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Removal and release of microplastics and other environmental pollutants during the start-up of bioretention filters treating stormwater

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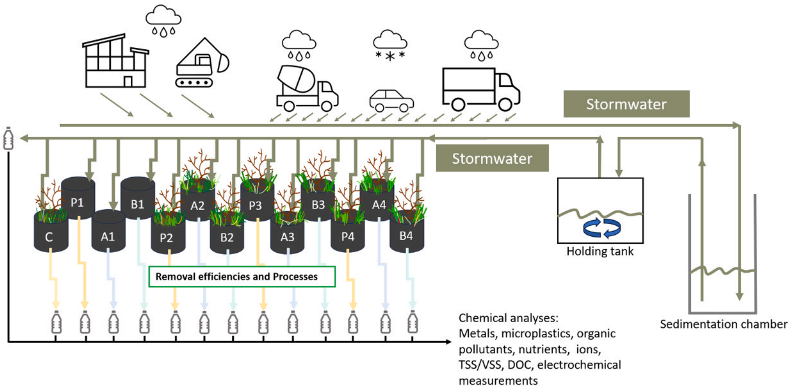
HIGHLIGHTS

- Bioretention filters with plants and sorbents effectively removed pollutants.
- The plants used were thrift, sea buckthorn, common rush, and red fescue.
- Microplastics >10 µm were effectively trapped in all bioretention filters.
- The leaching of nutrients and pollutants decreased from all bioretention filters.
- Biochar was the most efficient sorbent followed by ash and peat.

GRAPHICAL ABSTRACT



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ABSTRACT

Untreated stormwater is a major source of microplastics, organic pollutants, metals, and nutrients in urban water courses. The aim of this study was to improve the knowledge about the start-up periods of bioretention filters. A rain garden pilot facility with 13 bioretention filters was constructed and stormwater from a highway and adjacent impervious surfaces was used for irrigation for ~12 weeks. Selected plants (*Armeria maritima, Hippophae rhamnoides, Juncus effusus, and Festuca rubra*) was planted in ten filters. Stormwater percolated through the filters containing waste-to-energy bottom ash, biochar, or *Sphagnum* peat, mixed with sandy loam. Influent and effluent samples were taken to evaluate removal of the above-mentioned pollutants. All filters efficiently removed microplastics >10 µm, organic pollutants, and most metals. Copper leached from all filters initially but was significantly reduced in the biochar filters at the end of the period, while the other filters showed a declining trend. All filters leached nutrients initially, but concentrations decreased over time, and the biochar filters had efficiently reduced nitrogen after a few weeks. To conclude, all the filters effectively removed pollutants during

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the start-up period. Before being recommended for full-scale applications, the functionality of the filters after a longer period of operation should be evaluated.

1. Introduction

Stormwater from urban areas, and highly trafficked areas in particular, contains a large number of environmental pollutants, including metals, organic pollutants (OP) such as aliphatic hydrocarbons, aromatic hydrocarbons, polycyclic aromatic hydrocarbons (PAH), phthalates, and nutrients [6,54,74,77,86,96]. Recent studies have also shown that road runoff contains high concentrations of microplastics (MP) from road and tyre wear [45,46]. MP derived from road and tyre wear are predicted to become one of the major sources of microplastic loads globally [32,57]. These are also the largest quantified source of MP in Sweden and Norway [114,72] and represent the highest percentage of MP in European rivers [108].

Urban stormwater management has historically focused heavily on quantity control and combined sewer systems, but more recently, strategies have shifted towards blue-green infrastructure [26]. Further, the increased use of impervious surfaces has caused more pollutants being transported to receiving waters [97]. Recently, strategies have shifted towards blue-green infrastructure with an emphasis on both quantity and to reducing the discharge of pollutants and limiting the exposure of humans and the environment to hazardous substances [71]. Bioretention filters, often referred to as rain gardens, are sustainable treatment facilities, designed to prevent flooding but also to treat polluted stormwater in cities [21], by increasing surface permeability and providing processes such as sedimentation, filtration, sorption, biodegradation, uptake in roots and plants, and volatilisation [127,7,67]. The main purpose defines the design, including media depth, media composition, vegetation, addition of sorption materials, and volume of the submerged zone [39].

Many studies have shown that bioretention filters remove copper (Cu), zinc (Zn), phosphorus (P), and nitrogen (N) from stormwater [21,37]. Removal rates of Zn and Cu can reach around 80%, up to 95–99% [12,3,20,97], while dissolved and colloidal Cu seem harder to remove efficiently [60]. Removal rates for N and P are more variable [97]. N, P, and Cu can also leach from bioretention systems [16,98,97].

There are some studies indicating high removal efficiency of OP in bioretention filters ([19,47], although in-depth research is lacking. There are studies regarding removal of MP in stormwater using bioretention filters, e.g. [30,63,61,62,110], although there are still large knowledge gaps in this field and the results are difficult to compare, as there is great variation in the analytical methods, objectives, and other parameters in the existing literature, as well as a relatively low number of analysed samples [83]. The number of MP particles have been shown to vary between sites as well as with precipitation events [10], with larger stormwater volumes potentially diluting MP concentrations. It has been shown that the length of filters and the sizes and shapes of the particles affect the retention of MP in horizontal sand filters [106], and that MP particles mainly accumulate in the top layer in mature bioretention filters [63]. However, more studies investigating how MPs are removed, transported, and eventually degraded in biofilters are needed.

When treating stormwater from trafficked areas, the choice of plant species is of high importance as the plants must be able to resist long droughts as well as flooding, high pollutant, and chloride (Cl) loads. Plant selection also matters for infiltration rates, as different plants improve infiltration in different ways [116], and affect soil moisture, increase hydraulic conductivity and thereby the nitrogen removal compared to unplanted systems [18]. Vegetation can increase the rate of metal removal by bioretention filters, via plant uptake [79,101,75]. Phytoremediation techniques have shown that certain plants have a positive effect on the removal of OP from soil, but there is currently a lack of research on how plants affect OP in bioretention systems [18]. There are few studies addressing the potential effects of plants in bioretention systems regarding MP behaviour. When comparing removal of MP in the size range 100–300 μm, the performance of non-vegetated sand filters equalled that of a vegetated bioretention cell [61], while vegetation significantly increased the removal rates of smaller MP particles, 20–100 μm, compared to a non-vegetated sand filter.

Addition of sorption materials to the soil bed in biofilters has been researched, but there is a lack of studies testing the addition of sorbents in field conditions and examining how sorbents affect vegetation [118]. Coal fly ash has been studied for the removal of P [117,42]. Sand filters with a thin bottom layer of bottom ash from a refuse incineration plant were used in a laboratory study showed enhanced removal of nutrients from synthetic runoff [126]. Waste-to-energy bottom ash is not an established filter material for bioretention facilities, but rather an innovative option worth exploring because it is a residual product that does not need to be produced or mined. Residues from incineration of municipal solid waste incineration have large surface areas and high porosity [13], which both are desirable properties for filter material treating stormwater pollutants. A pilot plant using column bed filters of sand as a pre-filter, followed by a peat filter showed efficient removal of non-particulate OP from urban stormwater [74]. In a laboratory study, even in low concentrations metals showed a high sorption capacity on *Sphagnum* peat [51]. However, removal of copper might be affected due to leaching of organic matter, as filters constructed with a large amount of organic material, e.g., peat and compost, have been shown to leach nutrients and dissolved organic carbon (DOC) initially, which is correlated to leaching of copper [121,52]. In another study, peat was shown to be effective for uptake of nitrogen [29]. Biochar has received attention as a potential sorption material for stormwater management in recent years, showing positive results in removing various pollutants, mainly due to its high surface area and sorption capacities [5,102]. There are several reports regarding enhanced nutrient removal due to the addition of biochar in soil environments [33]. However, there is a lack of studies regarding the use of biochar in field conditions [5], as most previous studies were carried out as laboratory batch or column experiments.

There is also a lack of knowledge regarding the behaviour of bioretention filters during the start-up phase. A few studies indicate that there may be a so-called stabilisation phase as filters mature [113], affecting the hydraulic conductivity and removal efficiency (RE) for some pollutants. Initial low removal of total suspended solids (TSS) due to particulate washout has been observed [48], and increased concentrations of Cu, Zn, Cl, TSS, and total P in the effluent water after installation of rain gardens have been reported. However, to our knowledge, there are no studies on how other pollutants are affected during this stabilisation phase.

The aim of this study was to improve the knowledge about the start-up periods of bioretention filters and deepen the understanding of how different added sorption materials affect the function of the filters regarding removal efficiencies and processes, for nutrients and pollutants such as MP, OP, and metals in bioretention filters, with and without plants, and different sorption materials in field conditions during the start-up period. Pilot-scale bioretention filters planted with thrift (*Armeria maritima), sea buckthorn (Hippophae rhamnoides),* common rush *(Juncus effusus), and* red fescue *(Festuca rubra),* as well as biochar, *Sphagnum* peat, or ash from municipal solid waste incineration as sorption materials, were designed and constructed and irrigated with stormwater. Based on our current knowledge, this is the first study assessing the start-up and stabilisation of bioretention filters with different sorption materials in field conditions.

2. Materials and methods

2.1. Rain garden pilot facility location

The rain garden pilot facility was constructed in Gårda, Gothenburg, Sweden in spring 2022 and the study period lasted from May 30th to August 18th, 2022. The rain garden is located adjacent to the E6 Highway (Fig. 1). The bioretention filters in the rain garden were irrigated using stormwater. The stormwater originated from an area of 5.1 ha, of which 2.1 ha has impervious surfaces, made up of 82% roads, 6% roofs, and 12% other impervious areas [6,74]. The nearby E6 Highway is heavily trafficked and generates significant amounts of road wear/dust, from a daily mean of around 105,000 vehicles, of which 11% are heavy-duty vehicles according to the Swedish Transport Administration.

2.2. Overall experimental procedure

Fig. 2 illustrates the phases of the experimental procedure (program) applied to fulfil the aim of the study, i.e. to better understand of how different added sorption materials affect the removal efficiencies and processes, for nutrients and pollutants such as MP, OP, and metals in bioretention filters, with and without plants, in field conditions during the start-up period.

2.3. Method for selecting plants

The selection criteria for the plants used in this study are presented in Table 1. A list of appropriate criteria for this study was developed, and potential species candidates were compared to the chosen criteria. Four plant species were selected to be used together in the bioretention filters: *Armeria maritima*, *Hippophae rhamnoides*, *Juncus effusus*, and *Festuca rubra*. These plants were selected because they can stand in water for short periods of time, can withstand shorter droughts, tolerate increased salinity in the water, and can contribute to the removal of traffic-related pollutants and/or improve soil bed substrate conditions for microorganisms. Importantly, they are not considered invasive species in the specific environment where the rain garden is located.

2.4. Filter bed materials

The sorption material is either *Sphagnum* peat, biochar, or a combination of separate layers of peat, biochar, and metal-sorted and aged bottom ash from municipal solid waste incineration (MIBA) with added compost. The choice of peat as one of the filter materials for this study was based on previous research on peat and its sorption capacities [51, 73]. Biochar was selected as it has been shown to sorb a wide range of pollutants present in stormwater [5]. MIBA was mainly chosen due to its high metal content and could be seen as a secondary raw material for metals, Table 2 and [56]. Research about MIBA as filter material as well as cultivation in MIBA is limited, but initial studies show that MIBA is a promising material [56,104,109]) and it was therefore included in the present study.

All filter bed materials were characterised (see Table 2) for loss on ignition (LOI) 1000 °C according to SE-SOP-0060, and particle size distribution according to former SS027123 (modified) and former SS027124 (modified), water contents were analysed according to SS-EN 15934:2012 version 1. Total organic carbon (TOC) was analysed according to CSN ISO 10694 and CSN EN 13137:2002 CSN EN 15936 and total nitrogen (N) according to CSN ISO 1126d. Content of PAHs in sand, peat, sandy loam and compost were done according to US EPA 8270D+US EPA 8082 A+CSN EN 15527+ISO 18287+ISO 10382+CSN EN 15308. Content of PAHs in biochar and ash were done according to DIN ISO 18287: 2006–05. Content of specific metals were all done using ICP SFMS (inductively coupled plasma sector field mass spectrometry), but digestion and analysis methods differed. In sand, sphagnum peat, sandy loam, and compost, analyses were done according to SS-EN ISO 17294–2:2016 and US EPA Method 200.8:1994 after digestion with lithium metaborate ($LiBO_2$) smelt i.e. SE-SOP-0060 (ASTM D3682:2013; ASTM D4503:2008; An. Chem. 50:679–680) or SS-EN ISO 17294–2:2016 and US EPA Method 200.8:1994 after digestion using 7 M nitric acid (HNO3) according to SE-SOP-0021. For biochar SS-EN ISO 17294–2:2016 and US EPA Method 200.8:1994 were used after digestion using LiBO2 smelt i.e. SE-SOP-0060 (ASTM D3682: 2013; ASTM D4503:2008; An. Chem. 50:679–680) or SS-EN ISO 17294–2:2016 and US EPA Method 200.8:1994 after digestion according to SE-SOP-0721 i.e. microwave digestion using acid. Ash was analysed using SS-EN ISO 17294–2:2016 and US EPA Method 200.8:1994 after digestion using SE-SOP-0039 (SS-EN 13656:2003) i.e. acid digestion in HotBloc®. Mercury (Hg) was analysed according to SS-EN ISO 17852:2008 using the same digestion method. Other parameters of importance were compiled from previous studies of the materials or data from the literature.

The high LOI of peat (97%) and relatively high LOI of compost (23%) reflect the high organic content in the respective materials, compared to sand (0.42%), ash (7.5%), and sandy loam (3.9%). Biochar was not examined for LOI but had high concentrations of TOC and PAH-16 compared to the other materials. Biochar also had a much higher BET surface area than the peat material, even if the peat material was composed of smaller particles. Total N was highest in the compost and peat materials. According to the findings in the literature, biochar, peat,

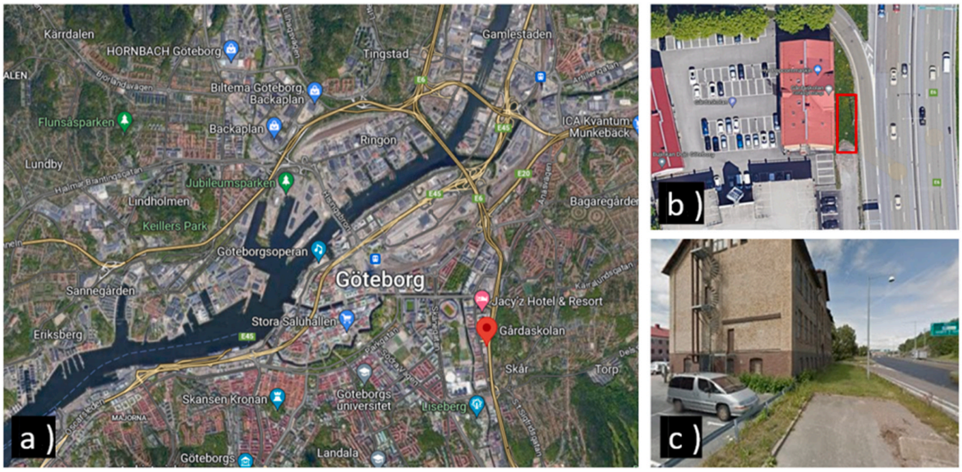


Fig. 1. (a) Map of Gothenburg where the Gårda area is marked with a red dot, (b) Aerial view of the construction site for the Gårda rain garden, (c) Photo of the site selected for the rain garden.

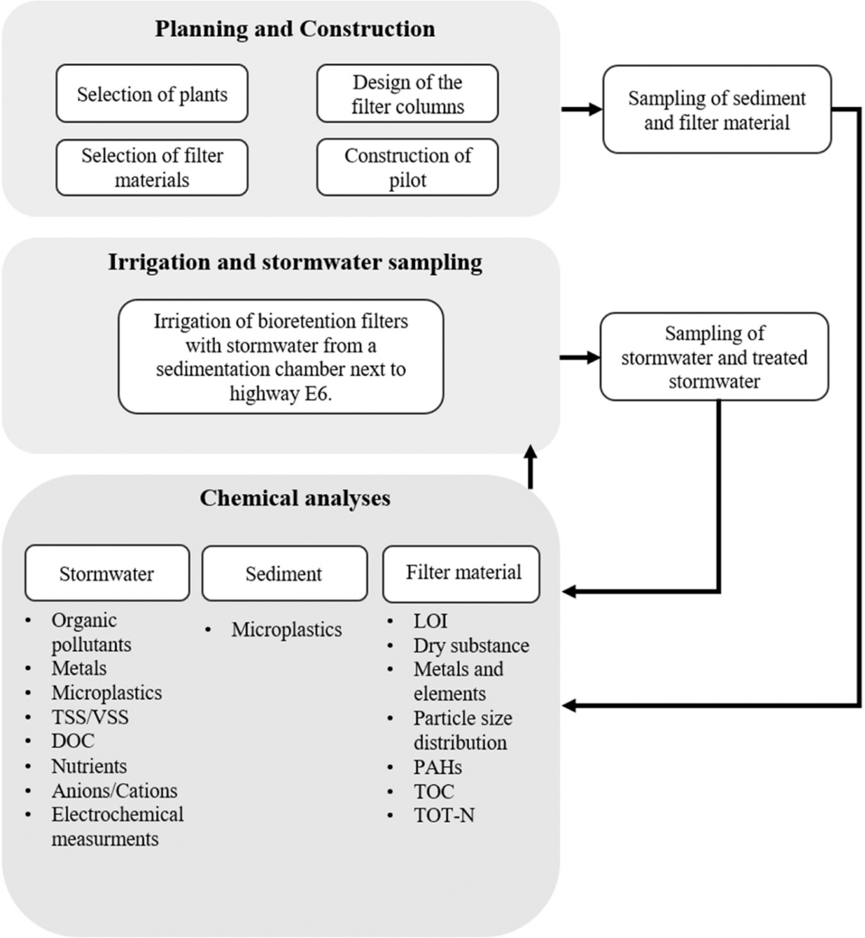


Fig. 2. Flowchart describing the phases of the experimental program.

and ash are all sorption materials with a high capacity to sorb metals, organic pollutants, and nutrients, see the references in Table 2.

2.5. *Design of filter columns and pilot facility*

In the rain garden pilot facility, 13 pilot-scale bioretention filters were established in columns made of polyethylene (~980 L). In the upper layer of all filters, there is a layer of sandy loam mixed with pumice stone, followed by a layer of sandy loam mixed with pumice stone and a sorption material, and a pure sorption material layer. Below the sorption layer, there is a layer of fine sand, followed by a gravel layer for drainage (Fig. 3).

The columns inner dimensions are 1250 × 1000 mm. The filter columns are designed with three effluent pipes at different heights (from the bottom: 110 mm, 400 mm, and 800 mm). During this campaign, only the lower effluent pipe was open in each filter. The lower effluent pipe is perforated within the filter bed and extends through the entire width of the bed, whereas the other two effluents pipes do not.

The part of the filter below the lower effluent pipe can be considered saturated with water. Previous research has shown that this is beneficial for nutrient removal, and for maintaining hydraulic conductivity and sustenance for plants and microbiome [105,120,124]. A 20 L container was used to collect the effluents. For each sorption material, there are three identical bioretention filters with plants and one without plants. There is also a control filter with plants but without a sorption layer. The location of the different columns, with and without vegetation, within the Gårda rain garden is displayed in Fig. 4.

2.6. *Operation of the pilot facility*

The Gårda area has a sedimentation facility for stormwater treatment, which consists of seven sedimentation chambers connected in series. The stormwater used for irrigation of the rain garden was pumped from the first chamber.

2.6.1. *Irrigation*

During the start-up period, the bioretention filters were irrigated 16 times, with volumes of 20–70 L stormwater per filter on each occasion. Water volumes for the irrigation were based on calculations of block rains, but also on the availability of stormwater. Dimensioning rain intensity was calculated according to Swedish Water's publication P110 eq. 4.5, p.65:

$$iå = [190\ x\ \sqrt[3]{Å}\ x\ (\ln\ (Tr)/Tr0{\cdot}98)]$$

where:

iå = rain intensity, l/s, ha

Tr = rain duration, min

Å = return time, months

Stormwater was pumped from the sedimentation chamber which receives the incoming water from the Gårda area and stored in Intermediate Bulk Containers (IBCs). Before irrigation, the water in the IBCs was mixed with a fast-rotating agitator to stir up sedimented particles. Irrigation was performed either by pumping water from the IBCs and filling watering cans or by pumping water directly onto the receiving columns.

Table 1
Selection criteria for the group of plants used in the bioretention filters in the Gårda rain garden and deemed suitable for this study. Selection criteria were based on reports from the Swedish Transport Administration [103], Adoption Guidelines for Stormwater Biofiltration Systems – Summary Report [89], Svenskt Vatten, which represents Sweden's municipal VA departments, [64], scientific reports e.g. [18,36,59,69,70,90,101], and a book by [38].

	Plants			
Criterion	***Hippophaë rhamnoides*** **Sea Buckthorn**	***Festuca rubra*** **Red Fescue**	***Juncus effusus*** **Common Rush**	***Armeria maritima*** **Sea Thrift**
Tolerance to Swedish climate and seasonal variances in temperature, light, precipitation, winds etc.	Mainly a coastal plant in Europe. Can be found in more arid places in Asia. Can tolerate sub-alpine environments. Can tolerate moderate droughts[49].	Widespread across the Northern hemisphere. Tolerates many different climates and habitats. Tolerant to shade [23].	Broad, nearly global distribution. Its habitats include wetland habitats such as marshes, swamps, wet pastures, and ditches. It is considered widely adaptable. Can tolerate short periods of drought.	Circumpolar distribution. Thrives in coastal areas in the Northern hemisphere. Drought tolerant once established. Tolerates part shade [123].
The plant should be native to Sweden and not be classed as invasive in this part of Sweden.	Native to Scandinavia (Sweden)	Native to Scandinavia (Sweden)	Native to Scandinavia (Sweden)	Native to Scandinavia (Sweden)
Tolerance to a cocktail of traffic-related pollutants from stormwater, including metals, organic pollutants, nutrients, salt (due to de-icing), and tyre and road wear particles (TRWP).	Resistant to salt and alkali stress[14]. Tolerant to terrains degraded by industrial activities as well as being close to heavily trafficked roads [78].	Tolerance to soils polluted with heavy metals[24] and petroleum[85].	High tolerance to metal pollution [93].	High tolerance to salt and heavy metals[123,99]
The plant should not directly compete with the other plants in the bioretention filter.	Mostly competes with other shrubs, bushes, and trees.	Does not compete directly for light. Tolerant to shade.	Have been observed to shade smaller plants under wetland conditions.	Does not compete directly for light. Partly tolerant to shade.
Should contribute to a bioretention filter with diverse types of plants.	Deciduous shrub.	Perennial grass that forms bunchgrass tufts.	Perennial herbaceous flowering plant.	Perennial flowering plant.
Should have a root system that increases the diversity of root types in the bioretention filter.	Rapidly growing, extensive root system.	Laterally spreading roots.	Forms a stoutbranching rootstock with short, finely divided rhizomes.	Woody taproot.
The plant should be perennial.	Can live between 30–50 years.	Perennial.	Perennial.	Perennial.
Should be able to perform some form of phytoremediation relevant to traffic-related pollutants and/or have qualities that improves the success rate of the biofilter.	Documented possible uptake of heavy metals[17,94]. Used for land reclamation[49].	Identified as suitable for phytostabilisation of heavy metal polluted soils.	Treatment with common rush can have positive effect on removal of nutrients and metals in polluted waters [35,80,115].	Hyperaccumulator for several metals,[99,123])
Other characteristic that motivated the choice of a specific plant.	Symbiotic N_2 fixation [25, 128]. This trait is not always suitable, e.g., for filters treating water with very high amounts of nutrients.	Considered suitable for revegetation of metal-contaminated soil[31].	Releases high amounts of oxygen into the rhizosphere[9].	Shown to survive up to 6 weeks of extended drought[66].

2.6.2. Sampling

After agitation, grab samples were collected from the water used for irrigation in the IBCs, henceforth referred to as influent, and from the outflow containers where the effluents from the filters were collected. The sampling containers were emptied before each irrigation, and samples were, when possible, not drawn until > 10 L had entered each outlet container to ensure a representative sample for the whole irrigation procedure. The total amount of outflow to the containers varied over the start-up period due to the amount of natural precipitation, the amounts used for irrigation, and the amount of water held by the filters prior to irrigation. A peristaltic pump was used to collect a sample from each effluent container for the chemical analyses. The samples were stored in glass bottles and kept in a cooling bag until transported to the laboratory, where they were stored in refrigerators until further analyses.

2.7. Analyses

2.7.1. Microplastics polymers

Ten different MP polymers (polyisoprene (PI), polybutadiene (PB), polyethylene (PE), polypropylene (PP), polyvinylchloride (PVC), polystyrene (PS), polyethylene terephthalate (PET), polyamide 6 (PA6), polymethylmethacrylate (PMMA), polycarbonate (PC)) were analysed in stormwater samples and one sediment sample at a commercial laboratory. The stormwater samples were first filtrated through a stainless-steel filter (SSF) with a pore size of 10 µm. The filter was then transferred to a beaker with water for 10 min ultrasonication. The filter was then rinsed and washed into the beaker before potassium hydroxide (KOH) was added to a final concentration of 10% KOH. The sample was then incubated at 40 °C overnight before a new filtration at 10 µm SSF. After incubation, the filter was ultrasonicated again, then removed before addition of calcium chloride ($CaCl_2$) for density separation. The sample were decanted at least three times before it was filtrated using a 1.6 µm GF/A filter prior to analysis. MP analysis was performed with a pyrolysis- Gas chromatography–mass spectrometry (GC/MS) setup (Pyrolyzer from Frontier-labs, Agilent GC 8890, Agilent MS 5977b). Samples were added to eco-cups which were put in the auto-sampler. Quality controls were weighed out manually from a mixture of 11 polymers with silicon dioxide (SiO_2) as diluent, with internal polymer standards added as well. The results were processed and interpreted in F-Search 2.0. For the sediment, at least a 25 g dry weight equivalent of the wet weight sample was weighed out into a beaker. If the dry matter percentage was high, small amounts of water were added before pouring $CaCl_2$ into the sample for density separation. Samples were decanted at least three

Table 2
Physical and chemical characterisation of filter bed materials.

Physical/Chemical parameters		**Filter bed materials**					
		Sand	**Biochar**	***Sphagnum* Peat**	**Ash**	**Sandy loam**	**Compost**
Product name, manufacturer		Filter sand, Rådasand	Biokol, Hjelmsäters Egendom	ScanPeat Blocktorv, ScanPeat	MIBA, Renova, Swedish WtE incineration plant	Hekla Regnbädd, Bara Mineraler	Peat-free potting soil, Plantagen
Sorbent information (from manufacturer)		Filter sand for water purification. Quartz, non-respirable.	Produced exclusively from local, FSC certified wood chips and local harvest residues. The wood chips come mainly from a local spruce forest infested with spruce bark beetle.	Raw peat is heated to approx. 350 °C in an anoxic environment, leading to granulation; humification degree H3–4	MIBA is a residual product from combustion and therefore has a low cost, both economically and environmentally, as it does not need to be mined or produced separately. MIBA is a draining material and has many active surfaces.	The product is an active substrate based on pumice stone, green compost, and sand.	Composed of fibre mulch, bark mulch, garden compost, and stone flour rich in silicon, iron, and magnesium. Other components are extracted from bark from sawmills, wood fibres from papermaking, and natural manure from farms.
pH		[a]	8.2[b]	2.6[c]	9.4[d]	-	-
Dry substance (DS) 105°	%	>99.5	93.3	88	80	71	41
Organic content, LOI 1000 °C	%	0.42	n.a.[e]	97	7.5	3.9	23
Total organic carbon (TOC)	%	0.23	91	47	1.3	2.2	12
Bulk density	kg/m^3	-	155[b]	77[c]	1080[d]	-	-
BET surface area	m^2/g	-	370[b]	1.3[c]	-	-	-
Cation exchange capacity (CEC)	meq/kg	-	11–14 [f]	309 [g]	-	-	-
Electrical conductivity (EC)	μS/cm	-	420[b]	800[h]	8200[d]	-	-
Effective particle size d10[i]	mm	n.a.	0.26	0.017	-	-	-
Effective particle size d60[j]	mm	n.a.	2.0	0.99	1.7	0.41	0.72
Effective particle size d90[k]	mm	n.a.	5.4	3.5	5.3	5.0	3.5
Metal sorption capacity		-	High[l]	High[g,m,n,o]	High[p]	-	-
Organic pollutants sorption capacity		-	High[l]	High[c,m,q]	High[p]	-	-
Nutrient sorption capacity		-	High-N[q]	High-N[r] and P[s]	n.a.	-	-
Content of ΣPAH-16	mg/kg	<1.25	12.5	<1.25	0.14	<1.25	<1.25
Content of total N		126	2140	6930	1080	1100	8700
Metal content							
As	mg/kg	0.47	<0.20	0.56	40	0.93	1.4

(*continued on next page*)

Table 2 (*continued*)

Physical/Chemical parameters		Filter bed materials					
Cd	mg/kg	0.029	<0.010	0.081	3.4	0.051	0.75
Co	mg/kg	3.4	0.086	0.54	86	4.0	4.8
Cr	mg/kg	<10	3.2	<10	1200	42	20
Cu	mg/kg	15	1.7	26	4300	18	30
Hg	mg/kg	<0.040	<0.020	<0.040	0.028	<0.040	0.044
Ni	mg/kg	8.5	0.18	6.6	330	11	9.1
Pb	mg/kg	3.1	<0.080	4.5	880	5.4	11
Zn	mg/kg	23	13	17	4200	34	130

[a]-=no data; [b]data from supplier; [c][73]; [d][56]; [e]n.a.= not analysed; [f][100]; [g][51]; [h][27]; [i]The particle size where 10% of the particles in a sample (by weight) are smaller, while 90% are larger; [j]The particle size where 60% of the particles in a sample (by weight) are smaller, while 40% are larger; [k]The particle size where 90% of the particles in a sample (by weight) are smaller, while 10% are larger [l][5]; [m][54] [n][52]; [o][55]; [p][109]; [q][53]; [q][100]; [r][29]; [s][82].

times before being filtrated with a 10 µm SSF. Note that for the characterisation in Section 3.2.1 the size was 27 µm. The filter was then transferred to a beaker with water for 10 min ultrasonication and followed the same procedure as for the water samples as described above. The chemical analysis and data processing were then performed for the sediment samples, using the same procedures and equipment as described above.

2.7.2. *Organic pollutants*

For the OP, six groups of aliphatic hydrocarbons (aliphatic $>C_5$-C_8, aliphatic $>C_8$-C_{10}, aliphatic $>C_{10}$-C12, aliphatic $>C_{12}$-C_{16}, aliphatic $>_{C5}$-C_{16}, aliphatic $>C_{16}$-C_{35}), five groups of aromatic hydrocarbons (aromatics $>C_8$-C_{10}, aromatics $>C_{10}$-C_{16}, methyl pyrenes/methylfluoranthenes, methyl chrysenes/methylbenz(a)anthracenes, aromatics $>C_{16}$-C_{35}), BTEX (benzene, toluene, ethylbenzene, m,p-xylene and o-xylene), 16 specific polycyclic aromatic hydrocarbons (PAH-L: naphthalene, acenaphthylene, acenaphthene; PAH-M = fluorene, phenanthrene, anthracene, fluoranthene, pyrene; PAH-H = benz(a)anthracene, chrysene, benz(b)fluoranthene, benz(k)fluoranthene, benz(a)pyrene, indeno(1,2,3,cd)pyrene, benz(g,*h*,i)-perylene, dibenz(a,h)anthracene), and 13 specific phthalates (dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-propyl phthalate (DPP), di-n-butyl phthalate (DBP), di-iso-butyl phthalate (DIBP), di-n-pentyl phthalate (DNPP), di-n-octyl phthalate (DNOP), di-(2-ethylhexyl) phthalate (DEHP), butyl benzyl phthalate (BBP), di-cyclohexyl phthalate (DCP), di-iso-decyl phthalate (DIDP), di-iso-nonyl phthalate (DINP), di-n -hexyl phthalate) were analysed by a commercial laboratory. The selection of OP for analyses was based on a list of priority pollutants in road runoff [74] and previous studies of stormwater and urban materials [44,6,96]. Aliphatic hydrocarbons and aromatic hydrocarbons were determined by GC-MS according to internal methods based on SPIMFAB's manual for quality. PAH-16 were determined by GC-MS, based on US EPA 8270D, US EPA 8082 A, CSN EN ISO 6468 and US EPA 8000D. Phthalates were identified with GC-MS according to the standard method DIN EN ISO 18856:2005.

2.7.3. *Trace metals*

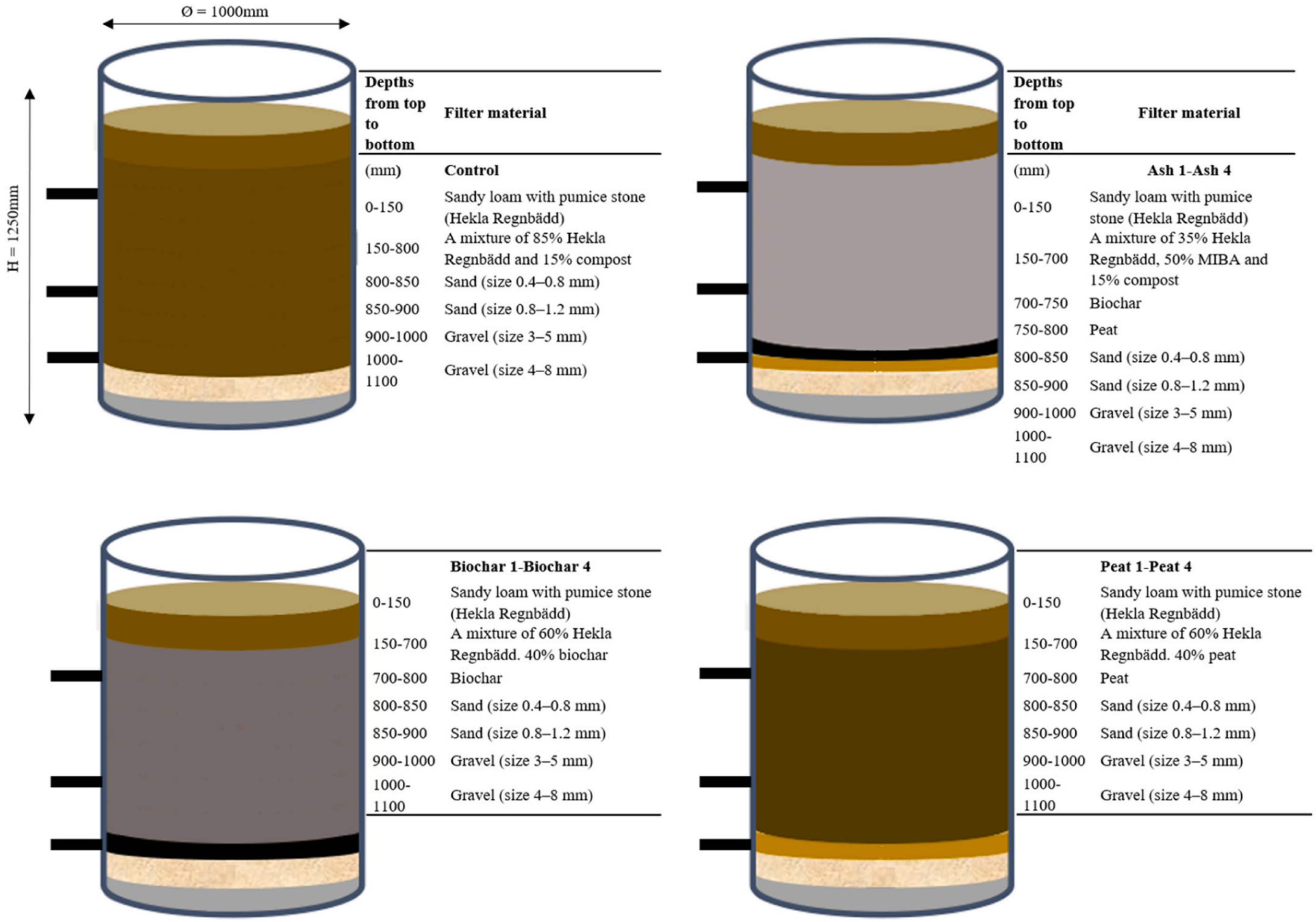
Arsenic (As), barium (Ba), lead (Pb), cadmium (Cd), cobalt (Co), copper (Cu), chromium (Cr), mercury (Hg), molybdenum (Mo), nickel (Ni), vanadium (V), and zinc (Zn) were determined in the water samples by a commercial laboratory. Prior to the metal analyses, the water samples were shaken thoroughly and then acid digested with 7 M nitric acid (HNO_3) (DuoPur). After digestion, the samples were analysed by inductively coupled plasma mass spectrometry (ICP/MS) following the standardised method SS-EN ISO 17294–2:2016.

2.7.4. *Other parameters*

Determination of total suspended solids (TSS) and volatile suspended solids (VSS) was carried out using the ESS Method 340.2: Total Suspended Solids, Mass Balance (revised June 1993). Anions and cations were analysed after filtration with a 0.45 µm filter in an ion chromatograph (Thermo-Fisher Dionex ICS-900). Dissolved organic carbon/ and nitrogen concentrations were analysed with a TOC analyser (Shimadzu TOC-V Series), after first being filtrated at 0.45 µm. General parameters such as pH, conductivity, turbidity, and redox were measured electrochemically with a multimeter (Hanna multimeter instrument (HI 9829)).

2.7.5. *Saturated hydraulic conductivity*

The saturated hydrological conductivity of the columns was tested in the field at the end of the campaign. First, the columns were filled with water until an outflow was observed (empty bed), and the required volume of water was noted. Secondly, the time from start to outflow and the time it took for 1 L of outflow to pass through the filter was noted. Once an outflow had been achieved, the outflow pipe was closed, the columns were filled to the brim with water, and the required volume was



Depths from top to bottom (mm)	Filter material
	Control
0-150	Sandy loam with pumice stone (Hekla Regnbädd)
150-800	A mixture of 85% Hekla Regnbädd and 15% compost
800-850	Sand (size 0.4–0.8 mm)
850-900	Sand (size 0.8–1.2 mm)
900-1000	Gravel (size 3–5 mm)
1000-1100	Gravel (size 4–8 mm)

Depths from top to bottom (mm)	Filter material
	Ash 1-Ash 4
0-150	Sandy loam with pumice stone (Hekla Regnbädd)
150-700	A mixture of 35% Hekla Regnbädd, 50% MIBA and 15% compost
700-750	Biochar
750-800	Peat
800-850	Sand (size 0.4–0.8 mm)
850-900	Sand (size 0.8–1.2 mm)
900-1000	Gravel (size 3–5 mm)
1000-1100	Gravel (size 4–8 mm)

	Biochar 1-Biochar 4
0-150	Sandy loam with pumice stone (Hekla Regnbädd)
150-700	A mixture of 60% Hekla Regnbädd. 40% biochar
700-800	Biochar
800-850	Sand (size 0.4–0.8 mm)
850-900	Sand (size 0.8–1.2 mm)
900-1000	Gravel (size 3–5 mm)
1000-1100	Gravel (size 4–8 mm)

	Peat 1-Peat 4
0-150	Sandy loam with pumice stone (Hekla Regnbädd)
150-700	A mixture of 60% Hekla Regnbädd. 40% peat
700-800	Peat
800-850	Sand (size 0.4–0.8 mm)
850-900	Sand (size 0.8–1.2 mm)
900-1000	Gravel (size 3–5 mm)
1000-1100	Gravel (size 4–8 mm)

Fig. 3. Filter column design for the bioretention filters in the Gårda rain garden pilot facility.

noted. When the columns were fully saturated, after about one hour and when bubbles were no longer observed on the surface, the outflow was opened and the time it took for the water to sink from the upper edge (buffer zone) to the soil level was recorded. The saturated hydraulic conductivity (K_{sat}) was calculated for each column using Darcy's Law.

$K_{sat} = [(\text{Volume}/\text{Time})/\text{Area})]$

2.8. Removal efficiency (RE)

For the removal efficiencies, it was assumed that the volume of water in the inlet is equal to the volume of water in the effluent. This is a rough estimation, as some water will get sorbed in the material in the columns.

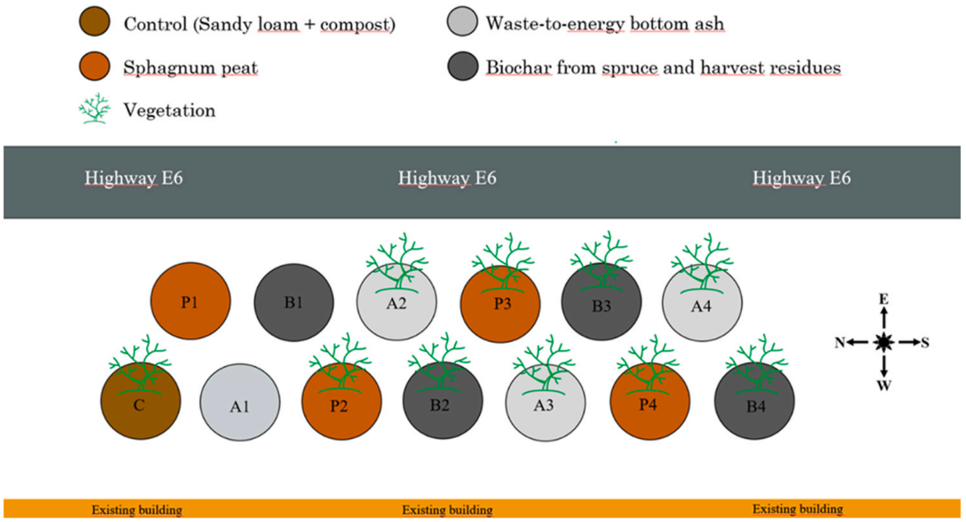


Fig. 4. The location of the different columns, with and without plants, within the Gårda rain garden. C = control filter, P = peat filter, A = ash filter, B = biochar filter.

The plants were assumed to take up no water, and evaporation was assumed to be zero.

RE % = [(Influent concentration – Effluent concentration)/Influent concentration] x 100

2.9. Principal component analysis (PCA)

The effluents from the filters were compared using a principal component analysis (PCA) using the Python package Scikit-learn [91]. Included parameters were pH, redox potential, turbidity, conductivity, dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), total dissolved nitrogen (TDN), ammonium (NH_4^+), nitrogen dioxide (NO_2^-), nitrate (NO_3^-), potassium (K^+), calcium (Ca^{2+}), magnesium (Mg^{2+}), and sulphate (SO_4^{2-}). All parameter values were standardised to a mean of 0 and standard deviation of 1 prior to the PCA.

3. Results and discussion

3.1. Comparison of effluent parameters using PCA

PCA was performed to evaluate if filters with the same filter materials behaved the same, and to get an overview of potential relationships between parameters and filter types. Bioretention filters with the same sorption material had similar effluent characteristics and the presence/absence of plants did not have an apparent effect on the effluents (Fig. 5). The control filter with compost was most similar to the peat filters. The PCA patterns were consistent during both the first and the second half of the start-up period. Biochar filters had higher pH and dissolved inorganic carbon (DIC) concentrations in the effluents. This can be explained by the high alkalinity of biochar, where carbonates make up a significant fraction of the alkalinity [28]. The ash filters had higher conductivity and concentrations of ions, which can be explained by the high salinity of MIBA [104]. The peat filters had higher effluent concentrations of TDN and DOC. Biochar has previously been shown to improve nutrient retention and plant uptake in soil [92]. Therefore, the absence of biochar in the peat filters may explain the higher effluent concentrations of TDN and NO_3^-.

3.2. Microplastics

3.2.1. Characterisation of MP in sediment and stormwater in Gårda

At the start of this study, a stormwater sample and a sediment sample from the sedimentation chamber used to irrigate the rain garden were analysed for MP. The results, including concentrations and percentage relative compositions of ten different microplastic polymers > 27 µm are presented in Fig. 6. A major part of the MP in the stormwater sample originated from tyre wear, as indicated by the high concentrations of polyisoprene (PI) of 147 µg/L (46%) and polybutadiene (PB) of 59 µg/L (18%). Urban stormwater from trafficked areas is known to contain large amounts of tyre and road wear particles [44,43] and the relative percentage of the polymers was similar to measurements carried out on snow samples from another urban area in Sweden [8]. Other detected polymers included polyvinylchloride (PVC) 48 µg/L (15%), polyethylene (PE) 37 µg/L (12%) and polypropylene (PP) 30 µg/L (9%). In the sediment sample, the relative percentage of PB was only 4.6% (11,500 µg /kg DS), and PI was 56% (142,000 µg/kg DS), indicating a high

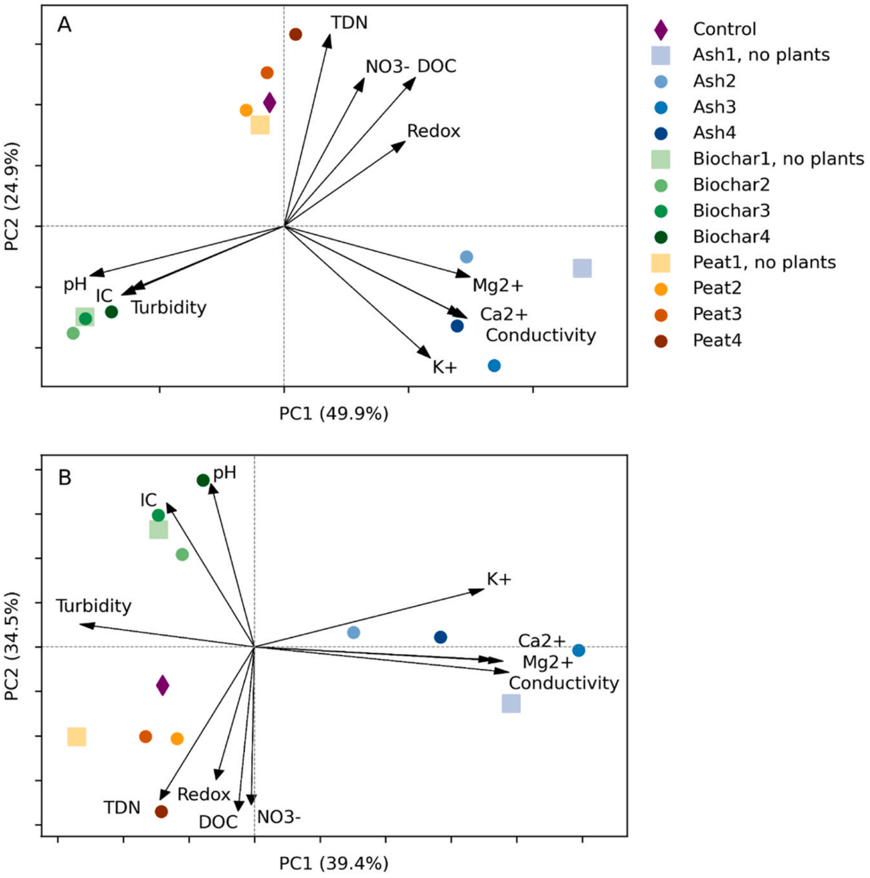


Fig. 5. Principal component analysis (PCA) showing similarity in effluent quality between the bioretention filters. The PCA is based on mean values of each parameter for the first (A) and second half (B) of the start-up period, respectively. The first half is defined as May 30th – June 30th and the second half is July 1st – Aug. 18th.

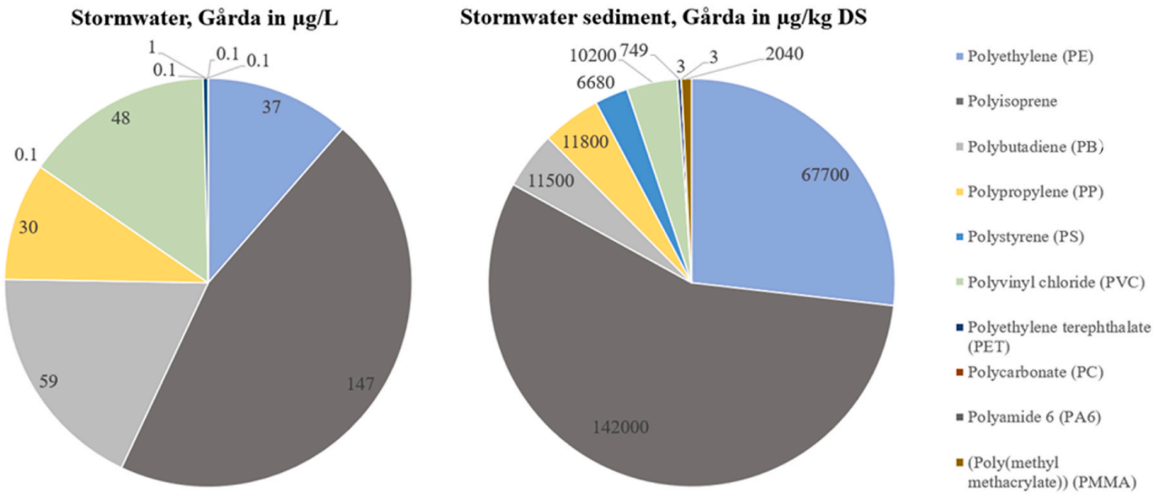


Fig. 6. Concentrations and relative compositions of microplastic polymers in stormwater used for irrigation and stormwater sediment from the chamber at the Gårda pilot rain garden, 2022-05-30.

occurrence of rubber wear particles with a high content of natural rubber in the sediment. In addition, other plastic polymers that are widely used in society, such as (PE) (27%), polypropylene (PP) (4.7%), (PVC) (4.0%), (PS) (2.6%), polymethyl acrylate (PMMA) (0.81%) and polyethylene terephthalate (PET) (0.30%) were found in a total amount of about 99,000 µg/kg DS.

3.2.2. *MP removal*

From the stormwater used to irrigate the bioretention filters, a total of 41 samples were analysed for MP, distributed between influent and effluent from filters C (control), A1, B1, P1, A4, B2, and P4. Polymers from microplastic particles >10µm were quantified in concentrations >1.0 µg/L, see Table 3and the most common MP, PE, were quantified in more than 50% of the influent water samples, while they were generally quantified in less than 10% of the effluent samples, regardless of filter type. Polyamide 6 (PA6) was not detected in neither the influent nor the effluent during any sampling occasion in this study. PMMA was detected once, in the effluent from P4.

The measured MP concentrations in the influent showed large variation in composition between sampling occasions, with a span of one detected type of microplastic and zero detected rubber components to six different types of microplastics and the presence of rubber components. Similar variations of MP levels in stormwater have been shown in other studies [110,61]. The individual amounts of different types of MP also varied significantly. Rubber components, as indicated by concentrations of PI and PB, were detected in the influent on four out of seven sampling occasions. PE was the only MP detected in all influent samples, at concentrations between 8.5–180 µg/L. PET and PC were detected once in the influent, at very low levels (1.3 µg/L and 1.2 µg/L, respectively). Note that the effluent water had been stagnant in the well before being pumped up to the IBCs, why the heaviest polymer particles are assumed to have settled into the sediment in the chamber, therefore not reaching the bioretention filters.

At the final sampling occasion, the removal capacity for polymers reached 98–100% for all polymers, except for PE and PP, for which the capacity was lower (>83%). Additionally, PC and PMMA were released from filter P4. For PP, the high concentrations in the effluent from most filters may be explained by contamination of PP residuals in the sampling containers after holes were drilled when assembling the pilot facility. Overall, the results from the start-up period showed very good removal efficiencies, indicating that rain gardens could potentially be used to decrease the spread of tyre wear and other MP. The differences in characteristics of the filter materials do not seem to affect the filter removal capacity of MPs during the start-up phase. Two recent studies examining the MP removal potential of bioretention filters showed similar results, although it is not common to analyse particles as small as down to 10 µm [61,62]. In a bioretention cell next to heavy car and foot traffic, on average 91% of all MP particles >20µm were removed during three storm events [30]. Another study showed a high reduction in MP particles (median 84%) in the fraction >106µm in a bioretention filter [110]. These measurements spanned over two years, and the number of MP particles in stormwater collected from a parking lot fluctuated greatly. The variations were correlated to dry days and rainfall intensity.

3.3. *Organic pollutants*

Specific aliphatic and aromatic hydrocarbons, PAH, and phthalates (see Methods) were analysed on several occasions in the influent and effluents from selected bioretention filters during the start-up period. The selected filters without plants were ash A1, biochar B1, and peat P1; and filters with plants were ash A2 + A4, biochar B2 + B4, and peat P3 + P4. Concentrations of all the specific OP that were higher than the limit of quantification in the influent and effluents of the filters during the campaign are presented in Table 4. Phthalates were not quantified in the influent nor the effluents. Also, the last sampling of filter P4 did not yield enough volume for all analyses, so the data set was supplemented with a sample from a later occasion (2022-10-11), for reference.

3.3.1. *Aliphatic hydrocarbons and PAH-16*

Aliphatic hydrocarbons C_5-C_{16} up to 81 µg/L, and C_{16}-C_{35} up to 110 µg/L, were analysed in the influent samples (Table 4.). In June, up to 99 µg/L of aliphatic hydrocarbons C_{16}-C_{35} leached from the peat filter P1, and from the biochar filters B1 and B2. However, these concentrations were much lower than the City of Gothenburg's guidelines of 1000 µg/L. In August, low concentrations of aliphatic hydrocarbons C_{16}-C_{35} were released from one of the peat filters, P1 (Table 4); for all the other filters, the RE was 100%. The main source of aliphatic hydrocarbons in stormwater is assumed to be vehicle exhausts [125], in particular exhausts emitted in the gas and particulate phase from diesel engines [1], but also engine oil and asphalt wear [41].

For PAH-16, the following specific compounds were detected in concentrations higher than the limit of quantification in the inlet samples; in concentration order: pyrene > fluoranthene > benzo(g,*h*,i)perylene > benzo(b)fluoranthene ≈ chrysene > phenanthrene > benzo(a) pyrene > indeno(1,2,3,cd)pyrene > benzo(k)fluoranthene, which is in line with the relative composition of specific PAH found in urban stormwater [96] and in stormwater sediments in the sedimentation facility at Gårda [74]. The PAH composition in this study suggests a

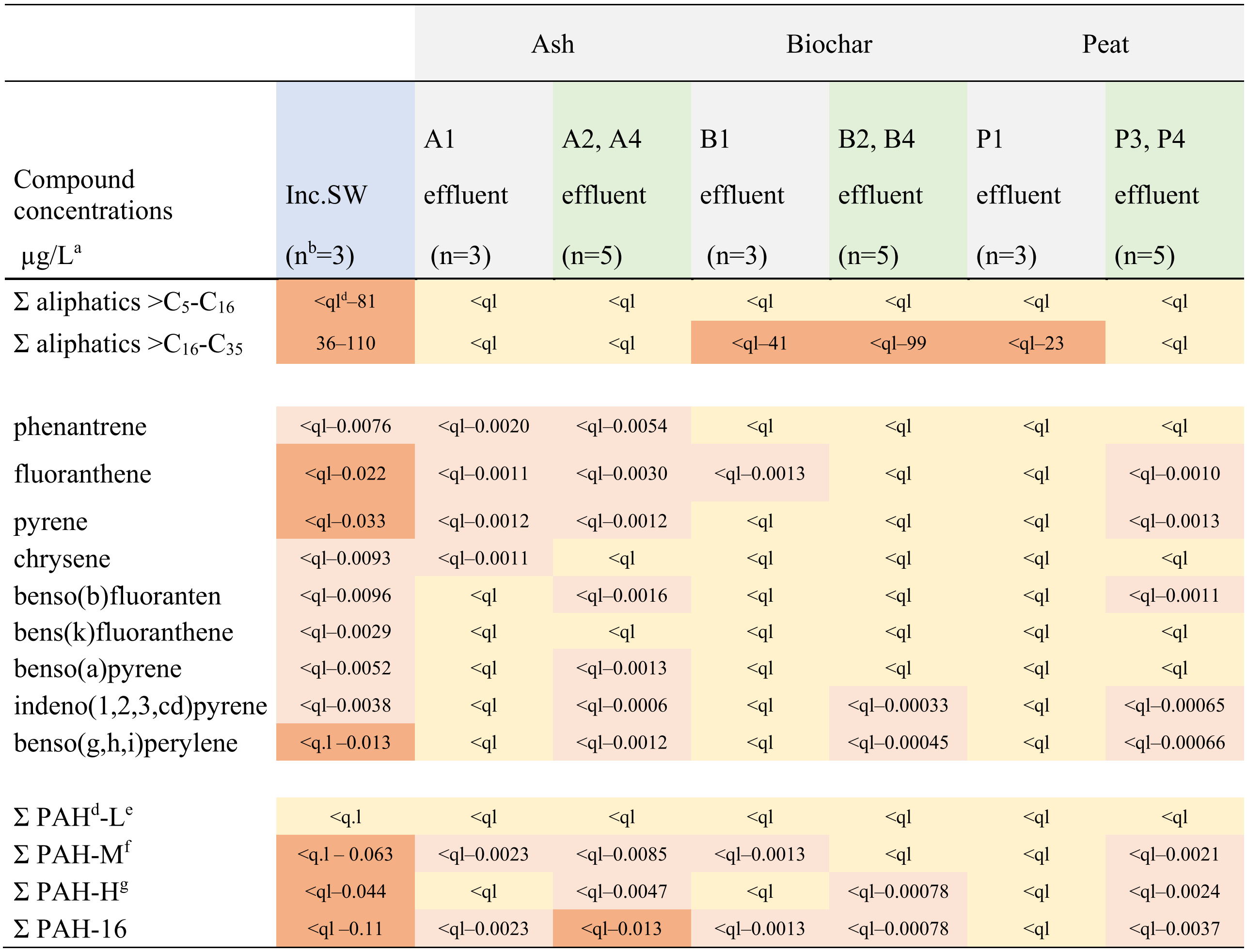
Table 3

Removal efficiencies (RE) of MP after 23 days and 53 days respectively from the start-up.

		Ash				**Biochar**				**Peat**				**Control**	
	Inc.SW	**A1**		**A4**		**B1**		**B2**		**P1**		**P4**		**C**	
Polymers µg/L[a]	influent n[b]= 7	effluent n = 5	RE 23;57 days %	effluent n = 5	RE 23;57 days %	effluent n = 5	RE 23;57 days %	effluent n = 5	RE 23;57 days %	effluent n = 5	RE 23;57 days %	effluent n = 4	RE 43;57 days %	effluent n = 5	RE 23;57 days %
PE	8.4–180[a] (74)	<ql[c]–> 750 (160)	-230;100	<q1–76 (28)	52; 83	<ql–180 (44)	9.0;100	<ql–44 (13)	73;100	<ql–>750 (15)	-220; 98	<ql–>750 (220)	-1200;100	<q.l–400 (110)	-130; 98
PI	<ql–150 (45)	<ql–1.3 (0.26)	99;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql–2.4 (0.6)	99;100	<ql	100;100
PB	<ql–88 (24)	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100
PP	<ql–30 (7.7)	<ql–33 (11)	88; −230	<ql–4.2 (1.9)	83;74	1.5.–14 (7.0)	59; −36	<ql–12 (5.9)	42; 32	<ql–18 (3.6)	44;100	<ql–43 (18)	-41; 23	<ql–17 (6.8)	42;45
PS	<ql–10 (2.2)	<ql	<ql; 100	<ql–7.9 (1.58)	-26*10^{4};100	<ql	<ql; 100	<ql–5 (1)	-17*10^{4};100	<ql–2.2 (0.44)	-7.3*10^{4};100	<ql–1.8 (0.45)	-49.9; 100	<ql–2.2 (0.44)	-7.3*10^{4};100
PVC	<ql–118 (25.66)	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100
PET	<ql–1.3 (0.19)	<ql	100;<ql	<ql	100;<ql	<ql	100;<ql	<ql	100;<ql	<ql	100;<ql	<ql	100;<ql	<ql	100;<ql
PC	<ql–1.2 (0.17)	<ql–12 (2.5)	<ql; 62.0*10^{4}	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql–4.6 (1.2)	<ql; −46*10^{4}	<ql–2.2 (0.32)	<ql; −80*10^{4}
PA6	<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql
PMMA	<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<q.l; 6.0 (1.5)	<ql; −20*10^{4}	<ql	<ql;<ql

[a] = minimum and maximum values; n[b] = number of samples analysed; <ql = below the limit of quantification; d removal efficiency = accumulated removal efficiency after 23 and 57 days, respectively. Following polymers are included in the Table: polyethylene (PE), polyisoprene (PI), polybutadiene (PB), polypropylene (PP), polystyrene (PS), polyvinylchloride (PVC), polyethylene terephthalate (PET), polycarbonate (PC), Polyamide 6 (PA6) and polymethyl acrylate (PMMA).

Table 4
Concentrations of specific organic pollutants quantified in inflow and effluents, in selected ash, biochar and peat bioretention filters during the start-up period, June – August 2022, at the Gårda pilot rain garden.



		Ash		Biochar		Peat	
Compound concentrations µg/L[a]	Inc.SW (n[b]=3)	A1 effluent (n=3)	A2, A4 effluent (n=5)	B1 effluent (n=3)	B2, B4 effluent (n=5)	P1 effluent (n=3)	P3, P4 effluent (n=5)
Σ aliphatics $>C_5$-C_{16}	<ql[d]–81	<ql	<ql	<ql	<ql	<ql	<ql
Σ aliphatics $>C_{16}$-C_{35}	36–110	<ql	<ql	<ql–41	<ql–99	<ql–23	<ql
phenantrene	<ql–0.0076	<ql–0.0020	<ql–0.0054	<ql	<ql	<ql	<ql
fluoranthene	<ql–0.022	<ql–0.0011	<ql–0.0030	<ql–0.0013	<ql	<ql	<ql–0.0010
pyrene	<ql–0.033	<ql–0.0012	<ql–0.0012	<ql	<ql	<ql	<ql–0.0013
chrysene	<ql–0.0093	<ql–0.0011	<ql	<ql	<ql	<ql	<ql
benso(b)fluoranten	<ql–0.0096	<ql	<ql–0.0016	<ql	<ql	<ql	<ql–0.0011
bens(k)fluoranthene	<ql–0.0029	<ql	<ql	<ql	<ql	<ql	<ql
benso(a)pyrene	<ql–0.0052	<ql	<ql–0.0013	<ql	<ql	<ql	<ql
indeno(1,2,3,cd)pyrene	<ql–0.0038	<ql	<ql–0.0006	<ql	<ql–0.00033	<ql	<ql–0.00065
benso(g,h,i)perylene	<q.l –0.013	<ql	<ql–0.0012	<ql	<ql–0.00045	<ql	<ql–0.00066
Σ PAH[d]-L[e]	<q.l	<ql	<ql	<ql	<ql	<ql	<ql
Σ PAH-M[f]	<q.l – 0.063	<ql–0.0023	<ql–0.0085	<ql–0.0013	<ql	<ql	<ql–0.0021
Σ PAH-H[g]	<ql–0.044	<ql	<ql–0.0047	<ql	<ql–0.00078	<ql	<ql–0.0024
Σ PAH-16	<ql –0.11	<ql–0.0023	<ql–0.013	<ql–0.0013	<ql–0.00078	<ql	<ql–0.0037

[a] minimum and maximum values; n[b] = number of samples analysed; [c]<ql = below the limit of quantification.; [d]polycyclic aromatic hydrocarbons; [e]Light; [f]Medium; [g]Heavy

Table 5
Removal efficiencies of specific organic pollutants quantified in influents and effluents, in selected ash, biochar, and peat bioretention filters during the start-up period at the Gårda pilot rain garden.

Removal efficiency of organic compounds (RE) %									
		Ash 1			Ash 4			Biochar 1	
Date	220615	220627	220818	220615	220627	220818	220615	220627	220818
aliphatics $>C_5$-C_8	<ql[a]	100[b]	<ql	<ql	100	<ql	<ql	100	<ql
aliphatics $>C_5$-C_{16}	<ql	100	<ql	<ql	100	<ql	<ql	100	<ql
aliphatics $>C_{16}$-C_{35}	100	100	100	100	100	100	56	100	100
sum PAH[c]-L[d]	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql
sum PAH-M[e]	100	98	100	100	98	100	-80	100	100
sum PAH-H[f]	-696	100	<ql	100	100	<ql	100	100	<ql
		Biochar 2			**Peat 1**			**Peat 4**	
Date	220615	220627	220818	220615	220627	220818	220615	220627	221011
aliphatics $>C_5$-C_8	<ql	100	<ql	<ql	100	<ql	<ql	100	<ql
aliphatics $>C_5$-C_{16}	<ql	100	<ql	<ql	100	<ql	<ql	100	<ql
aliphatics $>C_{16}$-C_{35}	100	100	100	100	100	36	100	100	100
sum PAH-L	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql
sum PAH-M	100	100	100	100	100	100	100	98	100
sum PAH-H	100	100	<ql	100	100	<ql	100	100	<ql

<ql[a] Below quantification limit for both influent and effluent; [b] Above quantification limit in influent, below quantification limit in effluent; [c]polycyclic aromatic hydrocarbons; [d]Ligh;, [e]Medium; [f]Heavy.

mixture of several traffic-related sources such as tyre wear, vehicle exhausts, brake linings, motor lubricant oils, and road surface wear ([74]; The concentrations of the total PAH-16 in the influent samples were up to 0.11 µg/L, which is low compared to the City of Gothenburg's local guideline value for benzo(a)pyrene of 0.27 µg/L. The concentrations of total PAH-16 were much lower in the effluents from the bioretention filters, but concentrations of up to 0.013 µg/L in the ash filter effluents were found in June, although the concentrations of PAH-16 in effluents from the same filter were ten times lower in August. This could possibly be explained by occurrence of PAH in the ash that were leached at the beginning of the project. In June, PAH-16 leached in low concentrations from the biochar and peat filters. In August, the only filter with detectable levels of PAH-16 in the effluent was the peat filter P1, and this may be explained by colloidal transport of the PAHs with DOC leaching from the peat filter [53,81]. Compared to toxic metals and nutrients, relatively little research has been conducted on the transport, sorption, phytoextraction, and degradation of OP in bioretention filters [68,122]. The studies conducted on bioretention filters have shown variable but relatively high removal efficiencies for OP. Organic pollutants is assumed to be removed by several different processes, including sedimentation on the surface of the filters due to sorption to larger particles, volatilisation from the surface, sorption in the solid materials, biodegradation in the filters, and/or phytoremediation by the plants; and of these processes, sorption has been identified as the most important [22, 25]. The high porosity and large surface area of biochar and MIBA would be beneficial for removal of OP, due to high the possibility of sorption while limiting the opportunities of colloidal transport with DOC [55]. *Sphagnum* peat is otherwise an effective sorbent for OP due to the high content of organic matter [50,73]. The peat used in this pilot facility had a much lower active surface compared to the biochar, Table 5.

3.4. Metals

All the results from the chemical analyses of metals, including the total concentration of selected major elements (Na, K, Ca, Fe, Mg, Mn, Al) and total concentrations of trace metals (As, Ba, Pb, Cd, Co, Cu, Cr, Hg, Mo, Ni, V, Zn), are presented in the Supporting material, Table S1. The total concentrations of several metal ions e.g., Co, Cr, Cu, Ni, Pb, and Zn, were compared to the City of Gothenburg's guideline values regarding release of polluted water to stormwater networks and recipients. Overall, metal concentrations in the effluents decreased with time, showing a stabilisation phase for the filters, or were continuously low (Cr and Hg). Initially, some metals were released. This release is believed to come from particles being leached from the materials or released as the materials settled, i.e., As and Cd from the ash filters, and Pb from the biochar filters. Especially for the biochar filters, which showed a large negative RE for certain metals at the beginning, the initial high turbidity and TSS show that many particles were flushed out as the filter materials were settling. However, the release from the filter materials decreased rapidly. After less than 20 days, As, Cd, and Pb were all below the stormwater guidelines for the City of Gothenburg. Average concentrations of several water quality parameters in stormwater, including Cu, Zn, TSS, and TN, have been shown to increase post-construction of bioretention filters [2]. These spikes in concentrations were contributed to the particles from the filter material being flushed out in the beginning, as the concentrations decreased after a few sampling occasions. In a study, two stages of stabilisation were observed to take place during drying and wetting cycles in a bioretention column [113]. One stabilisation phase was observed as filters were repeatedly wetted over time and the filter material settled. Another type of stabilisation occurred at the beginning of each wetting event, when concentrations from retained water in the filter was mixed with the new influent. This phenomenon further complicates the mechanisms behind the release and removal of several pollutants, in particular for pollutants being transformed from particulate to dissolved form between wetting and drying cycles. The speciation of Cu and Zn changed as stormwater passed through the bioretention filters [60], with an increase in dissolved and colloidal forms in the effluent compared to the influent.

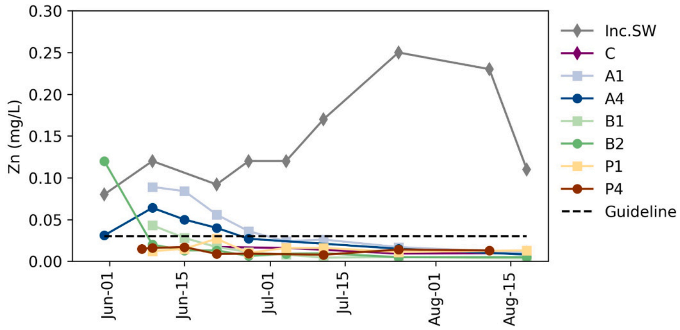


Fig. 7. Concentrations of Zn in influents and effluents of filters C, A1, A4, B1, B2, P1, and P4. In addition, the guideline value for the City of Gothenburg is shown.

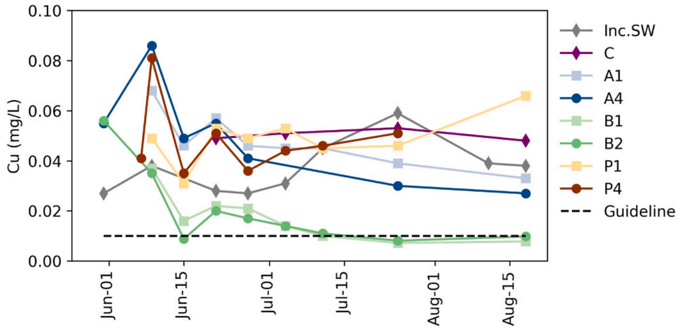


Fig. 8. Concentrations of Cu in influents and effluents of filters C, A1, A4, B1, B2, P1, and P4. In addition, the guideline value for the City of Gothenburg is shown.

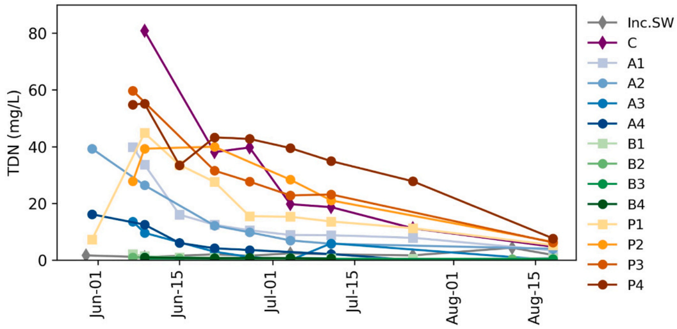


Fig. 9. Concentrations of total dissolved nitrogen (TDN) in the original stormwater used for irrigation, in comparison with concentrations after treatment, in the effluents from control, peat, ash, and biochar filters after 10 weeks of operation.

The results showed that the measured concentrations of Zn and Cu in the influent exceeded the City of Gothenburg's guideline values, and the highest concentration was found for Zn (0.25 mg/L). After 55 days the reduction of Zn was >90% in all filters, and after 80 days the reduction was close to 100%, irrespective of the concentrations in the incoming water (Fig. 7). When the rain garden had been active for 80 days, only Cu exceeded the guideline in the effluent waters (Fig. 8). However, the biochar filters, B1 and B2, reduced the Cu concentration to below the guideline, and a similar trend with declining concentrations in the effluents has been observed for the other filters. The variation in metal removal performance between the different filter materials could be due to several factors. Studies of filter materials chosen for their sorption capacities for dissolved metals have shown that low organic material content, higher pH, and large specific surface areas were beneficial for metal removal [112]. This is in line with our results, given that the biochar filters showed the overall best results regarding metal removal, high pH in the effluents, low leaching of organic matter and the large surface area of biochar. Cu and DOC in the effluents were found to be correlated, r (31 measurements) = 0.81, $p<0.01$, which is supported by several other studies [11,52,55,76]. Peat filters, with high organic content, had a lower removal capacity for copper, which may be explained by colloidal transport with DOC [55]. The addition of compost in the ash filters might explain the lower removal rate of copper depending on colloidal transport with DOC released from the added organic content. It should be noted that the control filter (C), which also contained compost, had the second highest release of Cu after 80 days, even higher than the influent, which shows that materials commonly used in rain gardens can also leach metals when subjected to stormwater. The sandy loam used in this study has been shown to leach Cu, Ni, Pb, and Zn in low concentrations [112]. It has been demonstrated, both in laboratory studies and in field studies, that Cu is difficult to remove using bioretention filters compared to other metals, particularly with filters containing higher amounts of added organic material [87]. The overall results show that bioretention filters are efficient for the reduction of several metals, which is in line with many other studies e.g., ([20, 49,79,88,111,119]).

3.5. Nutrients

Nutrients in the form of TDN were initially present in high concentrations in the effluents from all filters, however concentrations decreased with time (Fig. 9). Initial release of nutrients from materials containing soil is expected due to leaching [34]. All phosphate concentrations were below QL in all influent and effluent samples.

The control filter (filter C) showed the highest initial release of TDN (effluent concentration was about 77 times higher than influent concentration), which was comparable to the peat filters. Although the effluent concentrations decreased over time, both the control and peat filters still released TDN at the end of the study period, with removal efficiencies ranging from − 159% to − 321%. When comparing different bioretention design parameters, the addition of peat soil in systems for treatment of semi-synthetic stormwater led to a negative average removal rate for N (−178%) [121]. When stormwater, which is normally aerobic when entering a treatment system, percolates through bioretention filters, a change in conditions from aerobic to anaerobic can occur, which can affect certain filter materials [15]. Among the materials tested, peat and municipal compost made of leaves were shown to leach nutrients when conditions became anaerobic. It was also shown that previously sorbed ammonium was released from peat moss when the change to anaerobic conditions occurred. Several other studies show the leaching of nutrients from compost and compost-amended materials in bioretention systems [40,76], as well as the initial flush of nutrients from these types of materials [11,84].

Of the biochar filters, only filter B1 showed a negative removal rate for TDN, and only at the first measurement. During the latter part of the start-up period, removal efficiencies for TDN in the biochar filters were 69–80%. In previous laboratory studies of biochar columns without a submerged zone (SZ) for denitrification, N removal efficiencies of 45 ± 14% and 86%, respectively, have been reported [102,100]. Water retention in the filters is a key parameter for nitrate removal [117], due to increased contact time, reduced rate of peak outflow, and increased capture of first flush effluent. Another laboratory study of woodchip bioretention filters with SZs showed that biochar addition increased the denitrification rate due to increased pore water retention and decreased dissolved oxygen, thus creating more favourable conditions for denitrifying bacteria [4]. Biochar could also enhance denitrification by serving as electron donor [107].

The ash filters initially released high levels of TDN equivalent, at around 8–30 times the influent concentrations. However, the performance improved over time and at the last measurement in August, the RE varied from − 12.5% to 100%. Previous studies of bioretention columns containing ash have only reported on the performance of mature filters, not on the start-up period. Removal efficiencies of 82–97% and 58–70% were reported for phosphorus and total nitrogen, respectively [126].

Ammonium ions were present in the influent on several occasions, once in the early effluent of an ash filter, and several times in the effluents of all peat filters. For ammonium concentrations, see Supporting material, Table S2. The results from this study at the Gårda pilot facility seem to reflect the higher amounts of released nutrients from materials containing more organic matter than in other studies, but also the decline in nutrient release after the initial flush. The effluents showed redox potentials of 37–210 mV for the biochar filters, 13–210 mV for the ash filters, 82–230 mV for the peat filters, and (77–210 mV) for the control filter. Redox potentials are important for several transformation and removal mechanisms, e.g., transformation and removal of certain metals and nutrients [65]. The redox potential never exceeded 300 mV for any filter, which is important for denitrification processes [58]. Even when the fluctuations in redox potential are considered, there were favourable redox conditions for denitrification in all filter types during the start-up period, given that NO_3^- and carbon were available for denitrifying bacteria.

3.6. Vegetation

All plants of the chosen species, *Hippophae rhamnoides*, *Festuca rubra*, *Juncus effusus, Armeria maritima* grew well under the start-up period without first being established under less stressful conditions, and despite irrigation with polluted stormwater (Fig. 10). As mentioned before, there were no significant differences in pollutant removal between vegetated filters and un-vegetated filters during the start-up period, which can be explained by the fact that the plants were not

Fig. 10. Plant establishment during start-up period.

Table 6
Hydraulic conductivity in saturated soil of the different bioretention filter at the Gårda rain garden pilot.

		Control	Ash				Biochar				Peat			
Filter		C	A1[a]	A2	A3	A4	B1[a]	B2	B3	B4	P1[a]	P2	P3	P4
Date		220826	220923	221003	220926	220926	220926	220923	221003	220926	220826	221003	220926	220926
Ksat	cm/h	387	206	132	154	212	144	642	554	469	130	105	99	102

[a] filter without plants.

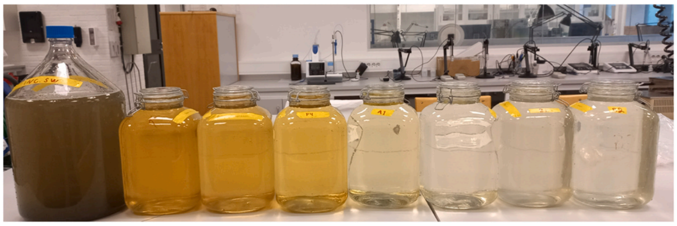


Fig. 11. Reduction in visible particles 2022-07-25. From the left; Influent, Control, P1, P4, A1, A4, B1, B2.

given enough time to establish their root systems sufficiently to be able to function as phytoremediators.

3.7. *Saturated hydraulic conductivity*

The saturated hydraulic conductivity (K_{sat}) was measured after the start-up period had ended (Table 6). The peat filters had the lowest K_{sat} value, in the range of 99–130 cm/h, followed by the ash filters which showed a greater span of 132–212 cm/h. This means that the water had a longer contact time for both sorption and desorption processes with the material in the peat filter compared to the ash filters. This is explained by the fact that the peat material had smaller particles than the ash and biochar materials. The measured K_{sat} in the control filter, 387 cm/h, was significantly higher than for the peat and ash filters. The biochar filters showed not only the highest overall K_{sat}, and the shortest contact time between water and material for three of the filters (B2: 642 cm/h, B3: 554 cm/h, B4: 469 cm/h), but also the greatest variation between filters, as the K_{sat} for filter B1 was much lower than the rest at 144 cm/h. The result is logical as the biochar also had more of the larger particles than the other sorption materials. It is not known why filter B1 had such low K_{sat} compared to the other biochar filters. Filter B1 is the one without vegetation, but this relationship was not observed for the other filter types. It should be noted that the hydraulic measurements were only performed on one occasion. The reason for not measuring K_{sat} for all filters on the same day, which would have been preferable, was that there was not enough stormwater available to saturate all filters simultaneously. This may have affected the results, as for example the vegetative and/or mycorrhizal development and settling of materials of the filters may have varied due to the gap between measurements. However, there were general similarities between filters with the same composition even though they were measured on separate occasions.

3.8. *General parameters*

A reduction in visible particles and coloured materials was clearly noticeable after treatment with all the bioretention filters (Fig. 11). Filters with biochar showed the highest removal of visible particles during the start-up period, except for the first few weeks when the reduction of visible particles increased with time in all filters. Until the end of the start-up period, no significant differences in visible particles or colours were noted between filters with and without plants for any of the filter types.

During the first few weeks, the pH of both the influent and effluents showed great variations. For pH and other field measurements, see Supporting material, Table S3. Effluents from the biochar filters had the highest pH overall (average 8.1) throughout the campaign; this was also noted in the PCA analysis (Fig. 5). The reason for this is likely the relatively high pH and high buffer capacity of biochar materials [28] compared to the other filter materials. The biochar filters showed lower effluent pH at the end of the study than at the start. This is assumed to be caused by leaching of alkaline materials and/or consumption of the materials' buffer capacity. The low initial pH for the ash filters was unexpected and is believed to be due to leaching of organic acids and carbonic acid from the compost material mixed with the ash and soil, and from the thin layer of peat below the main ash sorption layer. However, the main layer in the ash filters only contained 15% compost, and the layer of peat was relatively thin (50 mm) compared to the main sorption layer (550 mm). Effluents from the peat filters showed a steady decrease in pH over time, ending up with the lowest pH during this study together with the control filter. Peat has a naturally low pH, around 4.5, and in addition to a high organic content, also contains high levels of fulvic and humic acids, which are highly soluble in water [52]. The control filter had the greatest amount of sandy loam with compost, which was mixed with additional compost, explaining the low pH due to high organic matter content.

Filter B1, B2, B3, and C had high turbidity and TSS/VSS values compared to the other filters. Particularly high turbidity (FNU) was found in filter B1 (603 FNU) and B2 (403 FNU) at the beginning of the study. However, the initial turbidity for these filters dropped rapidly and was soon below the influent levels, which is in line with the visual impressions. Since the middle of July, all effluents have shown lower turbidity than the influents. As mentioned in Section 3.4 this phenomenon has been observed in other studies as well [2,95] and is due to a flush of particles as the materials in the filters settle. It is unclear why filter B4 did not express this phenomenon as strongly as the other biochar filters.

3.9. *Limitations*

Access to stormwater was a limitation during the start-up period. Dry weather led to occasions when irrigations volumes became low. However, the low amount of precipitation and available stormwater reflected the condition that would have affected a hypothetical newly constructed rain garden in the area, and reflecting field conditions was part of the purpose of the pilot. Another limitation that distinguishes the pilot facility in Gårda from an established bioretention facility is that the columns have a more closed environment, which can affect for example the exchange of microorganism, e.g., nematodes and earthworms, and fungi with the exterior environment. The lack of ability to efficiently analyse MP particles smaller than 10 μm is of concern for the MP research community, as the total masses calculated are underestimated. There are also no available methods for quantifying low concentrations of phthalates and PAH in water samples.

4. Conclusions

A stabilisation phase during which the filter materials settled occurred after the start-up of the bioretention filters, causing an initial flush of particles from all filters. This was especially noticeable for the filters that contained biochar, which showed high initial leakage of particles.

In the vegetated filters, all plants (thrift, sea buckthorn, common rush, and red fescue) survived the start-up period despite irrigation by highly polluted road runoff, and all plants grew well during this period. There were no significant differences between vegetated and non-vegetated filters regarding pollutant removal or hydraulic conductivity during the start-up period.

Microplastic particles >10μm were efficiently removed from the stormwater by all bioretention filters during the start-up period, however PE and PP were found in the effluents from all filters, especially during the start-up phase. This may be explained by the release of these polymers from the materials used when constructing the filters, as the effluent concentrations decreased considerably over time.

The results showed that aliphatic hydrocarbons and PAH were efficiently removed by all bioretention filters during the start-up period. At the end of the start-up period the total concentrations of all measured metals in the effluents were below the City of Gothenburg's guideline values for release of stormwater into recipients, except for Cu which were only below the guideline values in the biochar effluents. However, the effluent concentrations of Cu were decreasing with time.

Nutrients leached from all filter types at the beginning of the start-up period, however concentrations declined over time. Biochar removed nutrients from the incoming stormwater after a few weeks. Based on the results of this study, it can be concluded that all the bioretention filters with sorption materials added, both vegetative and non-vegetative, had a very good removal capacity for MP and all other pollutants measured, and that these types of bioretention filters may be used for full-scale applications. However, before full-scale application, studies to ensure the filters are still functional after a longer period in operation need to be performed.

Environmental implication

Stormwater from heavily trafficked areas contains a cocktail of pollutants originating from vehicle wear, tyre wear, road wear, and exhausts. The cocktail includes microplastics, metals, nutrients, and organic pollutants, which degrade the environment. Tyre wear also contains hazardous substances and may be a carrier of other pollutants, and there is an urgent need to find sustainable treatment solutions. This work investigates the fate of tyre wear particles and other stormwater pollutants during the startup of bioretention filters. Innovative sorbent materials such as biochar, *Sphagnum* peat and waste-to-energy bottom ash, which contains hazardous substances, are tested in the bioretention filters.

Future research

Future research should focus on investigating how the removal processes in the filters work more in-depth and investigate where in the different parts of the filters and plants these processes take place. It is also of great interest to investigate how extreme weather events affect different processes in bioretention filters. Changing weather patterns might, for example, result in prolonged droughts, or extreme flows during unfavourable conditions. This is important knowledge if bioretention filters are to be a part of a more resilient future infrastructure. There is also a need to investigate how bioretention filters remove and release MP smaller than 10 μm, and especially with a focus on tyre and road wear particles. There is a risk that the emission of MPs to stormwater is underestimated if small particles are not included which in turn affects mass balance calculations, risk assessment, and selection of treatment techniques. Regarding the pollutants trapped in bioretention filters future studies should be conducted undertaken to understand how these pollutants could be separated, and how metals can be recovered from the plants and soil-bed materials once the filters are used up.

CRediT authorship contribution statement

Glenn Johansson: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization, **Karin Karlfeldt Fedje**: Writing – review & editing, Writing – original draft, Visualization, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization, **Oskar Modin**: Writing – review & editing, Writing – original draft, Visualization, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization, **Marie Haeger-Eugensson**: Writing – review & editing, **Wolfgang Uhl**: Writing – review & editing, Funding acquisition, Conceptualization, **Yvonne Andersson-Sköld**: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization, **Ann-Margret Strömvall**: Writing – review & editing, Writing – original draft, Visualization, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.133532.

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