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Performance of a gross pollutant trap-biofilter and sand filter treatment train for the removal of organic micropollutants from highway stormwater (field study)

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ABSTRACT

This field study assessed the occurrence, event mean concentrations (EMCs), and removal of selected organic micro-pollutants (OMPs), namely, polycyclic aromatic hydrocarbons (PAHs), petroleum hydrocarbons (PHCs), nonylphenol (NP), 4-t-octylphenol (OP), and bisphenol A (BPA), in a gross pollutant trap (GPT)-biofilter/sand filter stormwater treatment train in Sundsvall, Sweden. The effects of design features of each treatment unit, including pre-sedimentation (GPT), sand filter medium, vegetation, and chalk amendment, were investigated by comparing the units' removal performances. Overall, the treatment train removed most OMPs from highway runoff effectively. The results showed that although the sand filter provided moderate (<50 % for phenolic substances) to high (50–80 % for PAHs and PHCs) removal of OMPs, adding a vegetated soil layer on top of the sand filter considerably improved the removal performance (by at least 30 %), especially for BPA, OP, and suspended solids. Moreover, GTP did not contribute to the treatment significantly. Uncertainties in the removal efficiencies of PAHs and PHCs by the filter cells increased substantially when the ratio of the influent concentration to the limit of quantification decreased. Thus, accounting for such uncertainties due to the low OMP concentrations should be considered when evaluating the removal performance of biofilters.

1. Introduction

Traffic-related activities are known to be one of the major sources of organic micropollutants (OMPs) in road runoff. Recent studies have shown that hazardous OMPs, such as phthalates, alkylphenols and polycyclic aromatic hydrocarbons (PAHs), have been frequently detected in road runoff at concentrations exceeding environmental quality standards (EQS) (Gasperi et al., 2022; Mutzner et al., 2022; Wicke et al., 2021). Many types of OMPs can resist biodegradation, bioaccumulate, and potentially cause ecological risk to receiving water bodies (Diblasi et al., 2009; Markiewicz et al., 2017). Although the dilution of runoff in receiving water bodies is expected to limit the risks, OMPs can still cause acute or chronic adverse effects (Rehrl, 2019; Spahr et al., 2019).

Stormwater biofilters (or bioretention) are one of the stormwater control measures (SCMs) developed to enhance the quality of runoff using infiltration processes through (often sand-based) filter media (Prince George's County, 2007). Some laboratory or pilot-scale studies have shown that OMPs such as PAHs, plasticizers, and bisphenol-A (BPA) (Bester and Schäfer, 2009; Leroy et al., 2015; Lu and Chen, 2018) can be effectively removed by biofilter systems. However, the behavior of micropollutants in laboratory-scale columns or mesocosm experiments under controlled conditions may not always be representative of that for field systems where there are more complex and varying environmental conditions (Flanagan et al., 2019). A few recent field studies have shown that biofiltration can be effective for OMP removal, particularly for hydrophobic and highly particulate substances such as polychlorinated biphenyls (PCBs), PAHs, and total petroleum hydrocarbons (PHCs) (David et al., 2015; Diblasi et al., 2009; Flanagan et al., 2018), though with a more limited effect for more hydrophilic and soluble substances such as plasticizers (phthalates and benzoates), BPA, polyfluorinated alkyl substances (PFAS), and herbicides (Boehm et al., 2020; Spahr et al., 2019; Zhang et al., 2016). There are still limited data about the treatment performance evaluation and validation of biofilter systems (specially treatment trains) under field conditions in relation to OMPs, in particular less-studied OMPs such as BPA and alkylphenols (compared with solids, nutrients, and metals).

Often, biofilter media targeting water quality treatment are sand or sandy-loam based engineered soils with a relatively low content of

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organic matter (to avoid nutrient leaching) (DWA-M 187, 2005). To enhance their performance, various techniques have been proposed. Usually, a more effective stormwater treatment can be achieved by combining complementary treatment processes (stormwater treatment trains (TT)). Biofilters can be equipped with a so-called forebay or gross pollutant trap (GPT) for coarse particle sedimentation and oil separation (Andersson et al., 2018). Although there have been successful examples of utilizing pre-sedimentation tanks to treat highway runoff (Andersson et al., 2018; Hunt et al., 2015; Purvis et al., 2019), some field observations (Greenway et al., 2012; Lange et al., 2021) have revealed that GPT may not perform well enough for removal of suspended solids (TSS), N nutrients, and microplastics. This study tried to determine the effectiveness of a GPT (including pre-sedimentation and oil separator) within a TT for removing OMPs from highway runoff.

Filter media amendment is another way to enhance the performance of biofilters. For example, chalk ($CaCO_3$) may compensate for low organic matter content and increase the pH of filter media, enhancing solute adsorption on the solid phase (DWA-M 187, 2005; Søberg et al., 2019). Originally, adding chalk was considered to enhance metal adsorption for highway runoff (DWA-M 187, 2005; Grotehusmann et al., 2016). However, there have been very few studies of the effect of chalk-amended biofilters on OMP treatment performance.

Vegetated biofilters can also have several positive benefits in relation to quality compared to non-vegetated sand filters. Plant cover can mechanically filter particulate pollutants, prevent clogging, and may remove pollutants directly by uptake or indirectly by increasing microbial activity (Chu et al., 2021; Le Coustumer et al., 2012; Muerdter et al., 2016). While most previous biofilter studies on the effect of vegetation species have focused on TSS, metal, and nutrient removal (Dagenais et al., 2018), a few have investigated removal of OMPs by biofilters, but mostly under controlled conditions using artificial runoff (Leroy et al., 2015; Randelovic et al., 2016; Zhang et al., 2014). Flanagan et al. (2018) studying OMP removal in two vegetated filtration facilities under real environmental conditions showed that particulate pollutants such as PAHs were removed efficiently (but not during a winter period), while dissolved fractions of OMPs had poor removal. However, there is still a need for further knowledge on the effect of vegetation as an independent factor for OMP removal under field conditions with real runoff.

The main objective of this study was to assess the performance of a full-scale GPT-biofilter/sand filter stormwater treatment train (TT) for the removal of target OMPs (i.e. sixteen PAHs, four fractions of PHCs, eight alkylphenols (APs), and BPA) from a highway bridge catchment. First, we evaluated the role of the GPT pre-treatment and then the effects of vegetation and chalk factors on the OMP treatment by three different biofilter cells: vegetated without chalk, vegetated with chalk, and non-vegetated without chalk. Finally, we investigated the performance of the TT in reducing potential environmental risks of OMPs and identified the most relevant OMPs in the highway runoff and in the effluent of different units, based on comparative ranking of the risk levels, something which has been rarely carried out in prior studies.

2. Methodology

2.1. Study site

This field study was carried out using a gross pollutant trap (GPT)-biofilter/sand filter treatment train (TT) located in Sundsvall, Sweden (62°23′0.5″N 17°20′50.5″E), which has Continental Subarctic Climate (Dfc) and cool summers. The system receives stormwater from an impervious catchment area of 4.7 ha including the 1.9 ha E4 highway bridge with an average traffic load of 13,000 vehicles/day, a highway exit way, main roads associated with a roundabout and sidewalk paths (Fig. S1).

The TT (Fig. 1) was designed according to German stormwater biofilter guidelines for treatment of highway runoff (DWA-M 187, 2005)

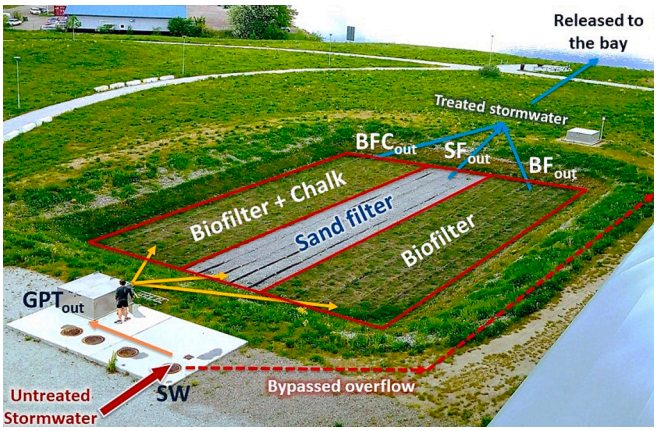

Fig. 1. GPT-biofilter/sand filter treatment train (TT) studied in Sundsvall, Sweden (SW, GPT_{out}, BFC_{out}, SF_{out}, and BF_{out} are the abbreviations for the locations where the samples are taken from stormwater inflow, and the outflow of gross pollutant trap and the filter cells respectively).

and was constructed in 2018 i.e. it was 3 years old at the time of sampling. Downstream of the road catchment, the collected stormwater is first transported by a 100-m-long underground pipe (slope 0.5 % and diameter 0.8 m) to the GPT section which includes a sedimentation chamber and an oil separator. The stormwater in the GPT is then discharged through a stepwise valve-controlled siphon system to three parallel filter cells divided by EPDM membranes where it infiltrates through the sand-based filter media. One of the filter cells is non-vegetated (*sand filter SF*), and the other two are vegetated (*biofilters BFC* and *BF*). The vegetation layer is made of salt-tolerant meadow sod (Veg Tech AB, Sweden) with 17 different plant species pre-cultivated in a 3–4 cm deep sandy or silty-sandy soil with 2.5–5 % *w*/w mulch (see Fig. S2). The filter media in one of the biofilters (*BFC*) is amended with 10 % w/w crushed grey chalk ($CaCO_3$). Afterwards, the treated stormwater is drained from the cells through a gravel drain layer with embedded drainpipes, then led to three sampling wells, and finally collected and released to the downstream recipient. If the runoff inflow exceeds the GPT's active detention volume (~23.3 m^3) during each charging and discharge step, the stormwater is bypassed from the TT at the GPT entrance. See Table S1 and Fig. S3 for more technical information on the TT compartments. In previous research, the same treatment train has been investigated for total, dissolved and truly dissolved metals by Lange et al. (2022), and microplastics by Lange et al. (2021).

2.2. Sampling procedure and strategies

The performance of the TT was investigated for 11 rain events between September 2020 and September 2021 (Rain A–K in Table S2). Samples were collected from five locations in the system:

- **SW**: stormwater received at GPT inlet from the catchment
- **GPT_{out}**: gross pollutant trap outflow (or filters inflow)
- **BFC_{out}**: chalk-amended vegetated biofilter outflow
- **SF_{out}**: non-vegetated sand filter outflow
- **BF_{out}**: vegetated biofilter outflow

Rainfall data were collected using a tipping bucket rain gauge (ISCO 674) next to the highway catchment (Fig. S1). Volume-proportional samples were taken during the rain events using ISCO-6712 automatic samplers. The samplers were programmed to collect a maximum of 8 volume-proportional subsamples estimated at each location. Flow was also measured at each sampling location: the GPT's influent and effluent by counting the number of discharge valve signals (supported by data from an ultrasound flowmeter at the inlet (PCM 4, NIVUS GmbH, Eppingen)), and the effluent of each filter cell by using pipe-insertion

electromagnetic flowmeters (MAG 5100 Siemens AG). The discharge valves are triggered by a floating water level meter installed in the GPT. When the valves are open, the GPT discharges 23.3 m^3 stormwater to the filter cells and simultaneously, a signal is sent to the samplers dedicated to the SW and GPT_{out} locations to take one subsample. The samplers at BFC_{out}, SF_{out}, and BF_{out} were triggered by the insertion flowmeter signals. Therefore, the total number of signals needed for programming the samplers at a certain rain event was estimated according to the maximum possible outflow volume of the filter cells to cover the entire outflow. Rain characteristics, including depth, peak/mean intensity (I_{peak} and I_{mean}), and antecedent dry period (ADP), number of subsamples taken, and the total and sampled volumes at each sampling location for all events are summarized in Table S2. Due to practical limitations such as uncertainties of rain depth and duration forecasts, as well as the time taken to deliver samples to the laboratory, it was not possible to cover the entire runoff volume in some events (see Table S2).

Each sampler was equipped with 24 lay flat Teflon™ PFA bags (Welch Fluorocarbon) (i.e. three bags per subsample), except for the sampler at BF_{out} where the subsamples were collected in a 10-l glass container (i.e. one composite sample). The Teflon bags and the container were washed with tap water before each sampling event. The samples collected were delivered to the laboratory within one day after sampling. In case of time delay in delivery (e.g. weekends), all OMPs samples were stored in a refrigerator (1–4 °C), and TOC samples in a freezer (<−15 °C).

2.3. Water quality analysis

All samples were analyzed for a number of selected OMPs and global parameters, including phenolic substances (bisphenol A, 4-t-octylphenol (OP), nonylphenol (NP), octylphenol ethoxylates (OPnEO; $n = 1, 2, 3$), nonylphenol ethoxylates (NPnEO; n = 1, 2, 3)), 16 PAHs, 4 fractions of PHCs, total organic carbons (TOC), total suspended solids (TSS), turbidity, conductivity, pH, and temperature. According to the literature, the selected OMPs have been ranked among the highest-priority or most-frequently micropollutants found in stormwater, particularly in road catchments (Gasperi et al., 2022; Lundy et al., 2012; Markiewicz et al., 2017; Masoner et al., 2019; Mutzner et al., 2022). A full list of the OMPs with their abbreviations and limits of quantification is given in Table 1. OMPs and TOC were analyzed by the accredited laboratory ALS

Table 1
List of selected organic micropollutants (OMPs) analyzed in the stormwater and treatment train (TT).

Category	Parameter	Abbreviation	Reporting limit (µg/L)
Phenolic substances	Bisphenol-A	**BPA**	0.05
	4-tert-Octylphenol	**OP**	0.01–0.25[a]
	Octylphenol monoethoxylate	**OP1EO**	0.01–0.03[a]
	Octylphenol diethoxylate	**OP2EO**	0.01–0.02[a]
	Octylphenol triethoxylate	**OP3EO**	0.01–0.033[a]
	Nonylphenol_mixture of isomers	**NP**	0.1–1.35[a]
	Nonylphenol monoethoxylate	**NP1EO**	0.1–0.3[a]
	Nonylphenol diethoxylate	**NP2EO**	0.1–2.54[a]
	Nonylphenol triethoxylate	**NP3EO**	0.1–3.12[a]
Polycyclic aromatic hydrocarbons (PAHs)	Naphthalene	**Nap**	0.03
	Acenaphthylene	**Acyl**	0.01
	Acenaphthene	**Acen**	0.01
	Fluorene	**Flu**	0.01
	Phenanthrene	**Phen**	0.02
	Anthracene	**Anth**	0.01
	Fluoranthene	**Flth**	0.01
	Pyrene	**Pyr**	0.01
	Benz(*a*)anthracene	**BaA**	0.01
	Chrysene	**Chry**	0.01
	Benzo(*b*)fluoranthene	**BbF**	0.01
	Benzo(*k*)fluoranthene	**BkF**	0.01
	Benzo(*a*)pyrene	**BaP**	0.01
	Dibenz(a.h)anthracene	**DahA**	0.01
	Benzo(*ghi*)perylene	**Bper**	0.01
	Indeno(1.2.3-cd)pyrene	**InP**	0.01
	Sum of all PAHs	**Σ16PAHs**	–
	Carcinogenic PAHs	**ΣCar.PAHs**[b]	–
	Non-carcinogenic PAHs	**Σnon-Car.PAHs**[c]	–
	Light-weight PAH molecules	**ΣLMW-PAHs**[d]	–
	Medium-weight PAH molecules	**ΣMMW-PAHs**[e]	–
	High-weight PAH molecules	**ΣHMW-PAHs**[f]	–
Petroleum hydrocarbons (PHCs)	Total PHCs	**C_{10} - C_{40}**	50
	PHC fractions	**C_{10} - C_{12}**	5
		C_{12} - C_{16}	5
		C_{16} - C_{35}	30
		C_{35} - C_{40}	10
Global parameters	Total organic carbons	**TOC**	500
	Total suspended solids	**TSS**	2200
	Turbidity	**Turb**	–
	Electric conductivity	**EC**	–
	pH	**pH**	–
	Temperature	**Temp.**	–

[a] The lower limit represents the min RL analytically expected and the upper limit represents the max RL reported by the laboratory for a certain substance (RL varies between the range due to matrix interference during chemical analysis.)
[b] Nap, BaA, Chry, BbF, BkF, BaP, BahA, Bper.
[c] Acyl, Acen, Flu, Phen, Anth, Flth, Pyr, IP.
[d] Nap, Acyl, Acen, Flu, Phen, Anth.
[e] Flth, Pyr.
[f] BaA, Chry, BbF, BkF, BaP, DahA, Bper, IP.

Czech Republic, and TSS by accredited ALS Scandinavia AB. It should be noted that reporting limits (RL) for phenolic substances (except BPA) were sometimes affected by matrix interference during chemical analysis, meaning that the RLs varied over the experiments (see Table 1). Global parameters were all measured during sampling events on site. Table S3 summarizes the analytical methods and equipment used in this study.

In order to assess any potential leaching of OMPs from the sampling bags and tubes, batch blank tests were carried out onsite and in the laboratory. The procedure for blank testing is described in our previous study in detail (Beryani et al., 2023). The results showed that the OMPs of concern were not found in the water in contact with sampling equipment. Nevertheless, after the first 6–7 events, we changed the Teflon bags of all samplers for a new series of bags. Moreover, to avoid cross-contamination throughout the experiments, we allocated each sampling bag to the same position in the identical sampler.

2.4. Data analysis

2.4.1. Event mean concentration (EMC)

To evaluate treatment performance, the event mean concentration (EMC) of OMPs at each sampling location was estimated using the subsamples' concentrations and flow records. The EMC is an expression of the total mass (M_T) conveyed by the total stormwater volume (V_T) at a specific location during the entire event period (Eq. (1)). In Eq. (1), n is the number of subsamples, m_i is the pollutant mass conveyed during the collection of the ith subsample, c_i is the ith subsample concentration, and v_i is the corresponding stormwater volume passed.

$$EMC = \frac{M_T}{V_T} = \frac{\sum m_i}{\sum v_i} = \frac{\sum_{i=1}^{n} c_i v_i}{\sum_{i=1}^{n} v_i} \quad (1)$$

For some events, no water was collected from a given sampling location due to practical reasons (see Table S2). In that scenario, to estimate a more accurate EMC following a suggestion made by Furuta et al. (2022), EMC calculations were adjusted by weighting the final subsample concentration with the missing volume in addition to the original volume sampled. In this adjustment, rain event G at SW and GPT_{out} was an exception where the first 17 % of stormwater was not covered. Thus, using the same analogy, the missing part of the rain event was assigned to the first subsample for EMC calculations.

2.4.2. Estimation of best EMCs and their associated uncertainties

To spread the uncertainty in the EMC due to analytical issues and missing data in individual analyses, a Monte-Carlo (MC) simulation was used in R software. In the MC method, Eq. (1) was used to find the EMC distribution at a given sampling point for a certain rain event. Details of the MC simulation are given in the supporting information. The median of the final EMC distribution was considered to be the best-estimate event mean concentration (EMC_{best}), and the range between 2.5 % and 97.5 % quantiles as the EMC's lower and upper limits of uncertainty ($-\Delta_l$, $+\Delta_u$).

The EMCs for PAH fractions (Σ16-, ΣCar-, Σnon-Car-, ΣLMW-, ΣMMW-, and ΣHMW- PAHs) and their uncertainties were estimated by running the MC method twice. In the first step, a concentration distribution for a certain PAH fraction was generated for each subsample using PAH substance group data under that fraction. In the next step, those distributions were used as input distributions to estimate the PAH fraction's EMC_{best} and its uncertainty at each sampling location (as explained before).

To visualize, assess, and compare the EMCs of OMPs at different sampling points, EMC non-exceedance probability (NEP) plots for all rain events were generated. To create these NEP plots, the Kaplan-Meier method, which is commonly applied to left-censored data analysis (Helsel, 2010), was used from the "EnvStats" V2.3.0 package for environmental statistics in R. Furthermore, to assess the stormwater quality at different treatment stages, the EMCs of OMPs shown in the NEP plots were compared with the existing lowest Predicted No-Effect Concentrations (PNECs) in freshwater (NORMAN, 2012). PNECs are considered as a basis for the environmental quality standards (EQSs) prioritized by European Union Water Framework Directive (EU WFD, 2013).

2.4.3. Removal efficiencies and uncertainties

Removal efficiencies (RE%) of different treatment sections were calculated using Eq. (2), where EMC_{in} and EMC_{out} stand for EMC_{best} at the section's inlet and outlet. Since the uncertainties in EMCs cause an error in the removal efficiencies, the absolute removal error (Err_{Re}%) was also calculated using Eq. (3) according to Taylor (1997), assuming that the EMC's uncertainties for the inlet and outlet (Δ_{in} and Δ_{out}) are independent. It should be noted that in the MC method used, the EMC_{best} for a censored EMC (i.e. if the concentrations of all subsamples were censored) was almost equal to half the LoQ. However, in cases where both inlet and outlet EMCs were censored for a given event, the calculated Re% and Err_{Re}% were excluded from our statistical analysis. Therefore, the number of data points for analyzing Re% and Err_{Re}% ranged from five to nine.

$$Re\ (\%) = 100 \times \frac{EMC_{in} - EMC_{out}}{EMC_{in}} \quad (2)$$

$$Err_{Re}(\%) = 100 \times \sqrt{\left(\Delta_{in} \cdot \frac{EMC_{out}}{EMC_{in}^{2}}\right)^{2} + \left(\Delta_{out} \cdot \frac{1}{EMC_{in}}\right)^{2}} \quad (3)$$

2.4.4. Statistical analysis

The significance of differences between the concentrations of a certain parameter ($N = 8$–11 rain events, except for ethoxylate alkylphenols) at the inlet and outlet of each treatment section were evaluated to gauge treatability. The differences between the EMCs of filter cells' outlets were also tested to compare statistically the filter cells' treatment efficiencies. Here, we used the Peto & Peto generalized Wilcoxon test, which is the most suitable way for assessing the significance of differences in left-censored log-normal data (our case) (Helsel, 2005). The Chi-square (X^2) calculated for the relative deviation of datasets was assumed statistically significant when the p-value (p) ≤ 0.05 (Null hypothesis, H_0: there is no difference between the concentrations of sampling points). For some points, correlation tests were carried out between TSS, turbidity, and other OMPs (refer to supporting information for details about the methods). All the statistical tests for censored data analysis were carried out using "NADA" (Nondetects and Data Analysis for Environmental Data) package in R (V4.1.3).

2.4.5. Risk analysis

According to Skivington (1997), risk refers to the combination of the occurrence likelihood of a defined hazard (presence of OMP) and the magnitude of the occurrence consequences. Eq. (4) was used to estimate the total environmental risk (R_T) associated with an OMP at a particular sampling point during the entire experiment. An OMP's environmental hazard, or potential criticality, was assessed using the risk quotient (RQ_j), which is calculated by dividing the observed concentration of the OMP by the chronic environmental quality standard (EQS) for freshwater (Mutzner et al., 2022). Here, the EQS was derived from the PNEC value (if available) for an OMP in an ecotoxicological database. For OMPs without a PNEC, no RQ was calculated; they were excluded from our risk ranking. Occurrence probability (P_j) of EMCs was quantified by dividing one by the number of events studied at a given sampling location. The calculated R_Ts were then used to compare the risk posed by OMPs at different stages in stormwater. Additionally, OMPs with an $R_T > 1$ were deemed to be potentially risky pollutants for the receiving water body, although the dilution effect in that recipient may mitigate the risks in reality.

$$R_T = \sum_j P_j.RQ_j = \sum_j P_j.(EMC_j \pm \Delta_j) \Big/ PNEC\ (j: \text{rain event}) \quad (4)$$

3. Results

3.1. EMC and occurrence analysis of OMPs

The EMC and occurrence analysis of OMPs in treated and untreated stormwater is presented below. A summary of the statistical analysis for the calculated EMCs of all OMPs is given in Table 2.

3.1.1. Phenolic substances

BPA was quantified in all samples at SW, GPT_{out}, and SF_{out}, but only in three of eight rain events at BFC_{out} and three of ten at BF_{out}, respectively (Table 2). The occurrence of alkylphenols (APs: OP and NP) was, however, lower in the stormwater (six and five of eight events for OP and NP, respectively) and the outlet of treatment sections, in general. At SF_{out}, the occurrences of OP and NP were identical to those at SW and GPT_{out} (5–6 of nine events), while OP was never found but NP was quantified in two and five events (out of eight) at BF_{out} and BFC_{out}, respectively, with relatively comparable concentration levels measured at SW (see Section 4.5).

As shown in Fig. 2(a)–(c), the median EMCs of phenolic substances (except alkylphenol ethoxylates) at both SW and GPT_{out} were often similar and exceeded PNECs for freshwater (BPA: 0.40 ± 0.10 μg/L, NP: 0.36 ± 0.15 μg/L, and OP: 0.08 ± 0.02 μg/L). Downstream, at the outlet of the vegetated biofilters (BF_{out} and BFC_{out}), the EMCs of BPA (median < 0.05 μg/L) and OP (all non-detects) significantly decreased to below corresponding PNECs (always <PNEC), but this was not the case for NP in 4–6 (the range takes uncertainty into account) of nine events (median: 0.41 ± 0.12 μg/L). The EMCs after the non-vegetated filter (SF), however, revealed that BPA and NP concentrations remained almost at the same levels as at the SF's inlet (i.e. no statistically significant difference in EMCs between GPT_{out} and SF_{out}: $X^2 < 0.6$, $p > 0.05$) so that the PNECs (0.24 and 0.3 μg/L, respectively) were still exceeded in seven of nine rain events for BPA and, accounting for uncertainties, 3–7 of nine events for NP. EMCs of OP at the SF_{out}, on the other hand, considerably decreased to low risk levels with a median of 0.035 ± 0.013 μg/L (EMCs < PNEC = 0.1 μg/L in eight of nine events).

The event-based results showed that OPnEO and NPnEO ($n = 1, 2, 3$) were not quantified at SW or in any effluent section, except NP2EO which was found at SF_{out} for one event (possibly due to leaching: see Section 4.5) Thus, the performance of the TT remained unclear for those substances.

3.1.2. Polycyclic aromatic hydrocarbons (PAHs)

Most PAHs (except Nap, Acyl, Acen, Anth, and Flu from LMW-PAH fractions) were frequently quantified in the untreated stormwater (SW) where EMCs exceeded PNECs. The occurrence assessment also showed that the occurrence and EMC levels of PAHs in the GPT_{out} followed similar patterns as observed in SW (NEP plots in Figs. 2d–h andS4a–l). No significant difference of EMCs was identified when comparing SW and GPT_{out}: $X^2 < 0.53$ and $p > 0.05$ for all PAHs except for Anth and Flu which were occasionally found in the GPT_{out}, but not in SW for the same event (discussed in Section 4.1).

Phen was the only LMW-PAH quantified in five and four of eight events at SW and GPT_{out}, respectively, (median EMC: 0.017 ± 0.05 μg/L) but never exceeded the PNEC (0.3 μg/L). Phen was never quantified in the filter outlets. MMW-PAH fractions including Flth and Pyr were almost always quantified in SW and GPT_{out} (seven or eight of eight events), while HMW-PAH occurrence at these two sampling points varied between four and eight of eight events in this order: BbF = Bper > BaP > Chry = InP > BaA > DahA. At SW and GPT_{out}, the EMCs of Flth and Pyr with estimated medians of 0.050 ± 0.008 μg/L and 0.079 ± 0.022 μg/L, respectively, were also frequently above the corresponding PNEC levels (7–8 of eight events). Six of eight HMW-PAHs exhibited risky concentration levels (EMC > PNEC) at SW and GPT_{out}. Of these, BaP, BaA, Chry, BbF, and DahA are classified as extremely or possibly carcinogenic (see Figs. 2d–e and S4a, b, e). In general, 41–43 % of Σ16PAH concentrations in the range 0.110–1.131 μg/L at SW and GPT_{out} could be attributed to carcinogenic substances.

PAHs were rarely observed at BFC_{out} and BF_{out}, except for one event (rain C) during which all MMW-PAHs and HMW-PAHs were found at BF_{out}, as well as Benzo(*ghi*)perylene in BFC_{out}. In this rain event, PAH EMCs were also observed at their maximum levels in SW and GPT_{out}. The non-vegetated biofilter SF results, however, varied for the different quantified PAHs in terms of occurrence and EMC. Both MMW-PAHs and HMW-PAHs (but none of the LMW-PAHs) were quantified during at least one up to nine of nine events at the SF_{out}: Pyr, BbF, and Bper with the highest occurrence, Chry and Flth with a moderate occurrence, and IP, BaA, BaP, BkF, and DahA with the lowest occurrence. Some of the hazardous PAHs (having relatively low PNECs (<0.01 μg/L)) found in SF_{out}, such as Pyr and Chry, were measured with considerably higher concentrations than their quality objectives in freshwater. Although more dangerous substances including BaP and DahA (with very low PNECs) occurred only a few times in SF_{out}, the actual risk levels after the treatment still remained unclear for such PAHs since their RLs (0.01 μg/L) were higher than the corresponding PNEC levels (this is also the case for all non-detects of BaP, DahA, Chry, Flth, and Bper at all other sampling points). Further detailed information about the occurrence and EMC of PAHs for all sampling points is provided in Table 2.

3.1.3. Petroleum hydrocarbons (PHCs)

Total PHCs (C_{10}-C_{40}) were quantified in all events at SW and GPT_{out}. Again, data analysis showed similar range/trend for the EMCs of all PHC fractions at both SW and GPT_{out} (Fig. 2i). EMCs of C_{10}-C_{40} (associated with the uncertainties) varied in the range of 145–1886 μg/L for SW, and 122–1431 μg/L for GPT_{out}, but both had a similar median of approximately 380 ± 70 μg/L ($X^2 = 0.39$ and $p > 0.05$: no significant difference in PHCs between SW and GPT_{out}). Considering Gothenburg's local guideline for aquatic receiving water bodies (Miljöförvaltningen, 2013), the observed C_{10}-C_{40} EMCs exceeded the maximum stormwater PHC concentration threshold of 1000 μg/L during two of eight and one of eight events at SW and GPT_{out}, respectively. After the TT, PHCs were repeatedly quantified at the SF_{out} (C_{10}-C_{40} median EMC: 148 ± 19 μg/L and always below the guideline value 1000 μg/L), while seldom at BFC_{out} and BF_{out} (only found in rain events C and D). C_{16}-C_{35} and C_{35}-C_{40} (i.e. heavier PHC molecules) were identified as the predominant PHC fractions in all samples, and C_{10}-C_{12} and C_{12}-C_{16} (lighter fractions) only accounted for a maximum of 1–4 % of total PHCs concentrations in different sampling locations. The EMCs of C_{16}-C_{35} were always higher than C_{35}-C_{40} with mean ratios of 3.6, 3.8, 3.5, 4.6, and 3.1 at SW, GPT_{out}, BFC_{out}, SF_{out}, and BF_{out}, respectively. See Table 2 for further details about PHC occurrence and EMCs.

3.1.4. Conventional water quality parameters

EMCs of TSS were in a similar range at SW and GPT_{out} (18.9–231.4 mg/L; median: 54.5 ± 6.6 mg/L and $X^2 = 0.08$, $p > 0.05$, i.e. no significant difference between SW and GPT_{out}) so that 9–11 of eleven events exceeded the European quality objective of 25 mg/L for TSS (the protective threshold against chronic effects in freshwater (EC, 2006)). The event mean turbidity at those two locations similarly varied between 40.4 and 236.8 NTU for all rain events (Fig. 2k). As shown in Fig. 2j, TSS levels considerably dropped into the range of 10.4–35.0 mg/L (median = 18.9 ± 2.3) after the non-vegetated SF, and even more significantly after the vegetated BF and BFC, within <2.5–26.7 mg/L (median = 3.0 ± 0.9). In general, the TSS measured at the outlet of filter cells rarely exceeded the threshold concentration of 25 mg/L recommended for freshwater (EC, 2006). Turbidity followed a similar pattern to TSS with mean values decreasing moving downstream from SW and GPT to the subsequent filter cells.

Table 2
Statistical summary of the calculated EMCs over events (N = $n_{quantified}$ + $n_{censored}$) at different sampling spots.

Parameter	Conc. unit	SW			GPT		
		n (quantified)	n (censored)	(min; mean; max; SD)	n (quantified)	n (censored)	(min; mean; max; SD)
Nap	μg/L	0	8	(–;–;<0.03;–)	0	8	(–;–;<0.03;–)
Acyl	μg/L	0	8	(–;–;<0.01;–)	0	8	(–;–;<0.01;–)
Acen	μg/L	0	8	(–;–;<0.01;–)	0	8	(–;–;<0.01;–)
Flu	μg/L	0	8	(–;–;<0.01;–)	1	7	(<0.006;–;0.01;–)
Phen	μg/L	5	3	(<0.015;0.026;0.065;0.017)	4	4	(<0.02;0.027;0.058;0.012)
Anth	μg/L	0	8	(–;–;<0.01;–)	1	7	(<0.009;–;0.01;–)
Flth	μg/L	8	0	(0.008;0.069;0.163;0.059)	7	1	(<0.007;0.062;0.142;0.053)
Pyr	μg/L	8	0	(0.038;0.12;0.262;0.088)	8	0	(0.03;0.102;0.235;0.08)
BaA	μg/L	6	2	(<0.008;0.021;0.044;0.014)	6	2	(<0.006;0.02;0.038;0.012)
Chry	μg/L	7	1	(<0.009;0.029;0.069;0.023)	6	2	(<0.008;0.028;0.057;0.019)
BbF	μg/L	8	0	(0.013;0.061;0.14;0.049)	8	0	(0.006;0.055;0.131;0.048)
BkF	μg/L	6	2	(<0.006;0.016;0.031;0.01)	6	2	(<0.006;0.015;0.031;0.01)
BaP	μg/L	8	0	(0.006;0.028;0.064;0.024)	6	2	(<0.007;0.025;0.054;0.019)
DahA	μg/L	5	3	(<0.006;0.011;0.027;0.006)	4	4	(<0.01;0.013;0.026;0.006)
BP	μg/L	8	0	(0.013;0.066;0.145;0.054)	8	0	(0.007;0.054;0.158;0.05)
IP	μg/L	7	1	(<0.006;0.029;0.072;0.023)	6	2	(<0.008;0.029;0.075;0.023)
Σ16PAHs	μg/L	8	0	(0.165;0.507;1.052;0.372)	8	0	(0.127;0.455;0.997;0.34)
ΣCarPAHs	μg/L	8	0	(0.061;0.208;0.438;0.155)	8	0	(0.051;0.195;0.409;0.145)
Σnon-Car.PAHs	μg/L	8	0	(0.104;0.3;0.631;0.218)	8	0	(0.074;0.26;0.588;0.197)
ΣLWM-PAHs	μg/L	8	0	(0.045;0.059;0.1;0.019)	8	0	(0.045;0.058;0.097;0.018)
ΣMWM-PAHs	μg/L	8	0	(0.047;0.19;0.41;0.147)	8	0	(0.037;0.163;0.377;0.135)
ΣHWM-PAHs	μg/L	8	0	(0.066;0.258;0.566;0.208)	8	0	(0.045;0.234;0.552;0.193)
C_{10} - C_{40}	μg/L	8	0	(172.2;613.2;1545.6;506.1)	8	0	(144.6;514.9;1213.9;385.6)
C_{10} - C_{12}	μg/L	2	6	(<2.772;3.283;5;0.512)	1	7	(<3.663;–;5;–)
C_{12} - C_{16}	μg/L	7	1	(<4.551;9.375;13.392;3.445)	7	1	(<3.913;7.707;13.343;3.328)
C_{16} - C_{35}	μg/L	8	0	(138.6;471.0;1167.7;380.8)	8	0	(119.3;399.8;941.3;293.9)
C_{35} - C_{40}	μg/L	8	0	(29.7;131;361.5;123.5)	8	0	(21.8;105.8;259.7;90.1)
OP	μg/L	6	2	(<0.041;0.131;0.338;0.104)	6	2	(<0.061;0.139;0.303;0.085)
OP1EO	μg/L	0	6	(–;–;<0.024;–)	0	6	(–;–;<0.01;–)
OP2EO	μg/L	0	6	(–;–;<0.45;–)	0	6	(–;–;<0.01;–)
OP3EO	μg/L	0	6	(–;–;<0.033;–)	0	6	(–;–;<0.028;–)
NP	μg/L	5	3	(<0.166;0.374;1.19;0.181)	5	3	(<0.2;0.397;1.11;0.201)
NP1EO	μg/L	0	6	(–;–;<0.18;–)	0	6	(–;–;<0.22;–)
NP2EO	μg/L	0	6	(–;–;<2.54;–)	0	6	(–;–;<1.49;–)
NP3EO	μg/L	0	6	(–;–;<1.27;–)	0	6	(–;–;<1.49;–)
BPA	μg/L	8	0	(0.247;0.542;1.179;0.314)	8	0	(0.169;0.48;0.933;0.249)
TOC	mg/L	11	0	(2.34;10.06;23.03;7.08)	11	0	(2.06;10.56;24.96;7.5)
TSS	mg/L	11	0	(22.4;74.3;205.8;56.9)	11	0	(18.9;69.0;200.8;55.1)
Turb.	NTU	9	0	(40.39;107.17;201.11;54.79)	9	0	(43.06;112.93;236.77;62.51)
EC	μS/cm	9	0	(32.77;138.39;299.49;82.08)	9	0	(38.54;141.83;318.53;87.16)
pH	-	9	0	(6.799;7.148;7.308;0.156)	9	0	(7.026;7.148;7.409;0.11)
Temp.	°C	9	0	(11.62;16.52;22.35;3.38)	9	0	(12.1;16.71;22.91;3.4)

[a] The water quality objectives refer to the lowest Predicted No-Effect Concentrations (PNEC) for freshwater from NORMAN Ecotoxicology Database, unless another guideline is mentioned.
[b] City of Gothenburg's guideline value for aliphatic and aromatic hydrocarbons (Miljöförvaltningen, 2013).
[c] WHO's recommendation for aromatic PHCs in drinking water (WHO, 2008).
[d] WHO's recommendation for aliphatic PHCs in drinking water (WHO, 2008).
[e] Gothenburg's stormwater annual average benchmark concentration at the point of discharge to receiving water bodies (Miljöförvaltningen, 2013).
[f] Protective threshold concentration against chronic effects on fish in freshwater (EC, 2006).

TOC was always quantified at all sampling points. The EMCs of TOC in untreated stormwater (SW) ranged between 2.3 and 26.2 mg/L with a median of 7.0 ± 0.6 mg/L for all rain events. The results (Fig. 2l) showed that TOC levels in SW did not significantly change after the GPT and SF treatment sections ($X^2 < 0.24$, $p > 0.05$). Therefore, a similar EMC variation over the events was observed at those sampling points as at SW (Table 2). Considering Gothenburg's guidelines for stormwater, TOC levels exceeded the threshold of 12 mg/L during (4–5) of eleven events at SW and GPT_{out}. TOC concentrations, however, slightly decreased after the vegetated biofilters (though not statistically significantly: $X^2 < 1.4$, $p > 0.05$) so that the EMCs varied in the range of 1.5–22.9 mg/L with medians of 7.4 ± 0.8 at BFC_{out} and 6.7 ± 1.0 mg/L at BF_{out}. Nevertheless, it exceeded the 12 mg/L recommended level in four of eight, five of nine, and five of ten events at BF_{out}, SF_{out}, and BFC_{out}, respectively.

The stormwater pH value at SW, GPT_{out}, BF_{out}, and SF_{out} remained unchanged in the neutral range of 5.6 and 7.4 (median: 7.15 ± 0.05), while noticeably increasing after BFC to 7.5–8.1 with a median of 7.9. All event mean pH values were within the recommended range (6.5–9) for stormwater discharged to freshwater (Miljöförvaltningen, 2013), except in rain event J at BF_{out} with the minimum pH measured 5.6.

3.2. Treatment train performance for OMP removal

3.2.1. Gross pollutant trap (GPT) removal efficiency

The results revealed that the GPT did not contribute to the stormwater treatment significantly, with median OMP removals <20 %. Fig. 3 shows a statistical summary of mean removal efficiencies and errors of the OMPs and global parameters for different treatment units. The overall median removal efficiencies (Re% associated with its Err_{Re}%) for the GPT section were 11.6 ± 14.2 % for Σ16PAHs, 13.2 ± 30.3 % for PHCs, 3.2 ± 42.6 % for BPA, 11.4 ± 26.4 % for TSS, and − 7.0 % for Turbidity. REs for OP, NP, and TOC were all near or below zero (see

BFC			SF			BF			Water quality objective[a]
n (quantified)	n (censored)	(min; mean; max; SD)	n (quantified)	n (censored)	(min; mean; max; SD)	n (quantified)	n (censored)	(min; mean; max; SD)	
0	8	(–;–;<0.03;–)	0	9	(–;–;<0.03;–)	0	9	(–;–;<0.03;–)	2
0	8	(–;–;<0.01;–)	0	9	(–;–;<0.01;–)	0	9	(–;–;<0.01;–)	1.3
0	8	(–;–;<0.01;–)	0	9	(–;–;<0.01;–)	0	9	(–;–;<0.01;–)	3.7
0	8	(–;–;<0.01;–)	0	9	(–;–;<0.01;–)	0	9	(–;–;<0.01;–)	0.25
0	8	(–;–;<0.02;–)	0	9	(–;–;<0.02;–)	0	9	(–;–;<0.02;–)	0.5
0	8	(–;–;<0.01;–)	0	9	(–;–;<0.01;–)	0	9	(–;–;<0.01;–)	0.1
0	8	(–;–;<0.01;–)	5	4	(<0.005;0.01;0.033;0.008)	1	8	(<0.01;–;0.027;–)	0.0063
0	8	(–;–;<0.01;–)	9	0	(0.017;0.026;0.065;0.015)	1	8	(<0.01;–;0.042;–)	0.0046
0	8	(–;–;<0.01;–)	2	7	(0.006;0.008;0.01;0.002)	1	8	(<0.01;–;0.016;–)	0.012
0	8	(–;–;<0.01;–)	6	3	(0.005;0.008;0.017;0.003)	1	8	(<0.01;–;0.024;–)	0.0029
0	8	(–;–;<0.01;–)	9	0	(0.008;0.013;0.037;0.009)	1	8	(<0.01;–;0.045;–)	0.017
0	8	(–;–;<0.01;–)	1	8	(<0.008;–;0.01;–)	1	8	(<0.01;–;0.011;–)	0.017
0	8	(–;–;<0.01;–)	2	7	(0.007;0.007;0.015;0.003)	1	8	(<0.01;–;0.025;–)	0.00017
0	8	(–;–;<0.01;–)	1	8	(<0.008;–;0.01;–)	1	8	(<0.01;–;0.011;–)	0.0014
1	7	(<0.007;–;0.01;–)	9	0	(0.008;0.014;0.042;0.011)	1	8	(<0.01;–;0.043;–)	0.0082
0	8	(–;–;<0.01;–)	3	6	(<0.005;0.008;0.022;0.005)	1	8	(<0.01;–;0.028;–)	0.27
1	7	(<0.04;–;0.095;–)	9	0	(0.114;0.143;0.301;0.062)	1	8	(<0.095;–;0.317;–)	–
0	8	(–;–;<0.035;–)	9	0	(0.052;0.065;0.131;0.025)	1	8	(<0.035;–;0.175;–)	–
1	7	(<0.026;–;0.06;–)	9	0	(0.061;0.078;0.17;0.035)	1	8	(<0.06;–;0.142;–)	–
0	8	(–;–;<0.025;–)	0	9	(–;–;<0.025;–)	0	9	(–;–;<0.025;–)	–
0	8	(–;–;<0.01;–)	9	0	(0.023;0.034;0.098;0.024)	1	8	(<0.03;–;0.069;–)	–
1	7	(<0.019;–;0.04;–)	9	0	(0.045;0.063;0.158;0.036)	1	8	(<0.04;–;0.203;–)	–
1	7	(<50;–;65.7;–)	9	0	(118.0;173.0;373.2;78.5)	1	9	(<50;–;426;–)	1000[b]
0	8	(–;–;<5;–)	0	9	(–;–;<5;–)	0	10	(<7.5;–;–)	90[c]; 300[d]
0	8	(–;–;<5;–)	5	4	(<2.819;3.223;5;0.565)	0	10	(<7.5;–;–)	90[c]; 300[d]
1	7	(<30;–;49.53;–)	9	0	(93.3;140.11;298.04;62.5)	2	8	(<30;59.5;319;86.5)	90[c] (Arom.)
1	7	(<10;–;14.16;–)	9	0	(20.2;30.6;71.2;15.6)	2	8	(<10;19.2;102;27.6)	–
0	8	(–;–;<0.02;–)	5	4	(<0.026;0.049;0.124;0.032)	0	8	(–;–;<0.02;–)	0.1
0	6	(–;–;<0.02;–)	0	6	(–;–;<0.01;–)	0	5	(–;–;<0.01;–)	0.9
0	6	(–;–;<0.02;–)	0	6	(–;–;<0.01;–)	0	5	(–;–;<0.01;–)	0.91
0	6	(–;–;<0.02;–)	0	6	(–;–;<0.04;–)	0	5	(–;–;<0.011;–)	0.91
5	3	(<0.22;0.428;0.673;0.167)	6	3	(<0.17;0.416;1.18;0.207)	2	6	(<0.089;0.127;0.392;0.1)	0.3
0	6	(–;–;<0.2;–)	0	6	(–;–;<0.18;–)	0	5	(–;–;<0.15;–)	0.64
0	6	(–;–;<0.36;–)	1	5	(<0.067;–;1.66;–)	0	5	(–;–;<0.38;–)	0.37
0	6	(–;–;<0.42;–)	0	6	(–;–;<3.54;–)	0	5	(–;–;<0.74;–)	0.3
3	5	(<0.042;0.051;0.097;0.018)	9	0	(0.158;0.365;0.58;0.164)	3	7	(<0.05;0.054;0.074;0.008)	0.24
8	0	(2.95;8.9;19.303;6.618)	9	0	(2.963;11.19;22.4;7.096)	10	0	(1.51;8.978;19.1;6.652)	12[e]
6	2	(<1.93;5.5;23.9;7.1)	9	0	(11.5;19.7;33.5;6.7)	5	3	(<2.5;5.6;15;4.1)	25[f]
8	0	(1.405;19.9;78.24;25.4)	9	0	(28.3;44.87;87.49;18.82)	8	0	(6.47;29.78;73.7;25.38)	–
8	0	(114.52;305.58;495.57;140.1)	9	0	(52.34;157.51;327;89.8)	8	0	(50.3;205.37;398;141.66)	–
8	0	(7.543;7.867;8.135;0.181)	9	0	(6.965;7.109;7.22;0.098)	8	0	(5.62;7.083;7.797;0.646)	–
8	0	(13.45;18.15;23.62;3.27)	9	0	(14.13;18.04;24.22;2.92)	8	0	(7.23;16.28;23.7;4.7)	–

Fig. 3). For C_{12}-C_{16} (one of the lighter and more volatile PHC fractions), GPT showed slightly better performance with a median removal of 32.1 ± 30.8 %, which might be related to the effect of the oil separator compartment in GPT designed to trap oils and volatile pollutants at the water's surface. Thus, GPT effluent quality followed almost the same level and pattern as observed at SW (thus, no significant difference in the EMCs of any OMPs between SW and GPT_{out}, as shown in Table S7) and did not considerably improve the stormwater quality (see NEPs in Fig. 2). Furthermore, OMP removal efficiencies by the GPT were often accompanied with large errors (most often median Err_{Re} between 30 and 50 %, Fig. 3).

3.2.2. Non-vegetated sand filter (SF) removal efficiency

In contrast to the GPT section, the SF moderately to substantially removed most of the OMPs from stormwater, although the removal efficiencies and errors for each OMP varied a great deal between events (Fig. 3). The observed variation in Re% was directly related to the ratio between the inflow EMC and LoQ of a given OMP. The closer the EMCs to the LoQ, the wider the range of Re% became (discussed in Section 4.6). The median Re%s were as follows: BPA: 43 ± 22 %, OP: 51 ± 24 %, Σ16PAHs: 61 ± 5 %, BaP: 73 ± 18 %, C_{10}-C_{40}: 60 ± 14 %, TSS: 70 ± 8 %, and turbidity: 57 %. Meanwhile, a negative and nearly zero median removal percentage was observed for NP (−5 ± 46 %) and TOC (9 ± 21 %). The difference in EMCs between GPT_{out} and SF_{out} were significant with relatively high Chi-squares ($X^2 > 4.9$ and $p < 0.05$) for OP, PAHs, C_{12}-C_{40}, TSS, and turbidity. However, the difference for BPA, BkF, and BbF (as well as NP and TOC) was insignificant (see Table S7) i.e. the SF performance was inadequate for these substances. Uncertainty analysis revealed that the absolute removal errors in SF_{out} were often higher for phenolic substances (median: 23–45 %), followed by TOC (21 %), PAHs (8–30 %, excluding Phen and DahA) and PHCs (13–20 %), and TSS (8 %).

In contrast to phenolic substances with lower treatment efficiencies, PAH and PHC removal by SF ranged between 55 and 90 % when their influent concentration (i.e. GPT_{out}) was at least four times bigger than the corresponding LoQ. Of the quantified PAH substances, the medium and high weight molecules Flth, BbF, Bper, and InP had the highest removal efficiencies (median > 70 %) with lowest errors (~10 %). On the other hand, Phen, and DahA had, relatively, the lowest efficiencies (median < 60 %) and highest Err_{Re} (≥30 %) (Fig. 3). Further, SF treated non-carcinogenic PAHs better than carcinogenic ones: ΣNon-car.PAHs ranged within 17–80 % (median 63 ± 7 %), whereas ΣCar.PAHs ranged within 0–78 % (median 57 ± 9 %). Among PHC fractions, a similar distribution of Re% among the events was observed for C_{12}-C_{16}, C_{16}-C_{35}, and C_{35}-C_{40} (like total PHCs) in SF, but the C_{12}-C_{16} removal errors were slightly (~6 %) higher than the other fractions (Fig. 3).

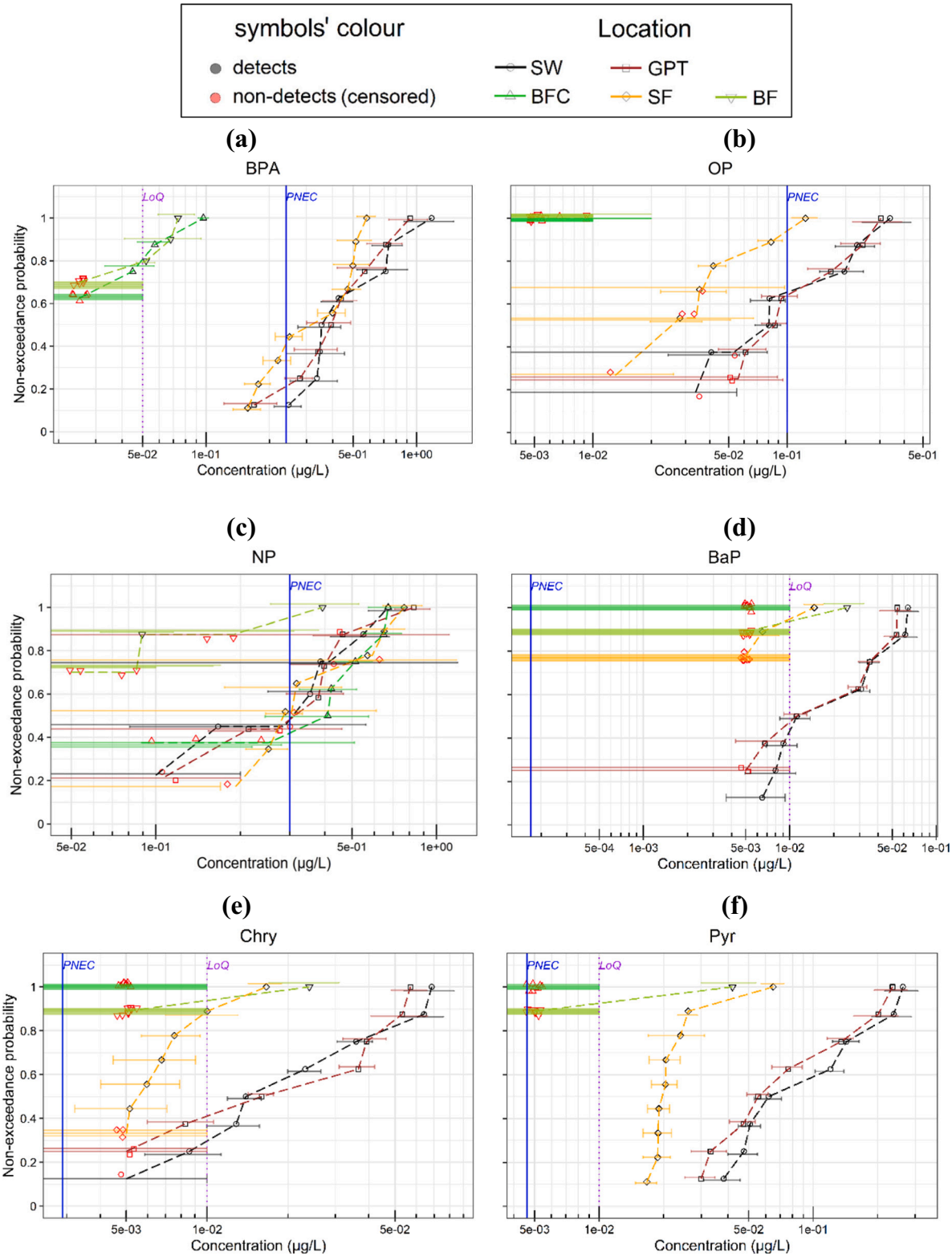

Fig. 2. Non-exceedance probability distribution (cumulative probability of the occurrence) of the calculated event mean concentrations (EMCs) at different sampling locations (SW, GPT_{out}, BFC_{out}, SF_{out}, and BF_{out}) for selected OMPs. Symbols represent the best EMC estimates connected by a dashed line for each sampling location, error bars represent the propagated uncertainties of EMCs, and the dotted blue lines represent the water quality objectives (dots and error bars have been moved a little (jittered) to avoid overlapping) (no single LoQ line could be applied for alkylphenols due to matrix interference).

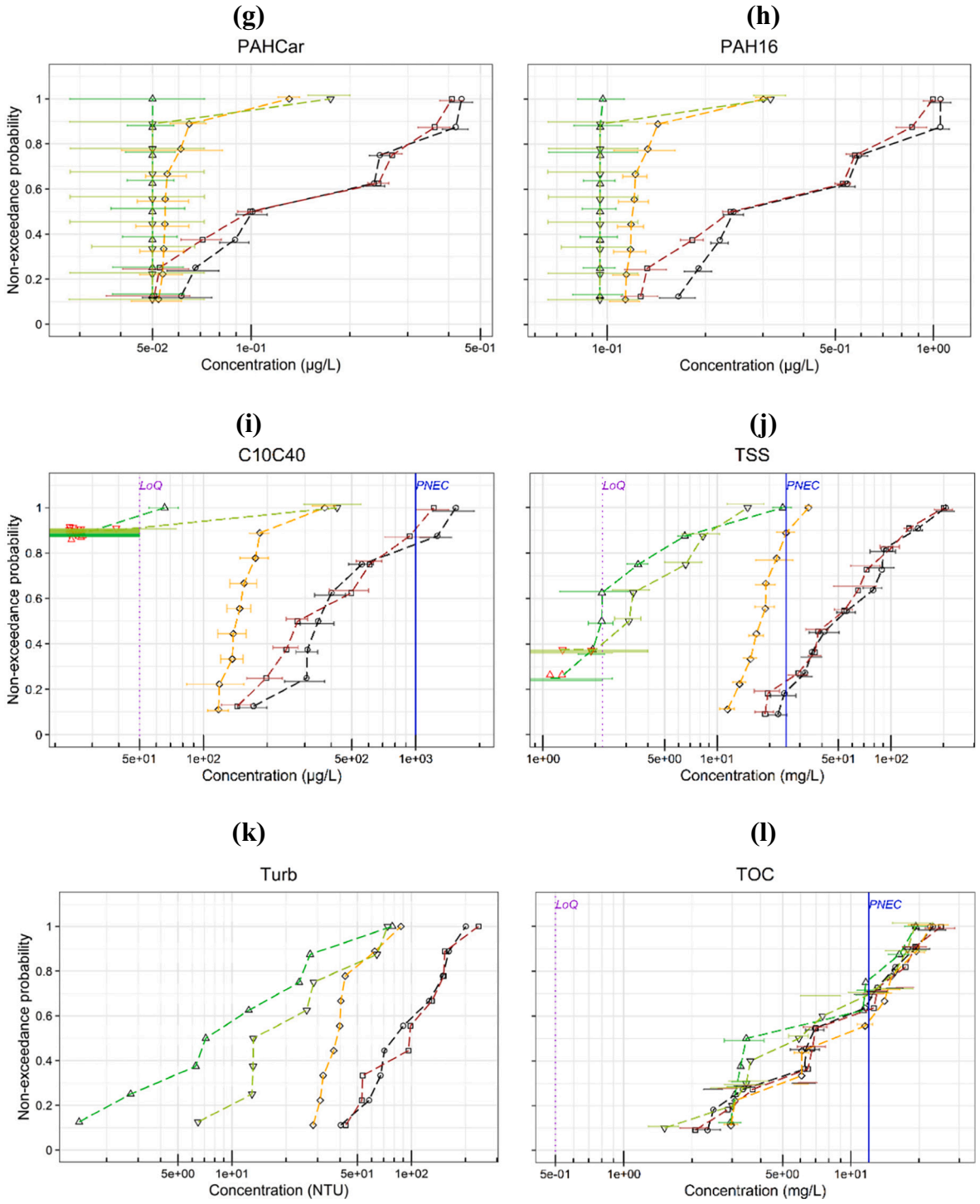

Fig. 2. (*continued*).

3.2.3. *Vegetated biofilters (BFC and BF) removal efficiencies*

Both vegetated filters (BFC and BF) performed better than the non-vegetated filter (SF) in improving stormwater quality. In general, both vegetated biofilters behaved similarly and were able to remove a majority of OMPs to a greater and better degree than SF. As shown in Fig. 3, the median Re%s for the BFC section were for BPA: 94 ± 6 %, OP: 95 ± 6 %, Σ16PAHs: 60 ± 6 %, BaP: 69 ± 33 %, C_{10}-C_{40}: 91 ± 9 %, TSS: 95 ± 3 %, and turbidity: 88 %. The corresponding treatment performances by the BF cell were for BPA: 91 ± 7 %, OP: 96 ± 4 %, Σ16PAHs: 75 ± 6 %, BaP: 69 ± 19 %, C_{10}-C_{40}: 91 ± 9 %, TSS: 95 ± 3 %, and turbidity: 79 %. The removal result showed that the differences between the performance of the vegetated and non-vegetated filters were significant for BPA, OP, NP (in BF only), Pyr, C_{16}-C_{40}, and TSS removals (Re% in BFC and BF > 90 %, which were at least 30 % higher than those in SF), but not as great for the rest of PAH substances and PHC fractions. As with SF, the BFC cell showed a negative or low removal efficiency for NP: −10 ± 66 % and TOC: 20 ± 18 %. Further, a lower TOC removal percentage (10 ± 27 %) was achieved by the BF cell. In contrast, BF treated NP efficiently but this was associated with a high uncertainty (median: 74 ± 19 %). NP was the only OMP for which BFC and BF significantly differed

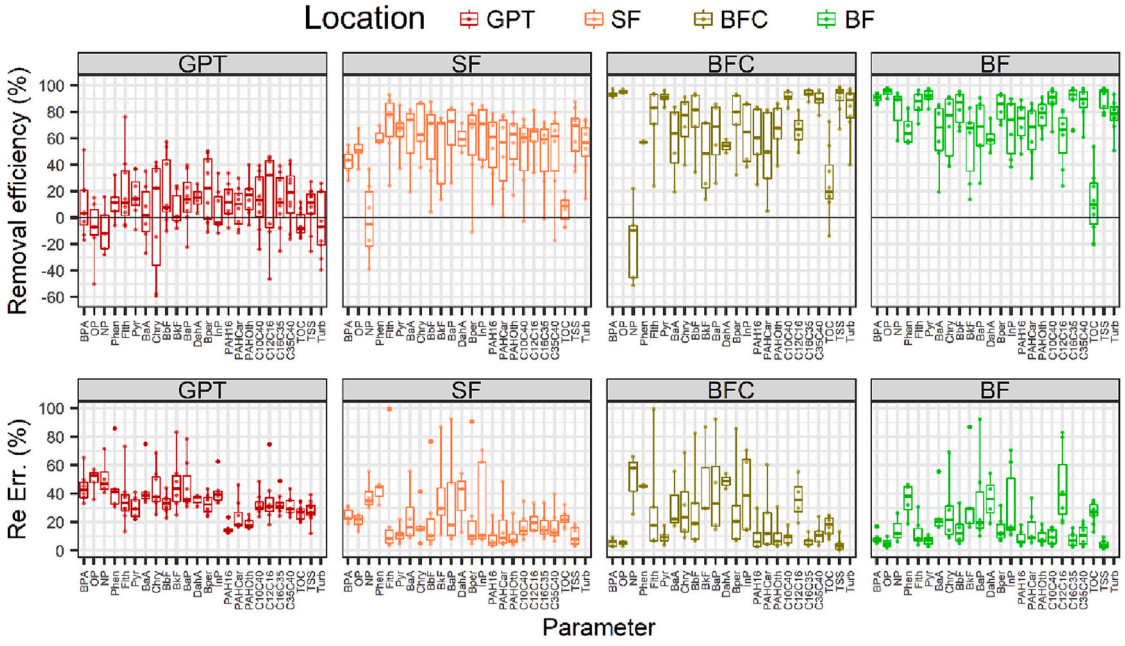

Fig. 3. Mean removal efficiencies (Re%) and errors (Err_{Re}%) of the parameters for various treatment sections.

regarding treatment performance ($X^2 > 6$ and $p < 0.05$) (discussed in Section 4.5). There were statistically significant differences between the EMCs at GPT_{out} and BFC/BF_{out} for phenolic substances, PAHs, PHC fractions, TSS, and turbidity, except for DahA (due to large analytical uncertainties leading to median $Err_{Re} > 38$ %) and TOC in the outflow of both vegetated cells, and for NP in BFC_{out} (see X^2 and p-values in Table S7). Although the maximum Re% for most OMPs reached between >70 % and 98 % (except for NP), the removal percentage for most OMPs varied in a wide range and was greatly affected by the influent EMC level with respect to LoQ (discussed in Section 4.6).

Furthermore, the uncertainty analysis revealed that, unlike the SF cell, BFC and BF exhibited low absolute removal errors (<15 %) for phenolic substances, except for NP in BFC which varied within a wide range of 20–67 %. As with the SF cell, the removal errors of PAHs varied greatly among the substances and events so that the medians oscillated between 8 % and 58 % but their maximum error could occasionally reach as high as 80 % (Fig. 3). Comparing both vegetated biofilters, PAHs' Err_{Re}% in BF was always moderately lower than that in BFC, although the errors calculated for PAH fractions (with medians <11 %) did not show a significant difference between the two cells. Likewise, BF and BFC had a similar error range of <20 % for total and heavier PHC fractions and TSS, while the errors for C_{12}-C_{16} reached 56 % in BFC and 83 % in BF (see discussion in Section 4.5).

3.3. OMP environmental risk analysis

The environmental risk of selected OMPs in untreated stormwater was determined and compared with that in the outflow of each treatment unit. Out of 32 parameters measured, 15 OMPs were not considered in our risk analysis due to: 1) no available PNEC (4 TPH fractions), or 2) all concentration values or a majority of them at any sampling location were below LoQ and LoQ < PNEC (i.e. no potential risk existed for Nap, Acel, Acen, Flu, Anth, and OPnEO and NPnEO; $n = 1, 2, 3$). Fig. 4 illustrates the total risk (R_T) for the other 17 parameters at different sampling locations.

The results revealed that, in the untreated stormwater, the R_T of TSS and 11 OMPs (BPA, OP, NP, and eight PAHs: Flth, Pyr, BaA, Chry, BbF, BaP, DahA, Bper) exceeded one, which may potentially pose a risk to the environment, while that was not the case for C_{10}-C_{40}, TOC, and other PAHs (i.e. Phen, BkF, and InP). Meanwhile, the associated errors for the total risk render our findings uncertain for OP and NP in SW since the error bars cross the critical risk threshold in those cases.

As shown in Fig. 4, GPT exhibited slightly a lower but almost equal R_T as observed in SW, meaning that GPT did not reliably reduce the environmental risk of OMPs. The SF section, however, did reduce the risk of OMPs and TSS compared to SW and GPT_{out} to some extent, except NP (discussed in Section 4.5). In the cases of TSS, OP, BaA, BbF, BkF, and probably Flth and DahA (considering the associated uncertainties), the SF section performed so efficiently that the risks with $R_T \geq 1$ dropped below one, to the safe level zone. On the other hand, SF did not reduce the risk levels of BPA, Pyr, Chry, BaP, DahA, and Bper sufficiently. For the vegetated biofilters, BFC and BF generally reduced the OMP risk levels to a greater extent than SF. In terms of environmental risk reduction, BFC and BF both performed equally efficiently for BPA, OP, Flth, BaA, BbF, Bper, and TSS, (R_T dropped down to safe levels below one), which previously had high risks at the SW and GPT_{out}. However, it cannot be concluded whether extremely hazardous OMPs (Pyr, Chry, BaP, and DahA) reached a safe level after BFC and BF treatment, as the very low PNEC concentration was below LoQ in the present study. As with the removal efficiency results, we did not see a significant difference between the R_T in BFC and BF outlets to identify the effect of chalk amendment on risk reduction.

4. Discussion

A comprehensive discussion of the concentrations, occurrence, and possible sources of OMPs in the untreated stormwater, as well as the comparison of our results with other major road runoff studies, can be found in (Beryani et al., 2023). The effect of each treatment unit in OMPs removal will now be discussed and compared with other studies.

4.1. Impact of gross pollutant trap (GPT)

The results revealed that the GPT section had, in general, a low removal efficiency which is in line with previous studies of sedimentation facilities, with a maximum 10–25 % removal for various substances such as TSS, metals, PAHs, NP, NP1EO, and NP2EO (Björklund et al., 2009; Lange et al., 2021, 2022; Pettersson et al., 2005). In some cases, the GPT effluent contained even higher concentrations for many OMPs than those in the stormwater (i.e. negative RE%) which showed that the

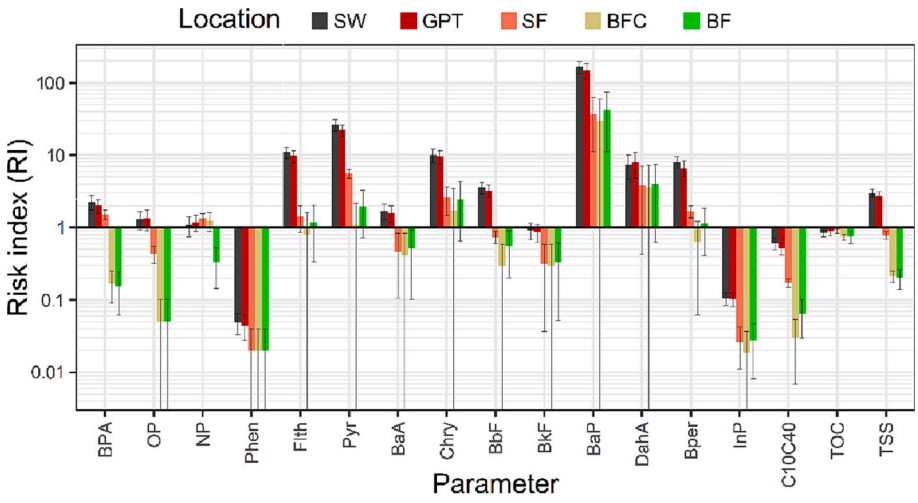

Fig. 4. Total risk (R_T) (with error bar) for selected parameters at different sampling locations.

GPT may sometimes release previously accumulated OMPs in the chamber. It is likely that the high turbulent inflow in the GPT might re-suspend the previously deposited sediments at the bottom of the chamber. In addition, the GPT's discharge system (i.e. pulse valve opening) could be a reason for sediment resuspension by causing a fluctuating and relatively high discharge flow from the facility when the valves were open.

Further, the GPT's low treatment efficiencies could be connected to the size of particles in relation to the GPT dimensions. Stormwater normally contains many fine particles which are not efficiently removed by such small sedimentation facilities (Li et al., 2005; Lieske et al., 2021; Pamuru et al., 2022). For this site, the GPT size was only about 15 % of Massachusetts's EPA minimum recommended capacity of ~6.3 mm runoff per ha impervious catchment for forebays (MDEP, 2022).

As a suggestion, larger ratio of GPT to catchment surface area alongside a continuous instead of stepwise discharge mechanism (so that the water is still evenly distributed over the filter cells) may improve the GPT performance.

4.2. Impact of sand filter media

In general, the SF cell improved the GPT's outflow quality (see Section 3.2.2), but it behaved differently in response to different OMPs depending on their physiochemical properties, as discussed below.

In this study, SF treatment performance was relatively less efficient for TSS (35–90 %; median: ~70 %) than previously reported for sand-based biofilters (>90 %) (e.g. Davis, 2007; Zarezadeh et al., 2018). The EMCs of TSS during five (out of nine) events were also higher than the SCM predevelopment target level of 20 mg/L suggested by Davis (2007). Turbidity removal by SF was even lower than TSS for most rain events (except E and J), which may suggest that a sand filter is less able to retain highly suspended, smaller colloids than larger/denser particles, as previously concluded by other studies (Davis, 2007; Lange et al., 2021). So, a high TSS removal may not be achieved by SF, especially for high flow intensities and high TSS loads. Conversely, there have been also non-vegetated facilities containing sand layers that had low TSS removal, due to fine material release from unestablished filter beds or long ADP adverse effects (Blecken et al., 2009; Davis, 2007), neither of which were the case in our study.

Regarding OMP removal, apart from the negative Re% for NP (separately discussed in Section 4.5), SF was mostly less efficient (and associated with slightly higher removal errors) for treating phenolic substances (i.e. BPA and OP), compared with quantified PAHs, PHCs and TSS. MWH-PAHs, HWH-PAHs and C_{16}-C_{40} fractions typically have a much higher particle-bound fraction compared to BPA, OP, and NP, because they have less solubility, higher hydrophobicity (larger K_{OW}), and more absorbability of organic content of sediments and Fe particles (larger K_{OC}) (see Table S4, Table S5, Table S6) (Andersson et al., 2018; Diblasi et al., 2009; Leroy et al., 2016). Therefore, the quantified PAHs and PHCs were more likely to be retained in the SF medium by a particle (TSS) filtering process (David et al., 2015; Furén et al., 2022) than the phenolic substances for which a higher dissolved/colloidal phase breakthrough may occur (Gasperi et al., 2022; Ruppelt et al., 2020; Shehab et al., 2020). Flanagan et al. (2018) also reported that alkylphenols are less efficiently removed by a bioswale facility than PAHs and PHCs, having higher partitioning to suspended solids. Consequently, the Re% of most PAHs (except Phen, Chry, BkF, DahA), and heavier PHC fractions (i.e. C_{16}-C_{40}) were strongly correlated with Re% of TSS and turbidity for the SF cell, whereas none of the phenolic substance removals were statistically associated with them. Our finding is consistent with the few available previous studies concluding that sand filters may not function adequately for treating more soluble OMPs such as triazine herbicides, biocides, and triclosan-methyl (Spahr et al., 2019).

We were not able to investigate the treatability of lighter PAH and PHC fractions by the SF (as well as the other filters) properly because they were not often quantified in the influent stormwater, probably due to partial loss through volatilization, and more limited use of gasoline with lighter fractions than diesel (Leroy et al., 2016). In addition, the ratio of C_{16}-C_{35} over C_{35}-C_{40} in SF_{out} increased for about 20 % of that in the inflow, which may again support our hypothesis that sand filtration basically performs better for retaining heavier hydrocarbon molecules due to their greater hydrophobicity and higher affinity to the solid phase.

Considering the results for different groups of OMPs, the predominant mechanisms responsible for the removal in a sand-based filter are most likely physical particle straining, colloidal attachment to the filter medium and, to a lesser extent, solute adsorption to the organic matter retained on the filter material surface. Therefore, a supplementary treatment unit with more reliable filtration and/or adsorption abilities will be required for treating more soluble OMPs such as phenolic substances as well as the dissolved fraction of all OMPs when a SF is the only existing filter cell in a biofilter facility. Although further detailed studies are needed to determine the exact size ranges of OMP particles that can be filtered using sand-based filtration, a vegetated biofilter may address the issue of more reliable treatment (see Section 4.3). As shown by other studies (Haritash and Kaushik, 2009; Mitchell et al., 2023), biological activities are presumably to be very low in the sand filter media (due to insufficient available carbon and nutrients to support a thriving microbial community and low bioavailability of many OMPs that have been quantified). Thus, the biodegradation contribution is probably very low

compared to the physiochemical filtration processes.

4.3. Impact of vegetation layer

To evaluate the effect of the vegetation layer, the OMP removal efficiencies of the vegetated sand filter (BF) were compared with the non-vegetated sand filter (SF). The presence of vegetation on the sand filter media substantially improved the removal of BPA, OP, NP, Pyr, Bper, Σ16PAHs, all PAH fractions except ΣLMW-PAHs, C_{10}-C_{40} (predominantly C_{16}-C_{40}), and, to a lesser extent, turbidity (Table S7). Two possible factors were suggested as the reason for the positive effect of vegetation during wet periods: plant-related processes (uptake, retention in root, preferential flow paths, and microbial degradation in the rhizosphere) and filtration processes by the (finer) topsoil layer on the vegetated filters.

Some laboratory biofilter column studies have already shown that certain plants can directly take up some hydrocarbons, such as naphthalene (up to 23 % for grass), and phenanthrene and pyrene to a certain extent (Kacálková and Tlustoš, 2011; Lefevre et al., 2012). However, the plant uptake is probably negligible here because most of the quantified OMPs were classified as organic substances with very low solubility and very high hydrophobicity (log $K_{OW} > 4$; see Tables S4–S6) which makes them unavailable for plant uptake through cell membranes (and therefore no subsequent processes such as phyto-accumulation/volatilization and metabolic transformation). However, they can still be retained and stabilized in the root epidermis (Leroy et al., 2015; Ruppelt et al., 2020). It is also believed that uptake, accumulation, and metabolic transformation of organic compounds by vegetation potentially occur when there is major contamination with contaminants more resistant to biodegradation (Imfeld et al., 2009), while generally low concentrations of OMPs were found in the highway stormwater and the GPT outflow. The plant-related effect is probably more relevant for the fate and treatment of OMPs during dry periods between the events (which was not specifically investigated in this study) where the surrounding area of plant root (rhizosphere) provides conditions that promote microbial decomposition (mineralization) of hydrocarbons retained in the soil (Dagenais et al., 2018; Muerdter et al., 2018). Nevertheless, further studies are needed to determine the direct effect of plant uptake by examining the plant tissues, especially for BPA and OP (and lower molecular weight PAHs if quantified also) with higher water solubility and less hydrophobicity (log K_{OW} around or less than four).

It is likely that filtration processes in the 3–4 cm vegetation topsoil are more relevant than direct vegetation uptake to better performance of the BF. As evidence, the presence of vegetation topsoil (sand to silty-sand) in BF and BFC noticeably decreased the infiltration rate and prolonged the filtration effect at the beginning of the runoff event (but not for most of the infiltration duration, following saturation towards the end of the event) compared to the SF. The OMP results agreed with Lange et al. (2021) and Fahlbeck Carlsson (2021) who observed slightly (but not statistically significantly) better removal efficiencies for microplastics (specially for the smaller particle size range 100–300 μm) and metals (both total and even more pronounced dissolved) by the vegetated filters (BF and BFC) compared to the SF, due to further particulate filtration capacity of the vegetation soil layer. During the filtration processes on particle-bound pollutants, a vegetation layer can greatly contribute to the removal of OMPs by trapping particles in the topsoil (especially larger ones, while smaller particles are further transported and removed in the underlying filter media (Chu et al., 2021)).

The vegetation soil layer was amended with 2.5–5 % organic mulch (supporting plant growth) which can contribute to the absorption of BPA, OP, NP, and colloidal/dissolved fractions of MMW-PAHs, HMW-PAHs and C_{16}-C_{40} that have higher log K_{OC} (see Table S4, Table S5, Table S6) (Duan et al., 2015; Furén et al., 2022; Hong et al., 2006). Beyond that, the organic matter from decaying vegetation (dead plant tissues) may enhance the removal by absorbing OMPs in the biofilters. The root surfaces may also play a role in OMP adsorption (Hutchinson et al., 2003).

Thus, the predominant processes responsible for the positive effect of vegetation in OMP removal can be linked to absorption by the vegetation soil and organic matter, as well as straining and sedimentation during filtration into the layer. In terms of practical applications, the results of this study supported the idea that vegetated biofilters are preferred over non-vegetated sand filters for OMP removal.

4.4. Impact of chalk amendment

Chalk ($CaCO_3$) is used to increase buffer capacity (to avoid acidification caused by microbial activities and consequent leaching) and compensate for low organic matter content of the filter media, both of which may improve solute adsorption on the solid phase (Søberg et al., 2019; Tondera et al., 2019). No comparable studies have been carried out so far to investigate the impact of $CaCO_3$ on bioretention for OMP removal. Studies of groundwater in geological chalk formations have suggested that the organic content of chalk grains can contribute to organic pollutant absorption (Graber and Borisover, 2003; Wefer-Roehl et al., 2001). However, previous tests on the filter materials of this site (Søberg et al., 2019) showed that chalk amendment does not considerably improve the organic carbon content of the BFC compared with SF and BF (Table S1). Hypothetically, chalk may also positively affect the physical deposition of particle-bound OMPs by changing water chemistry. Increasing pH and ionic strength (salt content) of stormwater greatly influences the precipitation of suspended solids (Behbahani et al., 2021; Diblasi et al., 2009; Randelovic et al., 2016).

Although the chalk amendment (10 % *w*/w crashed limestone in BFC) caused an increase in the event mean values of median pH (for about 0.8 with a significant difference between BF_{out} and BFC_{out}, Table S7) and median EC (for about 100 μS/cm, but not statistically significant), the results did not show any statistically significant difference between the performance of BF and BFC in OMP removal (except NP which was linked to a potential leaching; see Section 4.5). It is likely that chalk has no or only small practical importance in sand-based biofilter media (but if it does play a role, due to the significant impact of vegetation which already resulted in many non-detects in the BF and BFC effluents, its impact was not traceable). Thus, a general conclusion that chalk amendment is not useful for OMP treatment cannot be drawn based on this single study.

4.5. Potential leaching of alkylphenols

Comparing the results for the three different filter cells, inconsistent behavior was observed among phenolic substances exhibiting almost equivalent characteristics. While the removal of both BPA and OP were improved by the sand filtration and vegetation factors, NP exhibited negative removal efficiencies in BFC and SF, as well as a significant difference between BFC and BF. In general, negative removal, which suggests undetermined, additional sources of NP other than the inflow stormwater, can be either due to remobilization from the filter material or leaching from the facility's construction material and sampling equipment (Flanagan et al., 2019; Tondera et al., 2013). However, blank tests already showed no alkylphenols leaching from the sampling equipment. Remobilization was not probable from the filter media because a similar result to BF would be expected for SF and BFC if so. Therefore, it could be likely that there is NP leaching from the facility's construction material although this cannot be further identified at the site.

It was not possible to determine a specific source for potential OMP leaching from the facility's construction materials, including stormwater pipeline, filters' drainage pipes, cell separator membranes, and geotextiles. However, the three filter cells were separated by ethylene propylene diene monomer (EPDM) rubber membranes. EPDM could specifically be a potential source of alkylphenols, phthalates, and other

specified aromatics such as benzothiazole (highest potential), and perhaps PAHs, aliphatic $C_{24} - C_{35}$ hydrocarbons, and volatile organic carbons (VOCs) (Magnusson and Mácsik, 2017; Nilsson et al., 2008) even though the leaching from newer, underground EPDM rubber membranes is expected to be much lower. Other studies have also demonstrated that phenols and their derivatives were found in the water in contact with new and recycled EPDM (Magnusson and Mácsik, 2017; Nilsson et al., 2008). Furthermore, in this site, the geotextile used for covering the EPDM membrane is made of polypropylene which could be another potential source of NP leaching. Although polymers of low polarity such as polypropylene are less sensitive to chemical degradation, trace leaching of their additives into the surrounding soil water may occur mainly when microparticles have been formed from the geotextile (Wiewel and Lamoree, 2016). Previous field studies have shown potential emissions of BPA, NPs, and OPs (in the order of 10 ng/L) from the polypropylene geotextile and plastic drain pipe used in a biofilter swale and a green roof system (Flanagan et al., 2019; Gromaire et al., 2014).

As a general observation, it should also be noted that the conclusion regarding NP leaching might be influenced by the high uncertainties in NP removal efficiencies (due to matrix interference and LoQ fluctuation).

4.6. Uncertainty analysis

There were several sources of uncertainties in EMCs and removal calculations, which are discussed below.

4.6.1. EMC calculation

Analytical and sampling constraints linked the EMCs with uncertainties which may influence the accuracy and interpretation of EMCs. Our investigations clearly illustrated that the analytical measurement uncertainties (δ_i) and the number of censored data among subsamples of a certain OMP are critical in the estimated EMC error for a given event. Fig. S5 shows EMC errors associated with each OMP at different sampling locations. This reveals that EMC errors for all non-censored OMPs mostly varied between 10 % and 40 % (medians: 15–31 %). The median EMC errors of phenolic substances at SW and GPT_{out} were slightly higher than PAHs and PHCs due to higher δ_i (about 10 %) in subsamples. In SF, however, contrasting behavior was often observed because the number of censored EMCs for phenolic substances was lower than for most PAHs, which dominated the effect of the δ_i difference. It was not possible to analyze and compare the EMC errors of the different OMPs at BFC_{out} and BF_{out} due to the many censored mean concentrations ($0 < EMC < LoQ$). Furthermore, in the cases of NP and OP, matrix interference during the analysis of given subsamples adversely affected their LoQs and increased the EMC error range spread as a result of using the MC method. The EMCs of NP were affected by this, particularly at GPT_{out}, SF_{out}, and BFC_{out}.

A minor source of uncertainty in EMC calculations was the MC method itself. The EMC uncertainty level estimated by the MC simulation is always lower than the original δ_i in subsamples. Therefore, as shown in Fig. S5, the EMC errors for PAH fractions were about 10 % lower than those for PAH substances at SW, GPT_{out}, and SF_{out}, since the MC method was applied twice for PAH fraction EMC estimation.

Uncertainty in the volume calculation for the first subsample taken from SW and GPT (v_1) was another minor source of EMC errors. The uncertainty in v_1 was due to unknown, pre-existing stormwater in the GPT chamber (max. 21 m^3) before the start of the rain. Considering the number of subsamples, v_1 errors did not significantly contribute to the overall uncertainty of EMCs at those points (for more information refer to (Beryani et al., 2023).

4.6.2. Removal efficiencies

Censored and low concentrations of OMPs in stormwater samples may not only increase the EMC uncertainties but also influence the calculation of the removal efficiencies in the facility. The results showed that, since most EMCs at BFC_{out} and BF_{out} and almost half of those in SF_{out} were censored ($EMC_{out}/LoQ \approx 0.5 = const.$), then Re% and Err_{Re}% were considerably influenced by the ratio of influent concentration (EMC_{in}) to LoQ. As illustrated in Fig. 5, the closer EMC_{in} is to the LoQ, the lower the Re% and higher the corresponding Err_{Re}%. This trend was clearly observed for the filter cells when reaching a EMC_{in}/LoQ ratio below a certain threshold which was different for each filter cell i.e. 4 in SF, 12 in BFC, and > 15 in BF. This implies that the calculated Re% at the EMC_{in}/LoQ ratios below the threshold were underestimated in the filter cells, otherwise they can be assumed to be real percentages. As the removal performances of BF and BFC were higher than those of SF, a higher threshold was observed for BF and BFC. So, a higher EMC_{in} should have been recorded in order to calculate the real Re% in the vegetated filters (to be able to quantify OMPs in the effluent).

BPA influent concentrations were often above the ratio threshold defined for each cell. However, PAH substances and PHC fractions often fell below the threshold. This is the main reason why, for the BFC and BF cells, the removal efficiencies for most PAHs and PHCs (particularly lighter weight fractions due to lower EMC_{in}/LoQ ratios) were estimated at lower levels than expected with a wider range of errors. A few other studies have also stated that the removal efficiencies are lower and the errors higher when the OMP concentrations are closer to LoQ because of higher analytical uncertainties (Choubert et al., 2011; Diblasi et al., 2009; Flanagan et al., 2018; Ruppelt et al., 2020). Ruppelt et al. (2020) reported removal uncertainties of 30–100 % for low inflow concentrations of OMPs (<2.5 × LoQ), but <30 % for high concentration levels (>10 × LoQ) in pilot-scale sand columns, which are comparable with our results.

Another finding shown in Fig. 5 is that those points without censored effluent concentration data for a given filter cell ($EMC_{out}/LoQ > 1$) did not conform to the trend curve, meaning that the Re% represents a real removal percentage and has not been underestimated (or overestimated), so the corresponding removal error is less affected by the EMC_{in}/LoQ ratio. It is worth noting that due to the poor performance of GPT, no clear association between Re% and EMC_{in}/LoQ was observed.

The uncertainty analysis showed that the removal percentages/errors calculated for SF were less affected by EMC errors so that the efficiency values are more reliable for SF, while not for BFC and BF, since a greater number of data points were below the defined threshold in the two vegetated filters.

As a general suggestion, opting for analytical methods that have as low LoQs as possible and also sampling a larger number of rain events are recommended to lower the abovementioned uncertainties in OMP concentration/removal studies.

4.7. Risk analysis vs. removal efficiency

A study of runoff quality has already shown that BPA, OP, NP, and eight PAHs including Flth, Pyr, BaA, Chry, BbF, BaP, DahA, Bper (as well as TSS) frequently exceeded their PNECs, suggesting that they may potentially pose a risk to the receiving water body (Beryani et al., 2023). Previous road runoff quality studies have also reported that these OMPs are among the most frequently detected micropollutants in stormwater with concentrations typically exceeding their EQSs (Mutzner et al., 2022). Thus, in this study, two methods were used to examine the performance of the TT in removing/mitigating the effects of OMPs: 1) removal efficiencies in the separate TT units, and 2) total risk assessment and risk reductions by the TT units. Although calculating removal efficiency is the conventional way to evaluate the treatment capacity of a facility, it may not be enough to see the entire picture of SCM functionality in relation to environmental impacts of the actual concentration levels in the outlet. Just as with the removal performance results, the TT reduced the risks of OMPs differently – slightly to substantially – depending on OMP and treatment unit type. A few other data-driven or hypothetical studies on the risk assessment of biofilter outflows have shown that such systems can reduce health risks associated with heavy

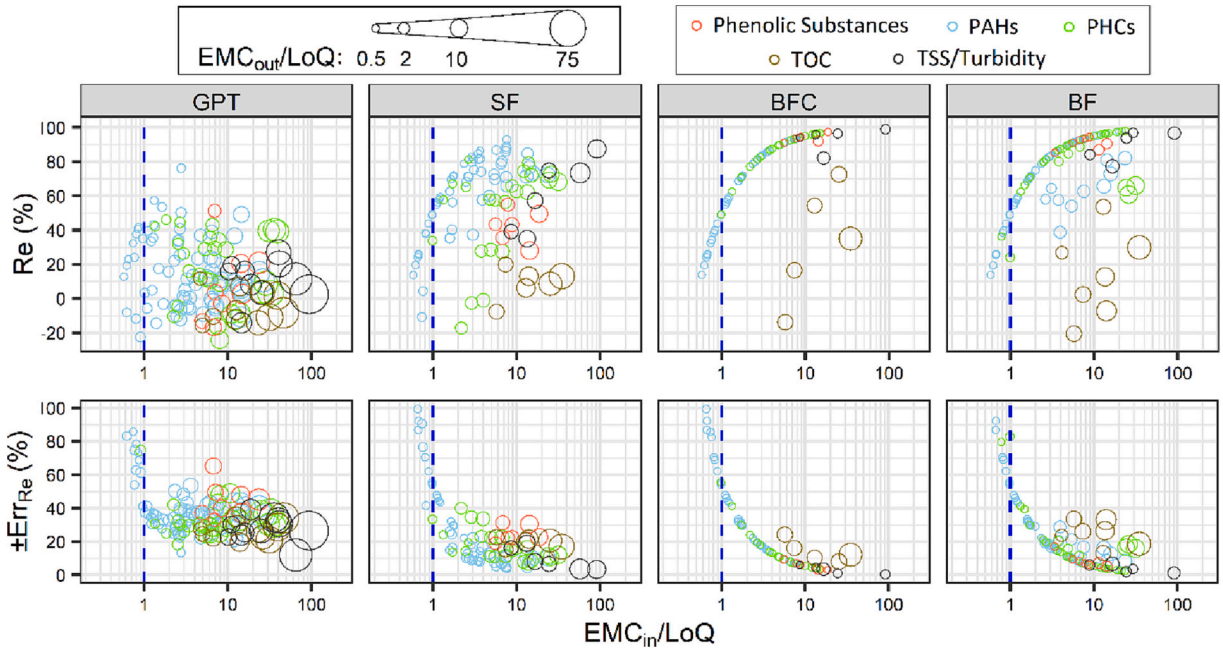

Fig. 5. Removal efficiencies (Re%) and their associated errors (Err_{Re}%) with respect to the ratio of influent/effluent mean concentration to the limit of quantification for different OMPs and rain events.

metals and pathogens (if combined with UV treatment) to a level such that the effluent water quality meets different reuse purposes (e.g. irrigation and toilet flushing) (Fang et al., 2021; Murphy et al., 2017).

Regarding the effectiveness of the evaluated factors (i.e. pretreatment, sand filtration, vegetation, and chalk amendment), the same results were achieved in terms of risk reduction, in general. However, for some OMPs, there were differences between removal efficiency results and what their risk ranking suggested. Sometimes, the EMCs in the outflows might pose a potential risk ($R_T > 1$), while a high Re% was often observed for them by a given treatment unit. This was the case for Flth and BaP in all filter cells and Pyr in BFC and BF. Conversely, Re% can be low, and thus assumed inadequate, while the outflow concentrations may not be a concern from an ecotoxicological perspective. This was the case for Phen, BaA, and TOC in all cells, NP in BF, as well as OP, BbF, C_{10}-C_{40}, and maybe TSS in SF. Other than these two situations, a similar conclusion was reached for OMPs in both methods, meaning that a low Re% agreed with a high risk ranking and vice versa.

The proposed risk-based approach can be a useful management tool that complements removal studies (Fang et al., 2021) and assists with decision-making and setting pollutant priorities on a comparative basis (Figuière et al., 2022). Meanwhile, to improve the level of confidence in the results of risk analysis, sampling more rain events would have been beneficial to obtain more accurate occurrence probabilities and to cover a wider range of rain depths and intensities, especially when the TT cannot cope with the excess amount of water and bypasses it during heavy rainfall.

5. Conclusion

This study contributed to the understanding of OMP removal performance of stormwater treatment trains by evaluating three biofilter cells combined with a pretreatment unit under field conditions. The findings of this study are important for future design modifications of such systems as well as strategies in choosing BMP for road catchments.

First, GPT removal pretreatment was not reliably efficient for any of the OMPs, TSS, and turbidity (medians often <20 %), mainly due to design shortcomings. Second, the non-vegetated sand filter (SF) performed moderately to sufficiently (median removal of 50–80 %) for PAH and PHC treatment, but weakly to moderately (<50 %) for the more hydrophilic phenolic substances. Third, a vegetation layer in biofilter cells (BFC and BF) improved the sand filtration removal substantially so that the EMCs of OMPs were most often below the LoQs, and TSS below 10 mg/L. The additional filtration and absorption capacity of the topsoil was assumed to be the main reason for the positive effect of the vegetation layer. Fourth, we could not quantify the effect of chalk amendment on OMP removal (due to many non-detects in BF and BFC outflows). Furthermore, NP leaching from the biofilters' construction material most likely occurred, as it was sometimes measured at higher levels in the filter cell outlets.

This study clearly demonstrated that uncertainties can be high for OMPs in field investigations under real conditions and low concentration levels; it is therefore important to consider them when analyzing the removal performance of biofilter treatment systems. As a general finding, by decreasing EMC_{in}/LoQ, the efficiency decreases and the error correspondingly rises. It was found that SF was less affected by the inflow concentration, therefore, showed lower removal errors compared with BFC and BF.

Finally, the pollutant risk analysis provided a more comprehensive tool for stormwater quality assessment which accounted for not only concentration analysis but also other influential factors in ecological impact assessments. According to the risk rankings, the overall treatment performance of the TT was reliable/adequate ($R_T r \ll 1$) and robust (>90 % risk reduction) for InP and C_{10}-C_{40}, moderate ($R_T < 1$ but probably not adequate enough risk reduction) for OP (only in SF_{out}), BaA, BbF, BkF, and TSS (only in SF_{out}), but insufficient ($R_T > 1$) or unreliable (low risk reduction) for BPA (only in SF_{out}), NP, Flth, Pyr, Chry, BaP, DahA, Bper, and TOC. Including such risk analyses in future stormwater treatment research is highly recommended.

CRediT authorship contribution statement

Ali Beryani: Conceptualization, Methodology, Investigation, Validation, Formal analysis, Visualization, Writing – original draft, Writing – review & editing. **Kelsey Flanagan:** Conceptualization, Methodology, Validation, Formal analysis, Writing – review & editing. **Maria Viklander:** Supervision, Writing – review & editing, Project administration,

Funding acquisition. **Godecke-Tobias Blecken:** Conceptualization, Methodology, Validation, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

Not applicable.

Data availability

All the data generated/analyzed in this study are included in the manuscript, supplementary information. The dataset is available at https://doi.org/10.5878/nny1-2045.

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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.2023.165734.

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