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Breakthrough and desorption of PFAS in aged GAC filters during drinking water treatment – a question of DOM composition

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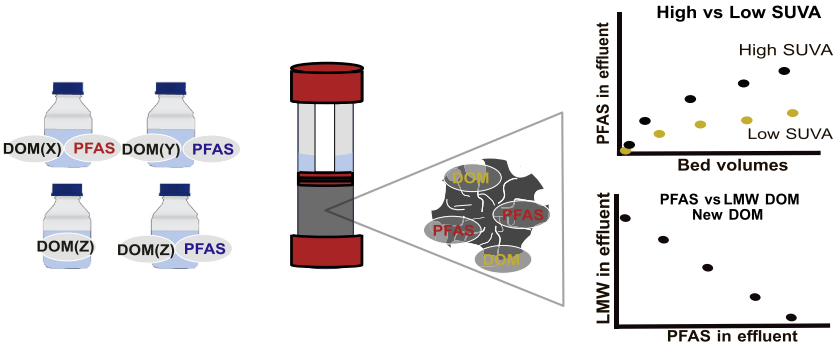
HIGHLIGHTS

- DOM composition affected PFAS adsorption and desorption in mature GAC filters.
- Increased feedwater aromaticity led to desorption of previously adsorbed PFAS.
- Increased feedwater aromaticity resulted in poorer PFOA removal.
- As lower molecular weight DOM adsorbed, shorter chained PFAS desorbed.

GRAPHICAL ABSTRACT



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ABSTRACT

Shifts in dissolved organic matter (DOM) composition, caused by seasonal or episodic events, can significantly alter the removal of per- and polyfluoroalkyl substances (PFAS) by granular activated carbon (GAC). To investigate how changes in feedwater composition impact PFAS removal in aged GAC, a 20-day experiment was conducted in eight parallel small-scale columns. Columns were filled with aged GAC from a full-scale drinking water treatment plant that still retained >80% efficiency for removing the sum of PFAS21 from groundwater. The GAC was exposed to feedwaters with varying DOM composition and aromaticity. The removal performance was evaluated using fluorescence spectroscopy combined with measurements of ^{14}C-labelled PFOA and unlabelled PFAS. This combination facilitated an assessment of changes in both DOM composition and PFAS removals. The results showed that a change in DOM composition of the feedwater resulted in the desorption of shorter-chained PFAS from the GAC and a simultaneous increase in the adsorption of lower molecular weight DOM, resulting in higher aromaticity in the outgoing water. These findings suggest a dynamic sorption process in which newly introduced DOM in the GAC feedwater competes for adsorption sites, potentially displacing previously adsorbed PFAS. Experiments with carbon–14 labelled perfluorooctanoic acid (^{14}C-labelled PFOA) indicated that an increase in feedwater DOM aromaticity, while maintaining a constant dissolved organic carbon

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(DOC) concentration, resulted in earlier breakthrough. This highlights the need to monitor and evaluate not only the quantity of DOM in feedwater, but also its composition.

1. Introduction

Granular activated carbon (GAC) has a long history of improving the taste and odour of drinking water by removing DOM and off-flavour compounds. The mechanisms for the removal include both adsorption and biological degradation/transformation by the biofilm formed on the GAC granules (Çeçen and Aktaş, 2011; National Research Council (US) Safe Drinking Water Committee, 1980; Yuan and Hofmann, 2022). GAC filtration can also be used for the removal of per- and poly-fluoroalkyl substances (PFAS) (Appleman et al., 2014; Belkouteb et al., 2020; Rahman et al., 2014). The health risks and prevalence of PFAS in the environment, including drinking water sources, have led to the implementation of strict regulations in many countries (Dimitrakopoulou et al., 2024; EPA United States Environmental Protection Agency, 2025; Eschauzier et al., 2011; European Commission, 2024; Kannan et al., 2004; Rahman et al., 2014). Owing to their stable chemical structures, PFAS are hardly biodegradable leaving adsorption as the primary mechanism for PFAS removal in GAC filters (Eschauzier et al., 2011).

Due to competition for adsorption sites, an increased abundance of dissolved organic matter (DOM) in the influent water expedites the ageing of GAC, resulting in earlier breakthrough of DOM and organic micropollutants (Chow et al., 2022; Corwin and Summers, 2012; Summers and Roberts, 1988a, 1988b). DOM encompasses a complex mixture of compounds with variable physical and chemical properties (Huguet et al., 2009; Kellerman et al., 2014; Minor et al., 2014; Stubbins et al., 2014; Thurman, 1985). Smaller DOM fractions have been reported to compete with organic micropollutants for adsorption sites through direct site competition (Li et al., 2003; Newcombe et al., 1997; Pranić et al., 2025). Larger, highly-conjugated aromatic DOM molecules (often referred to as humic substances) have been shown to hinder the adsorption of organic micropollutants through pore blockage (Li et al., 2003; Newcombe et al., 1997; Zietzschmann et al., 2014). However, some studies have indicated that higher molecular weight DOM fractions facilitate PFAS adsorption through hydrophobic interactions and agglomeration (Gagliano et al., 2020; Kothawala et al., 2017; McCleaf et al., 2017). Thus, the observations reported in literature indicate that the effect incoming DOM molecular sizes and composition has on PFAS removal by GAC is not yet fully understood. However, it is evident that not only quantity, but also quality of DOM appears to be an important factor.

The molecular size of DOM is closely associated with its bulk aromaticity, a chemical property representing the abundance of dissolved aromatic carbon in the water (Alberts et al., 2002; Ghabbour et al., 2001; Korak and McKay, 2024; Maizel and Remucal, 2017; Weishaar et al., 2003). Aromaticity is strongly correlated to specific ultraviolet absorbance (SUVA) (Croué et al., 2000; Mcknight et al., 1997; Weishaar et al., 2003) and to the ratio of fluorescence emissions measured at different wavelengths, since large, conjugated aromatic molecules emit at longer wavelengths than small, simpler molecules, including phenols (Aiken, 2014; Lakowicz, 2006; Wünsch et al., 2015). Recently, a fluorescence "aromaticity index" (ARIX) was shown to correlate linearly with both SUVA and the relative abundance of larger versus smaller DOM fractions separated using liquid chromatography (Murphy, 2025).

While numerous studies have investigated PFAS removal by GAC in

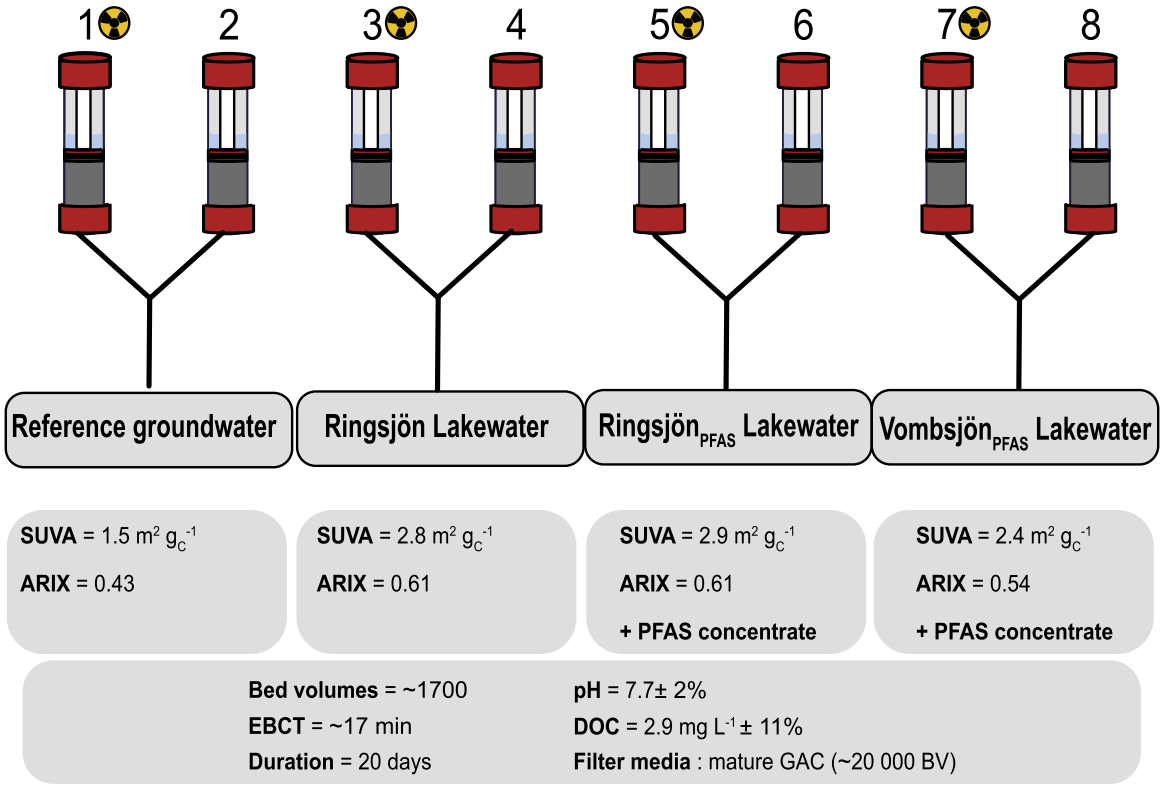

Fig. 1. Experimental set-up, operational parameters and feedwater characteristics. Each of the eight columns contained an identical mass of aged GAC sampled from a treatment plant ~20 000 BV and was exposed to feedwaters with similar DOC concentrations. One replicate of each feedwater was spiked with ^{14}C-labelled PFOA indicated by a radiation symbol. Half of the samples were additionally spiked with a diverse PFAS concentrate.

drinking water treatment (Appleman et al., 2014; Belkouteb et al., 2020; Chow et al., 2022; Kothawala et al., 2017; McCleaf et al., 2017; Newcombe, 1994; Park et al., 2020; Riegel et al., 2023; Tisler et al., 2025), the impact of altered DOM composition on PFAS adsorption in GAC filter media remains poorly investigated, as highlighted by Belkouteb et al. (2020) and Park et al. (2020). Additionally, the few studies we are aware of that investigated the effect of DOM composition on PFAS/micropollutant removal either investigated different feedwaters with new GAC, or were batch experiments, and only one study used aged GAC (Chen et al., 2022; Kennedy and Summers, 2015; Kothawala et al., 2017; Li et al., 2003; Newcombe et al., 1997; Summers et al., 1989). Continuous flow studies investigating the effect that sudden shifts in DOM composition have on aged GAC therefore address a critical research gap that may influence the way GAC filters are operated. Particularly in water treatment plants affected by fluctuations in incoming DOM composition.

The chemical composition of DOM, affecting bulk aromaticity and the relative abundance of higher molecular weight and lower molecular weight compounds, varies seasonally and due to climactic conditions in surface waters (Johnston et al., 2020; Murphy, 2025; Raymond and Saiers, 2010). Specifically, increased runoff during heavy precipitation and snowmelt facilitate the mobilisation of large, aromatic DOM molecules derived from soil and terrestrial vegetation into the water body (Aiken, 1993; Health Canada, 2019; Volk et al., 2002). Also, lake turnover events can mix small recently produced DOM molecules with large previously settled, recalcitrant organic compounds, altering the bulk DOM characteristics (Krzeminski et al., 2019; Volk et al., 2002). Similarly, periods of high algal and microbial productivity introduce low molecular weight compounds released during the metabolism and decay of algae (Goslan et al., 2017; Leloup et al., 2015). Shifts in DOM aromaticity signal a change in DOM reactivity that could have significant downstream effects on the performance of drinking water treatment (Weishaar et al., 2003). From this perspective, further investigation is needed to assess how changes in DOM aromaticity, and the resulting difference in DOM size distribution, in the feedwater affect PFAS removal by GAC under continuous-flow conditions, where these effects can be monitored over time.

The objective of this study was to examine how a change in feedwater DOM composition influence PFAS adsorption and desorption behaviour in aged GAC, representing a critical stage in the operation of many drinking water treatment plants, under continuous loading of DOM and PFAS. We hypothesise that PFAS removal by aged GAC filters is susceptible to changes in feedwater DOM composition, and specifically, that an increase in aromaticity in the feedwater will lead to a reduction in the PFAS removal efficiency and increase the risk of PFAS desorption. The results in this study highlight the importance of DOM quality on the ability of GAC to remove PFAS, especially for GAC nearing the end of its service life.

2. Materials and methods

Column experiments were operated with aged GAC for 20 days and ~1700 bed volumes (BV), to study the effect of feedwater DOM composition on the PFAS removal (Fig. 1). The GAC originated from a full-scale drinking water treatment plant that had previously treated groundwater containing ~80–100 ng L^{-1} PFAS over a period of approximately nine months, representing ~20 000 BV (Belkouteb et al., 2020; McCleaf et al., 2017). Despite the long service time, at the time of GAC retrieval the filter media was not yet exhausted (as detailed in Section 2.1). In the experimental setup, eight small-scale GAC columns were exposed to duplicates of four different feedwaters, differing in DOM composition and/or PFAS concentrations, where one replicate contained ^{14}C-PFOA, labelled at the carboxyl group (Figure S1).

2.1. Granulated activated carbon (GAC)

Granulated activated carbon, consisting of mainly Aquasorb6300® and traces of (<10%) of Filtrasorb400® and Aquasorb2000®, was collected in July 2024 from full-scale GAC filters used for PFAS removal at a drinking water treatment plant in Sweden. The properties of the GAC blend include an iodine number of ~1000 mg g^{-1}, effective size of ~0.6 mm and surface area of ~1000 m^2 g^{-1} (Table S1) (Belkouteb et al., 2020; Calgon Carbon Cooperation, 2018; Jacobi the carbon company, 2000, 2012). The treatment plant receives artificially infiltrated groundwater, whereby a groundwater aquifer is continuously replenished of extracted bore water using surface water percolated though an esker (Yu et al., 2022). The ground water is treated through aeration, softening, and sand filtration before reaching the GAC filters, as further detailed by Belkouteb et al. (2020). Samples of aged GAC were collected after a service time of ~20 000 BV at mixed depths from a full-scale groundwater treatment plant. Since the drinking water treatment plant sources water from PFAS-contaminated aquifers, it uses parallel filters containing GAC of different ages which are replaced before they are fully exhausted. At the time of GAC collection, the sum of PFAS4 and PFAS21 removals were >90% and >80% respectively. This represents the starting conditions for GAC performance during the experiments in this study. After sampling the GAC filter media was transported while moist to the laboratory, where it was quickly rinsed with tap water to remove any pulverised activated carbon, then stored at 8 °C until the beginning of the experiment. The GAC size distribution was determined by dry sieving, according to ISO 17,892–4:2016 (Table S2).

2.2. Feedwaters

The eight GAC columns were fed with replicates of four different waters: *i)* influent groundwater for the full-scale GAC filters, collected from the drinking water treatment plant at which the GAC was sampled (*Reference groundwater*); *ii)* lake water from Ringsjön (*Ringsjön*); *iii)* lake water from Ringsjön with the addition of PFAS (*Ringsjön$_{PFAS}$*); *iv)* lake water from Vombsjön with the addition of PFAS (*Vombsjön$_{PFAS}$*). Vombsjön is an active drinking water source for several municipalities in southern Sweden, while Ringsjön is a reserve drinking water source for the same region (Table S3).

To ensure focus on the effects of DOM quality rather than quantity, each source water was diluted with deionized (DI) water to obtain DOC concentration as similar as possible to the *Reference groundwater* (Table S4). All source waters were filtered in two steps, first through 1.2 μm glass fibre grade (GF/F) pre-filters (pre-combusted, 400 °C for 3 h) to remove large particles, followed by 0.45 μm nitrocellulose (NC) membrane filters to remove larger colloids. These filters were selected to allow high volume processing with minimal risk of leaching, which was subsequently confirmed via filter blanks (< 3% of sample DOC). Each filter was pre-rinsed with 100 mL DI water and 50 mL source water.

Half of the feedwater samples i.e., *Ringsjön$_{PFAS}$* and *Vombsjön$_{PFAS}$*, were spiked with a concentrate containing various PFAS, obtained by foam fractionation at a full-scale treatment plant in Denmark (RESC, Korsør). This plant treats runoff and shallow groundwater from a site with severe PFAS pollution from the use of firefighting foam (Table S5). PFAS concentrate was added (10 mL L^{-1}) to *Ringsjön$_{PFAS}$* and *Vombsjön$_{PFAS}$*, amounting to a PFAS concentration (17 compounds) of 45 μg L^{-1} ± 5% (Table S6). The above-described treatments resulted in four different feedwaters for the column experiments. All feedwaters were adjusted to pH 7.5–8.0 using 1 M NaOH or 1 M HCl. The resulting feedwaters varied in DOM composition and/or PFAS concentration, and were each named according to the type of source water and presence or absence of PFAS concentrate, i.e. *Reference groundwater, Ringsjön, Ringsjön$_{PFAS}$,* and *Vombsjön$_{PFAS}$*.

The four feedwaters were prepared and run in replicates, one with and without the addition of ^{14}C-labelled PFOA (American Radiolabeled Chemicals. Inc.). The addition resulted in ^{14}C activities of 0.1 μCi L^{-1},

corresponding to ~1 μg L^{-1} of radiolabelled PFOA. This arrangement resulted in eight different feedwaters: *Reference groundwater, Reference groundwater^{14C}, Ringsjön, Ringsjön^{14C}, Ringsjön$_{PFAS}$, Ringsjön$^{14C}_{PFAS}$, Vombsjön$_{PFAS}$,* and *Vombsjön$^{14C}_{PFAS}$*. The chemical compositions for the eight feedwaters are summarised in Fig. 1 and Table S7.

2.3. Experimental setup

Eight gas-tight glass columns (Cytiva) with adjustable bed heights and an inner diameter of 16 mm were used. This set up was similar to Betsholtz et al. (2024) with silicone tubing directing feedwaters separately to the eight columns. The columns were fed from the bottom via a peristatic pump, and the effluent was collected from the top of each column. Prior to filling the columns with GAC, the empty setup was rinsed with a weak acid (0.1 M HCl) followed by DI water. A mass balance calculation performed at the end of experiments confirmed 99 ± 5% averaged recovery of ^{14}C-labelled PFOA, indicating no significant loss of PFOA within the experimental rig.

After rinsing and drying the columns, 10 cm^3 of wet GAC, corresponding to 5.4 g ± 1% dry GAC (Table S8), was transferred to each column. Effluent water from the columns was sampled in glass bottles, which were collected and replaced daily (~80 bed volumes day^{-1}), to obtain flow-proportioned composite samples and to track the treated volume over a given time interval. Before bottles were put back into circulation, they were washed in a dishwasher (60 °C) once with detergent then twice with DI water. Two CO_2 traps were connected in series to the effluent bottles containing ^{14}C-labelled PFOA to monitor $^{14}CO_2$ formation, due to the degradation of ^{14}C-labelled PFOA. Each CO_2 trap contained 25 mL of 3 M NaOH. Figure S2 contains a schematic drawing of the experimental setup.

All columns were operated for 20 days at ambient temperature (18–22 °C) under oxic conditions (dissolved oxygen >8 mg L^{-1}), with no backwashing. The pH was maintained at 7.7 ± 2% and the EBCT was controlled at 17 min ± 2%, matching common EBCT for removal of organic micropollutants (Appleman et al., 2014; Corwin and Summers, 2012; Summers et al., 2010). Daily samples were collected for measurements of DOM composition, in the columns without the addition of radiolabelled PFOA, as well as for measurements of ^{14}C-activity in the columns with ^{14}C-labelled PFOA. Additionally, the concentrations of 20 PFAS were measured in columns without ^{14}C-labelled PFOA on nine occasions, as detailed in the sampling overview (Table S9).

2.4. Analysis

2.4.1. PFAS analysis

Selected per- and polyfluoroalkyl substances (PFAS) including compounds regulated under EU Directive 2020/2184 were determined in the PFAS concentrate, influent and effluent samples (Directive (EU), 2020). The list of PFAS comprised short- and long-chain perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkyl sulfonates (PFSAs), fluorotelomer sulfonates (FTSs), sulfonamides, and sulfonamidoacetic acids. The analytes detected at concentrations exceeding analytical detection limits consisted of perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA); linear sulfonates perfluorobutanesulfonic acid (PFBS), perfluoropentane sulfonic acid (PFPeS), perfluorohexanesulfonic acid (PFHxS), perfluoroheptanesulfonic acid (PFHpS), perfluorooctanesulfonic acid (PFOS), perfluorononanesulfonic acid (PFNS); fluorotelomer sulfonates 6:2-fluorotelomer sulfonic acid (6:2 FTS), and 8:2-fluorotelomer sulfonic acid (8:2 FTS); as well as N-methyl- and N-ethyl-perfluorooctane sulfonamidoacetic acids (N-MeFOSAA, N-EtFOSAA) and perfluorooctane sulfonamide (FOSA).

All analytes were quantified by means of solid-phase extraction (SPE, Oasis HLB cartridges (200 mg), Waters, Sweden) and internal standard calibration method. 50 μL isotope-labelled PFAS solution: MPFAC-24ES Mass-Labelled PFAS Extraction Standard Solution (Wellington Laboratories, Canada) in the concentration of (0.1 μg mL^{-1}) was added to the sample (30 mL). Samples were then loaded on the SPE columns, washed with 5 mL 25 mM ammonium acetate (aqueous solution), dried under vacuum (60 min), and the subsequent elution was performed with 5 mL methanol and 5 mL of 5% NH_4OH in methanol to polypropylene vials. Solvents were evaporated with nitrogen gas at 30 °C and reconstituted with 1 mL of methanol.

Instrumental analysis of the analytes was conducted using high-performance liquid chromatography coupled with tandem mass spectrometry (HPLC-MS/MS), using modified analytical procedures based on previously published methods (Jeong et al., 2022; Kamuf et al., 2024; Szopińska et al., 2026). In short, the analysis was conducted using an Agilent 1260 Infinity HPLC system interfaced with a triple quadrupole mass spectrometer (6470 LC/MS, Agilent Technologies). The mass spectrometer was operated with an electrospray ionization source in negative ion mode (Agilent Jet Stream ESI). Chromatographic separation of the target analytes was achieved on a polar-embedded reversed-phase C18 column (Zorbax Eclipse Plus C18 RRHD, 3.0 × 50 mm, 1.8 μm; Agilent), protected by a guard column of identical stationary phase. A delay column (InfinityLab PFC Delay Column, 4.6 × 30 mm; Agilent) was installed. Detailed information regarding sample preparation, chromatographic conditions, and method validation is available in Szopińska et al. (2026) and summarised in Section S1, and Table S10 - S11.

PFAS with concentrations below the limit of quantification (LOQ) in the chemical analysis (HPLC-MS/MS) were set to missing values except when estimating concentration increases over the filter. In the latter case, inlet concentrations were set to the LOQ as declared in the figure captions (when applicable). The LogD values of PFAS were determined at pH 7.5–8.0 *in silico* with Chemicalize® developed by ChemAxon.

2.4.2. ^{14}C measurements

The ^{14}C activity was measured in radiolabelled samples using a liquid scintillation counter (LSC), Tricarb 4910 TR (PerkinElmer Inc.), with a count time of 10 min. The ^{14}C activity was measured in the liquid phase, gas phase, and solid phase, as further described below.

Liquid phase: This phase included feed and effluent water. Each time feed or effluent water bottles were replaced, 4 mL of the sample was mixed with 12 mL of scintillation cocktail (Ultima Gold, PerkinElmer). In addition to a surfactant, the cocktail contains an aromatic solvent which absorbs energy from the β-particles emitted by the decay of the ^{14}C. The energy from the solvent is absorbed by an organic scintillator, which subsequently re-emits this energy as light which is detected by the liquid scintillation counter. *Gas phase:* 4 mL each from the primary and secondary CO_2 traps were separately mixed with 12 mL of scintillation cocktail. *Solid phase:* At the end of the experiment, the GAC from the columns was frozen (−20 °C) for later analysis of the ^{14}C activity adsorbed onto the GAC filter media of each column. Triplicate portions of 0.5 g ± 0.2% wet weight (0.2 g ± 2% dry weight, Table S12) were combusted in a Pyrolyzer^{genIII} (Raddec International Ltd.), while two CO_2 traps connected in series collected the $^{14}CO_2$ formed. The combustion program entailed stepwise heating and dwelling to 900 °C over a period of >5 h, as described by Betsholtz et al. (2024) and summarised in Table S13. For the analysis of the trapped gas formed during the combustion of the solid phase, 1 mL of the primary and secondary CO_2 traps were mixed with 15 mL of scintillation cocktail each, before analysis. The mass balance of the recovery of radioactivity introduced to the system as ^{14}C-labelled PFOA was calculated at the end of the experiment and included all phases (liquid, solid, and gas).

2.4.3. DOM chemical composition in feedwaters

The chemical compositions of the feedwaters were evaluated through DOC measurements, absorbance measurements at 254 nm

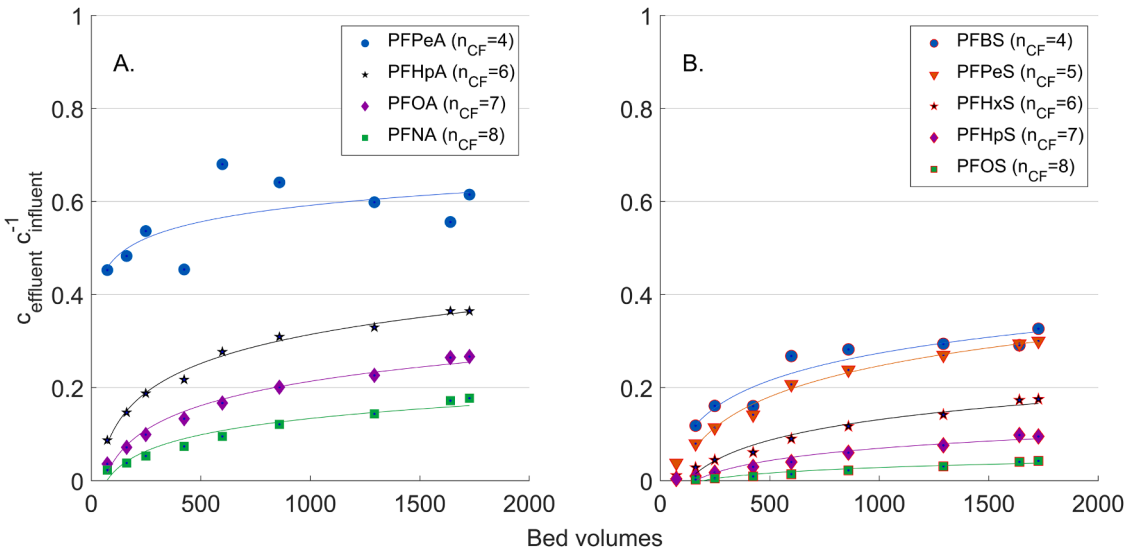

Fig. 2. Breakthrough curves of PFAS with increasing chain lengths (n_{CF}) and different headgroups - carboxylic acid (left panel, A) and sulfonic acid (right panel, B). The GAC columns were fed with lake water from Ringsjön spiked with PFAS concentrate (*Ringsjön$_{PFAS}$*).

(UVA_{254}) and fluorescence measurements. DOC was measured as non-purgeable organic carbon in baked (550 °C, 3 h) 40 mL glass vials and measured on a TOC-L (Shimadzu Scientific Instruments). UVA_{254} was measured on a Dr6000 spectrophotometer (Hach Company) in a 1-cm quartz crystal cuvette. Fluorescent DOM in the samples was measured on an Aqualog (HORIBA Ltd.) in a 1-cm cell. From the fluorescence measurements, an excitation and emission matrix (EEM) was obtained, giving a fingerprint of the DOM composition of the water, data corrections for spectral biases and inner filter effects were implemented according to standard methods (Kothawala et al., 2013; Murphy et al., 2013). Prior to performing optical measurements, samples were filtered through 0.45 μm syringe filters polyethersulfone (PES) fitlers (rinsed with 25 mL DI water, followed by 5 mL of sample).

The DOM aromaticity of the non-radiolabelled feedwaters was measured as SUVA and a recently introduced fluorescence aromaticity index (ARIX) (Murphy, 2025). ARIX tracks SUVA and the ratio of natural organic matter molecules having larger versus smaller molecular size, in this study referred to as higher molecular weight (HMW) and lower molecular weight (LMW) humic-like organic matter. The regions for HMW and LMW humic-like organic matter have been defined as emission at 520 nm at excitation 320 nm and emission at 390 nm at excitation 320 nm, respectively (Murphy, 2025). The ratio of emission intensity at 520 nm divided by the emission intensity at 390 nm at an excitation wavelength of 320 nm was computed, yielding ARIX. SUVA was measured as the ratio between UVA_{254} and DOC concentration (Weishaar et al., 2003). An advantage of determining aromaticity using fluorescence spectroscopy, rather than using a combination of absorbance spectroscopy and carbon analysis, is greater sensitivity, which is particularly important at low concentrations of DOC (Murphy, 2025; Philibert et al., 2022).

3. Results and discussion

Results were obtained from eight parallel GAC columns receiving feedwaters with varying DOM compositions. To facilitate the discussion, we refer to the PFAS chain length as the number of fluorinated carbons (n_{CF}). The discussion of PFAS adsorption onto aged GAC is based on data obtained from feedwaters spiked with the PFAS concentrate, whereas the discussion of desorption effects is based on feedwaters without added PFAS concentrate. Additional evaluations are based on results obtained from tracking ^{14}C-labelled PFOA and ratios of HMW fraction of DOM and LMW fraction of DOM extracted from fluorescence measurements.

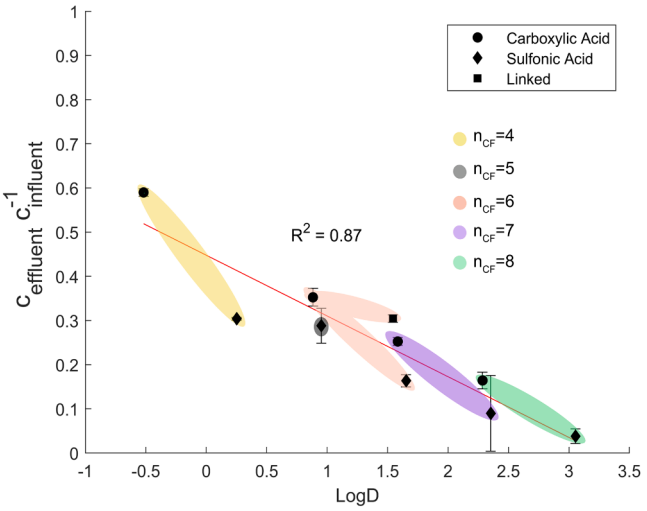

Fig. 3. PFAS breakthrough as a function of hydrophobicity for the GAC column fed with lake water from Ringsjön containing PFAS concentrate (*Ringsjön$_{PFAS}$*), average of day 15, 19, 20. Colour blocks represent different numbers of fluorinated carbons (n_{CF}). The data points are divided into different headgroups, (square) fluorotelomer, (circles) carboxylic acid and (diamonds) sulfonic acids. Error bars show propagated standard errors of mean of repeat measurements (n = 3).

3.1. Effect of changing feedwater DOM composition on PFAS adsorption hierarchies

Fig. 2 shows the breakthrough curves for PFAS with varying chain length and headgroups for columns fed with *Ringsjön$_{PFAS}$*. For PFAS with both carboxylic acid (CA)- and sulfonic acid (SA)- headgroups, an increased breakthrough of shorter-chained PFAS compared to longer-chained PFAS in the range of 4 to 8 n_{CF} was observed, similarly PFAS with a CA- headgroup were not as efficiently removed as PFAS with SA-headgroups with the same n_{CF} (Fig. 2A and 2B for *Ringsjön$_{PFAS}$* Figure S3A and S3B for *Vombsjön$_{PFAS}$*). Comparing PFAS breakthrough for columns fed with *Ringsjön$_{PFAS}$* and *Vombsjön$_{PFAS}$* which have small differences in aromaticity (SUVA 2.9 vs. 2.4 m^2 g_C^{-1}), revealed only

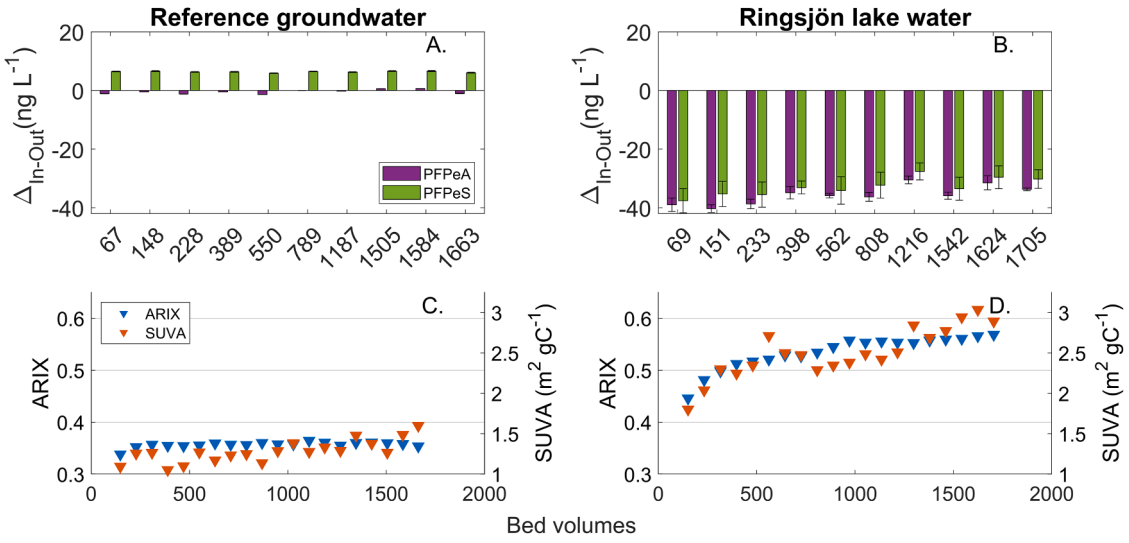

Fig. 4. Top row - Difference in PFAS concentration ng L^{-1} between influent (In) and effluent (Out) for two short chained PFAS (PFPeA n_{CF}=4 and PFPeS n_{CF}=5) in GAC columns fed with (A) *Original groundwater*; or (B) *Ringsjön* lake water. PFAS concentrations in *Ringsjön* lake water were < LOQ, influent concentrations were therefore set to LOQ for calculation purposes. Error bars show the propagated standard deviations of repeat measurements (n = 3). Bottom row – aromaticity measured using SUVA and ARIX in outgoing water for columns fed with (C) *Original groundwater* or (D) *Ringsjön* lake water.

minor differences in breakthrough behaviour. For bed volumes <700 deviations were typically <5%, while at bed volumes >700 the spread increased to <10%. The greater variability seen was primarily associated with shorter chained PFAS (n_{CF}= 4–6), which drove differences in breakthrough between the two feedwaters (Figure S3C-3F). However, the order of PFAS removal with respect to chain length and headgroup was conserved in relation to the hierarchy previously reported in the literature, even when the feedwater was altered and the aged GAC was exposed to feedwaters with higher DOM aromaticity (Appleman et al., 2014, 2013; Chow et al., 2022; Gagliano et al., 2020; McCleaf et al., 2017).

PFAS with higher LogD values (Table S14), and higher hydrophobicity, were more efficiently removed in the GAC columns than those with low LogD values (Figure 3 and S4). A strong negative correlation ($r(8) = -0.93$, $p < 0.001$, Table S15) was observed between PFAS breakthrough as a function of hydrophilicity for the two lake waters, *Ringsjön*$_{PFAS}$ and *Vombsjön*$_{PFAS}$, in line with previous studies on groundwater (Gagliano et al., 2020; McCleaf et al., 2017; Park et al., 2020; Tisler et al., 2025). The coloured blocks in Fig. 3 form clusters around PFAS with the same n_{CF}. Comparing PFAS with CA- and SA- headgroups, the trends in hydrophobicity were the same but with an offset equivalent to one fluorinated carbon atom. Thus, for the same number of n_{CF}, both the removal and LogD were lower for PFAS with CA-headgroups than SA-headgroups.

3.2. Effect of feedwater composition on the release of adsorbed PFAS

Fig. 4A and 4B show the difference in concentration between the influent and effluent waters of the two PFAS detected in the effluents: PFPeA (n_{CF}=4) and PFPeS (n_{CF}=5), for columns fed with the unspiked *Reference groundwater* and lake water *Ringsjön*. In the column fed with *Reference groundwater,* outgoing concentrations were ~ 0.5 ng L^{-1} for PFPeA and ~ 7 ng L^{-1} for PFPeS, which were below or similar to incoming feedwater concentrations (5 ng L^{-1} for PFPeA and 7 ng L^{-1} for PFPeS). However, in the column fed with lake water *Ringsjön,* outgoing concentrations of both PFPeA and PFPeS increased to 30–40 ng L^{-1} despite incoming concentrations below LOQs, 5.9 ng L^{-1} for PFPeA and 2.2 ng L^{-1} for PFPeS (Table S16).

The observed increases in PFAS concentration downstream of the aged GAC appear to result from desorption of historical loaded short-chained PFPeA and PFPeS, which is attributed to the change in DOM quality of the feedwater, since there was a much greater relative difference in SUVA (87%) between the two feedwaters compared to variations in DOC (16%, i.e. 2.5 ± 0.2 mg L^{-1} for *Reference groundwater* vs. 2.9 ± 0.1 mg L^{-1} for *Ringsjön*). Although it was attempted to bring all feedwaters to exactly the same DOC concentration at the beginning of the experiment, there was an unexpected difference in the measured DOC concentrations following dilution, possibly due to a combination of subsampling and measurement error. Still, given the relatively small difference in feedwater DOC concentrations, it is likely that the differences in DOM quality rather than quantity were responsible for the observed negative removal of PFAS from the aged GAC.

To further investigate this phenomenon, the relative size fraction and aromaticity of the outgoing waters were tracked using SUVA and ARIX fluorescence index. Changes in SUVA mirror changes in ARIX, albeit with more variability. For the column fed with *Reference groundwater,* ARIX remained relatively stable with a low to moderate increase (5%) for the whole duration of the column experiment ~1600 BV (Fig. 4C), whereas a more pronounced increase in ARIX (28%) was observed for the column fed with *Ringsjön* (Fig. 4D). The increase in ARIX for *Ringsjön* was connected to a systematic decrease in LMW fluorescent organic compounds in the outgoing water (Table S17), which suggests that the adsorption of LMW fractions of DOM could have contributed to the competition for adsorption sites previously occupied by PFPeA and PFPeS.

3.3. Water matrices: DOM quality and PFAS concentrations

Breakthrough curves of ^{14}C-labelled PFOA were determined for the investigated feedwaters. A good agreement (< ± 5%) between the breakthrough of ^{14}C-labelled PFOA and the non-radiolabelled PFOA was observed for *Ringsjön*$_{PFAS}$ ^{14C} and *Vombsjön*$_{PFAS}$ ^{14C} (Figure S5). This allowed for assessment of ^{14}C-labelled PFOA removal, in all GAC columns, including in cases when the concentrations of non-radiolabelled PFOA were below the LOQ (see the Supplementary data).

The mass balance for recovery of ^{14}C-labelled PFOA at the end of the column experiment was 101 ± 3% for *Reference groundwater*^{14C}, 104 ± 7% for *Ringsjön*^{14C}, 101 ± 3% for *Ringsjön*$_{PFAS}$ ^{14C}, 91 ± 4% for *Vombsjön*$_{PFAS}$ ^{14C}, with no formation of $^{14}CO_2$ in any of the columns (Table S18). This shows that the ^{14}C-labelled carboxylic headgroup of PFOA was recalcitrant to biodegradation during the column experiment. This observation is consistent with the poor biodegradability of PFOA

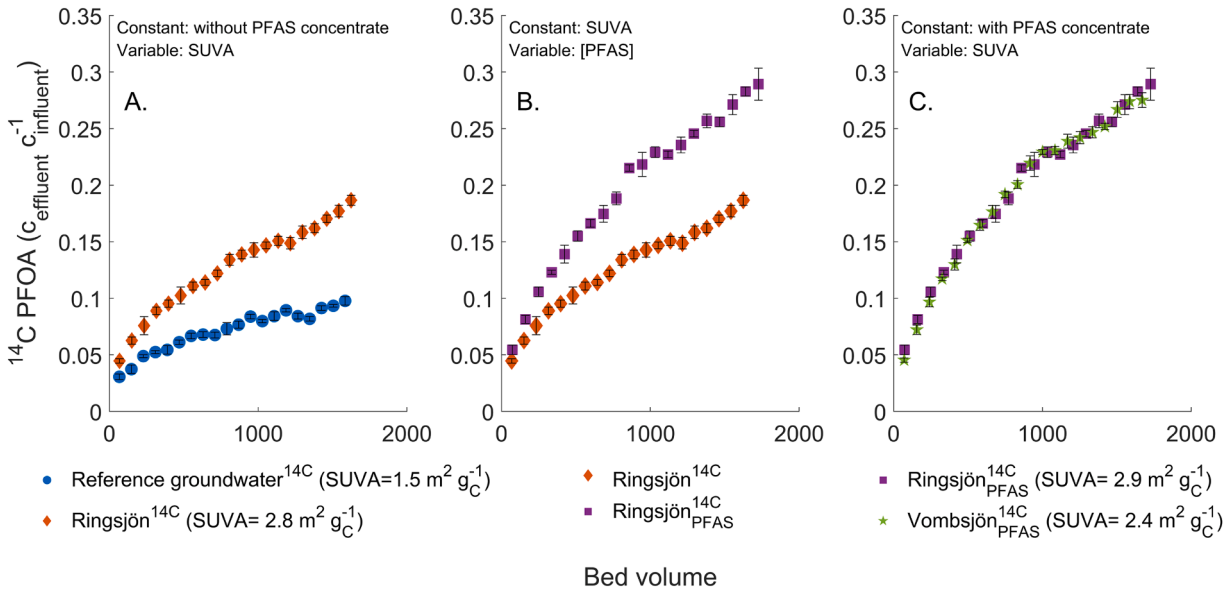

Fig. 5. Breakthrough curves of ^{14}C-labelled PFOA for waters with different SUVA and without PFAS concentrate (A), similar SUVA with and without PFAS concentrate (B), and variable SUVA with PFAS concentrate (C). Error bars show propagated standard deviation of repeat measurements (n = 3).

(Gellrich and Knepper, 2011; Rahman et al., 2014).

A higher breakthrough of ^{14}C-labelled PFOA was observed for the column fed with *Ringsjön^{14C}* than the one fed with *Reference groundwater^{14C}* (Fig. 5A). Thus, the removal efficiency of ^{14}C-labelled PFOA decreased as a result of the aromaticity in the feedwater increasing from what the GAC was accustomed to in the *Reference groundwater^{14C}* (SUVA=1.5 m^2 g$_C^{-1}$), to a higher aromaticity in *Ringsjön^{14C}* (SUVA=2.8 m^2 g$_C^{-1}$). A further deterioration of removal efficiency was observed in the presence of PFAS concentrate in *Ringsjön* lake water, despite the feedwaters having similar DOM compositions (Fig. 5B). These results indicate competition between the ^{14}C-labelled PFOA and the PFAS present in the feedwater after spiking with PFAS concentrate. However, for feedwaters containing PFAS concentrate (Fig. 5C) with moderate differences in aromaticity (SUVA 2.9 m^2 g$_C^{-1}$ vs 2.4 m^2 g$_C^{-1}$), small or no changes in removal efficiency were observed. This observation indicates that for feedwaters containing very high concentrations of PFAS, the competition between PFAS and the ^{14}C-labelled PFOA had a greater influence on GAC performance than the difference in DOM aromaticity.

4. Conclusions

By combining GAC column experiments with ^{14}C-labeling, analysis of 20 different PFAS, and fluorescence measurements, the following conclusions on the influence of feedwater composition on PFAS removal by aged GAC were drawn:

- Competition between LMW compounds in DOM and shorter-chained PFAS was observed when the aged GAC filter media was fed with water containing a new, more highly aromatic DOM matrix. Changes to feedwater DOM composition appeared to cause desorption of PFAS in conjunction with the adsorption of LMW fractions of DOM.
- For two different lake waters, the adsorption of longer-chained PFAS was favoured over shorter-chained PFAS, also SA- over CA- headgroups with the equivalent nCF, and higher hydrophobicity PFAS over lower. Changing the DOM matrix of the feedwater altered the extent of PFAS breakthrough but did not alter the relative trends in adsorption efficiency with respect to PFAS chain length and headgroup.
- Increasing the aromaticity of DOM in the feedwater, while maintaining nearly constant DOC concentration, substantially increased the breakthrough of 14C-labelled PFOA.

These findings highlight the importance of feedwater DOM quality for determining the performance of aged GAC filter media. In the current case, changing from lower to higher aromaticity resulted in earlier breakthrough of PFAS and favoured the adsorption of lower compared to higher molecular weight components in DOM. In treatment plants utilising GAC filters under variable source water conditions, it is important to consider the potential compromise in PFAS removal by aged GAC filters due to changes in the DOM composition of the feedwater.

CRediT authorship contribution statement

Aina McEvoy: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Validation, Visualization, Writing – original draft. **Małgorzata Szopińska:** Funding acquisition, Investigation, Resources, Writing – original draft. **Katarzyna Kozłowska-Tylingo:** Investigation, Writing – review & editing. **Kathleen Murphy:** Conceptualization, Funding acquisition, Methodology, Resources, Software, Supervision, Writing – review & editing. **Michael Cimbritz:** Conceptualization, Funding acquisition, Resources, Writing – review & editing. **Åsa Davidsson:** Conceptualization, Funding acquisition, Resources, Writing – review & editing. **Per Falås:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – review & editing.

Declaration of competing interest

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

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2026.126326.

Data availability

Data will be made available on request.

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