IMplementing MEasuRes for Sustainable Estuaries (IMMERSE)

WP 4. Measures: Assessments, tests, and pilots

Report for WP 4.7 Innovative Rain Gardens for Sustainable and Effective Treatment of Urban Runoff Polluted with Microplastics and other Pollutants

Authors: Glenn Johansson, Karin Karlfeldt Fedje, Yvonne Andersson-Sköld, Oskar Modin and Ann-Margret Strömvall

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1. Introduction

The aim and scientific challenge of this project were to design, construct and explore an innovative and sustainable rain garden with bioretention filters where microplastics and other pollutants from urban runoff are retained, degraded, or recovered. The overarching goal was to significantly reduce the transport of urban pollution to receiving waters and to contribute to green infrastructure and a circular economy in society.

The objectives of the project were to:

- Design and construct an innovative and sustainable pilot-scale rain garden for effective removal of microplastics and other pollutants and evaluate sorption materials such as peat, biochar, and ash as bed material, with and without plants.
- In the pilot, an in-depth study of the processes in the rain beds for removal, distribution, degradation, potential uptake in plants, and possible leaching of microplastics and other pollutants.

Here we present the results from the first part of the project, which includes the designs and start of the rain garden pilots and the initial results of the removal effectiveness and hydrologic performance. The calculations of removal efficiencies are based on sampling and analyses of metals, nutrients, organic pollutants, tyre wear and other microplastics in incoming and outgoing waters from the raingarden pilots. The report provides information on substrata and planting strategies identified to be the most optimum to reduce microplastics, metals, nutrients, and organic pollutants also applied in the pilot rain gardens. It also includes a quantification of the first annual reduction of microplastics, metals and organic pollutants in the leachates in relation to the incoming polluted water. The project will continue as a Ph.D. project until 2026 and will further include studies of the pollutant's distribution at different depths in soil beds, uptake in plants, the impact of mycorrhiza, and investigations of how the rain beds work under changing climate conditions. In the final phase of the project, the identification of sustainable, efficient, and innovative methods for the recycling or degradation of potentially residual microplastics, while recovering metals from plants, roots, and soil sorption materials used in the rain beds will also be carried out.

2. Background

Urbanization has contributed to a degradation in the quality of surface waters, where inadequate stormwater management and pollution control have played an important role. Stormwater management in urban areas has moved from quantity control and combined sewers to current strategies for quantity and quality source control, with an emphasis on the multiple benefits provided by blue-green infrastructure (Eckart, 2017). Though most stormwater discharges are still transported untreated to receiving waters, this has led to expanded opportunities, and that various technologies have been developed and used to treat stormwater locally. However, research supporting new development of innovative and more effective technologies and management strategies is urgently needed to meet the demands on the sustainable development of the urban environments. A cocktail of emerging environmental pollutants such as microplastics, metals, and organic pollutants is released into urban environments and emissions are particularly high in highly trafficked areas (Markiewicz et al., 2017; Polukarova et al., 2020; Järlskog et al., 2022b; Andersson-Sköld et al., 2020). The largest proportion of pollution is transported from roads by runoff and further by stormwater to receiving water courses. However, only a few percent of the stormwater generated in urban environments is treated, and the treatment systems available today are not designed to effectively remove the pollutants. In recent studies, we showed surprisingly high concentrations of both microplastics from tyre and road wear as well as other pollutants both on the street and in the nearby stormwater (Polukarova et al., 2020; Järlskog et al, 2020; 2021; 2022a; 2022b). Tyre and road wear microplastic particles in urban runoff is estimated to be the largest emission source of microplastics in Sweden (Andersson-Sköld 2020; Magnusson et al., 2016) and account for the highest proportion of microplastic loads into European rivers (Siegfried et al., 2017). So far, research has focused on clarifying the consequences and fate of microplastics in the environment; however, technological solutions that solve the problem are lacking. Here we have designed and started a rain garden as a pilot in the field. We have also started to investigate the removal efficiency of metals, organic pollutants, and tyre wear and other microplastics as well as the hydraulic performance for varying planting substrate and species assemblage. To our knowledge this is the first study of a cocktail of pollutants in pilot-scale rain gardens using municipal solid waste incineration ash, peat, and biochar as sorption material in mixtures in the soil-bed, and with and without plants.

Bioretention systems, also referred to as rain gardens or bio-infiltration, are one of the most versatile and widely used for the sustainable treatment of stormwater in many parts of the world Davis et al., 2009. The solutions will contribute to the development of green infrastructure in urban areas (Tzoulas et al., 2007). More recently, sorption filters made of bio-based materials have been installed in gully pots and have also been tested as a post-sedimentation treatment for organic pollutants (Markiewicz et al., 2020). If these techniques are introduced on a larger scale, large amounts of soil-bed and filter materials polluted with microplastics, metals, and organic pollutants will need to be managed in the future. In the design of a rain garden, it is of high importance to take into consideration soil bed material and the plants and all their interaction processes

(Skorobogatov et al., 2020). Very little is known about the fate of organic pollutants and microplastics in stormwater sewers and other management systems, and data on the effectiveness of stormwater treatment facilities to retain and degrade microplastics. Particles transported with runoff from road surfaces may accumulate in trapped sediments or in filter media, e.g., rain garden soil, or may be transported through the stormwater system to receiving waters. Despite many studies performed, research on processes for removal, distribution, potential degradation and/or up-take by plants or leaching of the pollutants in the various soil beds are missing for organic pollutants and tyre wear and other microplastics. Rain gardens may capture and efficiently retain nutrients and toxic metals (Li et al., 2009), but there are very few studies on the removal of organic pollutants (Diblasiet al., 2009) and there is only one recently published initial study on microplastics in bioretention filter (particles >100 µm) (Gilbreath et al., 2019). There are studies on removal efficiencies, but these lack in-depth investigations of the different removal processes, especially of cocktails of pollutants including microplastics.

In this report, we present results on removal efficiencies and leaching of the pollutants in the various soil beds, while in forthcoming parts of the project, the results will also provide research results on distribution, potential degradation, and/or uptake (phytoextraction) by plants. Such information is not only relevant to better understand the fate of pollutants and how to design rain gardens but also contributes to recycling opportunities, for example, if watering the phytoextraction plants with e.g., contaminated stormwater the metals, organic pollutants and microplastics present might be extracted by the plants, while organic pollutants and potentially also microplastics can be degraded by the plants or the solid material. Phytoextraction is used in a full scale to remediate contaminated soil, while the knowledge of cultivation in other materials like metal-rich bottom ash is very limited. By cultivating in bottom ash recovery of metals, that are not recovered today, is enhanced.

3. Materials and methods

3.1 Construction of the pilot rain garden at Gårda

Rain garden beds were established in columns made of polyethylene (~980 L). In the bottom of the columns is a drainage layer of fine gravel followed by a layer of course sand, a sorption material layer, a layer of sandy loam mixed with pumice stones and sorption material (in the ash filter mixed with compost), and finally a layer of sandy loam soil mixed with pumice stones is placed on top, see Figure 1. The sorption material is either peat, biochar, or a combination of separate layers with peat, biochar, and municipal solid waste incineration bottom ash. There are few studies on soil bed improvements in biofilters through the addition of various sorption materials, and only biochar and coal fly ash have been studied for more efficient removal of phosphorus (Tian et al., 2014) and biochar for organic pollutants (Zhang et al., 2008). However, it is important to note that there is a lack of studies where controls have been used without plants (Dagenais et al., 2018).

Each column type has three replicates with plants and one column without plants. There is also a control column without a sorption layer. Four plant species are used: Thrift *(Armeria maritima),* Sea buckthorn *(Hippophae rhamnoides)*, Common rush *(Juncus effusus),* and Red fescue *(Festuca rubra),* see Figure 2. These plants were selected because they are known to be able to stand in water for shorter periods of time, can withstand droughts, and because of the ability to absorb metals, and aid in the degradation of PAH (Borowik et al. 2019). Sea buckthorn is a hardy deciduous shrub, native from northwestern Europe to eastern Asia, capable of growing in arid regions (Kalia et al., 2011). Sea buckthorn can tolerate sub-alpine environments and moderate droughts. In Scandinavia, it is mostly a coastal plant and is considered to be salt tolerant. These attributes in combination with the ability to efficiently fixate nitrogen have made sea-buckthorn a prospect for soil improvement and land reclamation. Studies have shown that the uptake of toxic metals occurs in sea-buckthorn (Petrescu-Mag et al., 2021; Emre 2023). Red fescue is a perennial turf-forming grass that is native in Scandinavia and widespread across the Northern hemisphere (Dirihan et al., 2016). Red fescue tolerates many different climates and habitats and is known for its shade tolerance (Petrella and Watkins, 2020). This perennial grass has additional qualities, such as tolerance for toxic metals (Ma et al., 2003), tolerance to soils polluted with petroleum products (Palmroth et al., 2002), and an affinity for metals uptake (Prabha, 2009; Gajic et al., 2016) that makes it a relevant candidate for phytoremediation in environments burdened with traffic-related pollutants. Common rush is a perennial herbaceous flowering plant, and it is considered widely adaptable, which shows in its broad, nearly global distribution (Hurd et al. 1994). Common rush prefers wet habitats including marshes, wetlands, swamps, wet pastures, and ditches, but it can be found anywhere with moist soil, and it tolerates short periods of drought when established. Studies have shown that treatments with common rush can have a positive effect on the removal of nutrients and potentially toxic metals from polluted waters (Hernandez-Perez et al 2021; Deng et al., 2004;

Control

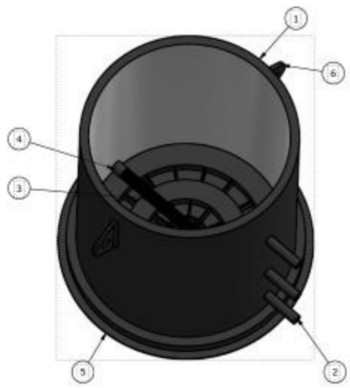
1. 0 – 15 cm Sandyloam with pumicstone (Top layer)
2. ***15 – 80cm Sandy loam mixed with 15% compost***
3. 80 – 90 cm Coarse sand
4. 90 – 110 cm Finegravel

Peat and Bio char

2a. Soil mixed with peat/bio char (40%)
2b. Peat/bio char

Ash

2a. Soil mixed with ash (50%) and compost (15%)
2b. Peat
2c. Bio char



Height: 125 cm Diameter: 100 cm

Figure 1. Soil-bed materials in the different bioretention filter columns in the pilot rain garden at Gårda.

Figure 2. The plant species used in the pilot rain garden at Gårda: Thrift *(Armeria maritima)*, Sea buckthorn *(Hippophae rhamnoides)*, Common rush *(Juncus effusus)*, and Red fescue *(Festuca rubra)*

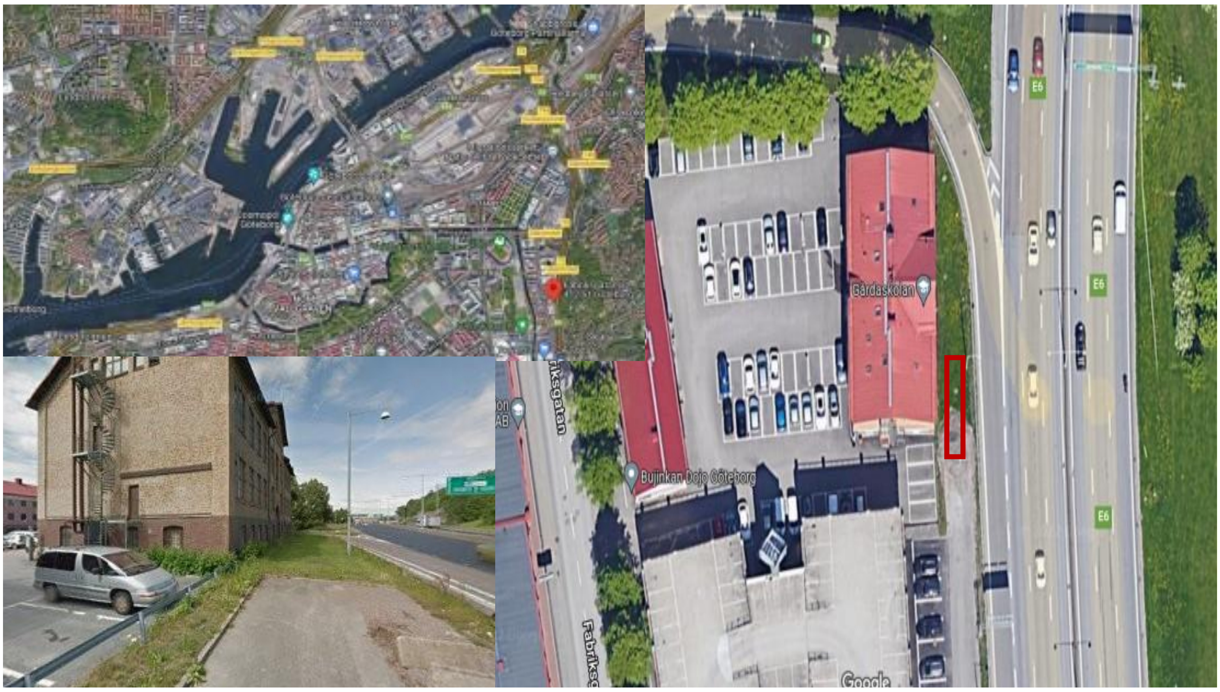


Figure 3 . The location for the pilot rain garden at Gårda, in the catchment area of the Göta River close to Highway E6 in the central part of Gothenburg city.

Syranidou et al., 2017; Beral et al., 2023). Thrift thrives in coastal areas and has a circumpolar distribution Wierzbicka et al. 2023). It is highly salt tolerant and tolerates droughts well when established. This perennial flowering plant is a known hyperaccumulator of toxic metals (Lange et al., 2020; Purmale et al., 2020; Dhamani-Muller et al., 2000).

The rain garden beds are placed next to highway E6 in Gothenburg in the Gårda area where impervious surfaces (94%) consist of roads, footpaths, and parking lots (Markiewicz et al., 2017; Björklund et al., 2009), see Figure 3. The beds are watered with highly polluted water from the inlet chamber at the sedimentation facility for stormwater mainly from the highway runoff.

Analyses of the water samples included analysis of microplastics, total metals, aliphatic hydrocarbons, polycyclic aromatic hydrocarbons (PAH), phthalates, nutrients, and general water quality parameters. The selection of the organic pollutants was based on the list of priority pollutants in road runoff (Markiewicz et al., 2017) and previous studies of urban material (Björklund et al., 2009); Polukarova et al., 2020; Järlskog et al., 2021). Metals were analysed by ICP/MS, and eight different microplastic polymers were analyzed by pyr-GC/MS, PAH and aliphatic hydrocarbons by GC/MS and sent to specialised laboratories for chemical analysis. At Chalmers, total organic carbon/nitrogen (TOC/N) and dissolved organic carbon (DOC) were also analyzed with a TOC analyzer. General parameters such as pH, conductivity, turbidity, oxygen, and redox were measured electrochemically with a multimeter. All these results will be presented in coming papers by Johansson et al., 2023a, b).

Figure 4. Measurement Campaign 1, the bioretention filters were operated with weekly irrigation, sampling, and chemical analysis from June 2022 to mid-August 2022.

3.2 Operation of the pilot rain garden

The pilot experiment has so far been run for a year and the performance was examined in two measurement campaigns. The concentrations and composition of tyre wear and other microplastics, organic pollutants, metals, dissolved organic carbon, and nutrients in influent and effluent water from the rain beds were measured, and the retention/sorption was calculated. The efficiency of the pilot rain gardens for removal of the pollutants was assessed. The hydraulic conditions and changes of water under the experiment were also examined. The growing rates of plants were determined through ocular inspection, plant height and weight measures. All these results will be presented in coming papers by Johansson et al., 2023a, b.

During Campaign 1 (Figure 4), the bioretention filters were subjected to irrigation 16 times, with volumes between 20–70L per stormwater/filter at each irrigation, depending on the availability of stormwater. Sampling and chemical analysis were carried out from the very end of May 2022 to mid-August 2022. This first measurement campaign aimed to deepen the knowledge of how the removal processes worked during the first three months of the start-up of the rain gardens. We hypothesized that the plant establishment and sorbent removal processes would need a relatively long time to reach equilibrium and an effective removal percentage. Nutrients and pollutants were expected to initially leach from the beds. During the autumn, September 2022 – December 2022,

Figure 5. Measurement Campaign 2. The bioretention filters were operated with irrigation, sampling, and chemical analysis from mid-January 2023 to March 2023 to study the removal processes when the plants are dormant.

only two occasions with sampling and chemical analyses were carried out, but the irrigation continued. During these periods, measurement data was evaluated, tables and figures were constructed, literature research was carried out and a draft of the first scientific article was written (Johansson et al., 2023a). *Measurement Campaign 2* (Figure 5), January – March 2023, was planned more in detail. In this second campaign, the aim was to understand how the bioretention filter works during winter when the plants are dormant but also to understand more deeply where in the bed the pollutants are removed during winter conditions. All the results from the two campaigns will be presented in coming papers by Johansson et al., 2023a,b.

4. Results and discussion

4.1 General parameters and nutrients

In Figure 6, the reduction of visible particles and colored material could be clearly seen after treatment in the bioretention filters. The visible particles in the stormwater were efficiently reduced after passing through the filters. The biochar filter showed the highest efficiency and the reduction of visible particles increased with time in all filters. Until now, no significant differences were seen between the filters with and without plants for any of the filter types and parameters.

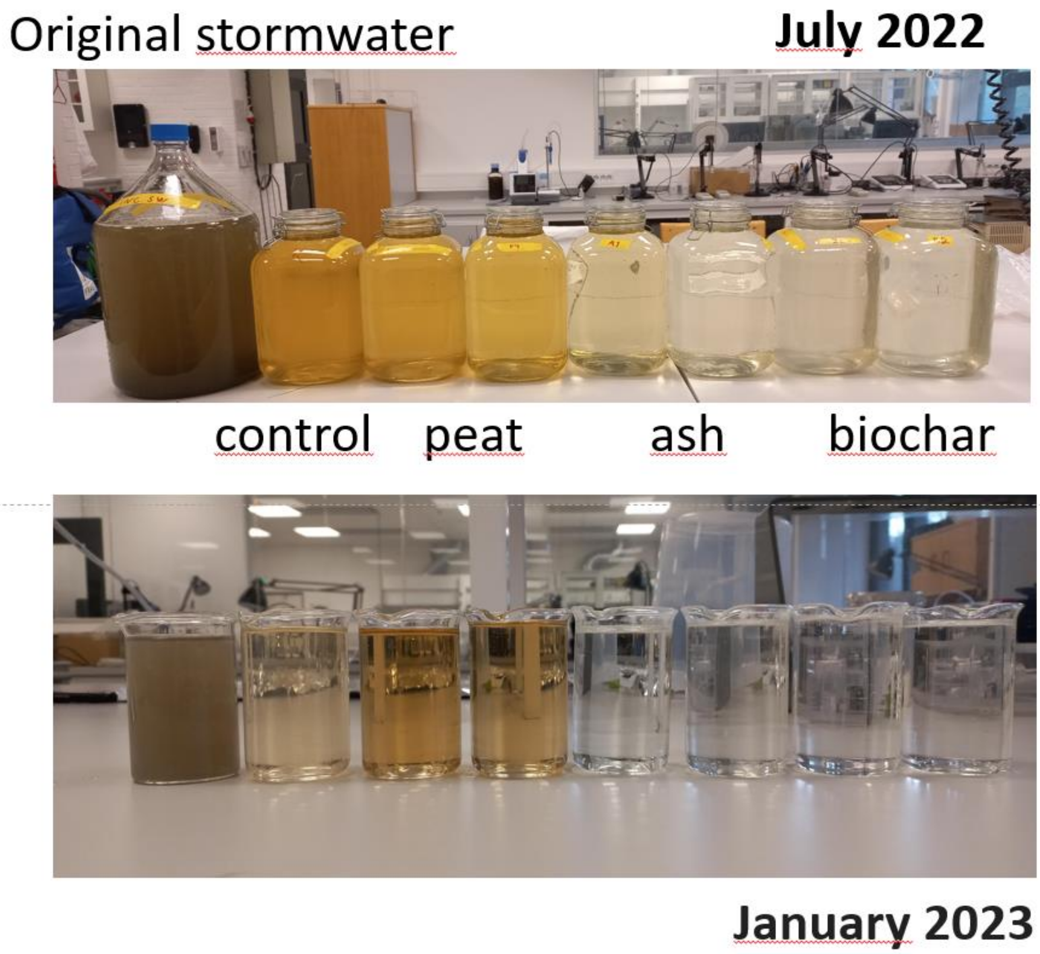


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Figure 6. The reduction of visible particles and colored material in original stormwater after treatment and in effluents from control, peat, ash, and biochar filters after 2 and 8 months of operation.

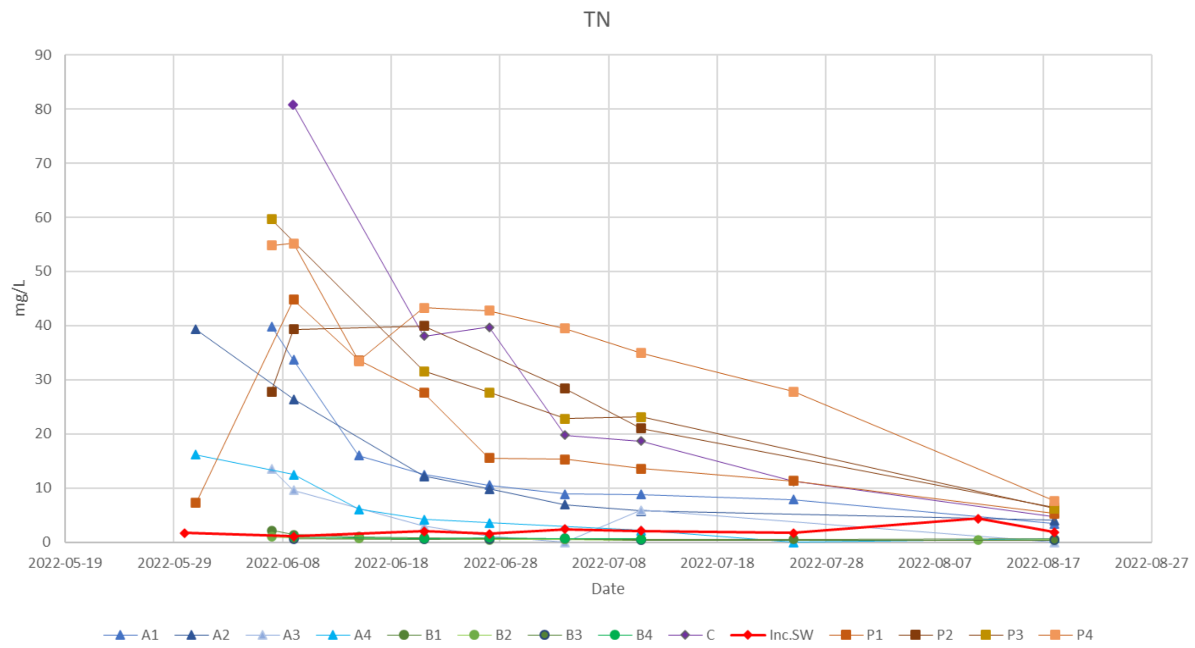

Figure 7. The concentrations of total nitrogen (TN) in original stormwater in comparison with the concentrations after treatment in effluents from control, peat, ash, and biochar filters after 10 weeks of operation.

As expected, nutrients in the form of total nitrogen (N-tot) were initially high in the effluent waters, as nitrogen was released from the filter materials themself, but the concentrations decreased with time in all filters, see Figure 7. The release of nutrients from materials containing soil is to be expected (Hatt et al. 2008). When trying to simulate real conditions, a so-called stabilization phase occurs, where unstable organic matter and salts are flushed out as the filter material settles. Biochar initially showed the lowest overall release of N-tot. Of the biochar filters, only filter B1 showed a negative removal rate for N-tot, and only at the first measurement. During the end of campaign 1, the removal efficiencies for the biochar filters were between 69–80% for N-tot. The ash filters managed to significantly lower the initial high release, about 8–30 times the influent concentrations of N-tot during campaign 1. At the last measurement in August, the performance of the ash filters varied (A1: -87%, A2: -13%, A3: <q.l., A4: 69%), although there was a significant improvement for all filters compared to the initial results. The peat filters released most N-tot at the end of the campaign with a negative removal efficiency from –160 to -320 %, but compared to the initial concentrations there was a significant lowering of released N-tot. The control filter (filter C) showed the highest initial release of N-tot, of about 77 times higher effluent concentration compared to influent, and was comparable to the peat filters during campaign 1. However, at the last measurement, the leaching of N-tot was still negative at about –160% removal efficiency but had improved about 48 times compared to the first acquired values, showing a clear decline in nutrients leaching from the control filter materials. However, it was only the biochar filters that managed to

reduce the nitrogen concentrations below the Gothenburg guideline value of 1250 µg/L for polluted stormwater release into recipients.

4.2 Removal of metals

Several metals (total concentrations) were analysed, e.g., Co, Cr, Cu, Ni, Pb, and Zn, and for the summer campaign, the highest concentration was found for Zn with 0.25 mg/L in the influent water, Figure 8. The overall results show that rain gardens are efficient for the reduction of several metals (total metal), which is in line with many studies before (Davis et al. 2003, Muthanna et al. 2007, Søberg et al, 2017, Lange et al 2020), even at the start of irrigation, before the filter beds were stabilised. As for the nutrients, the metal concentrations in the effluents decreased with time, showing a stabilization phase for the metals as well, or were continuously low (Cr and Hg). Initially, some metals were released from the filter materials e.g. As and Cd from the ash filters, and Pb from the biochar filters. The effluent concentrations were at maximum <0.002 mg/L for As and Pb and <0.002 mg/L for Cd compared to the maximum influent concentrations; 0.001mg/L for As, 0.005 mg/L for Pb, and <0.0001 mg/L for Cd, respectively, and the release from the filter materials decreased rapidly. After less than 20 days they were all below the stormwater guidelines for the city of Gothenburg.

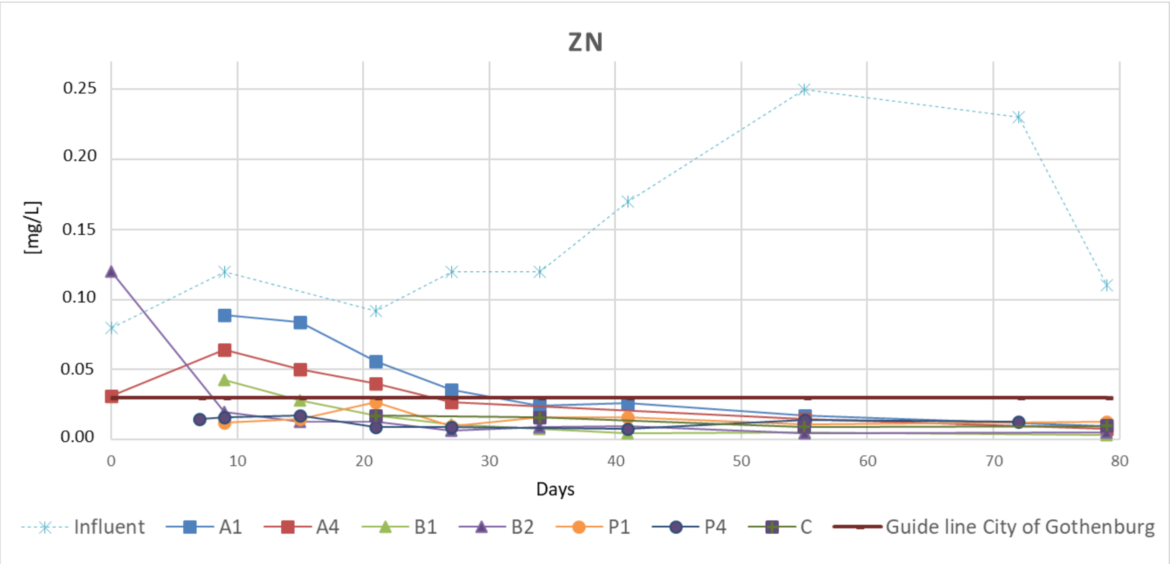

Figure 8. Concentrations of Zn in influent and effluent waters in the different filters including the control filter C. In addition, the guideline value for the City of Gothenburg is shown.

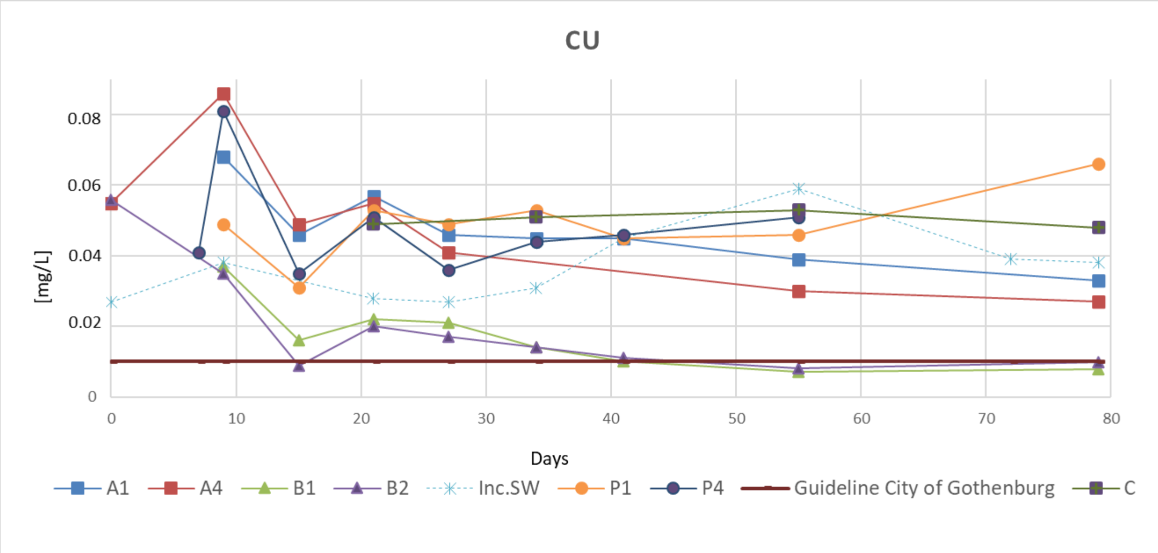

Figure 9. Concentrations of Cu in influent and effluent waters in the different filters including the control filter C. In addition, the guideline value for the City of Gothenburg is shown.

In general, the concentrations in the effluents were lower than the influent already after a few weeks for most metals and filters, i.e., the reduction efficiencies were high already from the starting up of the rain garden. As an example of the efficient reduction with time, Zn is shown in Figure 8. After 55 days the reduction of Zn was >90% in all filters, and after 80 days the reduction was near 100%, independently of the increasing concentrations in the incoming water, Figure 8. As discussed earlier, the city of Gothenburg has guidelines for maximum concentrations in stormwater for total concentrations of several metals and among the ones studied here, after 80 days running of the rain gardens, only Cu exceeds the guideline in the effluent waters, Figure 9. However, the biochar filters reduced the Cu concentration to below the guideline and there is a tendency that the other filter will follow. The general difference in metal removal performance between the different filter materials could be due to several factors. Søberg et al, 2019, tested different filter materials, including the sandy loam with pumice stones that is used in this study for adsorption capacities of dissolved metals, and found that low organic material content, higher pH, and large specific surface areas were beneficial for metal removal. They also found that this sandy loam material leached Cu, Ni, Pb, and Zn in low concentrations. This agrees with our initial results, given that the biochar filters showed the overall best results regarding metal removal, as well as high pH, low leaching of organic content in the effluents, etc. It is noticeable that the control filter has the second highest release after 80 days, even above the influent, showing that also commonly used soil materials for rain gardens can leach metals when subjected to stormwater. The results from the metal analysis from the winter campaign will be published in a coming paper (Johansson et al, 2023b).

4.2 Removal of microplastics

The efficiency of using rain gardens to prevent microplastics from reaching estuaries was examined by studying the influents and effluents of the pilot-scale rain garden, with 13 bioretention filters, in the catchment area of the Göta Älv river. Urban stormwater from a highly trafficked highway passing through Gothenburg is known to contain high amounts of pollutants, such as e.g., aliphatic hydrocarbons, phthalates, PAH (Markiewicz et al., 2017; Björklund et al., 2009), and tyre and road wear and other microplastic (Billsten et al., 2020; Chalmers unpublished data). In Table 1, results from a pyrolysis-GC/MS analysis of ten polymers in stormwater and stormwater sediment at the Gårda pilot rain garden are presented. As expected, microplastics originating from tyre and road wear, indicated by concentrations of polyisoprene and polybutadiene polymers, were dominating in the stormwater sediment with contents of more than 150 mg/kg DS. Additionally, other plastic polymers that are highly used in society such as polyethylene, polypropylene, and polyvinylchloride were found in amounts of about 100 mg/kg DS. All these polymers were also dominant in the corresponding stormwater, which without further treatment will reach the Göta Älv river estuary.

In the present project, stormwater from the sediment chamber mentioned above was used to irrigate the bioretention filters in the Gårda pilot rain garden. In the first campaign, polymers from microplastic particles >10 µm were quantified in concentrations >1.0 µg/L, see Table 2, and the most common microplastics were detected in more than 50% of the influent water samples, while

Table 1. Chemical analysis of ten microplastic polymers in stormwater and stormwater sediment at the Gårda pilot rain garden.

Microplastic polymers	Stormwater sediment Gårda 2022 µg/kg DS	Stormwater Gårda 2022 µg/L
Polyisoprene (PI)	142 000	126
Polybutadiene (PB)	11 500	88,4
Polyethylene (PE)	67 700	137
Polypropylene (PP)	11 800	11,6
Polyvinylchloride (PVC)	10200	118
Polystyrene (PS)	6680	10
Polyethylene terephthalate (PET)	749	<1.0
Polyamide 6 (PA6)	<30.0	<1.0
Polymethylmethacrylate (PMMA)	2040	<1.0
Polycarbonate (PC)	<30.0	<1.0
Sum Polymers	191 000	277

generally in less than 10% of the effluent samples, independently of filter type. The removal capacities of the polymers at the end of the first campaign and in August 2022 reached 98–100% for all polymers, except for PE which was lower, 83–100%, and for PC and PMMA which was leaking from the peat filter P4. For PP, the high concentrations in the outlet from most filters are explained by the contamination of PP residuals in the sampling containers after drilling holes when assembling the pilot facility. In total, the results from the summer campaign showed the potential of using rain gardens for decreasing the spreading of tyre wear particles and other microplastics. Other recent studies examining the microplastic removal potential for bioretention filters show similar results (Gilbreath et al. 2019, Smyth et al. 2021, Lange et al. 2021, Lange et al. 2022), although it is not common in the other studies to analyse particles as small as >10 µm. During the second campaign (January-March 2023), see Table 3, the filters also showed a good potential for removal of the microplastics during winter conditions (northern Europe), where there is a risk of freeze-thawing cycles in the filters. Microplastics (>10µm) and the polymers associated with the tyre wear particles were detected in all influent samples (n=9) during this period, while quantified only in two effluent samples (n=15). During the winter campaign, the analysis method was developed further to also include three additional polymers and the sum of rubber components.

Table 2. Chemical analysis of ten microplastic polymers in inflow and effluents, in selected ash, biochar, and peat bioretention filter during the summer campaign June – August 2022 at the Gårda pilot rain garden.

	SW	**A1 ash**		**A4 ash + plants**		**B1 biochar**		**B2 biochar + plants**		**P1 peat**		**P4 peat + plants**		**C plants**	
Compounds µg/L[a]	influent (n[b]=7)	effluent (n=5)	removal efficiency 23;57 days %	effluent (n=5)	removal efficiency 23;57 days %	effluent (n=5)	removal efficiency 23;57 days %	effluent (n=5)	removal efficiency 23;57 days %	effluent (n=5)	removal efficiency 23;57 days %	effluent (n=4)	removal efficiency 43;57 days %	effluent (n=5)	removal efficiency 23;57 days %
Polyethylene (PE)	8.4–180 (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
Polyisoprene	<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
Polybutadiene (PBD)	<ql–88 (24)	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100	<ql	100;100
Polypropylene (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
Polystyrene (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
Polyvinyl chloride (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
Polyethylene-terephthalate (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
Polycarbonate (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;$-80*10^4$
Polyamide 6 (PA6)	<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql	<ql	<ql;<ql
Polymethyl-methacrylate (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; [c]<ql=below the limit of quantification; [d]removal efficiency=accumulated removal efficiency after 23 and 57 days

Table 3. Chemical analysis of 13 microplastic polymers and rubber components in inflow and effluents, in selected ash, biochar, and peat bioretention filter during the winter campaign January - March 2023 at the Gårda pilot rain garden. [a]

	SW	A1 ash		A4 ash + plants		B1 Biochar		B2 biochar + plants		P1 peat		P4 peat + plants		C plants	
Compounds µg/L[a]	influent (n[b]=9)	effluent (n=3)	removal %	Effluents (n=3)	removal %	effluent (n=3)	removal %	effluent (n=3)	removal %	effluent (n=3)	removal %	effluent (n=3)	removal %	effluent (n=3)	removal %
Polyethylene (PE)	<ql–130	<ql	100	<ql–5.0	96–100	<ql–1.3	100	<ql–1.5	-29–100	2.9	98	<ql–1.8	98–100	<q.l	100
Polyisoprene	<ql–460	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100
Styrene butadiene rubber (SBR)	1.2–140	<ql	100	<ql	98–100	<ql	100	<ql	98–100	<ql	100	<ql–1.1	98–100	<ql–0.4	100
Acrylonitrile- butadiene styrene	<ql	<ql	100	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql
Polybutadiene (PBD)	2.6–250	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100
Polypropylene (PP)	<ql–95	49	-14000	<ql–8.0	-17–96	31	78	3.8–37	-131–67	6.5	-190	16–22	-38000-83	3.4–7.1	100
Polystyrene (PS)	<ql–1.7	<ql	100	<ql	100	<ql	100	<ql	99–100	<ql	100	<ql	100	<ql	99–100
Polyvinyl chloride (PVC)	<ql–47	<ql	100	<ql	99–100	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100
Polyethylene tere-phthalate (PET)	<ql–0.80	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100	<ql	99–100
Polycarbonate (PC)	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	<ql	100	<ql	<ql
Polyamide 6 (PA6)	<ql–0.30	<ql	100	<ql	100	<ql	100	<ql	95–100	<ql	100	<ql	100	<ql	100
Polyamide-6,6 (PA66)	3.2–9.2	<ql	100	<ql	100	<ql	100	<ql	99–100	<ql	100	<ql	100	<ql	100
Polymethyl methacrylate (PMMA)	<ql–2.8	<ql	100	<ql	100	<ql	100	<ql	96–100	<ql	100	<q.l	99–100	<ql	100
Sum plastic polymers	16–210	49	76	3.8–13	49–96	32	84	3.8–39	63–89	9.4	95	16–24	-23–88	3.4–7.1	100
Sum rubber components	7.5–850	<ql	100	<ql	100	<ql	100	<ql	100	<ql	100	1.1	100	<ql–0.4	100

minimum and maximum values; n[b] =number of samples analysed; [c]<ql=below the limit of quantification; [d]removal = removal efficiency after irrigation.

The removal efficiency for microplastics, >10µm, during the winter campaign (if excluding PP and PE that was most likely from the equipment), was for all filter types in the range between 95 – 100% (Table 3). High concentrations of rubber components 7.5 – 850 µg/L were detected in the inlet water but were only once detected in the effluent of the control filter. In the winter campaign, a few selected samples were also analysed for the concentrations of tyre wear particles >1.2 µm with a more developed pyrolysis-GC/MS technique for analysis of tyre wear (Rödland et al., 2022). With this technique, it was now possible to quantify tyre wear particles both in the inlet and outlet water from the columns, but still, the removal efficiency for tyre wear particles was >97% for all filter types. These results show that there are significant amounts of tyre wear particles in the interval 1.2 – 10 µm, that is not detected with the other, commercial, pyr-GC/MS method used in this project, and analysis methods to be able to analyse even smaller tyre wear particles are highly requested (Järlskog, 2022b).

4.4 Removal of organic pollutants

Six groups of aliphatic hydrocarbons, five groups of aromatic hydrocarbons, BTEX (benzene, toluene, ethylbenzene, m,p-xylene and o-xylene), 16 specific polycyclic aromatic hydrocarbons (PAH), and 13 specific phthalates were analysed at some occasions in inlet and outlet from selected bioretention filters during the summer campaign. The selected filters were without plants (A1, B1, P1) and with plants (A2, A4, B2, B4, P3 and P4). Concentrations of all the specific organic pollutants quantified in inflow and effluents, in the filters during the summer campaign are presented in Table 4. These organic pollutants were also analysed during the winter campaign January – March 2023 in selected filters, but these results will be evaluated later and published in a coming scientific paper (Johansson et al., 2023b).

Aliphatic hydrocarbons C_5-C_{16} up to 81 µg/L, and C_{16}-C_{35} up to 110 µg/L, were analysed in inlet water, pumped up and originating from the Gårda inlet stormwater chamber, and used to irrigate the bioretention filters in the rain garden (Table 5). From the biochar filter B2 in June, C_5-C_{16} leached, but were 100% removed in all the bioretention filters in August, see Table 5. Aliphatic hydrocarbons C_{16}-C_{35} leached from the peat filters P1, and from the biochar B1 and B2 up to 99 µg/L. However, these concentrations were far lower than the City of Gothenburg's guidelines for the release of polluted water to recipients of 1000 µg/L. Sources of the aliphatic hydrocarbons in the stormwater are exhaust and diesel fuel emitted in the gas and particulate phase from diesel engines (Alam et al., 2019; Anh et al., 2019), as well as engine oil and asphalt wear (Hwang et al., 2019). The aliphatic hydrocarbons leached only from the peat filter P1 in August (Table 5); for all other filters, the removal efficiency was 100% in August.

Table 4. Concentrations of specific organic pollutants quantified in inflow and effluents, in selected ash, biochar, and peat bioretention filter during the summer campaign June – August 2022 at the Gårda pilot rain garden.

Compounds concentrations µg/L[a]	Stormwater inlet filter (n[b]=3)	A1 ash effluent (n=3)	A2, A4 ash + plants effluent (n=5)	B1 biochar effluent (n=3)	B2, B4 biochar + plants effluent (n=5)	P1 peat effluent (n=3)	P3, P4 peat + plants effluent (n=5)
Σ aliphates >C_5-C_{16}	<q.l.[d]–81	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.
Σ aliphates >C_{16}-C_{35}	36–110	<q.l.	<q.l.	<q.l.–41	<q.l.–99	<q.l.–23	<q.l.
phenantrene	<q.l.–0.0076	<q.l.–0.0020	<q.l.–0.0054	<q.l.	<q.l.	<q.l.	<q.l.
fluoranthene	<q.l.–0.022	<q.l.–0.0011	<q.l.–0.0030	<q.l.–0.0013	<q.l.	<q.l.	<q.l.–0.0010
pyrene	<q.l.–0.033	<q.l.–0.0012	<q.l.–0.0012	<q.l.	<q.l.	<q.l.	<q.l.–0.0013
chrysene	<q.l.–0.0093	<q.l.–0.0011	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.
benso(b)fluoranten	<q.l.–0.0096	<q.l.	<q.l.–0.0016	<q.l.	<q.l.	<q.l.	<q.l.–0.0011
bens(k)fluoranthene	<q.l.–0.0029	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.
benso(a)pyrene	<q.l.–0.0052	<q.l.	<q.l.–0.0013	<q.l.	<q.l.	<q.l.	<q.l.
indeno(1,2,3,cd)pyrene	<q.l.–0.0038	<q.l.	<q.l.–0.0006	<q.l.	<q.l.–0.00033	<q.l.	<q.l.–0.00065
benso(g,h,i)perylene	<q.l –0.013	<q.l.	<q.l.–0.0012	<q.l.	<q.l.–0.00045	<q.l.	<q.l.–0.00066
Σ LMW PAH	<q.l	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.
Σ MMW PAH	<q.l – 0.063	<q.l.–0.0023	<q.l.–0.0085	<q.l.–0.0013	<q.l.	<q.l.	<q.l.–0.0021
Σ HMW PAH	<q.l.–0.044	<q.l.	<q.l.–0.0047	<q.l.	<q.l.–0.00078	<q.l.	<q.l.–0.0024
Σ PAH-16	<q.l –0.11	<q.l.–0.0023	<q.l.–0.013	<q.l.–0.0013	<q.l.–0.00078	<q.l.	<q.l.–0.0037

[a] minimum and maximum values; n[b] =number of samples analysed; [c]<q.l.=below the limit of quantification.

Table 5. Removal efficiencies of specific organic pollutants quantified in inflow and effluents, in selected ash, biocha,r and peat bioretention filter during campaign 1 at the Gårda pilot rain garden.

	A1			A4			B1		
Date	2022-06-15	2022-06-27	2022-08-18	2022-06-15	2022-06-27	2022-08-18	2022-06-15	2022-06-27	2022-08-18
	%			%			%		
aliphatics >C5-C8	<q.l.*	eff.<q.l.*	<q.l.*	<q.l.*	eff.<q.l.*	<q.l.*	<q.l.*	eff.<q.l.*	<q.l.*
aliphatics >C5-C16	<q.l.*	eff.<q.l.*	<q.l.*	<q.l.*	eff.<q.l.*	<q.l.*	<q.l.*	eff.<q.l.*	<q.l.*
aliphatics >C16-C35	eff.<q.l.*	eff.<q.l.*	eff.<q.l.*	eff.<q.l.*	eff.<q.l.*	eff.<q.l.*	56.4	eff.<q.l.*	eff.<q.l.*
sum PAH-L	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*
sum PAH-M	eff.<q.l.*	97.8	eff.<q.l.*	eff.<q.l.*	97.8	eff.<q.l.*	-80.5	eff.<q.l.	eff.<q.l.*
sum PAH-H	-696	eff.<q.l.*	<q.l.*	eff.<q.l.*	eff.<q.l.*	<q.l.*	eff.<q.l.*	eff.<q.l.*	<q.l.*

	B2			P1			P4		
Date	2022-06-15	2022-06-27	2022-08-18	2022-06-15	2022-06-27	2022-08-18	2022-06-15	2022-06-27	2022-10-11
	%			%			%		
aliphatics >C5-C8	<q.l.*	eff.<q.l.*	<q.l.*	<q.l.*	eff.<q.l.*	<q.l.*	<q.l.*	eff.<q.l.*	<q.l.*
aliphatics >C5-C16	<q.l.*	eff.<q.l.*	<q.l.*	<q.l.*	eff.<q.l.*	<q.l.*	<q.l.*	eff.<q.l.*	<q.l.*
aliphatics >C16-C35	eff.<q.l.*	eff.<q.l.*	eff.<q.l.*	eff.<q.l.*	eff.<q.l.*	36.1	eff.<q.l.*	eff.<q.l.*	eff.<q.l.*
sum PAH-L	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*	<q.l.*
sum PAH-M	eff.<q.l.*	eff.<q.l.	eff.<q.l.*	eff.<q.l.*	eff.<q.l.	eff.<q.l.*	eff.<q.l.*	98.0	eff.<q.l.*
sum PAH-H	*eff.<q.l.**	*eff.<q.l.**	*<q.l.**	*eff.<q.l.**	*eff.<q.l.**	*<q.l.**	*eff.<q.l.**	*eff.<q.l.**	*<q.l.**

<.q.l. Below quantification limit for both influent and effluent; eff. <q.l.** Above quantification limit in influent, below quantification limit in effluent.*

For the PAH-16 the following specific compounds were detected in concentrations higher than the limit of quantification in the inlet (Table 5), and in the concentration order: pyrene > fluoranthene > benso(g,h,i)perylene > benso(b)fluoranten ≈ chrysene > phenanthrene > benso(a)pyrene > indeno(1,2,3,cd)pyrene > bens(k)fluoranthene, which are in line with the relative composition of specific PAH found in urban stormwater (Polukarova et al., 2020) and stormwater sediment at Gårda (Markiewics, 2017). The PAH composition in this study suggests a mixture of several traffic-related sources: tyre wear, vehicle exhausts, brake linings, motor lubricant oils, and road surface wear (Markiewicz et al., 2017; Zhang et al., 2020). The concentrations of the total PAH-16 in the influent were up to 0.11 µg/L, which is low compared to the Gothenburg local guideline value for benzo(a)pyrene (0.27 µg/L). For the ash and peat filters the concentrations of PAH-16 were highest in the filters with plants indicating possible initial channeling in filter A2 and P4. However, there was no statistical significance to support this. The PAH-16 total concentrations were much lower in the effluents from the bioretention filters, but concentrations (up to 0.013 µg/L) in the ash filter effluents were detected in June. However, the concentrations from the same filter effluents were ten times lower in August. PAH-16 leached in lower concentrations from the biochar and peat filters during the summer campaign. The removal efficiencies in the filters in August were 99%, and the only filter without any leaching of PAH was filter P1 (peat filter, without vegetation). The last analysis of organic pollutants for P4 was done in October, instead of August, due to a lack of sufficient sample volume for all the analyses.

4.5 Hydraulic Conductivity

The saturated hydraulic conductivity (K_{sat}) was measured after the effluent sampling was finished for campaign 1, and the results are presented in Table 6. The peat filters had the lowest K_{sat} value, with values between 99–130 cm/h, followed by the ash filters which showed a greater span of 132–212 cm/h. 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} for three of the filters (B2: 642 cm/h, B3: 554 cm/h, B4: 469 cm/h), but also the greatest span between filters, as the K_{sat} for filter B1 was much lower than the rest and at 144 cm/h. Tt 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 could not be seen for the other filter types. It should be noted that the hydraulic measurements only were carried out on one occasion for campaign 1. The reason for not measuring K_{sat} for all filters on the same day, which would have been preferable, was due to not having access to enough amount of stormwater for saturating all filters simultaneously. This could have affected the results, due to the filters tested might have varied in vegetative and/or mycorrhizal development, settling of materials etcetera because of the time period between the measurements. However, there were general similarities between the filter types (ash, biochar, and peat). The saturated hydraulic conductivity will be measured again as the work with the pilot facility continues.

Table 6. Hydraulic conductivity in saturated soil in the bioretention filters at Gårda pilot rain garden after campaign 1.

Filter		C	A1	A2	A3	A4	B1	B2	B3	B4	P1	P2	P3	P4
Date		2022-08-26	2022-09-23	2022-10-03	2022-09-26	2022-09-26	2022-09-26	2022-09-23	2022-10-03	2022-09-26	2022-08-26	2022-10-03	2022-09-26	2022-09-26
(Ksat)	**cm/h**	**387**	**206**	**132**	**154**	**212**	**144**	**642**	**554**	**469**	**130**	**105**	**99**	**102**

5. Conclusions

Only a few percent of the stormwater generated in urban environments is treated and therefore reaches nearby watercourses untreated, and the largest proportion of microplastics found in Swedish urban stormwater derives from tyre and road wear. Protecting the environment from the cocktail of pollutants found in stormwater in the form of microplastics, potentially toxic metals, nutrients, and organic pollutants is of high societal value and a requirement to meet environmental goals. The project aimed to develop innovative rain gardens and to study their efficiency in treating stormwater from the cocktail of pollutants.

This work is part of an ongoing Ph.D. project, and the research will continue in the coming years with further results and conclusions. However, so far we can conclude that urban stormwater from the strongly trafficked highway E6 through Gothenburg is highly polluted because metals, organic pollutants, tyre wear and other microplastics, and nutrients are analysed in high concentrations. This water, with the only treatment of larger particles due to sedimentation in a chamber system, is released into the receiving creek Mölndalsån and thus enters and pollutes the Göta River Estuary. In this study, rain gardens with active filter materials i.e., biochar, peat, and ash, and plants identified to enhance bioremediation of the pollutants of interest, are used to address the pollutants. After one cultivation season, it can be concluded that all plants survived in all filters and all filters efficiently remove the microplastics, tyre and road wear particles and other pollutants i.e., metals, nutrients, and organic pollutants from the urban stormwater. All metal concentrations in the effluents are below the City of Gothenburg guidelines when passing the filters, except for Cu. However, the concentrations are decreasing with time. The results also showed that aliphatic hydrocarbons and PAH were efficiently removed in all bioretention filters. During the first campaign, plants were not shown to significantly affect the removal efficiencies of selected pollutants. This might be due to that the vegetation was planted shortly before the campaign started, and thus was still established during the timeframe of the campaign.

Concerning the microplastics, i.e., Polyisoprene (PI), Polybutadiene (PB), Polyethylene (PE), Polypropylene (PP), Polyvinyl chloride (PVC), Polystyrene (PS), Poly(methyl methacrylate) (PMMA), Polyethylene terephthalate (PET), Polyamide 6 (PA6), and Polycarbonate (PC), microplastic particles >10µm were efficiently removed from the stormwater after passing through the bioretention filters. PE and PP were detected in the effluents, especially at the start of the campaign, and this is depending on the release of microplastics from the material used when constructing the filters, as the effluent concentrations decreased with time. Some initial analyses on tyre wear particles >1.2 µm showed higher concentrations of microplastics in both influent waters as well as in the effluents. However, the removal efficiencies were similar to the efficiencies found for the larger particles. Further studies on these smaller sizes of microplastic particle sizes are planned in future studies.

6. Summary

Urbanization has contributed to a degradation in the quality of surface water, where inadequate stormwater management and pollution control have played an important role. Stormwater management in urban areas has moved from quantity control and combined sewers to current strategies for quantity and quality source control, with an emphasis on the multiple benefits provided by the blue-green infrastructure. Though most stormwater discharges are still transported untreated to receiving waters, various technologies have been developed and used to treat stormwater locally. However, research supporting the new development of innovative and more effective technologies, and management strategies, are urgently needed to meet the demands on sustainable development of urban environments.

A cocktail of emerging environmental pollutants such as microplastics, toxic metals, nutrients, and organic pollutants are released into urban environments and emissions are particularly high in highly trafficked areas. The largest proportion of pollution is transported from roads by runoff and further by stormwater to receiving watercourses. So far, research has focused on clarifying the consequences and fate of the cocktail of pollutants in the environment; however, the current technological solutions that address the problems are insufficient. Only a few percent of the stormwater generated in urban environments is treated, and therefore transported untreated to nearby aquatic environments, and the largest proportion of microplastics found in Swedish urban stormwater derives from tyre and road wear. Protecting the environment from the mixture of pollutants found in stormwater is of high societal value and a requirement to meet environmental goals.

The aim of this project was to develop an innovative rain garden pilot facility with different bioretention filters and to study their efficiency in treating stormwater from the above-mentioned pollutants. A pilot test rain garden with 13 bioretention filters was designed, and experiments were carried out to study beds with biochar, peat, and ash, with and without plants, including removal efficiency, hydraulic performance, treatment, and degradation processes. The project aimed to support the development of green infrastructure in urban environments. This project is the first study of microplastics and a cocktail of other pollutants in pilot-scale rain gardens, using a combination of plants and sorption materials such as municipal solid waste incineration bottom ash, peat, and biochar in the soil bed. The relevance and value of this project are to support the implementation of raingardens for the development of green and sustainable urban infrastructure. By treating polluted stormwater as close to the emission sources as possible, its nearby watercourses will be subjected to less pollutants.

The results showed that tyre wear and other microplastic particles >10μm were efficiently removed from the stormwater after passing through the bioretention filters, except for PE and PP. This was due to the release from the material used when constructing the filters, and both the release of PE and PP decreased with time. Some initial analyses on tyre particles >1.2 μm showed higher concentrations of tyre polymers in both influent waters as well as in the effluents. However, the reduction efficiencies were similar to the efficiencies found for the larger particles. Further studies on these smaller sizes of microplastic particle sizes are planned in future studies. The results showed that aliphatic hydrocarbons and PAH were also efficiently removed in all filters. All metal concentrations in the effluents are below the City of Gothenburg guidelines when passing the filters, except for Cu. However, the concentrations are decreasing with time. Nutrients in the form of total nitrogen were initially high in the effluent waters, as nitrogen was released from the filter materials themself, but the concentrations decreased with time in all filters, but it was only the biochar filters that managed to reduce the nitrogen concentrations below the Gothenburg guideline value for polluted stormwater release into recipients. This initial study of the removal efficiencies and removal processes for nutrients, metals, organic pollutants, tyre wear and other microplastics shows very promising results, especially for the biochar filters. Results for the coming years running and research of the filters will be presented in a Ph.D. thesis at the Chalmers University of Technology.

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