANOVA, where treatment was a fixed factor and the repeated measures were comprised of either nutrients or metals with time of sampling. The loads of nutrients and dissolved metals were calculated by multiplying concentrations by runoff volumes. Total loads after artificial stormwater irrigation were obtained by summing loads measured during the three sampling days. These results as well as average loads measured before irrigation were tested using multivariate ANOVA. Once statistically significant treatment effects appeared in the ANOVA models, the statistical significance of differences between treatments were interpreted using Tukey post hoc test. If a significant interaction between treatment and time was observed, the effect of treatment was analysed separately on each sampling event. ANOVA for multivariate measures was used to analyse the effects of treatment on the biomass and nutrient contents of grasses.

The homogeneity of variances and normal distribution of the data were tested using Levene's test and Shapiro-Wilk test, respectively. Data were log-transformed if the assumptions of normality were violated. If assumptions were not fulfilled even after log-transformation, data were tested using non-parametric Kruskal-Wallis and Mann-Whitney tests. All statistical analyses were carried out using the SPSS statistical package (IBM Corp. 2016).

3. Results

3.1. Nutrients

When comparing loads averaged across the stabilising period, TN load was higher from the two compost treatments than from Peat, while for TP loads it was vice versa (Fig. 2). Nitrogen leaching was also statistically significantly higher from compost (Comp) than from Peat (Table 2). However, the difference in TN loads between compost amended with biochar (Comp+bc) and Peat was insignificant (Table 2) indicating that biochar reduced the leaching of TN. The difference in TP loads between all treatments was also insignificant. In detail,

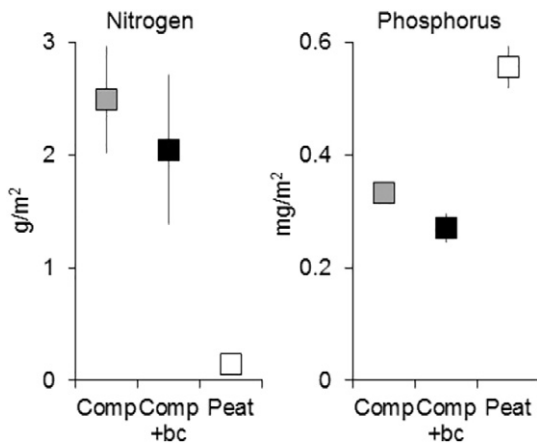


Fig. 2. Average (\pm SE) loads of dissolved total nitrogen (left) and phosphorus (right) in runoff during the stabilising period from November 2014 to December 2015. Note: nitrogen load in g/m², phosphorus load in mg/m². Comp & Comp+bc = compost treatments without and with biochar amendment, Peat = peat treatment.

Table 2

Repeated measures ANOVA results on the effects of sampling time and substrate treatment (C = compost, CB = compost with biochar, P = peat) on the loads of nutrients and metals in leachate waters during the stabilisation period, together with a post hoc Tukey's test of the statistical differences among treatments.

	Time		Time × Treatment		Treatment		Tukey's test
	F	p	F	p	F	p	
Nitrogen	3.56	0.020	1.66	0.160	6.09	0.036	C > P
Phosphorus	2.06	0.117	1.78	0.131	3.69	0.090	
Cu	22.45	<0.001	14.21	<0.001	14.99	0.005	P > C, P > CB
Cr	1.29	0.303	0.42	0.90	3.86	0.084	
Zn	45.19	<0.001	8.13	<0.001	6.02	0.037	C > P
Al	1.99	0.208	1.64	0.269	8.03	0.020	C > CB, C > P
Pb	8.64	<0.001	5.19	0.001	0.32	0.738	
Ni	72.06	<0.001	8.16	<0.001	6.57	0.031	C > P
Cd	56.08	<0.001	6.21	<0.001	4.75	0.058	
Mn	21.46	<0.001	3.98	0.004	4.18	0.073	

Bold statistically significant impacts of treatments on the loads of nutrients and metals.

TN leaching from Peat was 1–10 x lower than that from Comp or Comp+bc (Mann-Whitney: $p = 0.05$, Peat vs. Comp and Comp+bc in each sampling event). In the Comp treatments, TN loads declined from ca. 3000–5000 mg/m² at the beginning of the experiment to 500–2000 mg/m² in December 2015, and in the Peat treatment from 500 to 10 mg/m² and, thus, the impact of time on TN load was notable (Table 2). TP loads varied between 0.15 and 0.7 mg/m² in all treatments without temporal trends. Background loads of TN and TP in wet and dry deposition at the experimental area during the study were 4–19 and 0.3–1.9 mg/m², respectively.

Following irrigation with artificial stormwater, biochar reduced the leaching of TN, just like during the stabilisation period. Comp leached more TN than Comp+bc and Peat (Table 2), while TN load from Comp+bc was only marginally higher than that from Peat (Tukey $p = 0.056$). However, negative TN retention of –186% and –64% were observed in Comp and Comp+bc, respectively, indicating high leaching from the compost mixture itself. Peat also had low, yet positive TN retention (15%). Following stormwater irrigation, elevated load of TP was observed in Peat, ca. 50% higher than from Comp and Comp+bc, but differences between treatments were insignificant (Table 2) due to high variation (Fig. 3). TP leaching from all treatments was extremely low compared to the load in stormwater and, thus, all treatments retained TP by 99%.

The event mean concentrations of nutrients and metals are given in supplemental material.

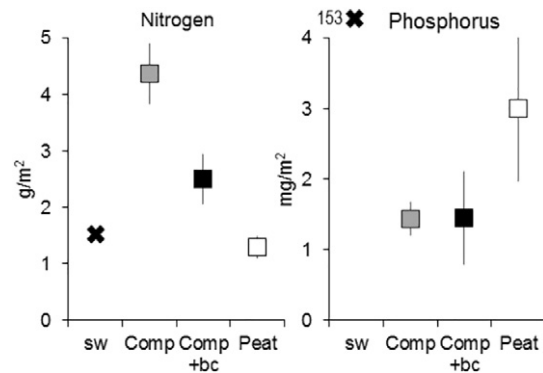


Fig. 3. Average (\pm SE) cumulative loads of dissolved total nitrogen (left) and phosphorus (right) in runoff following stormwater irrigation in May–June 2016. Note: nitrogen load in g/m², phosphorus load in mg/m². sw = loads in the artificial stormwater, Comp & Comp+bc = compost treatments without and with biochar amendment, Peat = peat treatment.

3.2. Metals

During the stabilisation period, the effect of treatment on the loads of all metals, except chromium (Cr) and aluminium (Al), depended on time (Table 2). In December 2015, an increase was detected in the loads of cadmium (Cd), nickel (Ni), lead (Pb) and zinc (Zn), especially in both compost treatments. When calculating average dissolved metal loads during the stabilisation period, significantly higher leaching of Zn, Al and Ni from Comp than from the other two treatments was evident (Table 2, Fig. 4) showing that biochar can increase the retention of these metals. On the other hand, Peat leached more Cu than the two compost treatments. Cr load remained at ca. 0.01 mg/m² during this period. Al loads, in turn, could not be determined due to low concentrations (below the detection limit of 92 µg/l) until December 2015 when concentrations suddenly increased in Comp up to 400–700 µg/l and in the other treatments well above 100 µg/l.

Following irrigation with stormwater, loads of dissolved Zn and Al were highest in Comp and lowest in Peat, the significant difference being between Comp and Peat (Table 3, Fig. 5). A similar pattern was evident in Cu loads, but the effects of treatments were marginally significant, as well as in those of Ni and Cd, which were, however, not added to the stormwater.

The retention of Cr, Al and Pb, which were added to the artificial stormwater, was 99–100% in all treatments. Also Cu and Zn were 91% and 87% retained, respectively, by Peat-containing lysimeters. Comp leached remarkably more Zn (on average 124 mg/m²) compared to the 76 mg/m² that was added to the artificial stormwater, resulting in negative retention (–63%). However, the addition of biochar improved the retention of Cu in compost close to levels leached from Peat.

Table 3

Multivariate ANOVA results on the effects of substrate treatment (C = compost, CB = compost with biochar, P = peat) on the average loads of nutrients and metals during the stabilisation period, as well as pH and the total loads of nutrients and metals following irrigation with artificial stormwater. Post hoc Tukey's test results of differences among treatments are also shown.

	Treatment		Tukey's test
	F	p	
Stabilising period			
Nitrogen	47.80	<0.001	C > P, CB > P
Phosphorus	3.99	0.079	
Cu	14.99	0.005	P > C, P > CB
Cr	3.86	0.084	
Zn	6.02	0.037	C > CB, C > P
Al	17.90	0.003	C > CB, C > P
Pb	0.32	0.738	
Ni	6.57	0.031	C > CB, C > P
Cd	4.75	0.058	
Mn	0.789	0.089	
Following irrigation			
pH	10.03	0.012	P > C
Electric conductivity	10.97	0.010	C > P
Nitrogen	36.26	<0.001	C > CB, C > P
Phosphorus	1.56	0.286	
Cu	4.50	0.064	
Cr	1.33	0.332	
Zn	7.35	0.024	C > P
Al	5.53	0.043	C > P
Pb	1.91	0.229	
Ni	5.85	0.037	C > P
Cd	5.98	0.038	C > P
Mn	4.72	0.059	

Bold statistically significant impacts of treatments on the loads of nutrients and metals.

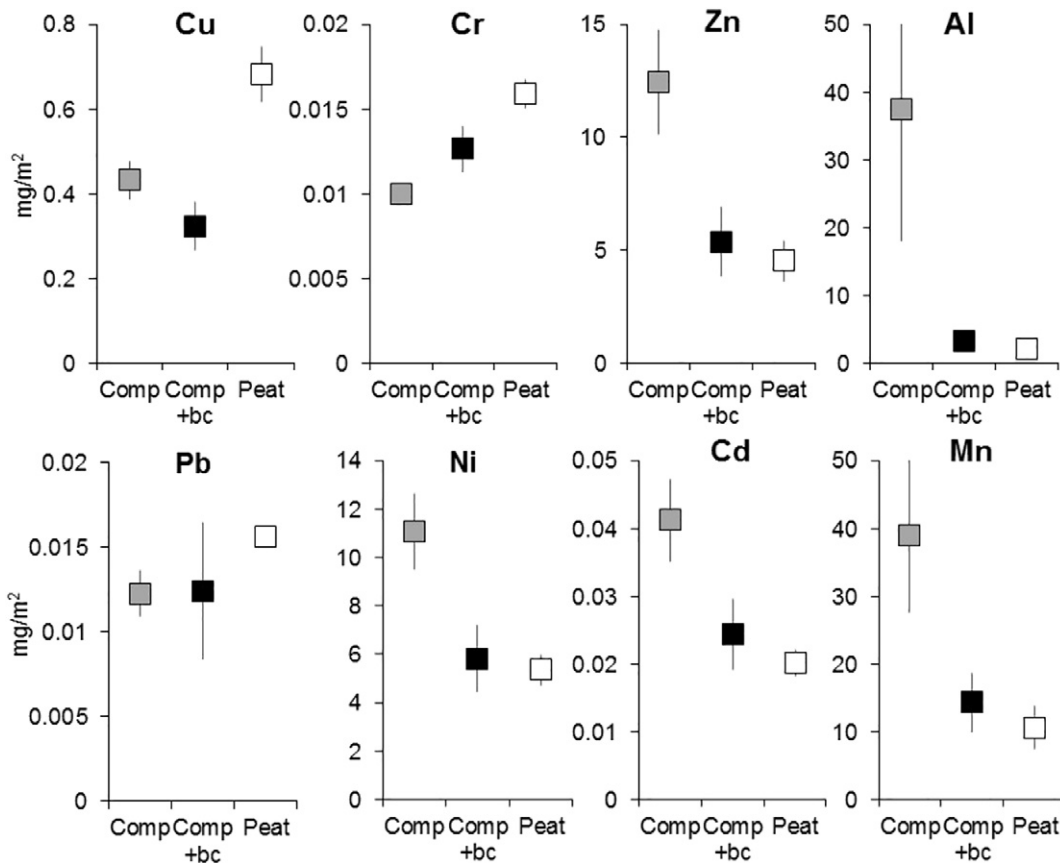


Fig. 4. Average (±SE) loads of dissolved total metals in runoff during the stabilising period from November 2014 to December 2015. Note that Al loads are only from December 2015 when concentrations were above the detection limit. Comp & Comp+bc = compost treatments without and with biochar amendment, Peat = peat treatment.

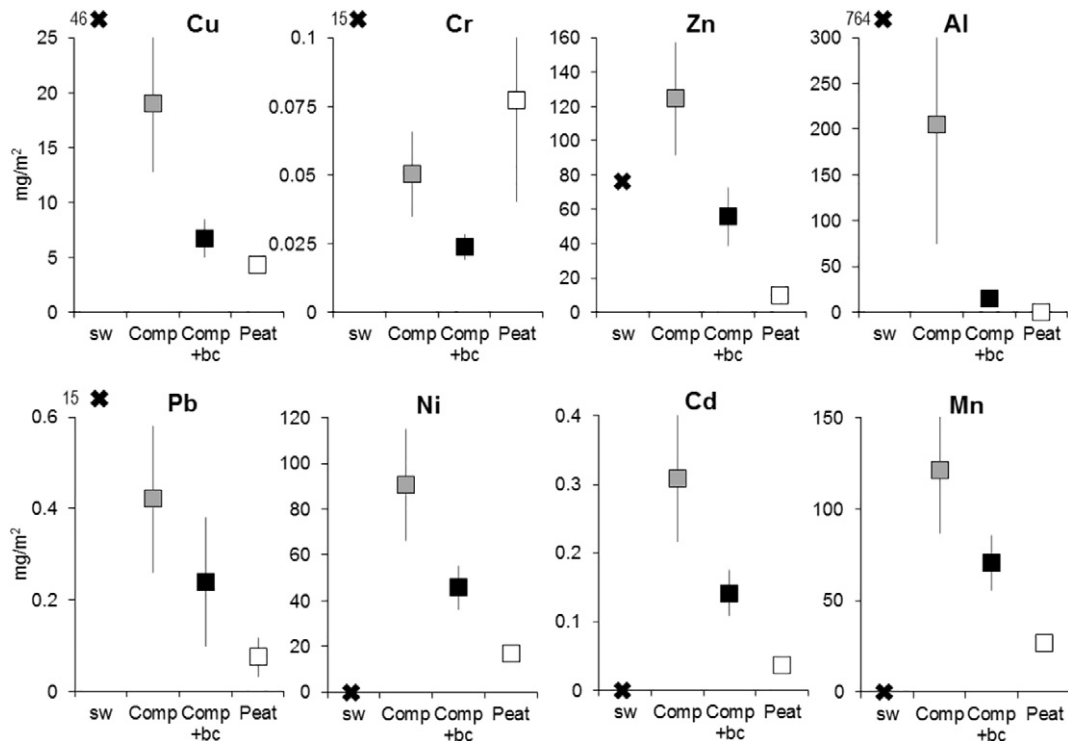


Fig. 5. Average (\pm SE) cumulative loads of dissolved metals in runoff following stormwater irrigation in May–June 2016. Note: stormwater did not contain Ni, Cd and Mn. sw = loads in the artificial stormwater, Comp & Comp+bc = compost treatments without and with biochar amendment, Peat = peat treatment.

3.3. Runoff pH and electric conductivity

Rainwater pH and irrigation water pH decreased from 6.7 to an average 4.7 when infiltrating through Comp, while significantly higher pH (average 6.1) was measured in water infiltrated through Peat (Anova $F = 10.03$, $p = 0.012$, Tukey $p = 0.01$; Fig. 6). However, pH declined less in water infiltrating through Comp+bc than in Comp, though the impact of biochar was not statistically significant. Opposite patterns were seen in electric conductivity that was 3 times higher in Comp compared to that in rain and irrigation water, while Peat did not change conductivity of the infiltrating water (Fig. 6). Biochar amendment attenuated the increase of conductivity. The effects of treatments on runoff pH and electric conductivity were significant, the difference being between Comp and Peat (Table 3).

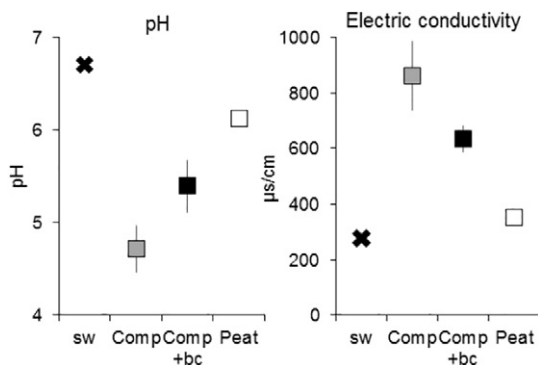


Fig. 6. Average (\pm SE) pH (left) and electric conductivity (right) in runoff following stormwater irrigation in May–June 2016. sw = loads in the artificial stormwater, Comp & Comp+bc = compost treatments without and with biochar amendment, Peat = peat treatment.

3.4. Retention of stormwater

The total retention of all water, including artificial stormwater and natural rainfall, was 21–26% without significant differences between treatments. More precisely, after the first stormwater irrigation (40 l), the two compost treatments retained 75% of water, while the retention in Peat was 64%. The corresponding values following the second stormwater irrigation (80 l) were 45% and 55%, respectively. Top soil moisture content increased from 10 to 20% to 50–60% in Comp and Comp+bc and 40–50% in Peat following irrigations with stormwater. Discharge velocity was on average 1500 ml/h. From the 2nd irrigation event until the last water sampling event a month later, total precipitation was 58 mm, resulting in an additional 45 l of water in each lysimeter. This volume was used in calculating the EMC values of the last sampling event.

3.5. Vegetation

During the first growth period, grasses germinated well in all lysimeters irrespective of treatment. However, at the end of the first growing period, the biomass of grasses growing in Peat was stunted, being on average 7 times lower compared to the two treatments with compost soil (Anova $F = 29.29$, $p = 0.001$) (Fig. 7). Percentage C in grasses was 44% in all treatments. This corresponds to ca. 137, 124 and 18 g C m² in Comp, Comp+bc and Peat treatments, respectively, i.e. significantly higher carbon mass was detected in the two compost treatments compared to Peat (Anova $F = 30.88$, $p = 0.001$), while concentrations of TN (Anova $F = 28.92$, $p = 0.001$) and TP (Anova $F = 30.17$, $p = 0.001$) of grasses growing in Comp and Comp+bc soils were 7.6 and 7.3 x higher (respectively) compared to grasses growing in Peat (Fig. 7).

3.6. Climatic parameters

The growing season in 2014 was slightly warmer (on average 16 °C) and rainier (205 mm) than in 2015 (14.4 °C and 175 mm, respectively),

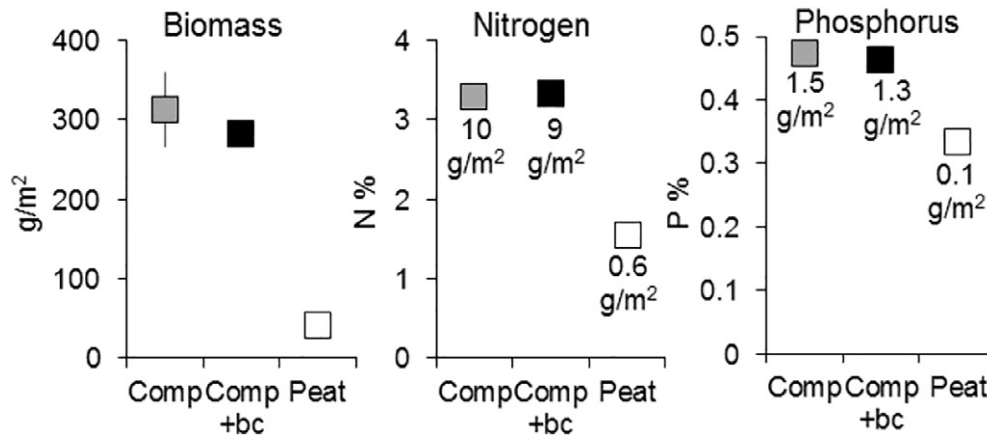


Fig. 7. Average (\pm SE) biomass and the contents of nitrogen and phosphorus in grasses in September 2014. Comp & Comp+bc = compost treatments without and with biochar amendment, Peat = peat treatment.

though the latter year was characterised by several days with higher precipitation amounts compared to the previous year (Fig. 8). In 2014 air temperature dropped below 0 °C earlier than in 2015 but was warmer than winter 2015–2016. The second winter was very cold with temperature below minus 10 °C during all January 2016. Consequently, in the first winter only top soil of lysimeters got frozen and only for short periods of time, while in the second winter the top soil was frozen for 7 weeks with the bottom soil also frozen for one week in mid-January (Fig. 8). Precipitation before sampling (during preceding month) was much higher in December 2015 compared to precipitation preceding the other sampling campaigns (Table 4). Samples taken in March 2016 included water that was melting from soils in the lysimeters and this water obviously represented several previous rain events spanning for time longer than just the preceding month. Sampling events in January 2015 and in February and March 2016 can be considered as representing winter conditions, given the average air temperatures below 0 °C during the preceding month (Table 4). Total amount of precipitation from the beginning of the experiment until the irrigation with artificial stormwater was 1200 mm.

4. Discussion

It is well established that the input of materials, such as nutrients and metals from watersheds to adjacent surface and ground water bodies are in decisive role determining the quality of invaluable water resources. Our results suggest that the application of sewage sludge-derived compost soil to road verges can substantially increase the leaching of TN, Al, Ni and Zn and thus potentially affect the ecology of the entire watershed. Our results further suggest that application of biochar can substantially reduce the leaching of TN and metals from compost soils, thus enabling its use at road verge areas in urbanised watersheds. However, biochar was not able to reduce the leaching of all contaminants.

We predicted that the leaching of nutrients and metals from compost mixture is higher than from peat. This prediction was partly confirmed as lysimeters with compost leached considerably more nitrogen (TN) and metals (Al, Ni, Zn and Cd) than peat - both during the 23-month stabilisation period and after stormwater irrigation. In contrast, there were no statistically significant differences in phosphorus (TP), Cr and Pb leaching between the treatments.

Further, we hypothesized that leaching of nutrients and metals from compost is reduced by biochar amendment. Biochar reduced TN leaching by 44% from compost soil during irrigation with stormwater when the soils had been stabilising for two years. During the stabilisation period, biochar decreased the loads of Al, Cu, Ni and Zn

significantly, while after stormwater addition the difference was apparent but statistically insignificant. We will next discuss in more detail the potential of the three soil types to retain nutrients and metals.

4.1. Nutrient retention

As algal productivity in freshwaters is usually phosphorus limited, reducing the loading of this nutrient from surrounding watershed is especially critical. During the almost 2-year stabilisation period, leaching of TP from the lysimeters was extremely low without consistent differences between treatments. Furthermore, after the two artificial stormwater irrigations, >99% of the added phosphorus was retained by all three treatments. As TP concentration of the top layer materials was quite similar in both Comp and Peat, this result was expected. Indeed, dissolved P carried by water is retained well by different soils types (Barrett et al., 2013; Coffmann and Clar, 2003; Pitt et al., 1999; Valtanen et al., 2017). The TP content of biochar was quite high, but Comp+bc did not leach more TP than the other treatments, suggesting that it was in insoluble form in biochar. On the other hand, although biochar increased leachate pH, this did not result in improved retention of TP. It is noteworthy that a considerable amount of TP was retained in grass biomass in the two compost systems: the grass biomass – being 5 x higher in the compost soils compared to peat soils – contained 1.5 times more TP (0.47 mg/m² and 0.46 mg/m²) in Comp and Comp+bc, respectively, compared to grasses growing in Peat (0.03 mg/m²). At the start of the study, the content of dissolved P in the top soil layers of Comp, Comp+bc and Peat was 5.9, 4.1 and 4.6 g/m², respectively. Consequently, on average 24, 32 and 3% of P in the top soil was bound to aboveground vegetation at the end of the 1st growing period, implying a remarkably large impact of vegetation on nutrient retention in the biofilter systems. This impact was much lower in Peat, where grasses had stunted growth compared to Comp and Comp+bc. When these values are compared to the amounts of TP leached through the lysimeter soils during the 1st growing period (Comp, Comp+bc and Peat: 0.6, 0.8, 0.4 g/m², respectively) the importance of TP immobilized by plant biomass as well as the soil layers becomes evident. The stunted growth of grasses in Peat probably partly explains the higher P leaching compared to the two compost treatments. In general, the high potential of biofilter structures to absorb P is in line with Valtanen et al. (2017) in northern conditions and by Coffmann and Clar (2003), Glaister et al. (2014), Barrett et al. (2013) and Pitt et al. (1999) under warmer climatic conditions. In all, the almost 100% ability of our bioretention systems to retain TP, irrespective of the top soil material, indicates that the biofiltration type used here is efficient to treat traffic-derived stormwater and thus mitigate eutrophication potential of adjacent

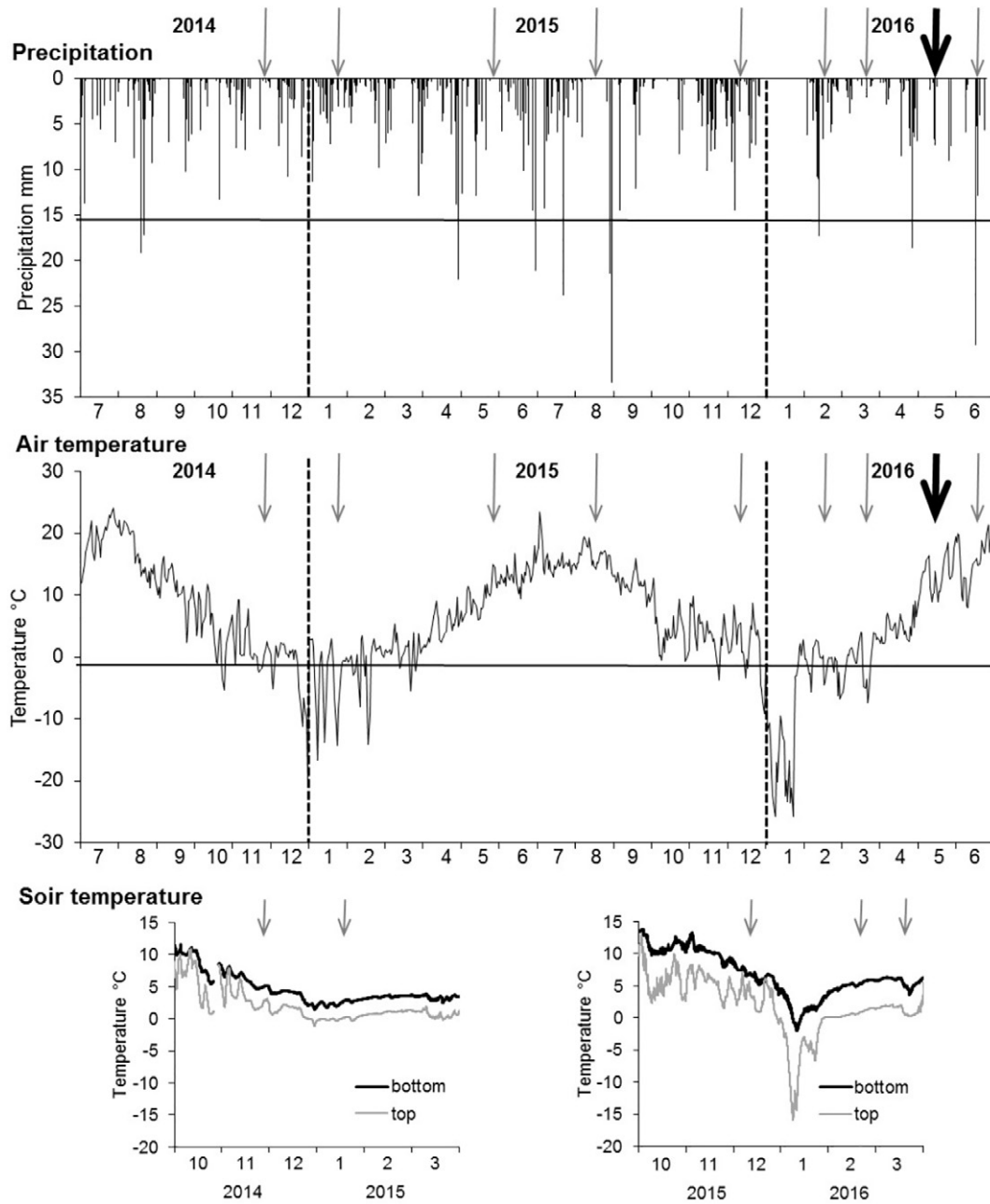


Fig. 8. Daily precipitation (top) and the average daily temperature (middle) during the experiment as well as average soil temperature in top and bottom layers of the lysimeters during the two cold seasons from October until March (below). Thin grey arrows indicate the time of runoff sampling and thick black arrows indicate the time of irrigation with the artificial stormwater.

water bodies. Importantly, decreasing the loading of P rather than N is a prerequisite to mitigate the eutrophication of lakes (Schindler et al., 2008).

In contrast to TP, TN was poorly retained by the biofiltration systems. At the beginning of the study (the stabilisation phase), the retention of TN by each treatment was negative, as indicated by the ca. 250–750 x higher TN loads measured in the water leachates compared to the amount detected in rainwater (via wet and dry deposition). As N-content in the mineral soils used in our study was negligible (H. Setälä, unpublished), TN in the water leachates were likely derived from the top layer materials. Moreover, N leaching from disturbed soil is common, for example at construction sites (Sillanpää, 2003; Wakida and Lerner, 2002). As such, studies that span over several years to allow for soil stabilising phases are required to determine the actual pollutant

retention capability of the soil. Thus, although our experiment lasted for two years, it may still be too short to demonstrate TN dynamics in biofilter systems. The leaching of TN from both compost soils was several times higher than from peat despite the added fertilizer in the latter. Loads of TN in the runoff decreased at the end of the stabilisation period, but the N content was still ca. 100 times higher compared to TN load in wet and dry deposition. After stormwater irrigation, TN loads increased again, which supports earlier findings about the inefficient retention of TN by biofiltration systems (Bratieres et al., 2008; Hatt et al., 2009; Pitt et al., 1994; Valtanen et al., 2014), but shows that sandy peat can well retain TN. However, extending the experiment longer and making more stormwater irrigations may reduce the capacity of also sandy peat to retain nitrogen. Somewhat unexpectedly, the pattern of TN in the leachate water was not affected by the cold period

Table 4

Mean air temperature, air moisture and rainfall depth during the preceding 30 days before each sampling.

Date	Temperature °C	Moisture %	Rainfall mm
Stabilisation period			
26.11.2014	1.7	86	34
22.1.2015	−4.9	82	61
27.5.2015	8.7	70	58
7.8.2015	15.0	75	64
10.12.2015	2.9	88	101
17.2.2016	−2.2	85	87
22.3.2016	−1.7	77	17
Stormwater irrigations			
12.5.2016	8.4	66	63
17.5.2016	9.3	65	66
1 month after stormwater irrigation			
17.6.2016	14.4	61	54

per se but more likely by the length of the time since the start of the experiment and irrigating the systems with stormwater. This suggests that biofiltration structures do not necessarily function badly during winter, as long as the soil is not completely frozen. During the first winter only the top soil got frozen for periods of few days and therefore the impact of cold season on the functioning of lysimeters was probably negligible. During the second, colder winter freezing reached the bottom of lysimeters, though only for one week, while top of soil was frozen for over 7 weeks. Nevertheless, no elevated leaching of TN was observed in the leachates taken after this period suggesting that TN dynamics was not much affected by freezing or thawing.

Despite substantial leaching of TN from the compost treatments, a remarkable proportion of TN was retained in the aboveground grass biomass: 9.8 and 9.4 g/m² in Comp and Comp+bc, respectively, corresponding to 19 and 20% of TN detected in the top 0.15 m layer soils. Grasses grown in peat contained only 0.6 g N m², which is 2% of TN in top layer soils. It is likely that the stunted growth of grasses was mostly responsible for the reduced their capacity to store TN. Corresponding TN leaching from the lysimeters between June to November 2015 was 11.6 (Comp), 6.3 (Comp+bc) and 1.2 g/m² (Peat), indicating similar amounts of TN in the leachates and in the green grass biomass. Under N deficient conditions, the ability of biochar to adsorb nitrogen can hamper the uptake of plant-available nutrients and thus have adverse effects on plant growth. However, in our study grass biomass and its N concentration was unresponsive to biochar addition, indicating sufficient nutritional conditions for plants, also in the Comp+bc treatment.

4.2. Retention of metals

Cr, Cd, Pb, Cu, Ni and Zn are metals frequently detected in runoff from urbanised watersheds (Kuoppamäki et al., 2014; Valtanen et al., 2014; Westerlund and Viklander, 2006) and they have been listed as elements that often cause failures of ecological status in water bodies (EAA, 2018). Loads of the most environmentally hazardous metals (Cr, Cd and Pb) in leachate waters discharging from our lysimeters were irresponsive to the type of soil in the current study. Interestingly, the retention of Cr and Pb derived from artificial stormwater was nearly 100%. These results show that Cr and Pb, as well as Cu, are fairly well retained by the biofilter structure and support many other studies reporting 72–99% retention of Cu and Pb in biofilters (Blecken et al., 2009; Bäckström et al., 2003; Feng et al., 2012; Muthanna et al., 2007; Reddy et al., 2014; Valtanen et al., 2017). However, the negative retention of Zn (−180%) after stormwater irrigation indicates leaching from composted sewage sludge with low pH in discharge. The mobility of Zn is promoted by low pH (Aastrup et al., 1995). After stormwater addition, only peat soil, with higher runoff pH, retained considerable

amounts (ca. 75% of the added amount) of Zn. Leaf compost has been shown to remove Al in stormwater biofilters (Feng et al., 2012), while in our study sewage compost had the lowest retention of Al, which may result from the very different origin of the compost material. Metal concentrations measured in the composted sewage sludge soil used in our study were far below threshold values (300, 1500 and 100 mg/kg dw for Cr, Zn and Pb, respectively) provided by Finnish environmental authorities. In spite of this, when comparing the metal concentrations of leachates to the classification of Stockholm Vatten (2001), the event mean concentrations (see supplemental material) of Ni (634 µg/l before stormwater irrigation, 1010 µg/l after irrigation), Zn (330–580 µg/l), Cu (25–206 µg/l) and Cd (2.4–3.4 µg/l) from compost treatments exceeded limit values of 225, 300, 45 and 1.5 µg/l, respectively.

Even though Cd and Ni were not present in our artificial stormwater mixture, these elements were measured in high concentrations in water filtrates - especially from the compost treatments after stormwater addition. These metals likely started to mobilise in the soil materials later on when the lysimeter soils became saturated with rain and stormwater. Especially lysimeters containing compost reached moisture up to 60% of volumetric content. From these systems the leaching of Cd and Ni were 3.5 and 5 x higher than from the peat systems. Since Cd is very mobile at low pH conditions in soil and soilwater (Aastrup et al., 1995), the low pH in Comp probably contributed to the leaching of Cd. As the discharge velocity was low, on average 1.5 l/h, there was enough time for various interactions between the soil and leaching water to take place, which, in turn, may have lowered the leachate pH. After two years, concentrations of Ni and Cd in the leachates from the compost containing treatments were still higher than those from the peat containing system. These results emphasise the long-term leaching of these metals even two years after the establishment of biofilters. Thus, our study highlights a notable time lag in the leaching of metals, especially as the loads suddenly increased 18 months after the start of the experiment following a very rainy preceding month. In contrast, the leaching of nutrients started immediately after the first rain event, suggesting that metals are more strongly bound to soil particles than nutrients and, thus, release more slowly. As elevated loads of metals were observed already well before air and soil temperatures dropped below 0 °C, freezing or thawing could not explain the sudden leaching of metals. Rather, the accumulated rainfall (1000 mm until mid-December 2015) and consequently the increased amount of infiltrating water (totalling ca. 55, 60 and 82 mm in Comp, Comp+bc and Peat, respectively) may had gradually transported metals down through the lysimeters.

4.3. Effects of biochar on nutrient and metal leaching

We have shown previously that biochar of the same type as used in this study, reduces the leaching of TP (Kuoppamäki et al., 2016). This effect was not evident in the current study, possibly due to extremely low TP concentrations in discharge. Instead, biochar reduced TN leaching, which is likely due to (i) sorption of N onto biochar (Ahmadvand et al., 2018; Gai et al., 2014) or (ii) biochar induced effects on soil abiotic and biotic properties (DeLuca et al., 2015). Biochar application modifies several soil physical properties such as cation exchange capacity, pH, redox conditions and water holding capacity, which, in turn, affect soil microbiological processes and thus impacts on the bio-availability and uptake of nutrients to plants (DeLuca et al., 2015). The mass export and retention of TN and TP in biofilter systems have been suggested to be primarily influenced by hydrological rather than biogeochemical mechanisms (reviewed in Jefferson et al., 2017). Since the water retention capacity did not differ between the three treatments at the end of the current study, the effect of biochar was apparently more related to biogeochemistry than hydrology.

Both increased (Saarnio et al., 2018) and decreased (Knowles et al., 2011) runoff of N from soils after biochar addition have been reported.

Biochar itself may also contain nitrogen in labile forms that are prone to leaching (Iqbal et al., 2015; Ulrich et al., 2015). However, according to Iqbal et al. (2015), Douglas fir originated biochar (650 °C) had no influence on the leaching of TN and TP from bioretention systems with compost material. It is well established that, depending on feedstock material and processing conditions, biochar characteristics may vary considerably, which should be taken into consideration when utilising biochar in environmental management (DeLuca et al., 2015; Clough et al., 2013). The short and long-term implications of biochar on N dynamics within and outside biofilter systems are specific to soil-biochar combinations, which call for further systematic studies to evaluate feedstock types and processing conditions for the successful management of water in urbanised watersheds.

Recent laboratory studies have shown the potential of biochar to remove contaminants from wastewater (Ahmad et al., 2014; Tan et al., 2015), but so far there have been no field studies to show the potential of biochar to purify stormwater. We showed that biochar reduced the leaching of sewage sludge originated Cd, Ni and Zn by ca. 50% compared to compost soil without biochar addition although after stormwater addition this effect was not statistically significant. Activated carbon is considered an effective adsorbent of various pollutants, and biochar can be used as a cheaper option in many applications (Thompson et al., 2016). The lack of consistency in published data makes comparisons between biochar types difficult. In addition to soil type in the biofilter structures, the properties of biochar also affects the fate of heavy metals (see Beesley et al., 2011; Egene et al., 2018; Wu et al., 2017). Our results suggest that the type of soil substrate used in biofiltration systems is important. However, mixing sewage sludge originated compost with biochar of birch origin, and adding a few cm thick layer of biochar underneath the organic soil layer can substantially reduce the leaching of N and heavy metals.

4.4. Concluding remarks

Given the substantial cover of roadside land at the urban fringe and, thus, the potential loading originating from such areas, the use of sewage sludge and other materials rich in nutrients and metals at the road verges should be considered carefully. The leaching of nutrients and metals from potential materials should be tested before using them in green infrastructure in urban watersheds. As with soils in other urban greenspaces (Pouyat et al., 2007), urban green swales are generally N saturated and thus the addition of extra N should be avoided to protect watersheds next to aquatic ecosystems. The retention of N and metals in biofiltration systems can be improved, as shown in our study, by biochar addition, but the capacity of biochar to retain both N and P can be context dependent, warranting careful verification before using. Synergistic or antagonistic effects of co-existing constituents under dynamic field conditions have been identified as an issue that needs further examination, especially when studying the impacts of biochar (Mohanty et al., 2018). We agree with this argument and call for more long-term controlled field studies on the performance of various biochars in affecting the retention of pollutants in stormwater biofilter systems. Long-term studies are needed also to understand the overall, long-term performance of various biofilters. In our study, the impacts of vegetation on the retention of both P and N was remarkable in both treatments containing compost, while in lysimeters with peat the stunted growth of grasses contributed to their negligible impact on nutrient dynamics. Thus, our results highlight also the need to understand the role of vegetation in the performance of biofilter systems more profoundly.

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Appendix A. Supplementary data

A photo of the lysimeters and figures showing event mean concentrations of nutrients and metals in leachates during the stabilisation period and after the stormwater irrigation are given in the supplemental material. Supplementary data to this article can be found online at doi:<https://doi.org/10.1016/j.wsee.2019.05.001>.

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