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Alternative sorption filter materials effectively remove non-particulate organic pollutants from stormwater

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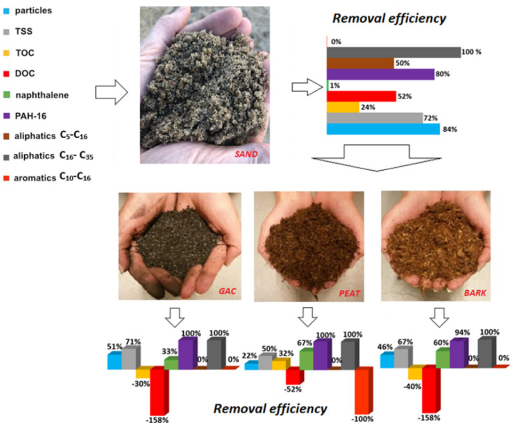
HIGHLIGHTS

- A sorption filter pilot plant for stormwater was successfully operated for 18 months.
- High removal was achieved for TSS in the GAC and for TOC in the peat filters.
- Bark and peat sorption filters effectively removed aliphatics C16–C35 and PAHs.
- All sorption filters performed very well under heavy pollution load.
- Emissions of nanoparticles were reduced by combining sand and sorption filters.

GRAPHICAL ABSTRACT



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ABSTRACT

Urban runoff contains a mixture of both particulate and non-particulate organic pollutants (OPs). Hydrophobic OPs such as higher petroleum hydrocarbons, phthalates, and polycyclic organic hydrocarbons (PAHs) are not exclusively bound to particles, but also present in runoff in colloidal and truly dissolved forms. These hydrophobic compounds can also form nano- and microsized emulsions that may carry pollutants in stormwater. Hence, it is of great importance to develop treatment technologies such as sorption filters that can remove non-particulate OPs from contaminated stormwater. A pilot plant using column bed-filters of sand as a pre-filter, in combination with granulated activated carbon, *Sphagnum* peat or *Pinus sylvestris* bark, was used to investigate the removal of non-particulate OPs from urban stormwater. Samples from the filter effluents were collected weekly; during or after rain events; and during stress tests when incoming water was spiked with contaminated sediment and petrol or diesel. All sorption filters showed efficient reduction of aliphatic diesel hydrocarbons C_{16}–C_{35}, benzene, and the PAHs phenanthrene, fluoranthene, and pyrene during most of the operation time, which was 18 months. During the stress test events, all sorption filters showed 100% reduction of PAH-16, petrol and diesel aliphatics C_5–C_{35}. All sorption filters released DOC and nanoparticles, which may explain some of the transportation of OPs through the filter beds. The recommendation is to use a combination of sand pre-filtration and all the studied sorption materials in stormwater filters in series, to achieve effective removal of different types of OPs. It is also important to improve the hydraulic conditions to obtain sufficient water flows through the filters.

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1. Introduction

Stormwater management in urban areas has moved from quantity control and combined sewers to the current strategies of quantity and quality source control, with emphasis on the multiple benefits provided by blue-green infrastructure (Eckart et al., 2017). Although this has led to expanded opportunities to treat stormwater locally, most stormwater discharges are still transported untreated to receiving waters. Studies show that organic pollutants (OPs) such as phthalates, alkylphenols and their ethoxylates, polycyclic aromatic hydrocarbons (PAHs), and aliphatic petroleum hydrocarbons are ubiquitous in urban runoff (Björklund et al., 2009; Markiewicz et al., 2017). Traffic represents a major source of OPs to the receiving environment through exhaust emissions, oil and grease spills, tyre and brake wear, and surface abrasion (Björklund, 2010; Markiewicz et al., 2017; Zhang et al., 2019). Because of the harmful effects of OPs on biota and humans, including a combination of genotoxicity, bioaccumulation, and resistance to degradation, stormwater treatment is becoming increasingly important (Haile, 2018; Mwanamoki et al., 2014; Rossi et al., 2004). Research and development that support innovative and effective technologies and management strategies for stormwater quality are needed to improve the health of aquatic ecosystems. Preventive measures may be an effective way to reduce the amounts of contaminants being transported with urban runoff; for example, in-situ treatment techniques such as street sweeping and vehicle wash have been proven to be successful pollution control measures (Markiewicz et al., 2020; Polukarova et al., 2020). Stormwater treatment is often based on sedimentation and generally not designed for efficient removal of OPs, metals, and inorganic substances, as the forms that attach to particles <1 μm do not settle (Haranas et al., 2012; Ilyas and Muthanna, 2017b). For this reason, there is an urgent need to develop efficient treatment methods that can prevent further transport and spread of OPs into the environment, where they may bioaccumulate in food chains.

Filtration of stormwater through a sorption material is one of the most promising techniques for removal of colloidal and dissolved OPs. Several field and laboratory studies have reported on the use of various alternative filter materials for stormwater treatment, originating from waste products, minerals, or plant-based materials (Björklund and Li, 2015; Ilyas and Muthanna, 2017a; Zhang et al., 2019). The efficiency of sorbents is characterised by physical-chemical properties such as polarity, aromaticity, surface area, pore size, and pore volume (Wenzhong et al., 2008; Xi and Chen, 2014). When selecting materials to use in filters, their sorption selectivity, mechanical stability, environmental impact and toxicity, cost-effectiveness, and reusability potential must also be considered. One of the challenges of stormwater treatment using sorption filters is to find sustainable materials in which the OPs are effectively degraded, while sorbed metals can be recovered (Fedje et al., 2015; Fedje and Strömvall, 2019), and un-degraded litter such as microplastics separated after use. Clogging is another challenge recognised as a key limiting factor of sorption filters for stormwater treatment (Kandra et al., 2015). Clogging is caused by physical, biological and chemical processes; clogging can for example be caused by fine-grained particles migrating into the pores of a coarser sorption material, biological growth on, or degradation of, the sorption material, or chemical precipitation of metal salts.

Peat and bark, readily available alternative low-cost sorption materials, have previously been proven useful for removing OPs and metals from contaminated waters (Björklund and Li, 2015; Kalmykova et al., 2014; Kalmykova et al., 2010). However, peat and bark have not previously been studied in pilot-scale filters, for longer time-periods with real stormwater, or for determining the effectiveness and sorption of non-particulate OPs. Peat is a complex organic material containing minerals and partially decomposed organic matter, where organic compounds such as lignin, cellulose, fulvic, and humic acids are major components (Ahmaruzzaman, 2008). The exact sorption mechanisms of OPs in peat are unclear and important to consider in future studies. The sorption may be a linear absorption process in which pollutants in high concentrations are partitioned into the peat's humic and fulvic substances (Kalaitzidis et al., 2006). The sorption in peat may also occur as a non-linear process, where OPs in low concentrations accumulate on parts of the peat with more condensed or oxidized organic matter, through adsorption, filling, and condensation within micropores.

Bark from pine (*Pinus silvestris*) is a waste product from the forest industry and can be used, without any additives or chemical preparation, for absorption of liquids such as petroleum products, emulsions, cutting fluids, glycol, paint, varnish, urine, and blood (Larsson, 2015). Pine bark is also effective in retaining hydrophobic OPs such as organochlorine pesticides (Ratola et al., 2003), pentachlorophenol (Brás et al., 2004), and PAHs (Björklund and Li, 2015). The low porosity and specific surface area of pine bark do not affect its sorption capacity, as most of its removal efficiency has been attributed to its content of organic compounds, such as lignin, which is hydrophobic and has a high aromatic content (Huang et al., 2006; Li et al., 2010; Xi and Chen, 2014). The chemical composition of bark is otherwise very complex and depends both on the tree species and on the morphology of the bark (Valentín et al., 2010). Pine bark consists mainly of lipophilic extractives, such as fats, waxes, terpenes, terpenoids, and higher aliphatic alcohols, and hydrophilic constituents, including condensed tannins (phenolic acids), but also contains insoluble compounds like polysaccharides, lignin, and suberin. All these compounds are likely to contribute to the OP sorption capacity of bark.

Many of the petroleum hydrocarbons frequently found in urban runoff, and particularly in road runoff, such as the BTEX compounds (benzene, toluene, ethylbenzene, and xylenes) and lighter aromatic hydrocarbons, are volatile and less hydrophilic, and not likely to be found in the particulate phase in stormwater (Fingas, 2016). On the other hand, many organic pollutants analysed in stormwater are hydrophobic by definition, e.g. higher petroleum hydrocarbons, phthalates, and medium and high molecular weight PAHs, and therefore assumed to be removed from the water phase through sedimentation of particles. However, it has been shown that hydrophobic OPs are also present in stormwater, in colloidal and truly dissolved forms (Nielsen et al., 2015). Moreover, hydrophilic-hydrophobic compounds, such as diesel hydrocarbons, alkylphenols (APs) and their ethoxylates (APEOs), diesel with APs and APEOS, phthalates, and stormwater containing OPs with humic acids (HA) and iron (Fe) colloids can form nano- and microsized emulsions that may carry pollutants in stormwater (Markiewicz et al., 2019). For this reason, it is of great importance to develop treatment techniques such as sorption filters that can remove both colloidal, truly dissolved, and emulsions of OPs from contaminated stormwater. These filters must also effectively remove the thousands of specific OPs with very different chemical properties, polarity, molecular weight, density, and volatility, which are released in urban and especially road environments (Markiewicz et al., 2017).

This is the first study aiming to research the treatment of road runoff polluted with non-particulate OPs using sorption filters. The chosen sorption materials have previously shown good sorption of different OPs in batch tests and are here studied, for the first time, on a larger scale in a pilot plant. The filters were exposed to stormwater from a catchment of mixed land use for 18 months. On several occasions, the filters' capacity was tested when incoming water was loaded with contaminated sediments and diesel or petrol.

2. Materials and methods

2.1. Characteristics of the sorption materials

The alternative sorption materials, *Sphagnum* peat and bark from *Pinus sylvestris,* were compared with granulated activated carbon (GAC). The sorption materials were analysed for physical-chemical properties (Table 1). These filter materials were selected because they: (1) contain natural organic compounds with potentially high sorption capacity for OPs, (2) have a high porosity, and (3) have a large active surface area for sorption, and in the case of bark also (4) a high hydraulic loading rate.

The pH of the GAC was basic whereas the bark and peat were acidic. The GAC exhibits a substantially larger surface area than peat and bark. The surface area is a key quality parameter for sorbents, and the reason for the reported excellent sorption capacity of many activated carbon materials. However, the content of organic matter was lower in GAC than in bark and peat. The hypothetical maximum flows in the pilot plant were determined to 3.2 L/min for peat and 26 L/min for bark (Table 1). Studies have found that the typical hydraulic loading rate of GAC in columns is between 7 and 10 m/h (Reed et al., 1996), which corresponds to a flow of between 8 and 12 L/min through the GAC column in the pilot plant. Hence, it was expected that the peat filter would have the lowest flow in the plant, and the bark filter the highest.

The particle size distribution (PSD) of the sorption filter materials was determined both before and after their use in the pilot plant. The PSD was determined by wet sieving, according to ISO 11277:2009: for the fractions >2 mm, 1–2 mm, 0.5–1 mm, 0.25–0.50 mm, 0.125–0.25 mm, and 0.063–0.125 mm. The smaller fractions, <0.063 mm, were determined with a laser particle size analyser using liquid dispersion mode.

2.2. Design and operation of the filter pilot plant

The pilot plant was located at the inlet of a stormwater sedimentation pond in Järnbrott, Gothenburg (Sweden), where stormwater is drained from a 480 ha catchment area which includes residential, commercial, and small-scale industrial land use, as well as a highway (annual average daily traffic is approximately 40,000 vehicles). Stormwater was pumped to the plant from a concrete basin containing standing stormwater mixed with groundwater (baseflow <10 L/s).

The pilot plant was designed to minimise maintenance and operational needs, why self-regulating gravity filtration was used. By pumping the stormwater from the concrete basin to an elevated level above the columns, the columns were fed with water through gravity flow (Fig. 1). The stormwater was first filtered through a sand column, then diverted into three parallel lines and divided between the GAC, peat, and bark columns. To enhance the performance of sorption filters, the addition of sand media is recommended. Sand filters are currently being used in combination with retention and wet detention ponds, to support treatment of stormwater surface runoff by reducing the suspended solids load and preventing clogging of stormwater filtering systems (Kumar et al., 2012).

All columns were modified pipes of rigid PVC (h 2200 mm, Ø 300 mm), with a 450 mm drainage layer of graded gravel at the bottom. The graded gravel underdrain was made up of different layers of sand and gravel in sizes, from bottom, of 64–32 mm, 32–16 mm, 25–10 mm, 10–5.0 mm, 5.0–3.0 mm, 3.0–2.0 mm, and 1.2–0.8 mm. The sand filter consisted of a 300 mm deep sand bed of particles in the size range 0.6–0.8 mm, and the bed height of the sorption filters was 600 mm. The height of the bed was calculated to achieve a flow of 2 L/min and a contact time of 20 min, using the empty bed contact time equation (Tchobanoglous et al., 2014). Before the adsorption materials were packed into the columns, the sieved fractions of peat, bark and unsieved GAC were soaked in water for a minimum of 24 h. The filter materials were then poured into the columns and allowed to settle for approximately one week before stormwater was applied. All four columns were designed to allow backwashing, to avoid clogging of the bed. The backwashing pressure was empirically determined to obtain a filter bed expansion of approximately 30%. During winter, the temperatures in the columns were kept above 0 °C.

The design of the pilot plant was proven functional but showed limitations in water flows through the filter media over time. Backwashing of the sand filter was required every 12 h to maintain the flows through the subsequent filters. The flow through the sorption filters declined within days of filter start-up; backwashing was carried out when necessary and depending on the influent water quality. Although backwashing of both sand and sorption filters was executed, the expected flow of 2 L/min through the filters was not maintained. During the first eight months of operation, the bark and carbon beds filtered the largest volumes (approximately 4500 and 4200 L, respectively)

Table 1
Properties of the tested sorption materials. Physical-chemical tests were performed on triplicate samples of the sieved materials (0.5–2 mm).

Sorbent	Granulated activated carbon (GAC)	Bark (*Pinus sylvestris*)	Peat (*Sphagnum*)
Product name, manufacturer	Filtrasorb 400, Calgon Carbon Corporation (USA)	Zugol Miljöskyddsmedel, Zugol AB Svensk Barkindustri (Sweden)	ScanPeat Blocktorv, ScanPeat (Sweden)
Sorbent information (from manufacturer)	For removal of dissolved organic compounds in water; made from bituminous coal	53% dried and granulated pine bark, 15% wood fibre	From Saltmyran (Arvidsjaur, Sweden); raw peat is heated to approx. 350 °C in an anoxic environment, leading to granulation; humification degree H3–4 [a]
Compounds with OP sorption capacity	Graphite (carbon atoms in a hexagonal structure) and carbon-oxygen, —hydrogen, and -nitrogen surface compounds (Bansal and Goyal, 2005)	Cellulose, hemicellulose, phenolic extractives, polysaccharides, lignin, suberin, tannin, terpenoids, resin acids, fatty acids (Valentín et al., 2010)	Lignin, cellulose, humic substances: humic acids, fulvic acids, humin (Ahmaruzzaman, 2008)
Particle size range analysed and used in filters	0.55–0.75 mm	0.5–2 mm	0.5–2 mm
pH [b]	11	3.2	2.6
Water content [c] (%)	Not analysed (usually <5%)	33	56
Loss on ignition (LOI) [d] (%)	87	99	97
Bulk density [e] (kg/m^3)	Not analysed [f]	140	77
BET surface area [g] (m^2/g)	780	0.2	1.3
Hydraulic loading rate [h] (L/min)	Not analysed	26	3.2

[a] According to the von Post scale.
[b] Swedish standard SS 02 81 22–2.
[c] Swedish standard SS-EN 12880.
[d] Swedish standard SS-EN 12879.
[e] International standard EN ISO 17892-2:2014.
[f] Apparent density 0.54 g/cm^3, according to Filtrasorb 400 Data Sheet by Calgon Carbon Corporation.
[g] Determined according to the *Brunauer–Emmett–Teller (BET) theory,* using liquid nitrogen as a saturator in a Micromeritics Tristar instrument.
[h] According to the Falling Head Method by Lewis (2016).

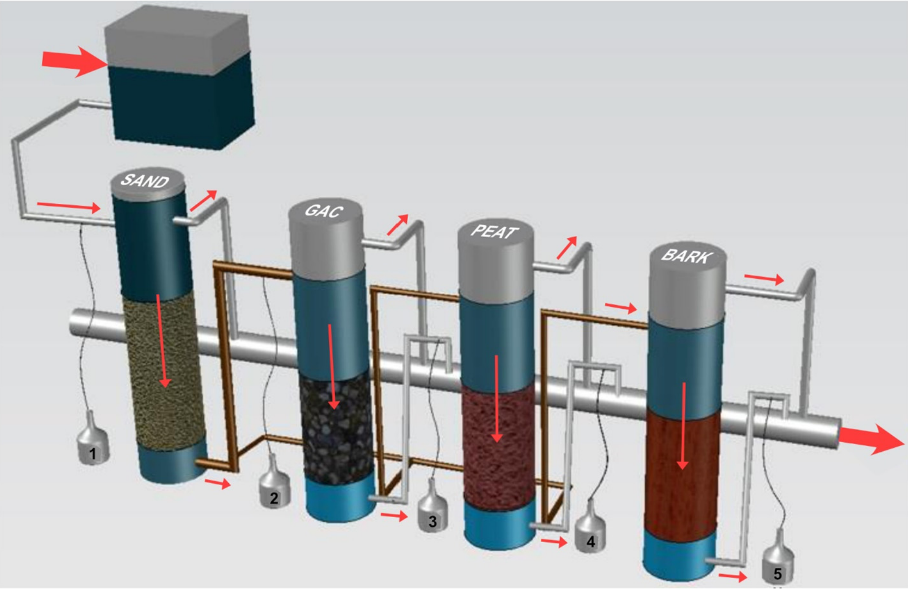

Fig. 1. Design of the stormwater pilot plant with inlet container, sand filter, followed by parallel lines of granulated activated carbon (GAC), peat, and bark filters. The effluent from the sand filter (indicated by brown pipes) is diverted into parallel lines feeding the GAC, peat, and bark columns separately. Water sampling points are labelled 1 to 5.

followed by peat (approximately 3500 L) of contaminated stormwater (Fig. S1, in Supplementary material). These results were expected based on the hydraulic load tests (Table 1), which indicated that the peat filter would have the lowest and the bark filter the highest flow.

2.3. Selection of OPs for chemical analysis

A total of 80 specific OPs had previously been analysed in the stormwater sedimentation pond at Järnbrott. As many as 40 specific OPs within the groups alkylbenzenes, aliphatics, nonylphenols and its ethoxylates, PAHs, phthalates, and several brominated flame retardants were found in concentrations exceeding guideline values (Strömvall et al., 2007). In a screening study of OPs in snow samples from the Järnbrott catchment area, the most frequently quantified OPs were PAHs, high molecular-weight phthalates, 4-nonylphenol, and 4-t-octylphenol (Björklund et al., 2011). Brominated flame retardants and chlorinated paraffins were sporadically quantified. The distribution of PAHs in different fractions in stormwater from Järnbrott has previously been determined (Nielsen et al., 2015); PAHs were found both on small particles (<0.7 μm) and in the colloidal fraction. The OPs selected for chemical analysis in this study were determined based on the aforementioned studies and results from a study where 1100 specific organic compounds emitted from road environments were identified, and the most important pollutants were prioritised, thereafter quantified and suggested for treatment (Markiewicz et al., 2017).

2.4. Collection and analysis of water and solid samples

Water samples were collected by weekly continuous-flow sampling (approximately 45 mL/h) at the inlet and outlet of the sand filter, and at the outlet of the GAC, peat, and bark filters, respectively (Fig. 1). Samples were also collected during specific rain events ($n = 4$) and after stress tests ($n = 9$), where polluted sediments from a stormwater pond, as well as petrol or diesel fuel (both from Preem), were added to the influent. In the stress tests, 20–30 L of sediment, newly extracted from the pond by grab sampling, was added to the inlet container of the plant (Fig. 1) in smaller portions and mixed by continuous stirring over several hours. In a few of the stress tests, approximately 1 L of petrol or diesel was also mixed with the sediment before addition. The purpose of the stress tests was to evaluate the filters' capacity to remove large loads of pollutants over a short time period, as opposed to the continuous-flow weekly sampling, where smaller loads of stormwater pollutants reached the filters.

Water samples were analysed for pH, conductivity, dissolved oxygen, and turbidity (using HANNA Multiparameter HI9829), total and dissolved organic carbon (TOC/DOC) (using Shimadzu TOC-V CPH Total organic analyser), total suspended solids (TSS) (filtration through 1.2 μm glass fibre filters), aliphatic C_8–C_{35} (five fractions) and aromatic C_8–C_{35} (three fractions) petroleum hydrocarbons, and US EPA 16 PAHs. Some weekly water samples were also analysed for 13 specific phthalates. The PAHs were divided into three groups based on their molecular weight: low weight (PAH-L) $= \sum$ (naphthalene + acenaphthylene + acenaphthene); medium weight (PAH-M) $= \sum$ (fluorene + phenanthrene + anthracene + fluoranthene + pyrene); and high weight (PAH-H) $= \sum$ (benzo(a)anthracene + chrysene + benzo(b) fluoranthene + benzo(k)fluoranthene + benzo(a)pyrene + dibenz(a, h)anthracene + benzo(g,h,i)perylene + indeno (1,2,3-c,d)pyrene). Analyses of the OPs were performed at a commercial laboratory by GC/MS, according to methods based on the quality manual from SPIMFAB (the Swedish Petroleum Institute Environmental Remediation Fund AB) (ALS, 2010).

Partitioning of OPs among the total, particle bound (trapped on 0.7 μm glass-fibre filters), colloid bound (passing through the C_{18} solid-phase extraction (SPE) disk), and truly dissolved (adsorbed to the C_{18} SPE disk) fractions was studied in water samples collected during one of the stress tests. This partitioning method is described in detail in Kalmykova et al. (2013). The distribution of micro- and nano-sized particles in total, filtered, and colloidal samples was measured with Dynamic Light Scattering (DLS) and Nanoparticle Tracking Analysis (NTA), using Litesizer™ 500 (Anton Paar Ltd., Austria) and NanoSight NS300 (Malvern Instruments Ltd., UK), respectively. The analytical techniques and instrumental settings are described in detail in Polukarova et al. (2020).

After 18 months of plant operation, the filter beds were replaced and portions of the upper 0–10 cm and the centre (approximately 10–35 cm from the top of the bed) of the used filter materials were analysed for OP concentrations, including aliphatic C_8–C_{35} and aromatic C_8–C_{35} hydrocarbons, US EPA 16 PAHs (all determined on GC/MS according to SPIMFAB's quality manual), specific oxygenated PAHs (using GC/MS), and phthalates (according to DIN 19742:2012–04). The PSD was also determined for the used sorption materials. The upper part of the used GAC filter material was also analysed through GC/MS screening, using a spectral deconvolution-based metabolomics method that automatically subtracts the background in the compound mass spectrum, making the fit with the NIST-library more accurate (Du and Zeisel, 2013). Only GAC was analysed, as naturally occurring organic compounds in peat and bark are released during solvent extraction, which inhibits identification of pollutants occurring at low concentrations. For more details on the GC/MS screening method, see Supplementary material page 2.

The structure of new and used filter materials was visualised using the scanning electron microscope (SEM) FEI ESEM Quanta 200. With SEM, secondary electrons are produced by incident of the primary electron from a surface of the sample, using a low-energy beam of electrons (1–30 keV), which creates an image with exceptional depth of field and magnification down to 1–3 nm (Dudkiewicz et al., 2011; Mohammadi et al., 2014). For more details on the SEM method, see Supplementary material page 2.

3. Results and discussion

3.1. Influent and effluent quality – general water quality parameters

Among all general water quality parameters, only TSS and turbidity were found in significantly higher concentrations in the sand filter influent (Kruskal-Wallis rank-based nonparametric test: TSS $\chi^2(4) = 45.635$, $p = .000$; turbidity $\chi^2(4) = 25.898$, $p = .000$), compared to the other filters, and were highest during the stress tests with added sediment; see maximum values in Table 2. On average, 72% of the influent TSS was removed in the sand prefiltration. The effluent from the sand filter was further treated in the sorption filters, and here the TSS removal was further removed by 71% for GAC > 67% for bark >50% for peat (Fig. S2, Supplementary material). Peat has the lowest bulk density (Table 1) and its lower TSS removal efficiency may be due to the release of bed materials or a sign of channelling. Other studies of TSS removal from stormwater using sand filters have shown similar removal percentages, ranging from 52% to 100%, although higher TSS influent concentrations (150–250 mg/L) have been tested (Hatt et al., 2008; Kumar et al., 2012). As the removal efficiency increases with increased sand media depth and reduced grain size, the TSS attenuation can be improved by optimising these parameters. Negative removal of TSS was recorded for all four filters in some of the weekly samples. However, particle release did not appear during the stress tests or rain events, which suggests that the filters functioned properly, even during heavy particle loads.

The TOC and DOC concentrations were highest in the weekly samples for all the filters, followed by samples from the stress tests and rain events. In samples from all filters, the TOC and DOC concentrations were very similar; on average, 93 to 100% of the TOC found were in dissolved form. Generally, only the sand filter was able to remove TOC and DOC and peat TOC (Fig. S2, Supplementary material), whereas GAC, peat and bark filters appeared to release DOC, and therefore the median removal efficiency was negative. This is alarming, because a loss of organic colloids could mean a loss of OPs, which bind to the colloids. This transport process was demonstrated in the leaching of metals from peat filters, where the presence of humic material and iron colloids led to colloid-facilitated transport of metals from peat filters (Kalmykova et al., 2010). Loss of DOC from peat and bark filters in the treatment of contaminated waters in laboratory tests has been reported in other studies, which also showed that leaching of DOC decreased over time (Kalmykova et al., 2009; Ribé et al., 2009). The continuous leaching of DOC in the current study may be caused by the decomposition of the peat and bark materials into smaller constituents, which is promoted by oxidation, higher temperatures, and a neutral to basic pH (Kalmykova et al., 2009). Microbial degradation may have taken place in the column beds, as the water quality indicated oxic conditions, neutral pH (Table 1), and stable temperatures as the filters were kept in a heated pilot plant. In contrary to the results found in this study, 87% of the DOC was removed by a micropeat mixture (<150 μm), which, however, can be explained by the ~10 times higher concentrations (Rosli et al., 2019). For drinking water treatment, a continuous and partial renewal of the GAC material is proposed to prevent leaching of DOC from GAC filters (Moona et al., 2018).

3.2. Removal of organic pollutants

3.2.1. Concentrations of OPs in water samples collected weekly, during or after rain events, and during stress tests

The concentrations of OPs collected during the different sampling events are presented in Table 3. Throughout the duration of the experiment, the weekly effluents from the peat and bark filters were more polluted than the influent stormwater entering the sand filter. This may seem strange but could be explained by leaching of already sorbed pollutants from the filter materials, and correlates well with the TOC and DOC leaching from the filters (Fig. S2, Supplementary material). Phthalates were analysed in four of the weekly samples but were not quantified in any of the samples, hence not discussed further.

The occurrence of PAHs, aliphatics, and aromatics in the influent may be due to their frequent occurrence in urban runoff (Kayhanian et al., 2007; Markiewicz et al., 2017) and their occurrence in polluted pond sediment (Strömvall et al., 2007), although the main source is assumed to be diesel and petrol fuels, which were applied to the pilot plant during stress tests. Diesel contains approximately 80% aliphatic compounds, mainly C_9-C_{23}, and 20% aromatics, of which <1% are BTEX compounds (Brewer et al., 2013; Fingas, 2016). Further, diesel contains mainly PAH-L and -M, but lower levels of PAH-H (de Souza and Corrêa, 2016). Petrol contains mainly alkanes, C_3-C_{13} and BTEX. The relationships of hydrocarbons in petrol and diesel are reflected in the abundance of aliphatics, aromatics, and PAHs in influent water samples (Table 3).

Among the aliphatics and aromatics, the highest concentrations quantified were related to petrol and diesel aliphatics C_5-C_{16} in the effluent from most of the sorption filters during all events (i.e. weekly, heavy rain, and stress tests). For all events, the bark filter released the highest concentrations of aliphatics despite the high concentrations found in the used filter material (Section 3.3.1) and it was therefore

Table 2
Minimum to maximum (median) values of general water quality parameters measured in weekly samples, rain event samples, and stress tests of the stormwater filter pilot plant.

Parameter	pH	Conductivity	Dissolved oxygen	Redox potential	Turbidity	TSS	TOC	DOC
Unit		[μS/cm]	[mg/L]	[mV]	[FNU]	[mg/L]	[mg/L]	[mg/L]
n	29	31	16	28	10	30	24	24
Influent (to sand filter)	6.5–8.2 (7.5)	310–2400 (520)	4.5–9.6 (6.8)	140–300 (230)	23–740 (160)	0.40–1100 (17)	7.8–830 (230)	6.8–890 (190)
Sand filter effluent (= inlet to GAC, Peat, Bark)	7.0–8.2 (7.6)	220–2600 (580)	1.0–9.7 (6.1)	120–320 (300)	6.2–73 (26)	0.40–66 (4.8)	6.5–890 (170)	6.0–720 (92)
GAC effluent	7.2–9.7 (7.7)	170–2700 (560)	3.3–10 (6.3)	100–330 (220)	2.8–52 (12)	<q.l.[a]-20 (1.4)	1.3–810 (230)	1.6–800 (240)
Peat effluent	7.0–8.0 (7.5)	290–1500 (550)	2.5–13 (6.3)	110–300 (220)	0.40–32 (12)	<q.l.-33 (2.4)	7.1–710 (120)	7.2–790 (140)
Bark effluent	7.0–8.8 (7.6)	220–2300 (570)	2.1–10 (6.7)	8–300 (220)	4.1–52 (15)	<q.l.-15 (1.6)	9.5–930 (240)	8.9–920 (240)

[a] Below the limit of quantification.

Table 3

Minimum to maximum (median) values of OPs measured in weekly samples, rain event samples, and stress tests of the stormwater filter pilot plant. Where no median is given, the value is <q.l.

OPs analysed in samples collected weekly						
Compounds [μg/L]	n_quantififed (S_{in}[a], S_{ut}[b] GAC, Peat, Bark)[c]	Sand filter influent	Sand filter effluent	GAC effluent	Peat effluent	Bark effluent
n_measurements[d]		5	5	5	5	5
Aliphatics C_5-C_{16}	2,2,2,2,2	<q.l.[e]-280	<q.l.-280	<q.l.-240	<q.l.-310	<q.l.-720
Aliphatics C_{16}-C_{35}	2,0,1,0,0	<q.l.-13	<q.l.	<q.l.-10	<q.l.	<q.l.
Aromatics C_8-C_{10}	0,0,0,0,1	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.-0.06
PAH-16	4,1,3,4,1	<q.l.-0.1 (0.01)	<q.l.-0.1	<q.l.-0.08 (0.03)	<q.l.-0.03 (0.01)	<q.l.-0.08
Total PAH-L	3,0,3,4,1	<q.l.-0.1 (0.01)	<q.l.	<q.l.-0.08 (0.03)	<q.l.-0.03 (0.01)	<q.l.-0.08
Total PAH-M	1,1,0,0,0	<q.l.-0.01	<q.l.-0.1	<q.l.	<q.l.	<q.l.
Total PAH-H	0,0,0,0,0	<q.l.	<q.l.	<q.l.	<q.l.	<q.l.
OPs analysed in samples collected during or after rain events						
n_measurements[d]		4	4	2	2	2
Aliphatics C_5-C_{16}	1,0,1,1,1	<q.l.- 110	<q.l.	<q.l.-110 (55)	<q.l.-10 (5)	<q.l.-990 (500)
Aliphatics C_{16}-C_{35}	3,0,0,0,0	<q.l.-290 (23)	<q.l.	<q.l.	<q.l.	<q.l.
Aromatics C_8-C_{10}	0,1,0,0,0	<q.l.	<q.l.-0.2	<q.l.	<q.l.	<q.l.
Aromatics C_{10}-C_{16}	0,0,0,1,0	<q.l.	<q.l.	<q.l.	<q.l.-0.13 (0.065)	<q.l.
PAH-16	4,4,0,0,1	0.03–0.4 (0.065)	0.02–0.04 (0.03)	<q.l.	<q.l.	<q.l.-0.02 (0.01)
Total PAH-L	3,4,0,0,1	<d.l.-0.06 (0.04)	0.02–0.04 (0.03)	<q.l.	<q.l.	<q.l.-0.02 (0.01)
Total PAH-M	3,0,0,0,0	<q.l.-0.2 (0.025)	<q.l.	<q.l.	<q.l.	<q.l.
Total PAH-H	1,0,0,0,0	<q.l.-0.16	<q.l.	<q.l.	<q.l.	<q.l.
BTEX	2,1,0,0,0	<q.l.-0.5 (0.14)	<q.l.-0.2	<q.l.	<q.l.	<q.l.
OPs analysed in samples collected during or after stress tests						
n_measurements[d]		9	9	8	8	8
Aliphatics C_5-C_{16}	5,3,3,3,3	<q.l.- 480,000 (0.07)	<q.l.-37,000	<q.l.-2700	<q.l.-5550	<q.l.-9400
Aliphatics C_{10}-C_{12}	3,0,0,0,0	<q.l.- 31	<q.l.	<q.l.	<q.l.	<q.l.
Aliphatics C_{12}-C_{16}	5,1,0,0,0	<q.l.- 80 (0.03)	<q.l.-120	<q.l.	<q.l.	<q.l.
Aliphatics C_{16}-C_{35}	6,3,1,0,0	<q.l.-840 (2.9)	<q.l.-350	<q.l.-11	<q.l.	<q.l.
Aromatics C_8-C_{10}	3,2,0,0,0	<q.l.-1.8	<q.l.-2.9	<q.l.	<q.l.	<q.l.
Aromatics C_{10}-C_{16}	1,0,0,0,0	<q.l.-0.8	<q.l.	<q.l.	<q.l.	<q.l.
PAH-16	8,4,1,3,3	<q.l.-3.6 (1.1)	<q.l.-0.08	<q.l.-0.02	<q.l.-0.03	<q.l.-0.02
Total PAH-L	7,5,1,3,3	<q.l.-3.6 (0.02)	<q.l.-3.6 (0.02)	<q.l.-0.02	<q.l.-0.03	<q.l.-0.02
Total PAH-M	5,0,0,0,0	<q.l.-0.8 (0.4)	<q.l.	<q.l.	<q.l.	<q.l.
Total-PAH-H	4,0,0,0,0	<q.l.-0.9	<q.l.	<q.l.	<q.l.	<q.l.
BTEX	1,2,0,0,0	<q.l.-2.1	<q.l.-2.1	<q.l.	<q.l.	<q.l.

[a] Sand influent.

[b] Sand effluent.

[c] Quantification frequency of OPs in each of the consecutive filters.

[d] No. of measurements of OPs performed for each consecutive sample.

[e] Below the limit of quantification.

assumed to have a good removal efficiency (Fig. 2). Concentrations of aromatics C_8-C_{16} were low (0.06–2.9 μg/L) and close to quantification limits in samples from all events. However, aromatics C_8-C_{35} were quantified in high concentrations in the used sorption filters (1.1–190 mg/kg DS), and in the highest concentrations in the top part of the bark filter (section 3.3.1).

Weekly sampling

In the samples collected weekly, the highest quantification frequency was observed for PAHs, of which low-molecular PAH-L was quantified in the highest concentrations. Naphthalene leached from the GAC filter in two of the weekly samples, but the concentrations were very low, 0.023 μg/L, and did not exceed the European Environmental Quality Standard (EQS) for receiving surface waters, which is 2.4 μg/L. Fluorene, phenanthrene, and fluoranthene (i.e. PAH-M) were quantified only in the effluent from the sand filter. The low sorption of PAH-M by the sand filter may be because these compounds are present in dissolved form, or because of binding to smaller particles and colloids that are not retained in the sand filter (Kalmykova et al., 2014; Nielsen et al., 2015). It is also possible that the PAH-M may be transported through the sand filter as emulsions formed by all the OPs present in polluted stormwater (Markiewicz et al., 2019). The PAH-H could not be quantified in any of the weekly water samples, as these often occur in low concentrations in stormwater due to their high partition to particles on the road surface (Polukarova et al., 2020). Further for the weekly samples, petrol and diesel aliphatics C_5-C_{16} (<q.l.-720 μg/L) were quantified in the highest concentrations, and the effluent from the bark filter showed the highest concentrations of these pollutants. Aromatics originating from petrol C_8-C_{10} (<q.l.-0.06 μg/L) were quantified only once, in the bark effluent.

Rain events

Aliphatics from diesel C_{16}-C_{35} (<q.l.-290 μg/L), PAH-L (<q.l.-0.06 μg/L), and PAH-M (<q.l.-0.2 μg/L) were quantified most frequently and in the highest concentrations in the sand influent samples collected during or after rain events (Table 3). Lower molecular weight aliphatics from petrol and diesel, C_5-C_{16} (<q.l.- 990 μg/L), were quantified in the bark filter effluent, at concentrations 10–100 times higher than in the samples from other sampling points. The peat filter also leached lighter OPs (i.e. aliphatics C_5-C_{16} and aromatics C_{10}-C_{16}), although at lower magnitudes than the bark filter. The higher leaching during rain events than in weekly samples could be due to the stress put on the filter capacity when treating water with higher incoming concentrations, which may lead to competition for active sites on the sorbents of the aliphatics and PAHs. During rain events, naphthalene was quantified both in the influent and in the effluents of the sand and bark filters. Naphthalene leached from bark on one sampling occasion. Most of the specific PAH-M and —H were quantified in the effluents during rain events, but only quantified in the influent on one occasion. The higher leaching of aliphatics, aromatics, and PAH-M during rain events with higher

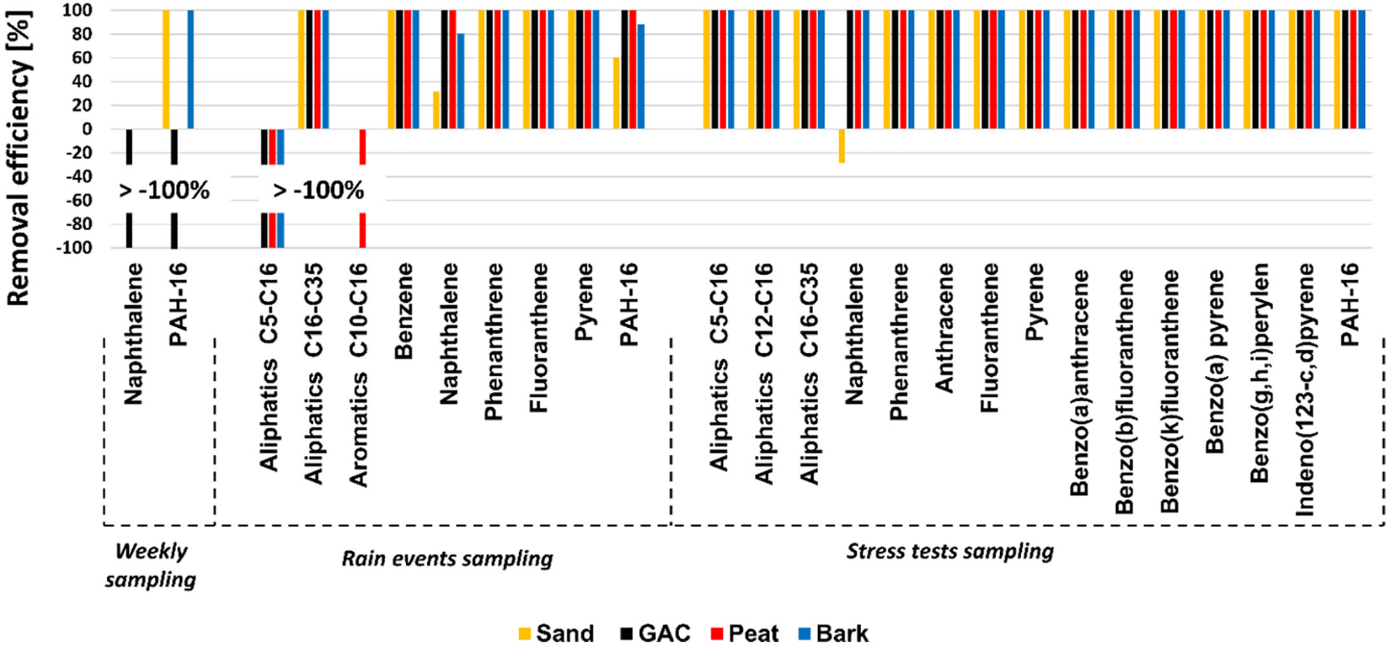

Fig. 2. Average removal percentage of aliphatics, aromatics, total PAH-16, and specific PAHs by the GAC, peat, and bark filters, in weekly samples, and during rain events and stress tests. NB! The removal efficiencies have been calculated from median concentrations.

concentrations of pollutants indicates competition for active sites in the sorbent materials. BTEX was quantified twice in the influent, in concentrations of 0.28–0.51 μg/L, and was completely removed by all sorption filters.

Stress tests

The samples collected during the stress tests showed the highest quantification frequency of different groups of aliphatics, aromatics, and PAHs (Table 3). However, the results show that the concentrations of OPs were not high in the effluents from the filters, except in relation to petrol and diesel aliphatics C_5-C_{16}. The maximum concentrations of aliphatics C_5-C_{16} in the effluents were 37,000 μg/L for sand, 2700 μg/L for GAC, 5500 μg/L for peat, and 9400 μg/L for bark, indicating a lower removal capacity for lower molecular weight aliphatics during periods of heavy pollutant loads. The diesel aliphatics C_{16}-C_{35} (11–840 μg/L) and C_{12}-C_{16} (80–120 μg/L) were found in the highest concentrations in the sand influent, and in effluents from the sand and GAC filters. PAH-H was quantified 4–5 times in the sand influent, in concentrations exceeding the EQS, but was successfully removed by all filters. Naphthalene was quantified at all sampling points. Pyrene was quantified in high concentrations in the spiked sand influent, 0.14–3.0 μg/L, however up to 100% of the pyrene was removed by all sorption filters. BTEX was quantified once in the influent water but twice in the effluent from the sand filter, indicating the release of pollutants that are relatively more water soluble than the other OPs measured in this study (Brewer et al., 2013; Heath et al., 1993). BTEX was efficiently removed by all sorption filters and was quantified on the used sorption materials, and in high concentrations in the GAC filter (Table 4).

Removal efficiency of OPs by the filters

In most **weekly samples** from influent water to the sand filter, concentrations of PAHs, aliphatics and aromatics were below the quantification limit. Samples collected during heavy rain events and stress tests showed high enough pollutant concentrations in the influent to allow calculation of the removal efficiency (Fig. 2).

Naphthalene was the only specific compound quantified frequently enough in the **weekly samples** to calculate the removal efficiencies, and leached from the GAC filter, resulting in negative removal efficiency (Fig. 2). PAH-16 was efficiently removed by the sand and bark filters, but the results for the peat showed a removal rate of 0%, and for GAC a release of 100% for the weekly samples. This is in contrast to the result from a study of activated carbon with a removal rate of 73–95% of PAHs, which can be explained by the much higher PAH concentrations used in the batch test (Lamichhane et al., 2016). The high PAH removal rate for bark was confirmed in a batch test in which approximately 80% PAH-M was removed within 30 min (Björklund and Li, 2015). The results from the **rain events** showed excellent (100%) removal efficiencies by all filters for diesel aliphatics C_{16}-C_{35}, benzene, phenanthrene, fluoranthene, and pyrene. However, during rain events the removal rate for naphthalene was only 32% by the sand filter and 80% by the bark filter, and were more efficiently sorbed in the peat and GAC filters. During rain events, concentrations of aliphatics C_5-C_{16} also increased after passing the sorption filters, and concentrations of aromatics C_{10}-C_{16} increased after passing the peat filter, indicating release of these pollutants from the filters. This may be caused by the competition for active sites in the sorbents, where lower molecular and relatively more water-soluble compounds are released first. During the **stress tests**, the detection frequencies were high for many of the OPs, and the removal efficiency was 100% for all filters, except in the case of naphthalene, which was less efficiently sorbed in the sand filter (Fig. 2).

Table 4
Loss on ignition (%) and quantified concentrations (mg/kg dry substance) of organic pollutants in the upper 0–10 cm and centre 10–35 cm of the bed of used filter materials. Full details on analysed compounds are provided in Table S2, Supplementary material.

Compound [mg/kg DS]	GAC top	GAC centre	Bark top	Bark centre	Peat top	Peat centre
Loss on ignition	68	82	78	75	79	80
Phthalates						
Dimethyl phthalate	0.69	n.a. [a]	<q.l. [b]	n.a.	<q.l.	n.a.
Di-(2-ethylhexyl) phthalate	0.25	n.a.	0.89	n.a.	1.4	n.a.
Aliphatic and aromatic hydrocarbons						
Aliphatics C_5–C_{16}	72	75	<q.l.	<q.l.	<q.l.	<q.l.
Aliphatics C_{16}–C_{35}	180	260	6600	250	1100	1200
Aromatics C_8–C_{10}	31	53	190	10	2.7	4.0
Aromatics C_{10}–C_{35}	1.1	<q.l.	3.1	<q.l.	<q.l.	<q.l.
BTEX						
Benzene	<q.l.	0.17	<q.l.	<q.l.	<q.l.	<q.l.
Toluene	<q.l.	0.81	<q.l.	0.097	<q.l.	0.051
Ethylbenzene	<q.l.	0.11	<q.l.	<q.l.	<q.l.	<q.l.
m,p-xylene	0.056	0.75	<q.l.	0.130	<q.l.	0.085
o-xylene	<q.l.	0.22	<q.l.	0.063	<q.l.	<q.l.
PAHs						
PAH-16	1.6	0.7	1.3	0.44	0.56	0.38
Total L-PAHs	0.80	0.57	0.74	0.15	0.14	0.092
Total M-PAHs	0.85	0.15	0.52	0.29	0.35	0.29
Total H-PAHs	<q.l.	<q.l.	0.074	<q.l.	0.085	<q.l.

[a] Not analysed.

[b] Below the limit of quantification.

3.2.2. Partitioning of OPs and PSDs in samples from a stress test

The effluent samples from one stress test (May 2017, 20 L sediment and 1 L diesel added to pilot plant influent) were subjected to partitioning studies and PSDs analysis. The results from the Nanosight (Fig. 4.) show that the influent contained a substantial number of nanoparticles in the size range 50–400 nm, with a mean diameter (MD) of 190 nm and average concentration (AC) of 2.1×10^8 particles/mL. The concentration of nanoparticles was reduced by one order of magnitude after passing the sand filter, to 3.3×10^7 particles/mL. Additionally, the MD increased to 240 nm, which may be due to the release of bigger particles from the sand filter. The ACs and MDs in effluents from GAC, peat, and bark filters were 1.6×10^7 particles/mL and 150 nm; 2.5×10^7 particles/mL and 210 nm; and 1.8×10^7 particles/mL and 290 nm, respectively. All sorption filters removed nanoparticles: GAC by 51% > bark by 46% > peat by 22%. The peat filter was least effective, suggesting the possible co-release of attached OPs, as the release of some aliphatics also increased after passing the peat filter subsequent to the rain event (Fig. 2). As seen in Fig. 3, all the sorption filters sorbed fractions of nanoparticles released from the sand filter, but also released nanoparticles in other size fractions.

Results from the Litesizer (Fig. 4a) show that the MD for particles in the sand influent was 120 nm, and the MD in the effluent was 80 nm. The MD in the sorption filter effluent for GAC was 0.5 nm, and the MDs for the peat and bark formed bimodal distribution peaks at 18 and 100 nm, and at 84 and 690 nm, respectively. The results from the different instruments are not comparable due to different settings (e.g. laser wavelength and sample volume), the parameters of the measurements, and because the results are given in different units (particle count/volume vs volume weight), although both instruments can confirm the occurrence of agglomerates. Peat and bark filters released larger nanoparticles than those present in the effluent from the sand filter, which shows a possible escape of the coarser fraction of the materials (Fig. 4a). Contrarily, the results for the GAC filter show that very small nanoparticles were released from the filter component. Filters consisting of finer particle sizes are assumed to achieve more effective adsorption, as the surface area increases with the smaller particle size. The hydraulic loads of such filters are, however, reduced and if installed in locations where surface runoff must be filtrated promptly to avoid flooding, a coarser filter would be a more obvious choice (Monrabal-Martinez et al., 2017).

All the investigated filters released nanoparticles; therefore, it is important to investigate in which phases the particles are likely to emerge. Fig. 4b shows that particles in the filtered samples were 0.1–10 μm in size, except in the GAC effluent where 82% of the particles were smaller

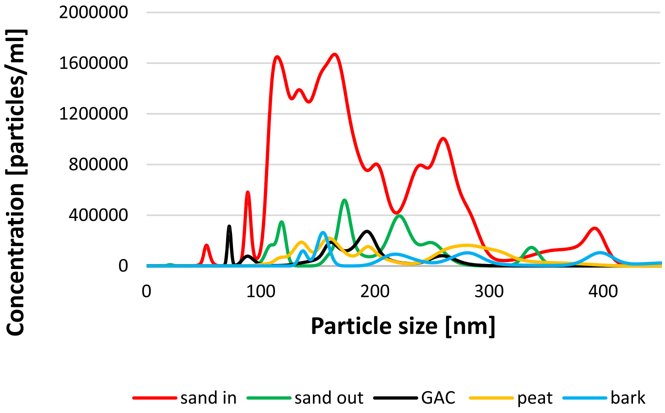

Fig. 3. Nanoparticle concentrations and size distributions (10–600 nm), measured with Nanosight on influent stormwater and effluents from sand, GAC, peat, and bark filters.

than 0.7 μm. In the sand influent and the bark filter effluent, only 2% of the particles were smaller than 0.7 μm. In the effluents from the sand and peat filters, the corresponding values were 14 and 31%, respectively. The occurrence of particles larger than 0.7 μm in the filtered samples could be explained by agglomeration of particles over time: this may occur as a result of low repulsion forces between particles.

Particles in the colloidal fraction of the influent, and in the effluents of the sand and bark filters, were 0.2–5 μm. The corresponding figure for the GAC and peat filter effluents was 0.2–1.4 μm.

In the phase separation, emulsions of OPs that may occur as particles in nm (Markiewicz et al., 2019) were trapped in the C_{18} sorbent used. Left in the filtrate from the C_{18} was the colloidal fraction extracted from the filtered phase. The particles in the colloidal fraction should be smaller, and this was observed with the Litesizer, but not in all samples analysed with the Nanosight (Fig. S3, Supplementary material). The peak around 0.1 μm in the peat filtered sample may be an emulsion that was trapped in the C_{18} sorbent as it disappeared in the colloidal fraction.

In the filtrated fractions of the influent, and of the effluents from the sand and peat filters, the ACs of nanoparticles were, as expected, higher than in the colloidal fractions (Fig. S3, Supplementary material). However, for the GAC and bark filters, the ACs in the colloidal fraction was one order of magnitude higher than in the filtrated fractions. One reason for this may be higher repulsion forces between particles in the effluent of GAC and bark, leading to less agglomeration than in the influent. The highest AC of nanoparticles were found in the colloidal fraction of GAC (2.4×10^8 particles/mL). For peat, the highest ACs were found in the filtrated (1.2×10^8 particles/mL) and colloid ($1.1 \times 10^{\mathbf{8}}$ particles/mL) fractions. These concentrations were one order of magnitude higher than those in the influent and effluent of the sand filter for both the filtrated (influent 7.1×10^7 and effluent 6.7×10^7 particles/mL) and colloidal fractions (influent 6.3×10^7 and effluent 2.4×10^7 particles/mL), which clearly shows that nanoparticles were released from the GAC and peat filters. The MDs for all samples were approximately equal for the filtrated (230–290 nm) and colloidal (260–290 nm) fractions, except for bark, which had a higher MD of 320 nm, indicating the release of slightly larger nanoparticles from the bark, which may be due to decomposition of the material. The results also show relatively high concentrations of BTEX, aliphatics C_5-C_{12}, aromatics C_8-C_{10}, and PAH-L in the truly dissolved and emulsion fraction, which may be due to the fact that these compounds are relatively more water soluble than the other groups of OPs analysed, see Table S1 in Supplementary material.

3.3. Evaluation of used filter materials

3.3.1. Organic pollutants in filter bed materials

Aliphatics and aromatics were quantified in all filter materials; the highest concentrations were found for long chain diesel aliphatics C_{16}–C_{35} (180–6600 mg/kg DS), followed by short chain petrol aromatics C_8–C_{10} (2.7–190 mg/kg DS) (Table 4). Among the PAHs, the -L and -M compounds were frequently quantified, and naphthalene, the lightest among the PAH-16 with only two rings, was found in the highest concentrations (0.092–0.69 mg/kg DS) in all samples (Table S2, Supplementary material). Like for the influent water samples, the quantified concentrations of PAH, aliphatics, and aromatics in the filter materials (Table 4) reflect the abundance of these compound groups in diesel (Brewer et al., 2013; Fingas, 2016).

Compounds with concentrations >q.l. are shown in Table 4; additional data are provided in Table S2, Supplementary material. Some OPs (phthalates, oxygenated PAHs) were not analysed in the centre section of the used filter bed materials due to low quantification frequency. The particle size distributions of the unused and used materials are presented in Fig. S4, Supplementary material.

The tops of the filter beds were exposed to influent water first, and therefore expected to be more contaminated than the lower sections of the beds. However, the quantification frequencies of the analysed OPs were fairly similar in the top and centre fractions of the GAC, bark, and peat beds (36% top, 47% centre; 38% top, 34% centre; 36%

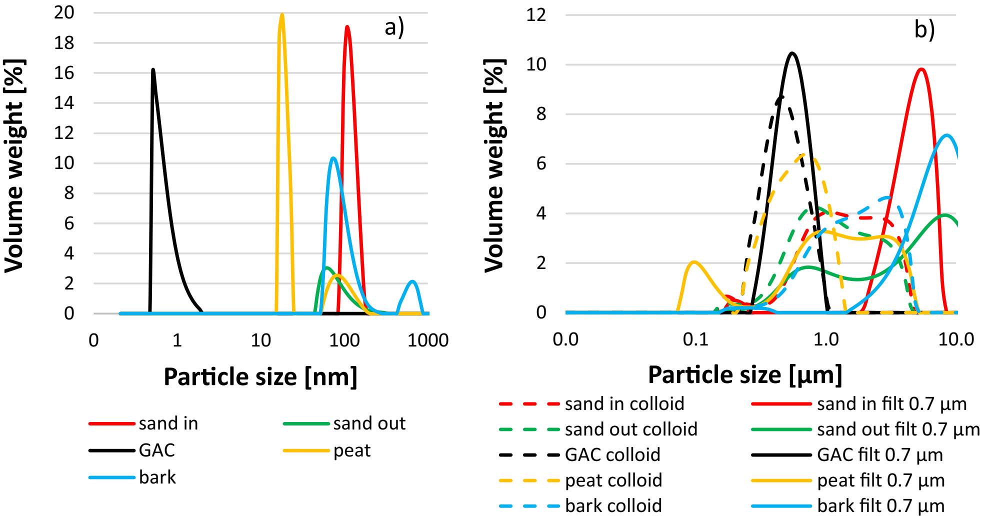

Fig. 4. Nanoparticle size distributions (0.3 nm–10 μm) measured with Litesizer for a) influent, as well as effluents from sand, GAC, peat, and bark filters and b) in filtered (0.7 μm) and colloidal phases in influent and effluents from sand and sorption filters collected during one stress test event.

top, 31% centre, respectively). The monocyclic aromatic BTEX compounds exhibit high water solubility (e.g. 1780 and 175 mg/L for benzene C_6 and o-xylene C_8, respectively) compared to their aliphatic counterparts (approximately 10 mg/L for C_5-C_8 aliphatics) (Brewer et al., 2013; Heath et al., 1993). The water solubility of both aromatic and aliphatic hydrocarbons decreases with an increased number of carbons (approximately 5–50 mg/L for aromatics C_8-C_{35}; 3.5×10^{-4} and 1.5×10^{-6} mg/L for C_{12}-C_{16} and C_{16}-C_{35} aliphatics, respectively). The same trend in water solubility is seen among PAHs (31 mg/L for 2-ring naphthalene, 0.1 mg/L for 4-ring pyrene, 0.004 mg/L for 6-ring benzo(a)pyrene) (Mackay and Shiu, 1977). The BTEX compounds are mainly hydrophilic ($\log K_{ow} < 3.2$), whereas other studied hydrocarbons are hydrophobic ($\log K_{ow}$ approximately 6–11 for $>C_8$-C_{35} aliphatics; 4–6 for $>C_8$-C_{35} aromatics; 3.3–7 for PAH-16) (Brewer et al., 2013; Heath et al., 1993; Mackay and Shiu, 1977). It can be assumed that the highly hydrophobic compounds (aliphatics, larger aromatics, PAH-M and —H) partition to particles in the influent stormwater, and/or are easily sorbed to the filter media, hence are removed in the top layer of the filter bed (Table 4). The BTEX and lighter aromatic compounds, on the other hand, are dissolved and tend to migrate through the column beds, as seen in Table 4.

Generally, higher concentrations of BTEX, aromatics, and PAH-16 were quantified in the GAC than in the bark and peat. Aliphatics were analysed in the highest concentration in bark, followed by peat, whereas higher concentrations of PAH-L were quantified in peat, followed by bark, then GAC (Table S2, Supplementary material). Although GAC is generally hydrophobic (Gonçalves et al., 2010; Mohammad-Khah and Ansari, 2009), the material exhibits higher concentrations of hydrophilic BTEX compounds, and lower concentrations of the hydrophobic aliphatic and aromatic hydrocarbons than peat and bark. In a previous study, where GAC and peat filters were used for treatment of landfill leachate, similar results were found: the more volatile and water soluble organic pollutants were effectively sorbed by GAC and the more hydrophobic pollutants were more effectively sorbed by peat (Kalmykova et al., 2014). The lignin of pine bark is full of aromatic groups that may attract hydrophobic compounds, but bark also contains hydrophilic surface groups that may attract hydrophilic compounds (Valentín et al., 2010). Peat also contains both hydrophilic and hydrophobic surface groups (Rezanezhad et al., 2016). The recommendation is therefore to use a combination of the studied sorption materials in stormwater filters connected in series to achieve effective removal of OPs with different molecular size, volatility, and water solubility, where GAC would sorb the small and relatively more water soluble compounds, bark the compounds in between, and peat the more high-molecular weight and lipophilic OPs. The suggestion is to place the sorption filters in a series, starting with prefiltration using a sand filter to remove TSS, DOC and nanoparticles, followed by a GAC filter that removes the most volatile hydrocarbons, then bark and finally peat for removal of the high-molecular weight OPs. The percentage of the OPs removed by each individual filter is expected to alter when several filters are connected in series, as the removal is often dependent on influent concentrations of pollutants (Björklund and Li, 2015; Tóth et al., 2012).

Approximately 100 specific organic compounds were quantified, in concentrations from 2.9 till 12,000 μg/kg, in the GAC material analysed using the GC/MS screening method. The dominant compounds in the sample were methylated monoaromatic hydrocarbons, sulfoxides, and both saturated and unsaturated aliphatic hydrocarbons. Traces of methylated polycyclic aromatic hydrocarbons and phthalates were also detected in the sample. The following compounds occurred in the highest concentrations from 12,000 till 500 μg/kg and with a NIST fit % >80: toluene > heptadecyl 3-chloropropanoate > p-xylene > cyclohexylmethyl pentadecyl sulfite > cyclohexylmethyl 2-methylpropyl sulfite > cyclohexylmethyl tetradecyl sulfite > ethylbenzene > pentyl (9E)-octadec-9-enoate > o-xylene. Toluene, o-, p-xylene, and ethylbenzene, together with benzene, are major components in petrol (Fingas, 2016), however they are also emitted from the exhausts of petrol-fuelled cars (EPA, 2006). Therefore, the main source for the GAC material was the stress test where petrol was added, showing the great capability of GAC to retain the more volatile aromatic hydrocarbons. The sources of heptadecyl 3-chloropopanoate and the cyclohexylmethyl alkyl sulfites are unknown, but the pentyl (9E)-9-octadecenoate (amyl elaidate) may be a fatty acid alkyl ester added to the diesel (Ribeiro et al., 2007) used in the stress test.

3.3.2. Electron microscope analyses

The structure of the sorption filter materials, before use and after 18 months of use in the pilot plant, are presented in Fig. 5. The objective of the SEM analyses was to observe the change in sorption capacity and the degree of deterioration of the material after long, continuous usage of the filters. The scale for each image is different and selected to achieve the best observable changes in the structure.

The adsorption capacity of GAC was found to be high, due to its large surface area, high pore volume, and porosity. In Fig. 5a, the microspores of unused GAC are clearly visible, and the image of the material after use (Fig. 5b) shows an attached particle presenting adsorption mechanism. Fig. 5b also shows that the pores in the used GAC material are much smaller and shallower than those found on the new material. This can occur as GAC is a polydispersed material (Fig. S5, Supplementary material) but is probably also depending on that the material surface has been covered with TSS, i.e. particles of different sizes >1.2 μm. The x-ray spectroscopy provided the composition of the pure GAC, where the elements C, Ca, and K were most abundant (Fig. S5, Supplementary material). After use, the major components of the material were the metals Ti and Mn, probably in the form of oxides, as the oxygen content also increased significantly (Fig. S5, Supplementary material). The C, Ca, and K content remained high and Si appeared to be a new major element after use. As in the case of Mn, this can be explained by the occurrence of small sand and clay particles that managed to pass the sand prefilter. The occurrence of Ti was unexpected, but may be explained by road paint wear releasing titanium dioxide, which is used as white pigment in road paint (Fatemi et al., 2006; Karlsson et al., 2019)

Pine bark is a material with low porosity, and with few pores categorised as macropores. For this reason, bark has a small specific surface area. Fig. 5c shows one of the porous parts of the bark, and the picture matches previously published pictures of pores in pine bark very well (Holmberg and Stenström, 2014). As much as 2–5% of the dry mass of bark relates to inorganic materials, and if the bark is combusted the resulting ash contains several metal oxides, including SiO_2, Al_2O_3, and CaO (Fedje et al., 2015). Data from the EDX elemental analysis showed that C, K, Ca, and O were the most common elements (Fig. S5, Supplementary material). After use, the bark filter material had a much more diverse composition, with the following elemental content order: Ti > O > Ca > K > Si > C > Al > Mg > Na > Fe > P > S (Fig. S5, Supplementary material). The new elements found in the used material were Ti and P, and Si, which occurred in high concentrations and originate from the stormwater, as explained above. The SEM image of the used pine bark shows that the pores are clogged and covered by TSS, i.e. small particles >1.2 μm from the treated stormwater. Possible decomposition is illustrated by the occurrence of micro fibrillated structures.

Peat moss has the unique combination of high porosity and low bulk density (Rezanezhad et al., 2016). Fig. 5e shows unused peat composed of fibres, pores, mineral particles, and void spaces. Fig. 5f is an image of used peat and shows that particles of different sizes have stuck between the voids, the pores are clogged, and smaller pieces of roots and fibres are shredded, which may be a sign of degradation of the material. According to the x-ray spectroscopy (Fig. S5, Supplementary material), unused peat had a similar mineral composition to bark, i.e. the major components were C, Ca, K, and O, however metals like Al, Mg, Fe, and Na were also quantified. The mineralogical formation of peat (Fig. S5, Supplementary material) changed slightly from the following

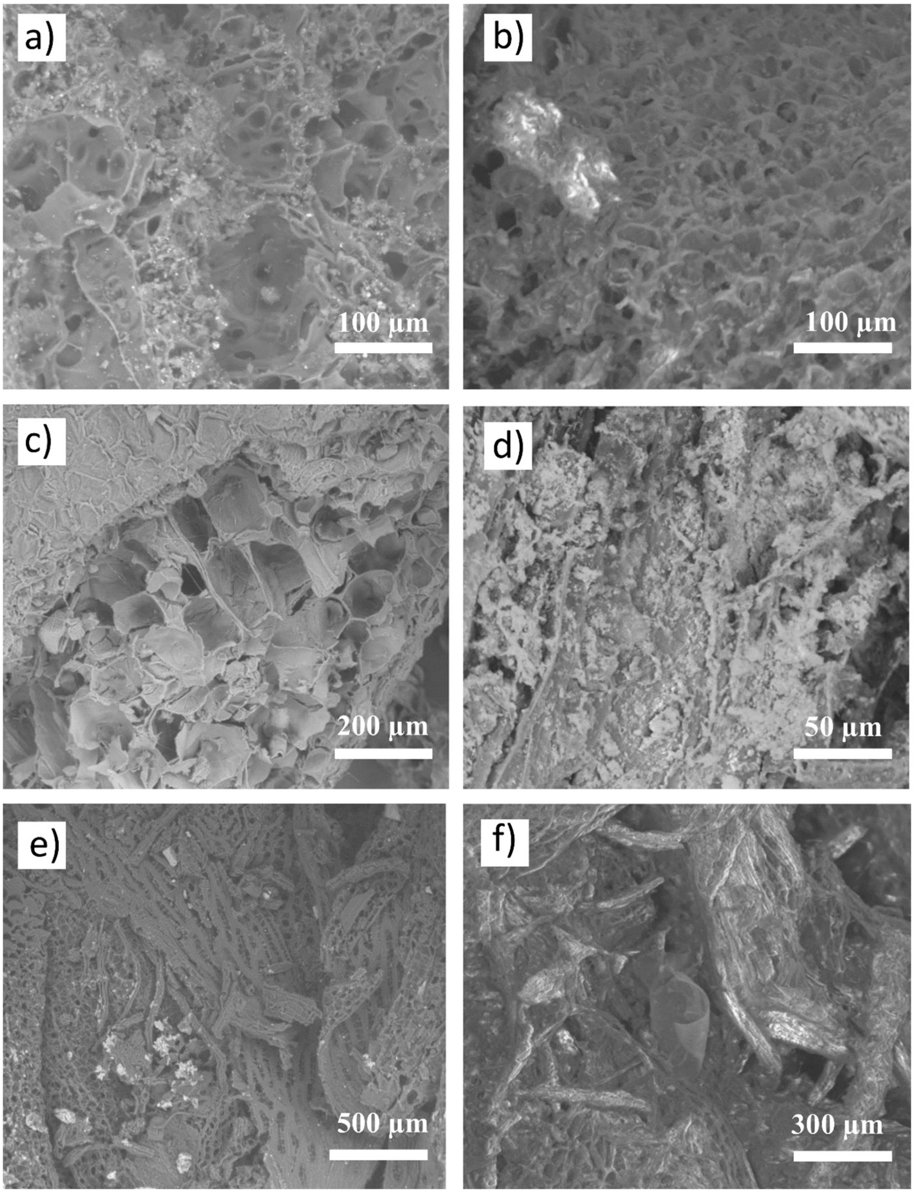

Fig. 5. Scanning electron microscopy (SEM) images of sorption filters a) GAC new material, image magnified 557 times, b) GAC after use, image magnified 555 times, c) bark new material, image magnified 361 times, d) bark after use, image magnified 1119 times, e) peat new material, image magnified 107 times, f) peat after use, image magnified 312 times.

order of elemental components in the new material: Ca > K > C > O > Fe > Si > Al > Mg > S > Na, to K > Ca > C > O > Fe > Si > Al > Na > Sin in the used material.

4. Conclusions

The design of the pilot plant was appropriate for assessing the ability of sorption materials to remove non-particulate OPs. The results show that all three sorption filters were removing OPs to a high degree, especially during the stress tests (high pollutant loads) when the removal efficiency was 100% for all three sorption filters. However, it was not possible to achieve the desired water flow through the filters. It is suggested that future research should focus on improving the hydraulic characteristics of the beds. All filters also effectively removed total suspended solids (particles >1.2 µm). Total organic carbon was removed by the sand and peat filters, but all filters leached dissolved organic carbon (<0.45 µm). This is alarming, as a loss of organic colloids may imply loss of OPs and other contaminants that bind to the colloids.

The OPs found in the highest quantified concentrations were petrol and diesel aliphatics C_5-C_{16}, in both the influent and effluents from most of the sorption filters during all events (i.e. weekly, rain, and stress tests). However, these aliphatics were not effectively removed by the sorption filters during rain events, indicating leaching of lighter pollutants from the filters during high pollutant loads. The results showed that GAC was effective for sorbing the OPs with the lowest molecular weights and highest water solubility, such as BTEX. Bark, and especially peat, was effective for the higher molecular weight OPs, such as PAH—H. The recommendation is therefore to use a combination of the studied sorption materials in stormwater filters.

All sorption filters sorbed some size fractions of nanoparticles while releasing nanoparticles in other size fractions. Overall, the sorption filters removed nanoparticles as follows: GAC by 51% > bark by 46% > peat by 22%. The low retention of nanoparticles in peat suggests a possible co-release of attached OPs, as the release of some aliphatics also increased in the peat effluent during rain events. The loss of nanoparticles correlates well with the loss of dissolved organic carbon; this is concerning as loss of OPs and other contaminants can bind to nanoparticles, or form nanoparticles in the form of emulsions, if they occur in high concentrations.

CRediT authorship contribution statement

Anna Markiewicz: Conceptualization, Methodology, Investigation, Writing - original draft, Writing - review & editing, Visualization. **Ann-Margret Strömvall:** Supervision, Conceptualization, Methodology, Investigation, Writing - original draft, Funding acquisition, Writing - review & editing. **Karin Björklund:** Supervision, Conceptualization, Methodology, Formal analysis, Investigation, Writing - original draft, Funding acquisition, Writing - review & editing.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.139059.

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