

Removal of perfluoroalkyl substances from drinking water with anion exchange resins and a divinylbenzene sorbent in column experiments

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ABSTRACT

The occurrences of per- and polyfluoroalkyl substances (PFAS) in drinking water may pose health risks to exposed consumers. PFAS removal with two anion exchange resins and a divinylbenzene sorbent in column experiments operated in parallel was investigated over a period of 520 days. For all adsorbents, removals of short-chain (C_3 to C_6 perfluorinated carbon atoms) PFAS were significantly lower than those of long-chain (C_7 to C_{10} perfluorinated carbon atoms) PFAS. The strong base resin removed the short-chain PFAS slightly better than the weak base resin. Sulfonates were removed better than carboxylates with the same number of fluorinated carbon atoms. Anion exchange resins exhibited a greater affinity for the aromatic fraction of dissolved organic carbon (DOC), resulting in a higher removal of UV-absorbing components than total DOC. The strong base resin outperformed the weak base resin and the divinylbenzene sorbent in terms of removing DOC concentrations and reducing UV_{254} absorbance. The reductions in UV_{254} and the elimination of short-chain PFAS exhibited correlations (Pearson's $r > 0.60$), indicating that UV_{254} abatement may be a possible surrogate parameter for short-chain PFAS elimination. Correlations (Pearson's $r > -0.60$) between the removals of nitrate and PFAS were observed.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a diverse group of persistent organic chemicals widely used in various industries over the past decades [1]. PFAS are suitable for a wide range of applications due to their distinct physical and chemical properties such as high surface activity, low surface tension, high biological persistence, high thermal and chemical stability, as well as being merely volatile and non-flammable [2–4]. PFAS contamination in water resources, along with the inefficacy of conventional treatment methods in removing them, poses a significant environmental challenge [5–8]. Drinking water contaminated with PFAS was reported to adversely impact human health including liver damage [9], testicular and kidney cancer [10], developmental effects [11], and endocrine-disrupting effects [12]. Sulfonic PFAS are considered to be more toxic than carboxylic PFAS, whereas short-chain PFAS exhibit less toxicity compared to long-chain PFAS [13–15].

Several treatment methods have been investigated for removing

PFAS, including coagulation (utilizing polyaluminum chloride, ferric chloride, or alum) [16], photocatalytic [17] and electrochemical oxidation [18] or adsorption (using ion exchange resins [19], or activated carbon (AC) [20,21]). Adsorption has been described as a comparably effective method for PFAS removal from contaminated water [4,19,22,23]. Effective adsorbents include activated carbon, β -cyclodextrin polymers, and fluorinated clays [4,22,24]. Ion exchange resins and granular activated carbon (GAC) have been widely applied for PFAS removal [22,25–28]. PFAS adsorption onto GAC, the most thoroughly investigated approach for treating PFAS contaminated water, has demonstrated efficient removals of long-chain PFAS, including perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS) [29,30].

Adsorption onto anion exchange resins is well-established for the removals of inorganic pollutants and natural organic matter from water [31,32] and also showed to be an effective method for the removal of PFAS from contaminated water [19,27]. Functional groups and carbon chain length of the respective PFAS were described to be very important factors for the adsorption characteristics. Long-chain PFAS adsorb better

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onto ion exchange resins than short-chain PFAS, and sulfonic PFAS with the same number of carbon atoms as carboxylic PFAS generally have a higher adsorption capacity onto ion exchange resins than the complementary carboxylic PFAS [26,33,34].

Although the scientific literature provides some insights about strong base anion exchange resins for PFAS removal, most studies have predominantly focused on the removals of PFOA and PFOS [19,27,35]. This study investigates the efficiency of a strong base anion exchange resin, a weak base anion exchange resin and divinylbenzene sorbent for the removals of short- and long-chain sulfonates and carboxylic PFAS. Specifically, this study aims to address 1) the long-term effectiveness of both anion exchange resins and divinylbenzene sorbent in continuously operated columns, 2) the impacts of chain length and functional groups on their removals, and 3) the competition among different PFAS and with organic and inorganic water constituents.

2. Materials and methodology

2.1. PFAS

In total, 10 PFAS were investigated, which were purchased from Th. Geyer GmbH (Renningen, Germany) with a purity level of at least 97% (Table 1). A stock solution with concentrations of 10 mg/L per PFAS was prepared by dissolving 10 mg of each PFAS in 1 L of ultrapure water.

2.2. Adsorbents

Two anion exchange resins and a divinylbenzene sorbent were examined characteristics of which are shown in Table 2.

2.3. Fixed bed column experiment

Three cylindrical glass columns with an internal diameter of 25 mm and a length of 270 mm were filled with MP, TP, and VP, respectively, with filter bed heights of 80 mm. A stainless-steel sieve of 150 μ m mesh was installed at the bottom and top of each column to avoid losses of filter material and the columns were closed with screw threads caps on both ends. Before starting the experiment, the columns were operated in an upward direction with drinking water (oxygenated groundwater) for one day to release any air bubbles entrapped in the filter beds (Fig. 1).

Influent drinking water was pumped into the columns in upstream direction using a peristaltic pump at a flow rate of 4 mL/min per channel. The pH of the test water ranged from 7.7 to 8.1, the average conductivity was 1233 ± 73 μ S/cm, the averaged dissolved organic carbon (DOC) concentration was 3.9 ± 1.0 mg/L, and UV absorbance at 254 nm (UV_{254}) was 0.050 ± 0.01 1/cm. The test water was contaminated with the stock solution to achieve an initial concentration of 2 μ g/L for each PFAS (total PFAS concentration 20 μ g/L). All PFAS were deprotonated under the operating conditions (Table 1).

Table 1
Characteristics of investigated PFAS.

Substance	CAS no.	Abbreviation	Molecular formula	Molar mass (g/mol)	ρ (g/cm ³)	pK _a	logK _{ow}	logK _{oc} (L/kg)	Functional group
Perfluorobutanoic acid	375-22-4	PFBA	C ₄ F ₇ O ₂ H	214.04	1.65	1.07	2.31	1.88	Carboxylic acid
Perfluoropentanoic acid	2706-90-3	PFPeA	C ₅ F ₉ O ₂ H	264.05	1.71	0.34	3.01	1.37	
Perfluorohexanoic acid	307-24-4	PFHxA	C ₆ F ₁₁ O ₂ H	314.05	1.76	0.78	3.71	1.91	
Perfluoroheptanoic acid	375-85-9	PFHpA	C ₇ F ₁₃ O ₂ H	364.06	1.79	2.29	4.41	2.19	
Perfluorooctanoic acid	335-67-1	PFOA	C ₈ F ₁₅ O ₂ H	414.07	1.8	4.20	5.11	2.35	
Perfluorononanoic acid	375-95-1	PFNA	C ₉ F ₁₇ O ₂ H	464.08	1.75	0.21	5.48	2.39	
Perfluorodecanoic acid	335-76-2	PFDA	C ₁₀ F ₁₉ O ₂ H	514.08	1.76	5.20	6.51	2.76	
Perfluoroundecanoic acid	2058-94-8	PFUnDA	C ₁₁ F ₂₁ O ₂ H	564.09	1.76	−5.2	7.21	3.3	Sulfonic acid
Perfluoroheptane sulfonic acid	375-92-8	PFHpS	C ₇ F ₁₅ SO ₃ H	450.13	–	–	–	3.0	
Perfluorooctane sulfonic acid	1763-23-1	PFOS	C ₈ F ₁₇ SO ₃ H	500.13	1.25	3.32	5.43	3.53	

<http://www.chemaxo.com> [21,30].

Table 2
Characteristics of adsorbents.

Treatment substance	Adsorbent		
	Lewatit MP 62 WS	Lewatit TP 106	Lewatit VP OC 1064 MD PH
Abbreviations	MP	TP	VP
Type	Anion exchange resin	Anion exchange resin	Ion exchange resin
Category	Weak-base	Strong-base	–
Ionic form	Free base (OH [−] ions)	Cl [−]	–
Functional group	Tertiary amine	Quaternary ammonium	None
Matrix	Styrene	Styrene	Divinyl-benzene (DVB)
Structure	Macroporous	Gel	Porous
Appearance	Beige, opaque	White, opaque	White, opaque
Total exchange capacity (eq/L)	1.7	0.65	–
Bead size (mm)	0.4–1.25	0.4–0.55	0.49 (± 0.05)
Water content (%)	44–52	37–47	54–63

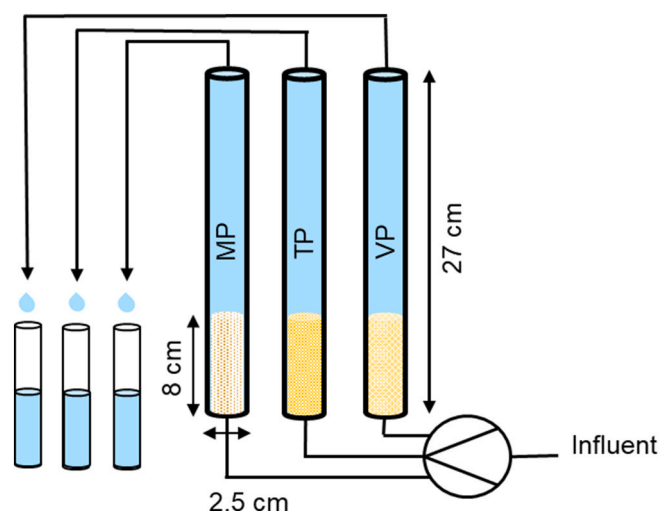


Fig. 1. Schematic diagram of the experimental setup with a volume flow of 4 mL/min resulting in a filter velocity of 0.48 m/h and 10 min empty bed contact time (EBCT).

2.4. Sampling

The influent and effluents were sampled with a sample volume of 100 mL each in glass bottles twice a month. Samples were filtered with membrane filters (Chromafil Xtra PET-45/25, Macherey-Nagel,

Germany) with 0.45 μm pore size, and stored at 4 °C until analysis.

2.5. Analytical methods

Conductivity and pH were measured with conductivity (Cond 340i, WTW, Germany) and pH (pH 3310, WTW, Germany) probes, respectively. DOC was measured with a total organic carbon analyzer (Vario TOC cube, Elementar Analysensysteme, Germany). A spectrophotometer (Lambda 25, UV/Vis PerkinElmer, USA) was used to measure UV_{254} . Specific UV absorbance (SUVA) was calculated using UV_{254} absorbances and DOC concentrations. Concentrations of chloride (Cl^-), bromide (Br^-), fluoride (F^-), nitrate (NO_3^-) and sulfate (SO_4^{2-}) were quantified using ion chromatography (Metrohm, Herisau, Switzerland). PFAS concentrations were measured by high-performance liquid chromatography coupled with mass spectrometry (QTRAP 6500+, AB Sciex) according to the method described by Ingold et al. [7].

2.6. Data analysis

Treatment efficacies were assessed using treated bed volumes (BV) and empty bed contact time (EBCT) according to Eq. (1) [36,37]. EBCT represents a theoretical contact time of water within the filter volume of the adsorbents (residence times are shorter).

$$\text{BV}_{\text{treated}} = \frac{V_{\text{treated}}}{V_{\text{ads}}} = \frac{t \times Q}{V_{\text{ads}}} \quad (1)$$

V_{treated} is the treated water volume [m^3], t [min] is experimental time, Q is the flow rate [mL/min] and V_{ads} is the volume [m^3] of the filter bed.

Pearson correlation coefficients (r) were calculated to determine linear correlations between different water quality parameters and PFAS.

3. Results and discussion

3.1. Removals of DOC, UV_{254} and anions

TP and MP resins achieved maximum 50% and 42% reductions of DOC, respectively, within the initial 6 days (880 BV) of operation, whereas VP exhibited 17% DOC removal (Fig. 2). UV_{254} abatements reached a maximum 91% using anion exchange resins (both TP and MP), and VP showed a maximum 46% reduction of UV_{254} within the same period of operation (Fig. 2).

The DOC removal of TP became constant (approx. 20%) after 20×10^3 BV. At this throughput, UV_{254} abatement was at ca. 30%, which continuously decreased further until the end of the experiment. Until ca. 40×10^3 BV, percentage of UV_{254} abatement was higher than DOC

removal for TP. For MP, constant DOC removal in a corridor between ca. 30 and 20% was reached already after approximately at 36×10^3 BV. UV_{254} abatements decreased progressively over the course of the experiment. Until ca. 40×10^3 BV, the UV_{254} abatements with anion exchange resins was higher than DOC removal. The high removal of the UV_{254} absorbing DOC fraction with the anion exchange resins was likely due to stronger ionic interactions with negatively charged organic fractions [38]. Previous studies also suggested that strong-base resins exhibit high removal efficiencies of UV absorbing DOC components, with some studies reporting up to 93% removal of such compounds [39–42]. TP and MP showed similar UV_{254} abatements until ca. 15×10^3 BV, after that MP achieved ca. 10 percentage points more UV_{254} abatement until the end. VP had a weaker starting phase for UV_{254} abatement, but from ca. 15×10^3 BV onwards it showed an almost identical course (ca. 5 percentage points lower) as TP.

Removals to small extents were observed for F^- and NO_3^- over the course of the experiments (Fig. 3). All adsorbents showed increases in NO_3^- removal after 20×10^3 BV (for MP) and 30×10^3 BV (for TP and VP) and the total reduction of NO_3^- was less than 15%. F^- removals were constant, except for MP, which showed a slight increase (within the 0–10% range) towards the end of the experiment.

3.2. PFAS removals

Removals of the short-chain (C_3 to C_6 ; perfluorinated carbon atoms) and the long-chain (C_7 to C_{10} ; perfluorinated carbon atoms) PFAS in the column experiments varied among the three different materials, as illustrated in Fig. 4. The removals of PFBA and PFPeA exhibited a more rapid decline than removals of other short-chain PFAS for MP (Fig. 4a). The order of removals for short-chain PFAS with anion exchange resins (MP and TP) followed the sequence 1) PFBA, 2) PFPeA, 3) PFHxA and 4) PFHpA. Within the test period of 520 days, complete breakthroughs occurred for three PFAS (PFBA, PFPeA and PFHxA) with MP (Fig. 4a), while two PFAS (PFBA and PFPeA) exhibited complete breakthroughs with TP (Fig. 4b). After breakthrough, effluent concentrations of PFBA, PFPeA and PFHxA (for MP) surpassed their influent concentrations resulting in negative removals (Fig. 4a and b). The chromatographic effect of the short-chain PFAS was likely due to the displacement of the shorter chain PFAS by natural organic matter or longer chain PFAS [37].

No removal was observed for PFBA for the adsorbent VP (Fig. 4c), and PFPeA and PFHxA showed quickly decreasing removals. However, no chromatographic effect was observed, probably due to the very low loading of the two PFAS on the adsorbent. For VP, PFHpA showed a complete breakthrough after ca. 30×10^3 BV and a desorption period. MP and TP illustrated better removals than the VP for the short-chain PFAS (including PFPeA, PFHxA and PFHpA) (Fig. 4a, b and c). The TP

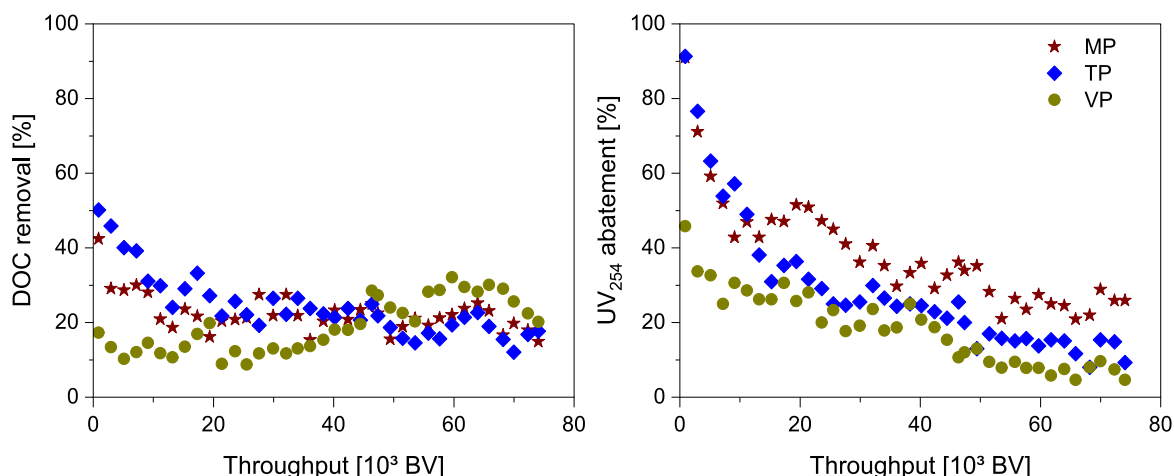


Fig. 2. DOC and UV_{254} removal of the anion exchange resins (MP and TP) and the adsorbent (VP).

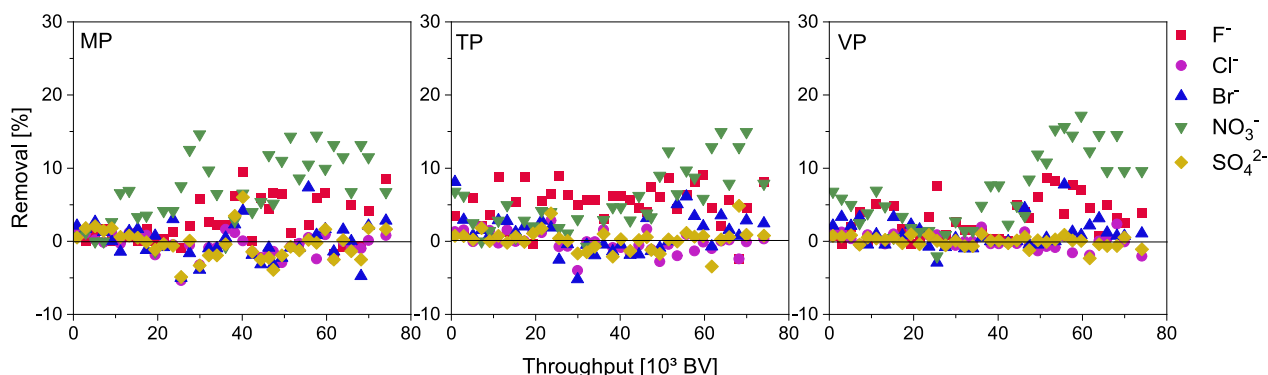


Fig. 3. Anion removal with the anion exchange resins (MP and TP) and the adsorbent (VP). Concentration profiles are shown in Fig. S1.

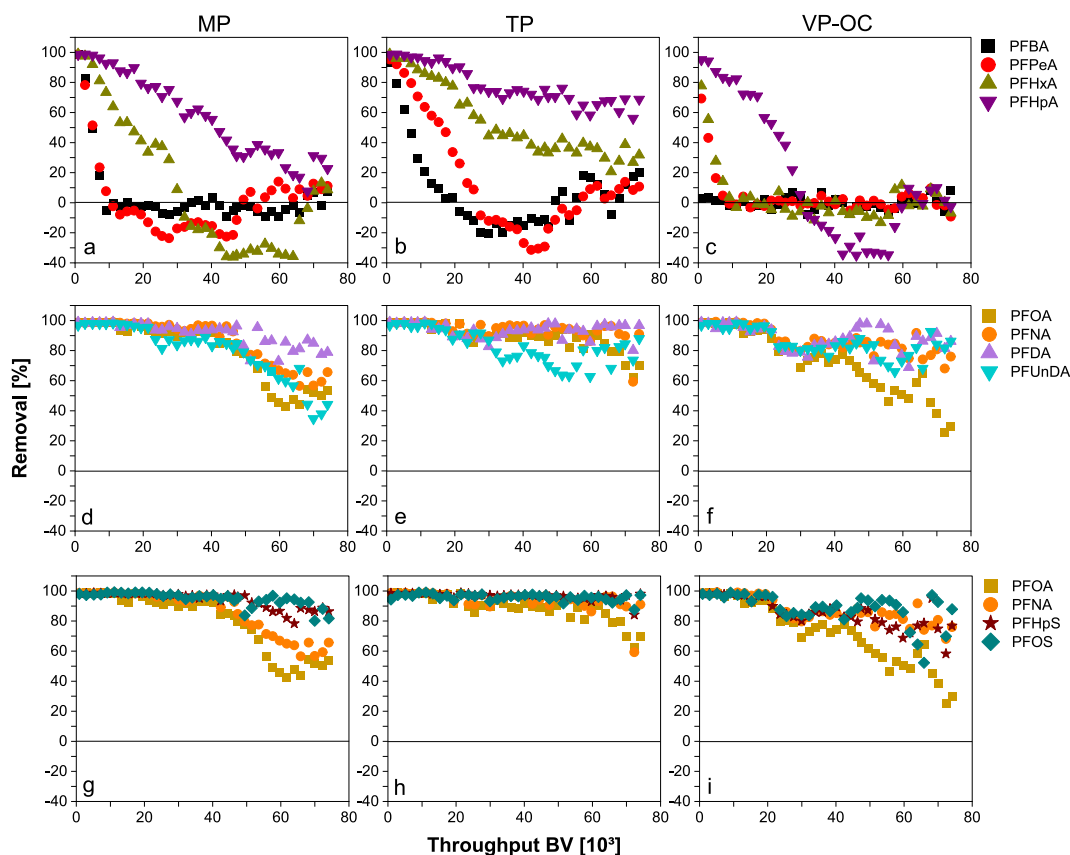


Fig. 4. PFAS removals with MP (a, d and g, left column) TP (b, e and f, middle) and VP (c, f and i, right) for short-chain carboxylates (first row), long-chain carboxylates (second row) and different functional groups but with similar numbers of perfluorinated C-atoms (third row).

resin exhibited a better performance than the MP resin for the removal of short-chain PFAS (such as PFHxA and PFHpA). The short-chain (PFBA and PFPeA) PFAS broke through relatively quickly, making their removal challenging. Similarly, Riegel et al. [43] reported that adsorbent breakthroughs were achieved rapidly when dealing with short-chain PFAS.

As expected, the adsorbents showed better performances for the removals of long-chain PFAS than for short-chain. Complete breakthroughs of long-chain PFAS (perfluorinated carbon atoms C_7 to C_{10}) did not occur during the course of our experiments until 80×10^3 BV (equivalent to 520 days of operation) (Fig. 4d, e and f). The enhanced removals of long-chain PFAS is attributed to a decreasing polarity of PFAS with increasing chain length and hence increased hydrophobic-hydrophobic interactions with the adsorbents [43–45]. TP showed the greatest removals for long-chain PFAS followed by MP and VP.

Similarly, previous studies have reported that anion exchange resins had higher adsorption capacities for PFAS than other adsorbents, including granular activated carbon [41,46,47].

Compared to PFHpS, the removals of PFOA (i.e., with the same length of perfluorinated carbon chain) decreased earlier for all adsorbents (Fig. 4g, h, i). For the anion exchange resins, PFOS was completely removed over the entire operation and for VP, a substantial decrease in removal started not before 60×10^3 BV. PFOA removals decreased after 40×10^3 BV for MP and after 60×10^3 BV for TP. Sulfonic PFAS (including PFHpS and PFOS) of the same number of C atoms as the corresponding carboxylic PFAS (PFOA and PFNA) showed higher removals than their carboxylic acids equivalents for MP (Fig. 4g) TP (Figs. 4h), and VC (Fig. 4i). Similarly, Zeng et al. [34] reported that sulfonic PFAS are adsorbed more easily compared to carboxylic PFAS with the same lengths of perfluorinated carbon chains.

The efficacy of TP in adsorbing PFAS, including short-chain PFAS, surpassed that of the MP. The higher efficiency of the TP resin for short-chain PFAS removal likely stems from its quaternary ammonium functional groups and thus its stronger electrostatic attraction to anionic the functional (carboxylate and sulfonate) groups of the PFAS compared with tertiary amine as functional group of MP. In PFAS molecules, highly electronegative fluorine atoms pull electron density away from the functional group, facilitating the acidic character of carboxylic and sulfonic acids, respectively. This also stabilizes the negative charge of the corresponding anions and could be the origin of their increased affinity for positively charged resins [48]. Although the MP resin with tertiary amine functional groups demonstrated lower removals of short-chain PFAS, it notably excelled in eliminating sulfonic PFAS, even those of short-chain length (Fig. 4g). According to the manufacturer's information on the TP resin it is especially suitable to retain soft anions, to which perfluoroalkyl carboxylates and sulfonates could be counted.

3.3. Correlations between quality parameters and PFAS removal

Pearson's correlations were calculated between the reductions of different parameters (as a function of time) and PFAS removals for all materials (Fig. 5). Moderate to strong correlations (Pearson's $r > 0.60$) were observed between the abatements of UV₂₅₄ and the removals of short-chain PFAS for the anion exchange resins. These correlations appeared stronger for TP (Pearson's $r > 0.80$) than for MP. This suggests that UV₂₅₄ abatements may provide information as surrogate parameter for short-chain PFAS removals [49]. The high abatement of UV₂₅₄ with

the strong-base TP was likely due to its higher ion exchange capacity [39,42]. No clear correlations were found between UV₂₅₄ abatements and removals of PFHpS and PFOS (Pearson's $r < 0.50$). In case of VP, no correlations could be found between short-chain PFAS removals and UV₂₅₄ abatements (Fig. 5c), however stronger correlations (between PFAS removals and UV₂₅₄ abatements) were observed as the chain length increased (for example, with PFOA and PFNA Pearson's $r > 0.80$).

The removals of inorganic ions as unexpectedly nitrate removals correlated negatively (Pearson's $r > -0.60$) with the removals of PFAS (above perfluorinated chain length C₄) for the weak-base resin MP and the correlations appeared weaker for TP. For the VP, concentrations of nitrate seemed to influence the removal of PFOA. Several correlations (Pearson's $r > 0.70$) could be derived for MP between the removals of PFAS (above chain length C₅) and other parameters, whereas, for TP fewer correlations occurred. For example, removals of PFHxA (Fig. S2) and PFUnDA were significantly correlated (Pearson's $r = 0.81$). For VP, the least number of correlations were derived. Such correlations can be practically useful as they suggest the potential for using surrogate parameters to monitor PFAS breakthrough in real-time, especially when direct PFAS analysis is not feasible.

4. Conclusion

The study investigated the effectiveness of three absorbents for the removal of 10 different PFAS in column experiments. The two anion exchange resins showed higher capacities for UV₂₅₄ absorbing constituents than for the bulk DOC most likely because of higher affinities of

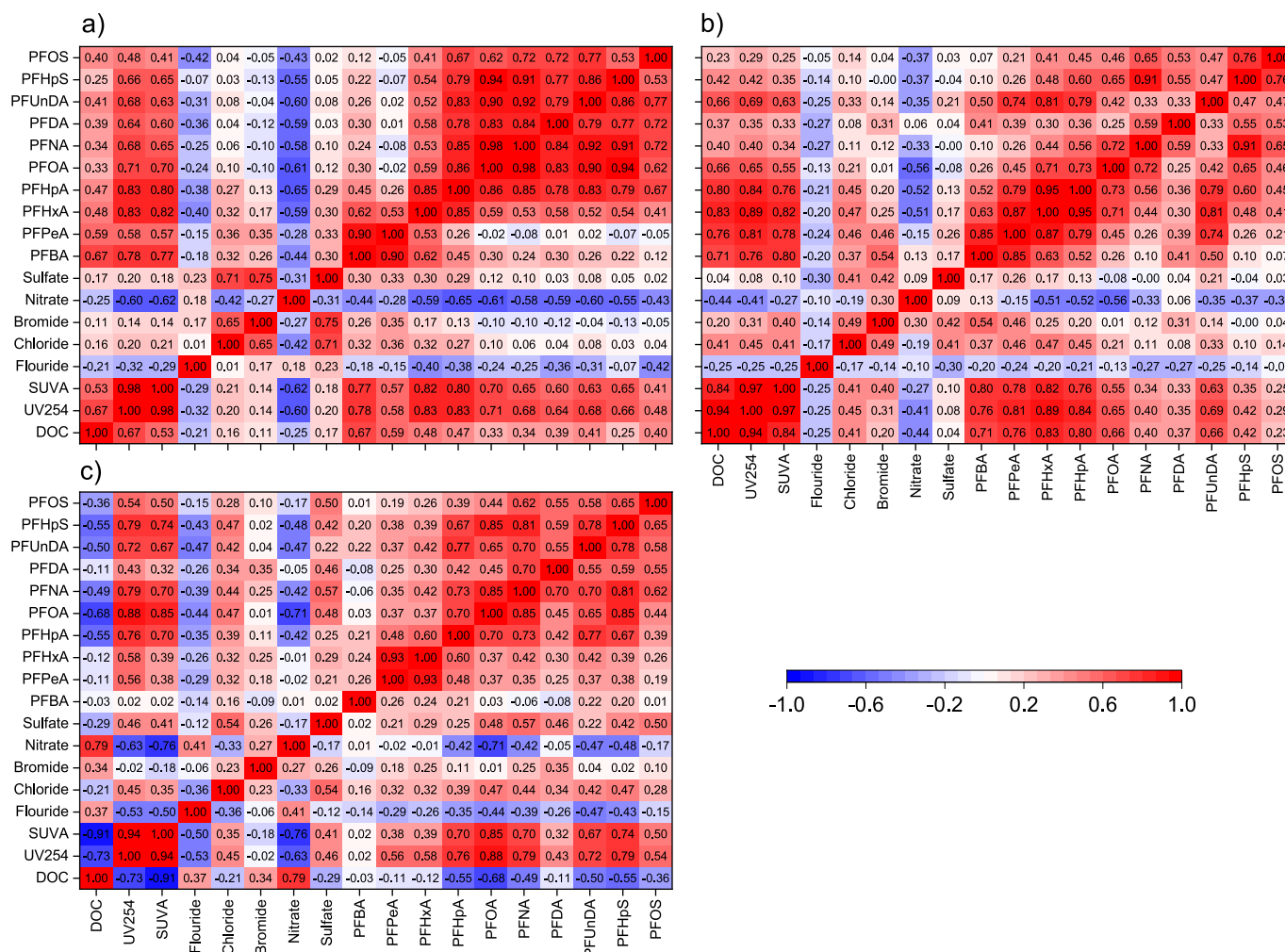


Fig. 5. Heat map of Pearson's correlations between reductions of different parameters and the removal of PFAS with a) MP b) TP and c) VP.

anion exchange resins for the aromatic fractions of the DOC compared to other fractions. The strong base resin (TP) showed substantial reductions of DOC and UV₂₅₄ compared to MP and VP.

Both anion exchange resins showed better removals for short-chain PFAS than VP. Furthermore, the strong base resin (TP) exhibited slightly better performances than the weak base resin (MP) in removing short-chain PFAS. Long-chain PFAS were effectively removed by both anion exchange resins and the adsorbent. In general, TP outperformed MP in terms of all tested PFAS and VP showed the least potential for PFAS removal. The UV₂₅₄ abatement revealed significant Pearson's correlations with short chain PFAS removals and is therefore a promising surrogate parameter.

CRedit authorship contribution statement

Omamah Ali: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Muhammad Zeeshan:** Writing – review & editing. **Daniel Mahringer:** Writing – review & editing. **Alexander Kämpfe:** Investigation. **Aki Sebastian Ruhl:** Writing – review & editing, Supervision, Resources, Project administration.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Omamah Ali reports administrative support and equipment, drugs, or supplies were provided by German Environment Agency Berlin City Campus. Omamah Ali reports a relationship with German Environment Agency Berlin City Campus that includes: non-financial support. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jwpe.2026.110201>.

Data availability

Data will be made available on request.

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