



# A case study of organic micropollutants in a major Swedish water source – Removal efficiency in seven drinking water treatment plants and influence of operational age of granulated active carbon filters

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## HIGHLIGHTS

- 163 organic micropollutants (OMPs) were analysed in river, raw and drinking water from Sweden.
- Total concentrations of 27 detected OMPs increased downstream the river.
- Removal efficiency of OMPs was affected by treatment strategy and operational age of GAC.
- Breakthrough in GAC filters occurred in the order DOC, PFASs and other OMPs.
- Topping up with a portion (~10%) of fresh GAC appeared to improve removal of DOC and PFASs.

## GRAPHICAL ABSTRACT



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## ABSTRACT

A wide range of organic micropollutants ( $n = 163$ ) representing several compound categories (pharmaceuticals, pesticides, *per*- and polyfluorinated alkyl substances, flame retardants, phthalates, food additives, drugs and benzos) were analysed in water samples from the Göta Älv river (Sweden's second largest source water). The sampling also included raw water and finished drinking water from seven drinking water treatment plants and in addition a more detailed sampling at one of the treatment plants after six granulated active carbon filters of varying operational ages. In total, 27 organic micropollutants were detected, with individual concentrations ranging from sub  $\text{ng L}^{-1}$  levels to  $54 \text{ ng L}^{-1}$ . The impact of human activities along the flow path was reflected by increased concentrations downstream the river, with total concentrations ranging from  $65 \text{ ng L}^{-1}$  at the start of the river to  $120 \text{ ng L}^{-1}$  at the last sampling point.

The removal efficiency was significantly ( $p = 0.014$ ; one-sided *t*-test) higher in treatment plants that employed granulated active carbon filters ( $n = 4$ ; average 60%) or artificial infiltration ( $n = 1$ ; 65%) compared with those that used a more conventional treatment strategy ( $n = 2$ ; 38%). The removal was also strongly affected by the operational age of the carbon filters. A filter with an operational age of 12 months with recent addition of ~10% new material showed an average removal efficiency of 92%, while a 25-month old filter had an average of 76%, and an even lower 34% was observed for a 71-month old filter. The breakthrough in the carbon filters occurred

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in the order of dissolved organic carbon, *per*- and polyfluorinated alkyl substances and then other organic micropollutants. The addition of fresh granulated active carbon seemed to improve the removal of hydrophobic organic compounds, particularly dissolved organic carbon and *per*- and polyfluorinated alkyl substances.

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## 1. Introduction

In Sweden, a large proportion of drinking water (DW) is produced from surface water, either directly (~50%) or *via* artificial infiltration (~25%) (Svenskt Vatten, 20160823). It is therefore of high importance to protect surface waters used as drinking water sources from chemical pollution. The widespread use of synthetic organic chemicals in society (Benotti et al., 2009; Geissen et al., 2015; Padhye et al., 2014; Richardson and Ternes, 2014; Ruff et al., 2015; Schwarzenbach et al., 2006; Stuart et al., 2012) entails the potential release of organic micropollutants (OMPs) with toxic, persistent and bioaccumulative properties into source water areas, and hence deterioration of raw water quality.

Conventional drinking water treatment processes (e.g. flocculation or rapid sand filtration) are not efficient for the removal of many commonly occurring organic micropollutants (OMPs) (Margot et al., 2013; Stackelberg et al., 2007; Ternes et al., 2002; Tröger et al., 2018). Although granulated active carbon (GAC) filtration may improve the removal rate (Margot et al., 2013; Stackelberg et al., 2007; Ternes et al., 2002), this is not always the case (Tröger et al., 2018) and complete removal is often not achieved (McCleaf et al., 2017; Tröger et al., 2018). GAC removal efficiency for OMPs often decreases quickly with time (Kennedy et al., 2015; McCleaf et al., 2017) and is negatively affected by the presence of dissolved organic matter (Lavonen et al., 2015; McCleaf et al., 2017). Even at low levels (ng L<sup>-1</sup> range) of OMPs (e.g. endocrine-disrupting compounds), insufficient removal could lead to human exposure to hazardous chemicals (Falconer et al., 2006). A recent example is the human exposure to *per*- and polyfluorinated alkyl substances (PFASs) through drinking water (Hu et al., 2016; Sharma et al., 2016; Zafeiraki et al., 2015) and drinking water-related bioaccumulation of PFASs in the exposed population (Gebbinck et al., 2015; Gyllenhammar et al., 2015). Other DW micropollutants of concern include flame retardants (Garcia-Lopez et al., 2010; Wang et al., 2011), pesticides (Badach et al., 2007; Fava et al., 2010; Mekonen et al., 2016; Petrovic et al., 2003) and pharmaceuticals and personal care products (PPCPs) (Stackelberg et al., 2007; Webb et al., 2003).

The concentrations of OMPs in surface waters are largely influenced by upstream point sources, such as effluents from wastewater treatment plants (WWTPs), hospitals or industries (Gago-Ferrero et al., 2017; Loos et al., 2013; Söregård et al., 2019). In recent years, there has been growing awareness of the occurrence of OMPs in surface waters in Europe, leading to improved water management, e.g. the 20-year-old Drinking Water Directive (98/83/EC) is under review, and the list of priority substances in the Water Framework Directive (WFD) was expanded in 2011 (Directive, 20190620). In Switzerland, the presence of OMPs in raw waters has led to legislative action requiring improved post-treatment at municipal WWTPs (McArdell, 2016), while in Sweden policymakers are stimulating the development and implementation of additional (advanced) full scale post-treatment steps (e.g. ozonation) for the removal of OMPs. Prior to the installation of advanced treatment steps and potentially unnecessary but costly upgrading of existing DWTPs, it is of interest to obtain a better quantification of the presence of OMPs in raw water sources to provide a sound basis for future decisions.

The combination of a cold climate, large forest cover and mostly thin soils poses a challenge to accessing high-quality raw water for drinking water production in many Nordic countries due to high levels of dissolved organic carbon (DOC), with levels typically between 4 and 15 mg L<sup>-1</sup> (Lavonen et al., 2015). The presence of DOC degrades the efficient run time of the GAC filters with respect to the removal of OMPs,

as has been shown in experimental studies (Pramanik et al., 2015), but little is known about the relationship between GAC age and OMP removal efficiency in full-scale plants.

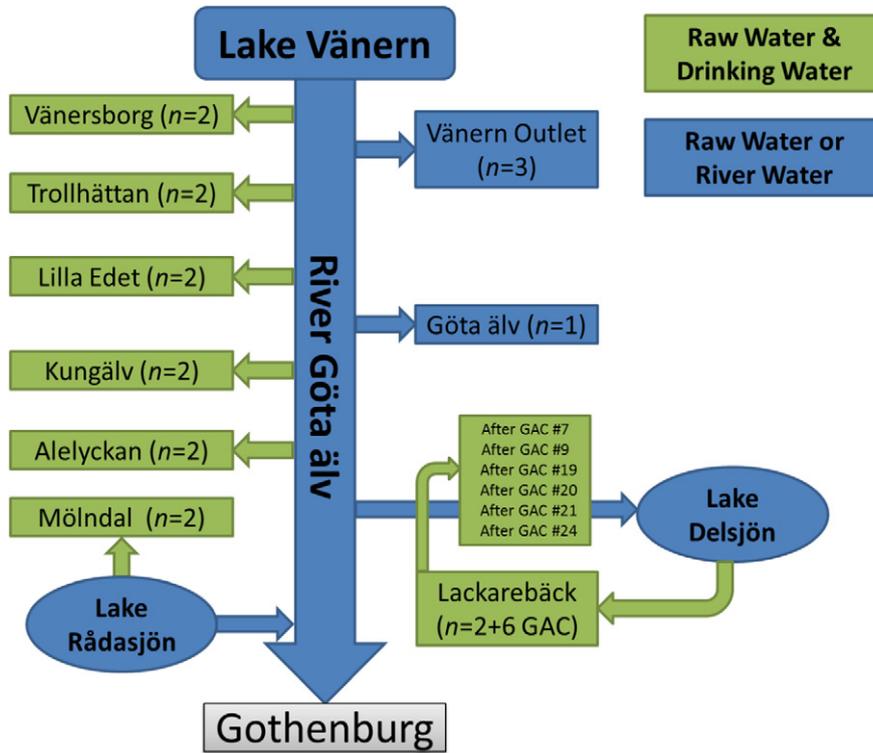
In light of the current situation, a better understanding of the presence and behaviour of OMPs in full scale treatment processes is needed. Most previous studies have focused on either pilot-scale studies or a single full scale DWTP (Appleman et al., 2014; Escher et al., 2011; Kennedy et al., 2015; Pramanik et al., 2015; Stackelberg et al., 2007). The overall aim of this study was to investigate the occurrence and removal of a large variety of OMPs in municipal drinking water produced from surface water and how different full-scale treatment strategies impact the quality of the finished drinking water. Seven DWTPs along the Göta Älv river were studied. The river is Sweden's second largest raw water source, but also the recipient of contaminated water, e.g. treated sewage water, storm water discharges, leakage from areas with contaminated soils, and direct contamination from boat traffic. Besides a number of regulated OMPs (e.g. pesticides), little is known about OMP levels either in raw water or the finished drinking water. More than 160 OMPs were measured in river water, raw water and finished drinking water from the selected DWTPs, which use a variety of different pre-treatment and treatment strategies, including artificial infiltration and GAC filtration. One of the major DWTPs included in the study has a multi-parallel GAC-filtration system with different GAC ages, which allowed investigation of the influence of the operational age of GAC filters on OMP removal efficiency. To the best of the authors' knowledge, this study is the first to evaluate the relationship between the operational age of parallel GAC filters and OMP removal efficiency in a full-scale DWTP.

## 2. Materials and methods

### 2.1. Sampling locations

The Göta Älv river originates from Lake Vänern, Sweden's largest lake, which covers an area of 5650 km<sup>2</sup>. At ambient flow conditions (ca. 565 m<sup>3</sup> s<sup>-1</sup>), water travels the 93-km-long flow path in around 50 h. The catchment area for the river is 3500 km<sup>2</sup>, with a population density of 270 people per km<sup>2</sup> (Göta Älvs Vattenvårdsförbund, 2015). Due to the large upstream Lake Vänern, the total dissolved organic carbon (DOC) concentration in Göta Älv shows only slight seasonal variations, with the level generally staying within the range of 3.5–4.5 mg L<sup>-1</sup> (Miljödata, 20190816). The river passes through six different municipalities, including Gothenburg (Sweden's second largest city), and ultimately flows into the Kattegat, a strait that forms part of the connection between the Baltic Sea and the North Sea. There are several heavy industries located along the river, including manufacturing and chemical production plants, which could be of concern for the overall water quality (Göta Älvs Vattenvårdsförbund, 2015). Along the path included in the current study area (upstream of Gothenburg; Fig. 1), wastewater from ~80,000 people is discharged along the river from >20 small and large sewage treatment facilities. The two largest point sources of OMPs are the Trollhättan WWTP (~50,000 people) and Vänersborg WWTP (~30,000 people). The wastewater accounts for ~0.1% of the water flux in the river (*i.e.* a 1000-fold dilution).

In total, seven different DWTPs produce drinking water (Fig. 1) for approximately 750,000 people, with a supply capacity ranging from 7500 to 300,000 people per plant. The DWTPs use different strategies to obtain raw water. Lackarebäck DWTP and sometimes Alelyckan DWTP, use water that has been pumped from Göta Älv to a lake system (Lake



**Fig. 1.** Schematic overview of the sampling sites (green boxes). Raw water and drinking water were sampled at all DWTPs. At Lackarebäck DWTP, six additional samples were collected after six different GAC filters. Lake Delsjön receives its water mainly from Göta Älv and constitutes the raw water for Lackarebäck DWTP. Lake Rådasjön is a separate water body that connects to Göta Älv downstream of the intake for Mölndal DWTP.

Delsjön), Mölndal DWTP takes its raw water from a lake upstream in the river system (Lake Rådasjön), while the remaining plants use water directly from the river all year round (Fig. 1 and Table 1 for details).

The treatment strategies also vary between the DWTPs (Table 1 and Fig. A1 in the Appendix). All the plants use classical coagulation, flocculation and/or filtration as major microbial barriers. Three plants had additional barriers: Trollhättan used slow sand filtration, Kungälv used artificial infiltration and Lackarebäck used ultrafiltration. Four of the DWTPs had GAC filters to improve the removal of taste, odour and unwanted chemical compounds.

In total, 24 water samples (12 L each) were collected along the river and from the seven DWTPs during a two-day sampling campaign (20–21 September 2016). The samples included river water collected near the outlet of Lake Vänern (Vänern outlet,  $n = 3$ ) and from Garn (~40 km downstream the outlet; Göta Älv  $n = 1$ ), raw water and finished drinking water from the seven DWTPs ( $n = 2 \times 7 = 14$ ), and

detailed sampling inside the largest DWTP (Lackarebäck;  $n = 6$ ). The additional Lackarebäck samples were taken after the water had passed through parallel GAC filters. On one occasion, four of these filters were each provided with a small portion (~10%) of new GAC material (Table A1 in the Appendix). To account for this addition, an adjusted operational age was calculated according to the equation:

Adjusted operational age

$$= \frac{(Age_{original} * Volume_{original} + Age_{addition} * Volume_{addition})}{(Volume_{original} + Volume_{addition})}$$

This correction resulted in an adjusted operational age of 1–3 months less than the original operational age. The timing of the addition of the new GAC material influences the filter's removal efficiency of organic compounds (Moona et al., 2018) including OMPs and DOC. The consequences of this in the current study are discussed further in Section 3.3.

**Table 1**

Sampling sites and information on the DWTPs. Treatments are divided into the following categories: (i) using GAC, (ii) using artificial infiltration and (iii) using neither GAC nor artificial infiltration. Detailed treatment schemes are displayed in Fig. A1 in the Appendix, and the operational ages of the individual GAC filters at Lackarebäck are given in Table A1 (Appendix). N/A = not applicable.

Sampling site	Raw water	Production (m <sup>3</sup> day <sup>-1</sup> )	# of users	GAC filter: # of filters; operational age and type	Treatment category
Vänersborg DWTP	Göta Älv	7700	28,000	No GAC	iii
Trollhättan DWTP	Göta Älv	13,200	50,000	No GAC	iii
Lilla Edet DWTP	Göta Älv	2000	7500	1 filter; 12 months Filtrisorb 200	i
Kungälv DWTP	Göta Älv	5000	36,000	No GAC	ii
Alelyckan DWTP	Göta Älv (Lake Delsjön)	91,000	250,000	14 filters; 33 months (average age) 12 * Filtrisorb 200 and 2 * cocoa nutshell	i
Mölndal DWTP	Lake Rådasjön	14,400	63,000	2 filter; 18 months (average age) Filtrisorb 200 and cocoa nutshell	i
Lackarebäck DWTP	Lake Delsjön	87,000	300,000	16 filters; 42 months (average age) 2 * Filtrisorb 200, 8 * TL830 and 6 * cocoa nutshell	i
Vänern outlet	Göta Älv	N/A	N/A	N/A	N/A
Göta Älv	Göta Älv	N/A	N/A	N/A	N/A

Triplicate samples for determination of DOC were collected in rinsed (Milli-Q water), pre-burnt (400 °C) glass bottles at each site, and in addition a triplicate sample was collected just before the GAC filters in Lackarebäck to be able to calculate the removal efficiency for DOC.

Triplicate samples (Vänern outlet) were also collected for quality control of the chemical analysis (analytical precision of the method using real samples). The samples were collected in stainless steel containers (Sharpsville Container Corporation), which were pre-cleaned with ethanol followed by Milli-Q water. The containers were rinsed with the sample water three times prior to water collection. At locations where it was not feasible to collect the sample directly into the container, a stainless steel bucket was used to transfer the sample to the container. The samples were stored at +4 °C until extraction, which was carried out within one week of sampling. The location of the sampling sites and DWTP details are given in Table 1.

## 2.2. Reference standards and chemicals

The analytical method targeted 163 OMPs from a wide range of compound classes including pharmaceuticals ( $n = 48$ ), pesticides ( $n = 79$ ), PFASs ( $n = 13$ ), flame retardants ( $n = 11$ ), phthalates ( $n = 3$ ), a food additive ( $n = 1$ ), drugs ( $n = 3$ ), and benzos (benzotriazoles/benzothiazoles) ( $n = 5$ ) (Table A2 in the Appendix). The selection of micropollutants was based on compounds previously suspected to be present in drinking water (Benotti et al., 2009; Ivancev-Tumbas, 2014; Kumar and Xagorarakis, 2010; Mekonen et al., 2016; Padhye et al., 2014; Petrovic et al., 2003; Segura et al., 2011; Webb et al., 2003; Westerhoff et al., 2005; Hu et al., 2016) and the availability of reference standards. All the pesticides included were part of the Swedish national surface water screening programme (Jansson and Kreuger, 2010). The instrumental methods have previously been used (Tröger et al., 2018), but were expanded by 30 additional OMPs for the current study. A mixture of isotope-labelled internal standards (ISs;  $n = 27$ ; Table A2 in the Appendix) was prepared in methanol at a concentration of 1 µg mL<sup>-1</sup>. The native target compounds were assigned the corresponding co-eluting isotope-labelled IS where possible, otherwise with the IS with the closest retention time on condition that detection was also robust, as described below (Section 2.5).

All pesticide standards (native and isotope-labelled) were supplied by Teknolab Sorbent (Kungsbacka, Sweden) as accredited mixtures, and PFAS standards were supplied by Wellington Laboratories (Guelph, Canada). The remaining standards were supplied by Sigma-Aldrich (Buchs, Switzerland) as individual compounds, either as solutions or in powder form, at analytical purity or higher, and were made into stock solutions in methanol or acetonitrile at a concentration of 1 mg mL<sup>-1</sup>.

Ultrapure water (Milli-Q) for both solid phase extraction (SPE) and mobile phases was generated in-house by a water purification system (Millipore; Bedford, USA). The buffers, acids and bases used for mobile phases were of LC-MS grade and purchased from Sigma-Aldrich (Buchs, Switzerland). The methanol and acetonitrile (LC-MS grade) were from Merck (Darmstadt, Germany) and the ethanol (AnalaR quality) was purchased from VWR International (Fontenay-sous-Bois, France).

## 2.3. Extraction of water samples

The samples were extracted by means of a semi-automated extraction system (4790 SPE-DEX®, Horizon Technology, Salem, New Hampshire, USA) using 47 mm Atlantic HLB-M SPE disks (Horizon Technology). In total, 5 L of the sample were transferred to an amber glass bottle before extraction and the IS mixture was added to achieve a concentration of 20 ng L<sup>-1</sup> of each IS. The samples were filtered using an in-line 1-µm glass fibre filter (1 Micron - Fine, Fast Flow Sediment Pre-Filters Horizon Technology) placed before the SPE disk. The system was preconditioned using 2 × 25 mL methanol followed by 2 × 25 mL Milli-Q water. The sample was applied onto the disk through the filter, the SPE disk was then washed with 2 × 25 mL 5% methanol,

and finally air-dried for 10 min before elution. The elution of the filter and the SPE disk was performed using 3 × 25 mL methanol. For full details on the SPE-DEX programme, see Table A3 in the Appendix.

The eluate was then reduced to ~1 mL using a TurboVap Classic II system (Biotage, USA) in a 200 mL evaporation tube. The extract was transferred to a 12 mL glass tube and the TurboVap tube was rinsed twice with ethanol, which was combined with the extract. The extract was then reduced to ~0.5 mL (TurboVap) and transferred to an amber glass vial. The 12 mL glass tube was rinsed twice with ethanol, which was combined with the extract. Finally, the extract was reduced again to ~0.5 mL and then diluted to 1 mL with Milli-Q water. The final extracts were stored at -20 °C until instrumental analysis.

## 2.4. Instrumental analysis

The instrumental analysis was carried out using a time-of-flight mass spectrometer (QToF; Xevo G2-S, Waters, Manchester, UK) coupled to a UPLC system (Acquity H-Class with FTN injector, Waters, Milford, USA). The column was an Acquity UPLC HSS T3-C18 (Waters, 2.1 × 100 mm, 1.8 µm particle size) for the positive ionisation mode analysis and a UPLC BEH-C18 column (Waters, 2.1 × 100 mm, 1.7 µm particle size) for the negative mode. The mobile phase was a gradient containing Milli-Q water and acetonitrile. In positive mode, 5 mM of ammonium formate was added to the water phase and 0.01% of formic acid was added to both the water and acetonitrile. In negative mode, 5 mM ammonium acetate was added to the water phase and 0.01% of ammonium hydroxide was added to both phases. Both ion modes used the same linear gradient with a flow of 0.5 mL min<sup>-1</sup>, starting at 5% acetonitrile and ending at 99%. The injection volume was 10 µL. All data were collected in MS<sup>E</sup>-mode, with a resolution of ~30,000 at 556.28 *m/z* using leucine enkephalin for the lock spray and UNIFI v1.8.2 as the software for data collection and evaluation. More details of the analytical procedure are given in Tröger et al. (Tröger et al., 2018).

DOC was analysed via combustion using a Shimadzu TOC-VCPH carbon analyser following the procedures set up by Lavonen et al. (2015). Laboratory blanks for repeated Milli-Q samples varied between 0.20 and 0.25 mg L<sup>-1</sup> and were corrected for. The analytical precision of DOC, based on triplicate analysis, was ±0.05 mg L<sup>-1</sup> for the large majority of samples (details in Table A4 in the Appendix).

## 2.5. Quality assurance and method performance

The target compounds were primarily identified using accurate mass screening (10 ppm mass error data extraction window) and precise retention time (one-minute time window). In addition, isotope patterns, additional adducts (other than the ±H used for quantification) and fragmentation spectra were considered for a positive identification. A database of the target compounds, including the molecular structures, was created. UNIFI automatically performs an *in silico* fragmentation prediction of the compound and matches this to the fragmentation spectra of a detected peak. If matching fragments are found, this further increases the identification certainty.

Method detection limits (MDLs) are given in Table A2 in the Appendix. The MDLs were estimated using the following formula (previously used by Tröger et al. (2018)):

$$MDL = \frac{(C_{spiked} - C_{blank}) * [R_{cutoff}/R_{average}] + 3 * [RSD_{response}]}{(C_{spiked} - C_{blank}) * [R_{cutoff}/R_{average}]}$$

$C_{spiked}$ : concentration of the target compound in spiked sample

$C_{blank}$ : concentration (if detected) in blank sample

$R_{average}$ : average response (detector counts) from the mass spectrometer

$R_{\text{cutoff}}$ : minimum response (detector counts) used to not discard a peak

$RSD_{\text{response}}$ : relative standard deviation in the response in the spiked sample.

This formula was used for the MDLs since it includes both the measurement uncertainty and the response cut-off. A cut-off response of 200 counts was used based on expert judgment from the normal noise conditions during an analytical run. The MDL was estimated from a triplicate of control samples using drinking water (from the facilities at SLU, Uppsala, Sweden) spiked to  $20 \text{ ng L}^{-1}$  of each individual targeted compound ( $n = 163$ ), and a triplicate of the same unspiked drinking water for blank subtraction. The SPE extraction recovery was calculated by dividing the average response from a triplicate of control samples spiked before extraction with the average of a triplicate of control samples spiked after extraction. Matrix effects were calculated by dividing the average response in the triplicate of control samples spiked after extraction by the response for an external calibration point at the same concentration (without matrix). For recovery and matrix effect calculations, any response from the unspiked control sample was subtracted. The average recovery of the native compounds was 72%, with an average relative standard deviation (RSD) of 8.2% (more details in Table A2 in the Appendix). The average RSD for all compounds detected ( $n = 26$ ) in the triplicate samples from the Lake Vänern outlet was 19% (for details, see Table A5 in the Appendix).

Finally, a correction factor ( $CF$ ) was calculated for each compound. The  $CF$  was calculated from the triplicate control samples by dividing the spiked concentration ( $20 \text{ ng L}^{-1}$ ) by the average measured concentration (see Table A2 in the Appendix), where a  $CF$  of 1 indicates that no correction is needed. The median  $CF$  was 1.1. There are two main reasons why the  $CF$  might deviate from 1. It could be either a difference in recovery or a difference in ion suppression/enhancement between the selected IS and the native compound being quantified. The following formula was used to calculate the final (corrected) concentration in the water samples:

$$C_{\text{corrected}} = C_{\text{sample}} * CF$$

$C_{\text{corrected}}$ : corrected concentration

$C_{\text{sample}}$ : measured concentration in the sample

$CF$ : corrected factor calculated from the control samples.

### 3. Results and discussion

#### 3.1. Micropollutants in the raw water

In total, 27 compounds out of the 163 analysed were detected at levels above MDL in one or more of the water samples (Table A5 in the Appendix). The detected compounds come from the following categories; pharmaceuticals ( $n = 8$ ), PFASs ( $n = 7$ ), pesticides ( $n = 7$ ), a food additive ( $n = 1$ ), flame retardants ( $n = 1$ ), a drug ( $n = 1$ ), and benzos ( $n = 2$ ). The average total concentration ( $\pm$ SD) in the river and raw water was  $84 \pm 15 \text{ ng L}^{-1}$  ( $n = 9$ ). The main contributors to the total concentration in the river water were caffeine ( $23\text{--}54 \text{ ng L}^{-1}$ ), followed by lidocaine ( $9.5\text{--}13 \text{ ng L}^{-1}$ ), carbamazepine ( $5.2\text{--}13 \text{ ng L}^{-1}$ ) and metoprolol ( $4.5\text{--}9.5 \text{ ng L}^{-1}$ ) (Fig. 2), making up on average  $74 \pm 5\%$  of the total concentration.

There was an increase in total concentration along the flow path of the water from Vänersborg to Alelyckan, with total levels ranging from  $65 \text{ ng L}^{-1}$  to  $116 \text{ ng L}^{-1}$  (Fig. 2). The raw water of Mölndal and Lackarebäck DWTPs showed lower total concentrations ( $89$  and  $88 \text{ ng L}^{-1}$ ), probably because they use alternative water sources (Fig. 1). The increasing trend was primarily driven by an increase in caffeine (an increase of  $30 \text{ ng L}^{-1}$ , 130%), and on a relative (and absolute) scale also in the other three main contributors (carbamazepine 12%,

lidocaine 14% and metoprolol 57%), atenolol (410%), benzos (330% for both), lamotrigine (164%), and diclofenac (77%). All of these are known to be indicator compounds of WWTP effluents (Gago-Ferrero et al., 2017; Loos et al., 2013; Tröger et al., 2018). For many other compounds, the concentrations stayed more or less constant along the flow path, e.g.  $\sum$ PFASs (0.3%). These findings are in good agreement with Gago-Ferrero et al. (Gago-Ferrero et al., 2017), who noticed that elevation of OMP concentrations downstream of the WWTPs was highly pronounced for many pharmaceuticals, personal care products and artificial sweeteners, but less so for PFASs, and suggested that that these substances follow different routes of entry into the aquatic environment.

The observed concentrations were mostly higher than those found in the source water and raw water of a major DWTP in Lake Mälaren (Tröger et al., 2018), the most important drinking water supply in Sweden. For the 19 compounds detected in both studies, the average ( $\pm$  SD) total concentration was  $74 \pm 15 \text{ ng L}^{-1}$  in the Göta Älv ( $n = 9$ ; Table A5 in the Appendix) and  $40 \pm 1.1 \text{ ng L}^{-1}$  in Lake Mälaren ( $n = 2$ ) (Tröger et al., 2018), and all four major contributors in the current study were found at lower concentrations in the Lake Mälaren study (caffeine:  $6.5 \pm 0.2 \text{ ng L}^{-1}$ , lidocaine:  $1.0 \pm 0.04 \text{ ng L}^{-1}$ , carbamazepine:  $1.8 \pm 0.09 \text{ ng L}^{-1}$ , and metoprolol:  $2.9 \pm 0.08 \text{ ng L}^{-1}$ ). In contrast, the concentrations of  $\sum$ PFASs ( $n = 7$ ) were lower in Göta Älv ( $1.4\text{--}1.9 \text{ ng L}^{-1}$ ) than in the Lake Mälaren study, where the average was  $5.7 \pm 0.1 \text{ ng L}^{-1}$  (Tröger et al., 2018).

The OMP concentrations detected in the Göta Älv river/raw water were also compared to those in the river Rhine (Ruff et al., 2015), which is one of the most important source waters in central Europe, providing drinking water to approximately 22 million people in six countries. The study comprised ten sampling sites along the path of the river from Switzerland to the Netherlands, and showed overlapping, but often higher top concentrations than in the Göta Älv study. For example, for the four dominating compounds in Göta Älv, the concentration ranges in the Rhine were  $42\text{--}1086 \text{ ng L}^{-1}$  for caffeine (compared to  $23\text{--}54 \text{ ng L}^{-1}$ ),  $6\text{--}87 \text{ ng L}^{-1}$  for carbamazepine (compared to  $5\text{--}13 \text{ ng L}^{-1}$ ),  $2\text{--}15 \text{ ng L}^{-1}$  for lidocaine (compared to  $9.5\text{--}13 \text{ ng L}^{-1}$ ), and  $3\text{--}78 \text{ ng L}^{-1}$  for metoprolol (compared to  $4.5\text{--}9.5 \text{ ng L}^{-1}$ ).

#### 3.2. Overall removal efficiency of micropollutants in the seven different DWTPs

The removal efficiency of the individual OMPs at the different DWTPs was investigated by calculating the ratio between the concentration of the OMP in the finished drinking water ( $C_{\text{finished}}$ ) with the concentration in the raw water ( $C_{\text{raw}}$ ) ( $TE (\%) = C_{\text{finished}}/C_{\text{raw}} \times 100$ ). If the OMP concentration was <MDL (in total 15% of the data points), a value of MDL/2 was used for the calculation. The average removal efficiency in all seven DWTPs for all detected OMPs was  $55 \pm 12\%$ . However, the efficiency varied greatly between compound groups, with PFASs showing lower values (on average  $18 \pm 11\%$ ) than the remaining OMPs ( $67 \pm 12\%$ ) (details in Table A6 in the Appendix).

Fig. 3 displays a flowchart detailing how many of the detected compounds ( $n = 27$ ) had a removal efficiency  $\geq 80\%$  at the studied DWTPs, categorised in three groups by treatment category (Table 1): (i) using GAC ( $n = 4$ ), (ii) artificial infiltration ( $n = 1$ ) or (iii) neither GAC nor artificial infiltration ( $n = 2$ ) (assigned *Other* in Fig. 3). A cut-off value of  $>80\%$  was chosen as it can be considered a reasonable barrier effect value.

PFASs were not removed above 80% regardless of the treatment strategy. In contrast, all the DWTPs removed atenolol, carbendazim and cyprodinil to levels below the MDL. Overall, the DWTP using artificial infiltration indicated a  $> 80\%$  removal for ten compounds, while seven compounds were removed  $>80\%$  in the GAC-DWTPs (Fig. 3). In the DWTPs without artificial infiltration or GAC, only one compound was removed  $>80\%$  (lidocaine). The DWTP in Kungälv (artificial infiltration) showed the highest overall removal efficiency ( $67 \pm 37\%$  for all

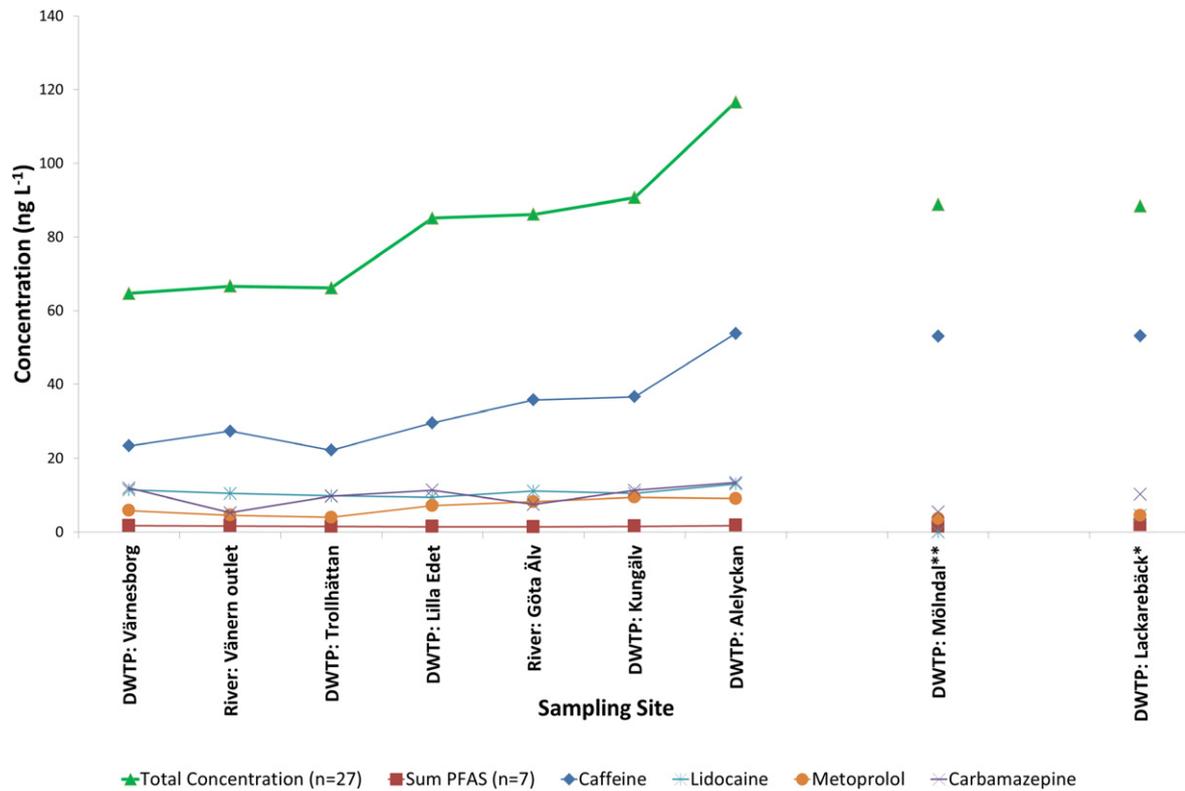


Fig. 2. Total concentrations of the 27 detected compounds in the river water and raw water of the DWTPs along Göta Älv, together with the concentration of the four most abundant OMPs and the  $\Sigma$ PFASs. The graph follows the water's flow path (sampling sites from north to south). \*not using Göta Älv as raw water directly; \*\*not using Göta Älv as raw water at all.

OMPs) followed by the four DWTPs with GAC, which had an average removal efficiency of 60% (range 49%–63% and SDs 34%–37%). The DWTPs that employed GAC or artificial infiltration had a significantly higher

(one sided  $t$ -test;  $p = 0.014$ ) removal efficiency than the DWTPs that used a more conventional treatment strategy (Vänersborg  $41\% \pm 38\%$  and Trollhättan  $35\% \pm 37\%$ ). These results suggest that using GAC and

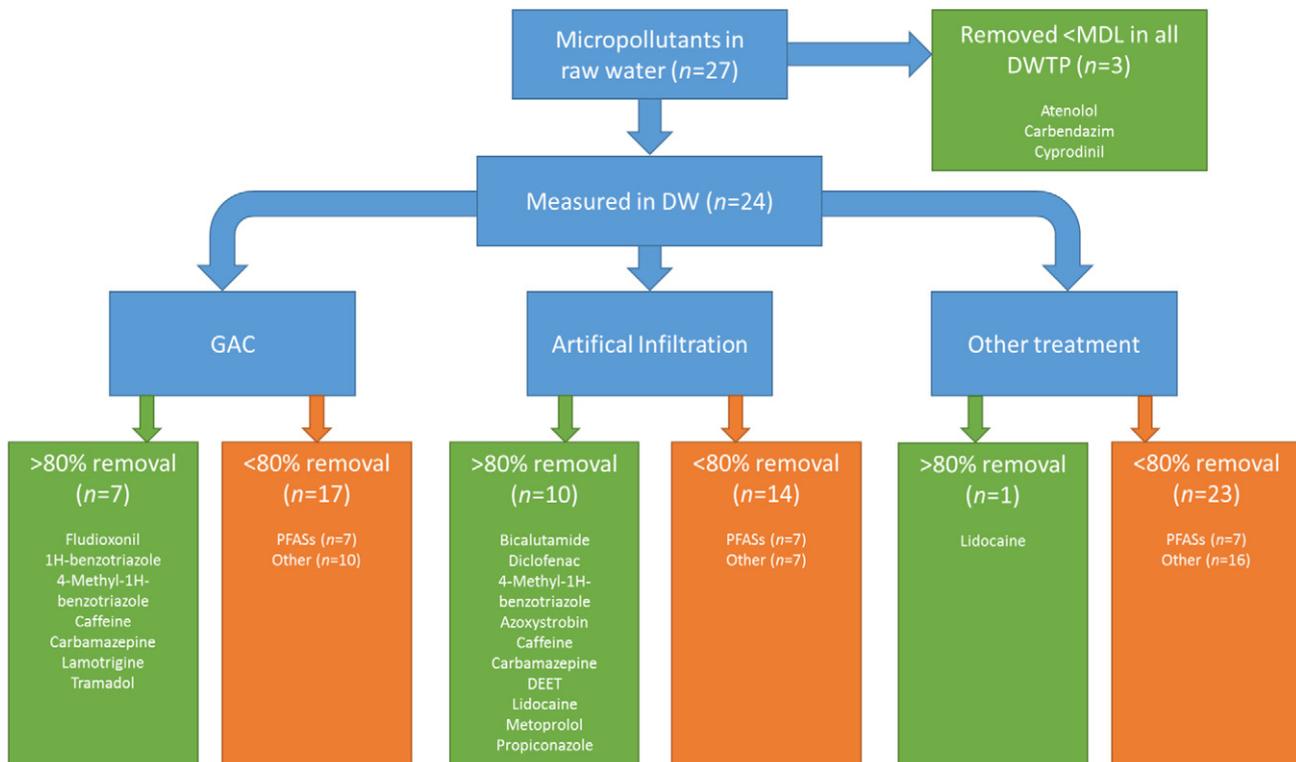


Fig. 3. Flowchart showing the average number of compounds removed by >80% (green box) or lower (orange box) at DWTPs (i) using GAC ( $n = 4$ ), (ii) artificial infiltration ( $n = 1$ ) or (iii) neither GAC nor artificial infiltration ( $n = 2$ ).

artificial infiltration is advantageous for OMP removal, with the exception of PFASs, which require treatment with fresh GAC or even more advanced treatment techniques (McCleaf et al., 2017). There is greater uncertainty in the results for the DWTP using artificial infiltration due to this strategy being used by only one facility and due to the long residence time between the intake of the raw water and the finished drinking water (7–30 days), which was not considered in the sampling (all the samples were collected over two days). It should also be highlighted that the inclusion of GAC filter(s) in the treatment process is not a guarantee of high removal efficiency of OMPs (Tröger et al., 2018) *per se*. The importance of using relatively fresh GAC is discussed in the following section.

### 3.3. Influence of GAC operational age on the removal efficiency for micropollutants and DOC

The influence of the operational age of GAC filters on OMP removal efficiencies was investigated using the samples taken at Lackarebäck DWTP ( $n = 6$ ). The removal efficiency was calculated for all compounds with concentrations >MDL in the raw water and in at least three of the six of the samples collected after the GAC (18 compounds in total). Concentrations below MDL were substituted with a value equal to MDL/2 (see Table A7 in the Appendix for more details). As there was a lower removal efficiency for the measured PFAS substances than for other compounds, the compounds were separated into two different groups: PFASs ( $n = 7$ ) and non-PFAS compounds ( $n = 11$ ). Fig. 4 shows the removal efficiency for  $\Sigma$ OMP (average), for the two groups of OMP (averages for PFASs and non-PFAS respectively), and for DOC (average of triplicate samples) in relation to the adjusted operational age of the GAC.

The removal efficiency showed a decreasing trend with operational age of the GAC for both OMP categories (PFASs and non-PFAS) and for DOC. The average removal efficiency for all  $\Sigma$ OMP was 92% and 90% after the newest GAC filters (12 and 15 months respectively) but only 46% and 34% for the oldest (54 and 71 months respectively) (see Table A7 in the Appendix for full details). The two oldest filters exhibited even lower efficiencies for PFASs (23% and 5.4% respectively) (Fig. 5), while remaining more effective (61% and 51%) for the non-PFAS compounds (Fig. A2 in the Appendix). The removal efficiency for DOC showed a similar trend, but with lower removal rates (and quicker breakthrough) than for the OMPs, with a removal efficiency of 42% for the newest filter, 7% for the 25-month old filter and levels below 4% for the oldest filters.

It should be noted that the timing of the topping up of the new GAC was not fully reflected in the adjusted operational age, and this may have led to an artificially high removal efficiency for the four newest

filters (that were topped up once in the past) in relation to their adjusted operational ages (12, 15, 23 and 25 months; Table A1 in the Appendix). For the OMPs, the topping up seemed to have a small or no effect for the 23-month-old and 25-month-old filters, which were topped three and 12 months prior to the sampling respectively, but still showed almost identical removal efficiencies. DOC, which is known to have a short breakthrough time in GAC filters (Moona et al., 2018), was affected differently to the OMPs, with a clear drop between the 23-month-old and the 25-month-old filter, indicating a positive DOC removal effect through the topping up that lasted approximately three months. For the 12-month and 15-month old filters, the topping up was performed less than two weeks before the sampling. Consequently, for the DOC removal values for the two newest filters, it is highly probable that these values indicate higher removal efficiencies than expected in relation to the adjusted operational ages. Similarly, it can be expected that OMP removal was also affected positively by the recent topping up, particularly for the PFAS substances, which showed a larger drop between the two newest filters and the 23-month-old and 25-month-old filters. The asterisks in Fig. 4 highlight the samples where extra care should be taken when interpreting the treatment capacity. These high removal values can probably not be obtained without topping up with new GAC material (McCleaf et al., 2017).

Most of the PFASs showed a similar decreasing trend, but for some of the substances the removal efficiency was more drastically reduced over time (Fig. 5). Perfluorobutane sulfonic acid (PFBS) was retained to 96% in the two newest filters, but after the 54-month-old and 70-month-old filters an increase in concentration was found, resulting in negative removal efficiencies (−49% and −19% respectively). These findings are in line with observations for PFBS in the pilot-scale experiment by McCleaf et al. (McCleaf et al., 2017), who also noted negative removal efficiency values and suggested that this phenomenon could be related to desorption of PFBS previously captured by the GAC. For both the sulfonic PFASs (PFBS, PFHxS and PFOS) and the carboxylic PFASs (PFOA, PFNA, PFDA, and PFUnDA), shorter chain length correlated with lower removal efficiency (Fig. 5). This behaviour has previously been observed (Appleman et al., 2014) and is suggested to be due to the higher hydrophobicity of the longer-chain PFASs (McCleaf et al., 2017), which has a stronger interaction with the GAC material. Again, it is likely that removal efficiency values for the two newest filters are higher than expected in relation to the adjusted operational ages.

PFASs are generally more poorly retained by GAC (Appleman et al., 2014) when adhering to typical GAC regeneration cycles, and pilot-scale experiments have shown that removal efficiency decreases with the number of bed volumes and operational GAC age (McCleaf et al., 2017). Other studies have shown that the operational age of activated carbon is also important for the removal of other OMPs (Stackelberg

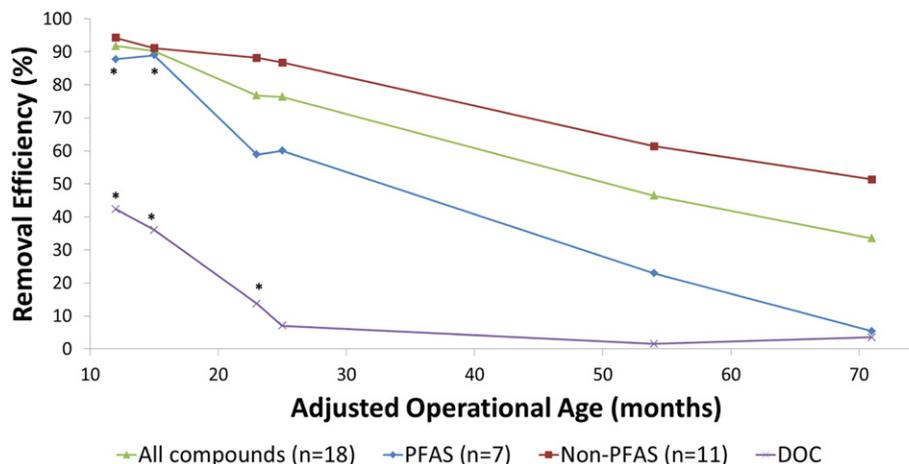


Fig. 4. Removal efficiency as a function of adjusted operational age of the GAC filters for all compounds (average for  $\Sigma$ OMP,  $n = 18$ ),  $\Sigma$ PFASs (average,  $n = 7$ ), non-PFAS compounds (average,  $n = 11$ ) and DOC (average, triplicate samples). \*Artificially high removal efficiency values due to the recent addition of fresh GAC.

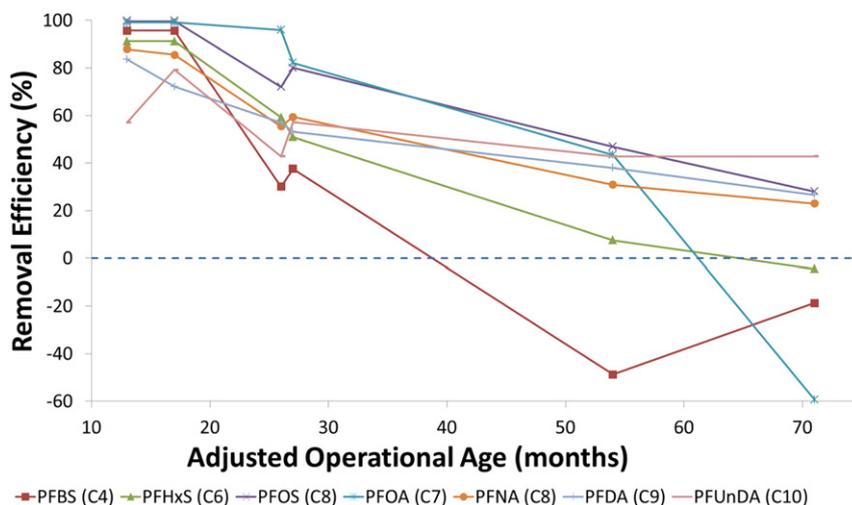


Fig. 5. Removal efficiency (%) by GAC filters of different operational age for seven individual PFAS compounds. The length of the carbon chain of each PFASs is indicated in parentheses (C4 etc.). \*Artificially high removal efficiency values due to the recent addition of fresh GAC.

et al., 2007; Tröger et al., 2018; Westerhoff et al., 2005). For example, a GAC filter that had been in operation for three years showed a poor removal efficiency of OMPs at a full-scale DWTP (Stackelberg et al., 2004). In a subsequent study (Stackelberg et al., 2007) at the same DWTP, where the GAC filter had been replaced with a fresh GAC filter (two months' operation time), the removal efficiency of the OMPs was significantly improved.

#### 4. Conclusions

The source water for the Göta Älv drinking water treatment plants contains a range of OMPs, including pharmaceuticals, pesticides, PFASs and other compounds. In total, 27 OMPs out of 163 targeted were detected in one or more of the samples, which consisted of river water, raw water and drinking water samples from seven DWTPs along the water path. The concentration of  $\sum$  OMPs in the river and the raw water ranged from  $\sim 60$  ng L<sup>-1</sup> at the start of the river and increased to  $\sim 120$  ng L<sup>-1</sup> further downstream. The  $\sum$  OMP concentrations were comparable, but mostly higher, than those previously found in the source and raw water of a DWTP in Lake Mälaren (Tröger et al., 2018), the most important source water in Sweden, with the exception of PFASs, which were higher in Mälaren, while the concentrations were typically within the low-end concentration range found in the European river Rhine (Ruff et al., 2015).

The seven DWTPs used different treatment strategies to remove OMPs. There was a clear benefit in terms of overall OMP removal efficiency to have GAC filters implemented in the treatment process, and the plant using artificial infiltration showed a comparable OMP removal to those using GAC filters. DWTPs with and without GAC filters had average removal efficiencies of 60% and 38% respectively, while the DWTP with artificial infiltration showed an average removal of 65%.

The present study shows that it is important to replace, topping up or regenerate GAC filters frequently. A GAC filter with an adjusted operational age of 12 months and the recent (0.5 months prior to sampling) addition of fresh GAC showed an average removal efficiency of 92% for  $\sum$  OMPs, while for a 25-month-old filter the average was 76% and an even lower value (34%) was observed for a 71-month-old filter. DOC showed the quickest breakthrough in the GAC filters, followed by PFASs. There were clear indications that the addition of fresh GAC enhanced the removal of DOC and PFASs and this strategy could be used as a cost effective way to keep a high removal efficiency even with older GAC filters. This positive removal effect provides DWTPs with a flexible option for both continuous and emerging treatment needs.

Predicting the removal of OMPs, and especially PFASs, is a complex and site-specific task. Many DWTPs rely on fixed schemes for regenerating or, as in the current case, filling up individual GAC filters. Regular OMP measurements at individual filters are costly and alternative breakthrough measurement methods are needed. As the breakthrough of DOC occurs earlier than for the OMPs, DOC measurements are not an optimal indicator, and it remains to be solved how breakthrough can be diagnosed in a cost-efficient and practical way.

#### Declaration of competing interest

All authors hereby declares no conflict of interest.

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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.2019.135680>.

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