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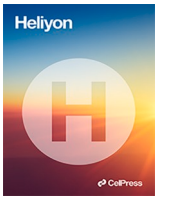

Research article

Transformation of potentially persistent and mobile organic micropollutants in column experiments

Muhammad Zeeshan [a,b,*], Pia Schumann [a,b], Silke Pabst [c], Aki Sebastian Ruhl [a,b]

[a] German Environment Agency, Section II 3.3, Schichauweg 58, 12307, Berlin, Germany
[b] Technische Universität Berlin, Water Treatment, KF4, Str. des 17. Juni 135, 10623, Berlin, Germany
[c] German Environment Agency, Section II 3.1, Schichauweg 58, 12307, Berlin, Germany

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ABSTRACT

The occurrence of potentially persistent and mobile (PM) organic micropollutants (OMP) in the aquatic environment is recognized as a severe threat to water resources and drinking water suppliers. The current study investigated long-term fate (persistency and bio-transformation) of several emerging contaminants in a simulated bank filtration (BF) for the first time. In parallel, four sand column systems were operated with groundwater and continuously spiked with an average concentration of 1 μg/L for 24 OMP. Each column system consisted of two sand columns connected in series. Presumably, biological activities in the first column were higher than in the second column, as dissolved oxygen utilization, dissolved organic matter (DOM) and UV absorbance at 254 nm (UV_{254}) reduction rates were high in the first column. This study revealed that 9 out of 24 OMP were persistent and mobile throughout the study under oxic conditions and within a hydraulic retention time (HRT) of 12 days. However, 2 (out of 9) OMP were persistent but showed sorption behavior. 15 (out of 24) OMP displayed bio-transformation, 4 were eliminated entirely within 4.5 days of HRT. Others showed constant or improved degradation with the adaptation (or operation) time. Improved degradation with adaption was high in the bioactive sand columns. However, 8 OMP showed improved elimination at high HRT, even in low biologically active columns. In addition, no significant effect of the DOM on the eliminations of OMP was found except for 4-hydroxy-1-(2-hydroxyethyl)-2,2,6,6,-tetramethylpiperidine (HHTMP), 2-methyl-2-propene-1-sulfonic acid (MPSA) and sulfamethoxazole (SMX). The eliminations of HHTMP (Pearson's $r > 0.80$, $p < 0.05$), MPSA (Pearson's $r > 0.70$) and SMX (Pearson's $r > 0.80$) correlated with the removals of humic substances in the sand columns. Overall, adaptation time and HRT play a crucial role in the elimination of emerging OMP through BF, yet at the same time several OMP exhibit persistent behavior.

1. Introduction

Water pollution is an emerging issue due to the release of industrial chemicals, pharmaceuticals and personal care products to the aquatic environment, mainly by incompletely treated wastewater disposal. Consequently, surface water contains multiple organic micropollutants (OMP). A highly polar and persistent OMP is referred to as a persistent mobile organic compound (PMOC) and alternatively known as a persistent and mobile substance (PM) that only degrades very slowly (with half-lives in fresh water above 40

* Corresponding author. German Environment Agency, Section II 3.3, Schichauweg 58, 12307, Berlin, Germany.
E-mail addresses: muhammad.zeeshan@uba.de, engr.m.zeeshan@live.com (M. Zeeshan).

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days) and show a low tendency to sorb to surfaces or organic matter in soil sediments and thus might reach to the drinking water [1,2]. Persistency and mobility are two intrinsic properties of PMOC. Persistency refers to low biodegradation, and mobility refers to low sorption potential in soil, sediments and water. Therefore, these PM substances have been prioritized as pollutants of concern for drinking water, particularly in Europe, to protect drinking water [3]. Recent studies showed the presence of emerging contaminants such as cyanoguanidine (CG), n-(3-(dimethylamino)-propyl)methacrylamide (MAPMA), 2-methyl-2-propene-1-sulfonic acid (MPSA), benzyldimethylamine (BDMA), benzyltrimethylammonium (BETMAC), adamantan-1-amine (ATA), 1,3-di-*o*-tolylguanidine (DIOTOG), 1,3-diphenylguanidine (DPG), 4-hydroxy-1-(2-hydroxyethyl)-2,2,6,6,-tetramethylpiperidine (HHTMP), melamine (MEL), 2-acrylamido-2-methylpropane sulfonate (AAMPS), dimethylbenzenesulfonic acid (DMBSA), *p*-toluenesulfonic acid (PTSS) and trifluoromethanesulfonic acid (TFMSA) in surface and groundwater with concentrations in the ranges of ng/L and μg/L [4–10]. In addition, 53% of the detected emerging PM substances were above 0.1 μg/L [4,8,11]. In Europe, there is an ongoing discussion to register PM substances under REACH legislation (registration, evaluation, authorization and restriction of chemicals (EC 1907/2006) regulation Annex XIII), similar to the previously regularized PBT (persistence, bioaccumulative and toxic) substances. PM substances persist and accumulate in water with consequent exposure to humans and organisms.

Bank filtration (BF) is a managed aquifer recharge system and is effectively used to supply drinking water in Europe and many other counties [12,13]. In BF, surface water infiltrates into the aquifer. Thereby, infiltrating water undergoes a series of physical, chemical and biological changes resulting in the attenuation of contaminants. Previous studies revealed the attenuation of several OMP through BF [14–18]. Different factors such as redox condition, temperature, pH, porosity of the medium and travel time, play an essential role in the performance of the BF system [15,19,20]. Controlled sand columns are generally used to evaluate the fate of OMP by simulating BF processes.

This study was conducted 1) to elucidate the long-term behavior of 24 potentially persistent and mobile OMP such as CG, MAPMA, MPSA, BDMA, BETMAC, ATA, DIOTOG, DPG, HHTMP, MEL, AAMPS, DMBSA, PTSS and TFMSA through simulated BF (the fate of these OMP is reported for the first time here), and 2) to determine the influence of dissolved organic matter (DOM) on the elimination of OMP.

2. Methodology

2.1. Experimental setup

A quadruplicate experiment was performed by connecting two columns in series (Fig. 1). Each column with a length of 1 m (and sampling ports at 0.25, 0.50 and 0.75 m) and an internal diameter of 0.14 m (0.015 m^2 surface area) was filled with sand from a full-scale infiltration basin for managed aquifer recharge (Saatwinkel, Berlin). The sand was sieved to remove particles larger than 4 mm. The first columns of each system were filled with homogenized sand from 0 to 100 cm depth, and the second columns were filled with sand from 20 to 100 cm depth (excluding the top colmation layer with higher POC content, sand properties are provided in Table S1) [21]. In the current study, the columns were operated at a downstream flow rate of 60 mL/h, corresponding to a filter velocity of ca. 0.1 m/d (approximate pore water velocity 0.2 m/d) and a hydraulic retention time (HRT) of 6 days in each column (based on previous experiments by Hellauer et al. [22] with the same column setup). The influent was delivered to the top of first columns using a 4-channel Ismatec pump.

Initially, the columns were acclimatized using groundwater for six months. The pH of the influent groundwater was between 8.0 and 8.3 and averaged values of conductivity, dissolved organic carbon (DOC), dissolved oxygen (DO) and UV absorbance at 254 nm (UV_{254}) were 787 ± 12 μS/cm, 2.9 ± 0.1 mg/L, 9.1 ± 0.9 mg/L and 9.9 ± 0.4 1/m, respectively. Concentrations of nitrate (NO_3^-), ammonium (NH_4^+) and sulfate were 3.59 ± 0.17 mg/L, 0.02 ± 0.01 mg/L and 97.3 ± 2.3 mg/L, respectively. The influent water temperature was between 12 and 14 °C during the experimental period. Previously, Hellauer et al. [22] operated these columns in a similar arrangement but using different intermediate oxidation techniques (between the first and second columns).

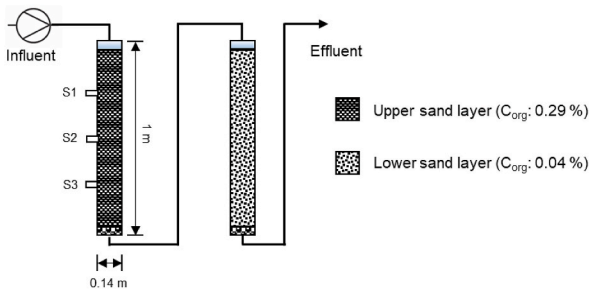

Fig. 1. Schematic diagram of the experimental setup with sampling ports S1 (0.25 m), S2 (0.50 m) and S3 (0.75 m). Samples at the end of the first column correspond to 1 m depth or 6 days HRT and at the end of the second column to 2 m depth or 12 days HRT.

2.2. Organic micropollutants

A micropollutant stock solution of 10 mg/L was prepared by dissolving 10 mg of 24 compounds (all analytical grade) in 1 L of ultrapure water by mixing for a minimum of three days. The stock solution was dosed (target concentation 1 μg/L) into the influent

Table 1
Physio-chemical properties and elemental compositions of the OMP.

Name	Abbreviation	CAS. NO.	Weight (g/mol)	Formula	Composition	pK_a	K_{oc}^b
Cyanoguanidine	CG	461-58-5	84.08	$C_2H_4N_4$	C (28.57%), H (4.80%), N (66.63%)	5.15	0.38
Benzotriazol	BTA	95-14-7	119.12	$C_6H_5N_3$	C (60.50%), H (4.23%), N (35.27%)	9.04 0.22	1.25
Sulfamethoxazole	SMX	723-46-6	253.28	$C_{10}H_{11}N_3O_3S$	C (47.42%), H (4.38%), N (16.59%), O (18.95%), S (12.66%)	1.83; 1.85; 5.57; 5.60; 5.65 [a]	
Carbamazepine	CBZ	298-46-4	236.27	$C_{15}H_{12}N_2O$	C (76.25%), H (5.12%), N (11.86%), O (6.77%)	2.25	
Primidon	PRI	125-33-7	218.25	$C_{12}H_{14}N_2O_2$	C (66.04%), H (6.47%), N (12.84%), O (14.66%)		
N-(3-(dimethylamino)-propyl) methacrylamide	MAPMA	5205-93-6	170.25	$C_9H_{18}N_2O$	C (63.49%), H (10.66%), N (16.45%), O (9.40%)	9.3	
2-Methyl-2-propene-1-sulfonic acid sodium salt	MPSA	1561-92-8	158.15	$C_4H_7NaO_3S$	C (30.38%), H (4.46%), Na (14.54%), O (30.35%), S (20.27%)	-0.99	1.26
Benzyldimethylamine	BDMA	103-83-3	135.21	$C_9H_{13}N$	C (79.95%), H (9.69%), N (10.36%)	8.9	2.01
Benzyltrimethylammonium chloride	BETMAC	56-93-9	185.7	$C_{10}H_{16}ClN$	C (64.68%), H (8.69%), Cl (19.09%), N (7.54%)		2.95
Adamantan-1-amine	ATA	768-94-5	151.25	$C_{10}H_{17}N$	C (79.41%), H (11.33%), N (9.26%)	10.71	2.23
1,3-Di-*o*-tolylguanidine	DIOTOG	97-39-2	239.32	$C_{15}H_{17}N_3$	C (75.28%), H (7.16%), N (17.56%)		2.91
1,3-Diphenylguanidine	DPG	102-06-7	211.26	$C_{13}H_{13}N_3$	C (73.91%), H (6.20%), N (19.89%)		2.86
4-Hydroxy-1-(2-hydroxyethyl)-2,2,6,6,-tetramethylpiperidine	HHTMP	52722-86-8	201.31	$C_{11}H_{23}NO_2$	C (65.63%), H (11.52%), N (6.96%), O (15.89%)		1.23
Melamine	MEL	108-78-1	126.12	$C_3H_6N_6$	C (28.57%), H (4.80%), N (66.63%)	9.56	0.72
Sodium 2-acrylamido-2-methylpropane sulfonate	AAMPS	5165-97-9	229.23	$C_{10}H_{11}N_3O_3S$	C (47.42%), H (4.38%), N (16.59%), O (18.95%), S (12.66%)		0.99
Acesulfame K	ACE	55589-62-3	201.24	$C_4H_4KNO_4S$	C (23.87%), H (2.00%), K (19.43%), N (6.96%), O (31.80%), S (15.93%)	3.02	0.86
Dimethylbenzenesulfonic acid	DMBSA	25321-41-9	186.23	$C_8H_{10}O_3S$	C (51.60%), H (5.41%), O (25.77%), S (17.22%)	-1.94	1.66
Amidotrizoesäure/Diatrizoic acid	DCA	117-96-4	613.91	$C_{11}H_9I_3N_2O_4$	C (21.52%), H (1.48%), I (62.01%), N (4.56%), O (10.42%)		
p-Toluenesulfonic acid	PTSS	104-15-4	172.2	$C_7H_8O_3S$	C (48.83%), H (4.68%), O (27.87%), S (18.62%)	-2.14	1
Saccharine	SAC	81-07-2	183.18	$C_7H_5NO_3S$	C (45.90%), H (2.75%), N (7.65%), O (26.20%), S (17.50%)	1.94	1.2
Trifluoromethanesulfonicacid	TFMSA	1493-13-6	150.08	$C_2F_6O_6S_2$	C (5.52%), H (0.00%), F (26.18%), O (22.04%), S (14.72%)		-0.0
Dimethylbenzenesulfonic acid sodium salt	XSA	1300-72-7	208.21	$C_8H_9NaO_3S$	C (46.15%), H (4.36%), Na (11.04%), O (23.05%), S (15.40%)		1.66
Diclofenac	DCF	15307-79-6	318.1	$C_{14}H_{10}Cl_2NNaO_2$	C (52.86%), H (3.17%), Cl (22.29%), N (4.40%), Na (7.23%), O (10.06%)	4.50 4.02 [c]	
Valsartan acid (2′-(2H-tetrazol-5-yl)-[1,1′-biphenyl]-4-carboxylic acid)	VSA	164265-78-5	266.25	$C_{14}H_{10}N_4O_2$	C (63.15%), H (3.79%), N (21.04%), O (12.02%)	4.0	

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a [23].
b [24].
c [25].

groundwater. The physico-chemical properties of all the OMP used in this study are summarized in Table 1.

2.3. Sampling

Sampling started after one month of addition of the stock solution and samples were collected weekly from the influent (0 m), after the first column (1 m) and from the effluent (2 m) in glass bottles of 100 mL. Samples from the intermediate sampling ports (0.25 m, 0.50 m and 0.75 m, compare Fig. 1) were collected after three months because some OMP were fully eliminated within the first column (1 m). Collected samples were filtered (0.45 μm, CHROMAFIL Xtra PA-45/25, Macherey-Nagel, Germany) and partly analyzed (pH, conductivity, DOC, UV_{254} and fluorescence) within 24 h, while filtered samples were stored at 4 °C for cation/anion and LC-MS/MS measurements.

2.4. Analytical methods

pH and conductivity were measured using a pH probe (WTW pH 3310) and conductivity probe (WTW Cond 340i). A total organic carbon (TOC) analyzer (Vario TOC cube, ElementarAnalysensysteme, Germany) was used to analyze DOC. UV_{254} was measured with a spectrophotometer (Lambda 25, UV/Vis spectrometer, PerkinElmer, USA). The ratio of UV_{254} to DOC was used to calculate the specific UV absorbance (SUVA).

Concentrations of anions and cations were measured by ion chromatography (930 Compact IC Flex, Metrohm, Switzerland).

DO and the temperature was measured optically in flow-through cells (Fibox 4 trace, PreSens, Germany).

Fluorescence of 90 samples were analyzed using a FluoroMax-4 (HORIBA Jobin Yvon, Edison, NJ, USA) with a scan speed of 4800 nm/min, 5 nm bandwidth, excitation wavelength of 220–500 nm (5 nm interval) and emission wavelength of 250–600 nm (2 nm interval). Blank excitation-emission matrix (EEM) datasets were acquired using ultra-pure water.

OMP were analyzed using high-performance liquid chromatography (HPLC; Agilent 1290 Infinity, Agilent, Waldbronn, Germany) coupled to a 5500 triple-quadrupole mass spectrometer (MS/MS) with Turbo V ion source (Sciex, Darmstadt, Germany). An HPLC separation was performed using Atlantis (Waters, Milford, USA) T3 column (3.0 μm, 2.1 mm × 100 mm). The mobile phase flow rate was 0.3 mL/min with a gradient (Table S2) of eluent A (water containing 0.1% (v/v) formic acid) and eluent B (methanol containing 0.1% (v/v) formic acid). The injection volume of the sample was 50 μl and the column temperature was set to 35 °C. The mass spectrometer was operated in positive and negative electrospray ionization (ESI) mode and in multiple reaction monitoring (MRM). Method performance evaluation, limits of quantification and experimental source parameters are provided in Tables S3 and S4 in the supplementary information.

2.5. Data evaluation

The staRdom package for R [26] was used to process raw EEM datasets by blank subtraction, inner-filter effect correction, Raman normalization, scattering removal and excitation and emission correction factors. Fluorescence indices and PARAFAC was applied to corrected EEM datasets.

EEM datasets were used to estimated fluorescence index (FI), biological index (BIX) and humification index (HIX). The FI was calculated as ratio of emission intensities (450/500 nm) for an excitation wavelength of 370 nm. The ratio of emission intensities (380/430 nm) for an excitation wavelength of 310 nm was used for the estimation of BIX. The HIX was evaluated as ratio of the sum of emission intensities ((435–480)/(300–345) nm) for an excitation wavelength of 254 nm [27,28].

Rate constants for OMP (*k*) were calculated as the slope of the natural logarithms plotted against the time following first-order kinetics (Equation (1)).

$$y = y_o e^{-kt} \tag{1}$$

where, **y** is the concentration at time **t**, $\mathbf{y_o}$ is the initial concentration and **e** is Euler's number. The half-life was calculated using

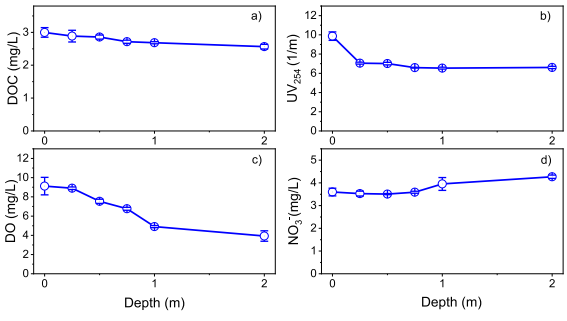

Fig. 2. Averaged values of (a) DOC, (b) UV_{254}, (c) DO and (d) NO_3^- at different depths in the quadruplicate sand columns, error bars indicate standard deviations (for each sand column, n > 10).

Equation (2).

$$t_{1/2} = \frac{Ln(2)}{k} \quad (2)$$

where, $t_{1/2}$ is the half-life of the OMP.

3. Results and discussion

3.1. Redox condition and bulk organic parameters

The quality of water at different sampling ports in the sand columns was not significantly different (ANOVA, $p < 0.05$) throughout the experimental operation time. Hence, averaged values of DOC, UV_{254}, DO and NO_3^- were calculated (Fig. 2). The columns were oxic throughout the study and no significant change in pH (8.0–8.3) was observed between the influent and the effluents of the columns. However, oxygen consumption was higher in the first columns than in the second columns (Fig. 2c). DOC removal was significantly higher in the first columns (10%) compared to the second columns (4%; Fig. 2a). A significant drop of UV_{254} was depicted in the first 0.25 m of the columns with only slight decreases afterward (Fig. 2b). SUVA reduction was also significant (Fig. S1) and similar to UV_{254} in the first 0.25 m of the columns. However, a slight increase in SUVA was observed in the second column. These results indicate that in the first 0.25 m, aromatic carbon sources are preferentially used, whereas aliphatic carbon is degraded afterward [13,29]. NO_3^- concentrations were constant in the first 0.75 m depth and then increased along with the depth (Fig. 2d), whereas NH_4^+ concentrations were below 0.03 mg/L and a slight variation along the columns was observed (Fig. S1).

The rate constants for different parameters were also higher in the first columns than in the second columns (Table 2). The DO utilization rates (1/d) were higher in the first columns than in in the second columns. High DO utilization in the first columns supported that organic content in the first columns was higher than in the second columns. In addition, the high rate constant for DOC, nitrification (increase in NO_3^- concentration) and UV_{254} reduction in the first column indicated high biological activity in the first columns compared to the second columns.

In previous experiments (with the same column setup), Hellauer et al. [22] described a significant difference in redox conditions and bulk organic carbon removal in the second columns under different operation conditions (intermediate oxidation). DOC removal was constant in the first columns, but in second columns DOC removal increased from 2 to 33% with aeration with air, pure oxygen or ozone. Redox conditions were also changed from anoxic to oxic in second columns. However, this study observed constant (10%) removal of DOC in the first and in the second coulmns (4%). An insignificant difference in DOC removal and redox conditions were noted in both columns after an adaptation time of six months with groundwater. Thus, an adaptation time shows the robustness of the systems and the potential influence of different treatments.

3.2. DOM characterization using EEM-PARAFAC

Two fluorescence dissolved organic matter (fDOM) components were identified using EEM-PARAFAC (Fig. 3a) and the model was validated by split-half analysis (Fig. 3b). The first component (C_1) showed two peaks; the first was at an excitation and emission wavelengths of 280 nm and 440 nm, respectively. The second peak was at an excitation wavelength of 325 nm and an emission wavelength of 440 nm. The second component (C_2) displayed a peak at an excitation wavelength of 250 nm and an emission wavelength of 425 nm. The fluorescent components were compared with other studies using the Open Fluor database [30] and the results were interpreted according to the similarity >95% with the database. Based on the comparison, C_1 was identified as humic-like, terrigenous material [31–33]. The peak at the high excitation wavelength of compound C_1 corresponds to highly processed organic material [34,35]. C_2 was also identified as terrestrial humic-like compounds with excitation maxima in an ultraviolet range [32,36,37].

A slight and uniform decrease in the loading of C_1 was noted. However, a significant decrease in C_2 was observed in the first columns, but C_2 was almost constant in the second columns (Fig. 3c).

FI values were between 1.60 and 1.67 Raman unit (R.U.) and showed a slight increase along with the depth of the columns (Fig. S3). BIX values were lower than 0.8, indicating fewer authigenic components in DOM. However, the BIX values showed an increasing trend along with the column's depth showing a slightly increased DOM freshness. The HIX was below 0.94 and a slight decrease in HIX was noted in the first 0.5 m of the columns. Similarly, SUVA showed reduction in the top layer, indicating the removal of aromatic DOM. A

Table 2
Rate constants (based on averaged values of replicates) for DOC, DO utilization and nitrification in two sand columns connected in series, data evaluation (first-order kinetics) is shown in Fig. S2.

Parameter	Rate constant *k* (1/d)	
	First sand columns (0–1 m)	Second sand columns (1–2 m)
DOC	- 0.018	- 0.008
DO utilization	- 0.103	- 0.037
Nitrification	0.027	0.001

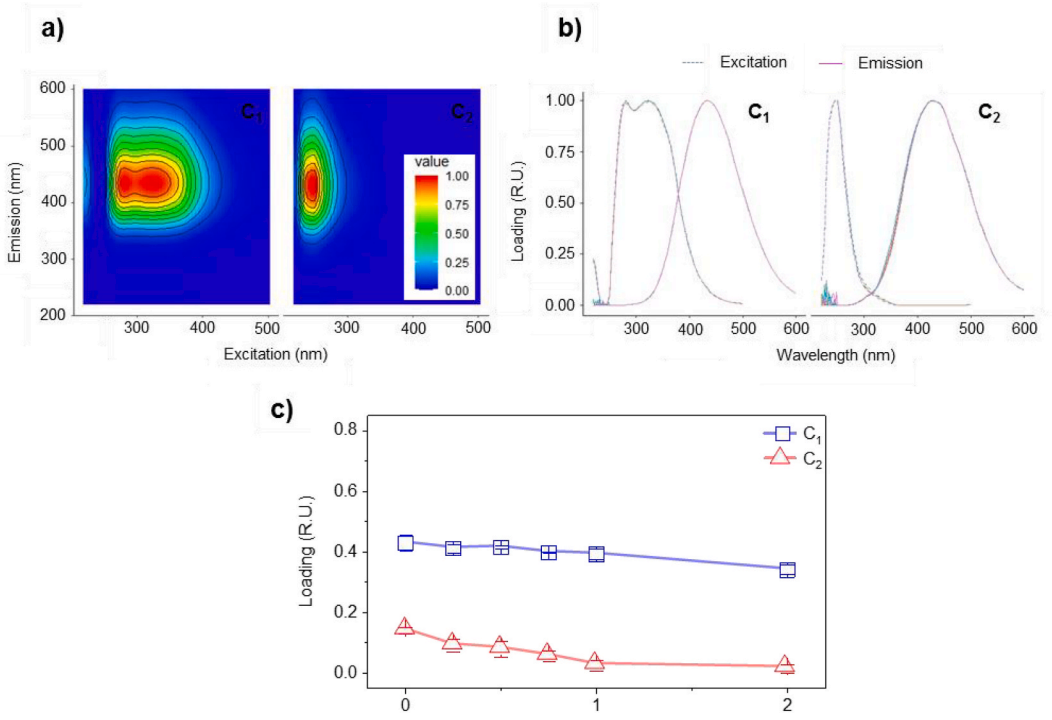

Fig. 3. (a) The contour plot shows two fDOM components, identified using EEM-PARAFAC and (b) validated using split-half analysis, line plot and (c) loading of two fDOM components in the columns, error bars show standard deviations.

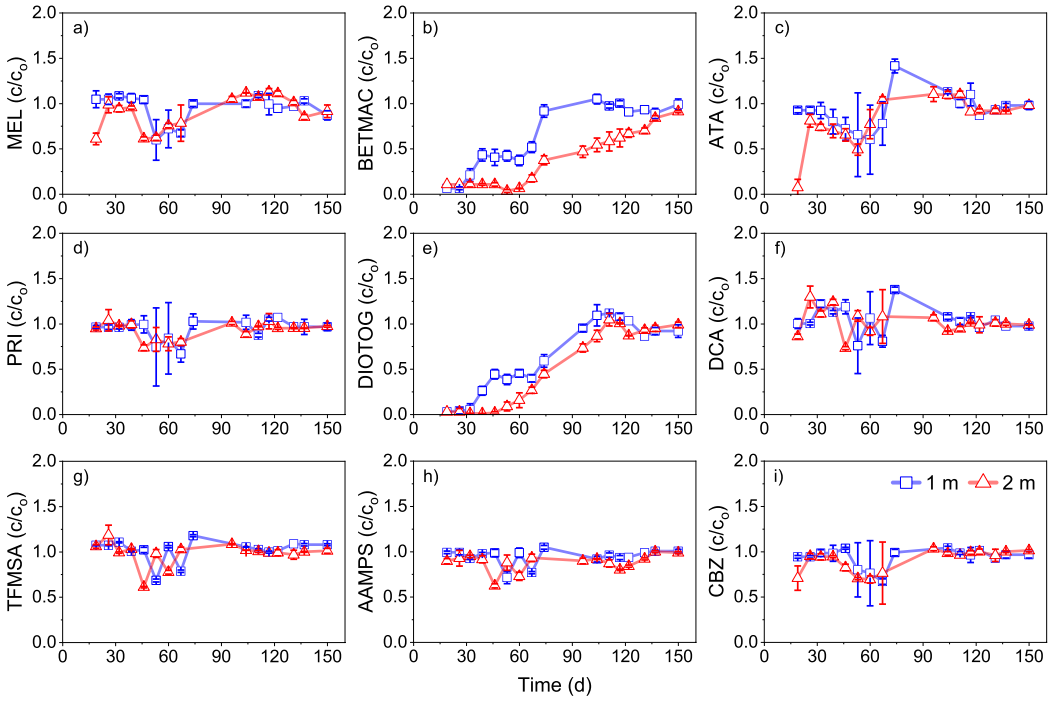

Fig. 4. Relative concentrations (calculated using averaged values of the replicates) of OMP showing sorption ((b) BETMAC and (e) DIOTOG) and persistence ((a) MEL, (c) ATA, (d) PRI, (f) DCA, (g) TFMSA, (h) AAMPS and (i) CBZ) throughout the study period in the sand columns at 1 m and 2 m depth (error bars indicate standard deviations for replicates).

slight increase in HIX from 0.5 to 1.0 m was observed. The increase was presumably due to the decrease in aliphatic DOM or the leaching of humic substances from the sand [38,39]. According to previous research, the fluorescence indices indicate that DOM mainly stems from aquatic bacteria [40,41].

3.3. Fate of OMP

OMP showed similar concentrations (ANOVA, $p < 0.05$) in the four parallel column systems throughout the experimental period. Thus, averaged values were calculated.

3.3.1. Persistent OMP

9 of the 24 OMP (MEL, ATA, PRI, DCA, TFMSA, AAMPS, BETMAC, DIOTOG and CBZ) showed persistent behavior throughout the study in both of the columns (Fig. 4). Low rate constants for these 9 OMP support the fact that these OMP were persistent in the BF under oxic conditions (Fig. S4). Rate constants for MEL (Fig. 5c) and CBZ (Fig. 5d) were very low throughout the study. However, BETMAC (Fig. 5a) and DIOTOG (Fig. 5b) showed a decreasing rate constant with ongoing operation time. An HRT of 12 days was insufficient to attenuate all 9 OMP. Hence, HRT in the sand columns has not shown any effect on the biodegradation of all 9 OMP.

The persistent and mobile behavior of BETMAC, ATA, AAMPS, TFMSA and DIOTOG has been widely reported in surface water and groundwater [5,8,42,43]. However, no data was reported about the fate of these substances during BF processes.

Similar to our study, a persistent behavior of MEL in bank filtrate was reported [5,44–46]. In addition, An et al. [47] described that MEL was not removed by biodegradation during biological wastewater treatment.

In previous studies, PRI and DCA were reported as persistent OMP [15,23,48–50]. The persistent behavior of DCA and PRI was likely because of the presence of amine and amide functional groups in the structure, respectively. Bertelkamp [51] reported that degradation of OMP decreased due to amide and amine groups.

CBZ was reported to be a highly persistent compound and the behavior is independent of the redox conditions [15,52,53]. It showed incomplete removal or persistent behavior under oxic conditions [54] and combined oxic/nitrate-reducing conditions [51]. However, Munz et al. [48] reported that CBZ substantially degraded under iron-reducing conditions.

BETMAC showed sorption for about 100 and 160 days (equivalent to ca. 9 and about ca. 14 bed volumes (BV)) in the first columns and second columns, respectively (Fig. 4b). In addition, BETMAC was fully retained due to adsorption in the two sequential columns for about 60 days (ca. 5.5 BV). On the other hand, DIOTOG showed sorption phenomena in the first and second sand column for about 100 (ca. 10 BV) and 110 days (ca. 11 BV), respectively (Fig. 4e). BETMAC is a cation and it was reported that cationic ammonium-based surfactants with low solubility showed sorption to organic solid [55,56].

3.3.2. OMP showing transformation

15 OMP (out of 24) were biologically transformed during this study. CG, PTSS, MAPMA and DCF were eliminated entirely in the first sand columns (Fig. 6). The intermediate sampling ports of the first column showed that CG and MAPMA were completely eliminated within 0.5 m (ca. 3 days HRT) of the first columns (Fig. S6). The rate constant for CG was 0.27 per day. Schwarzer et al. [57] revealed that soil bacterial consortia were responsible for the rapid degradation of CG. MAPMA showed improved degradation over time ($R^2 = 0.83$); similarly, the rate constant increased from 1.31 to 1.52 per day (Fig.S4). MAPMA is a secondary fatty acid amide which were previously reported to be biologically degraded by microorganisms under aerobic conditions [58].

DCF was entirely eliminated within 0.75 m (ca. 4.5 days HRT) in the first columns and the rate constant was 1.99 per day. Degradation of DCF in a BF process under oxic conditions was reported in previous studies [52,59]. The degradation of CG, MAPMA and DCF was presumably due to high microbial activities in the first columns, as indicated by DOC and UV_{254} reduction (Fig. 6).

PTSS showed complete elimination in the sand columns (Fig. 6j). However, improved removal was observed with adaptation. The rate constant increased from 0.25 to 0.54 with an adaptation time of 96 days. After about 60 days (ca. 5.5 BV) PTSS was eliminated entirely after 0.75 m (ca. 4.5 days HRT) of the first sand column. The biodegradation of PTSS is consistend with previous findings reporting PTSS biodegradationby and desulfonation [60,61].

BTA, BDMA, ACE, SAC, DMBSA and XSA were also bio-transformed (Fig. 6). For these OMP, the bio-transformation rates strongly

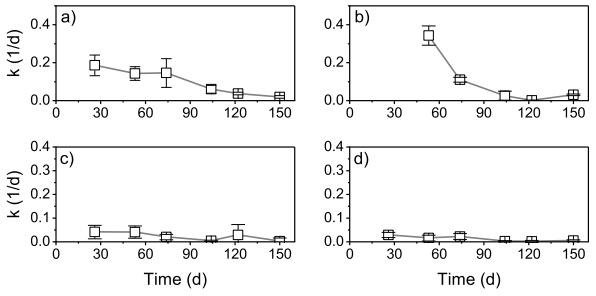

Fig. 5. Rate constants (k) for (a) BETMAC, (b) DIOTOG, (c) MEL and (d) CBZ at different times during the experiment (data evaluation (first-order kinetics) is shown in Fig. S5).

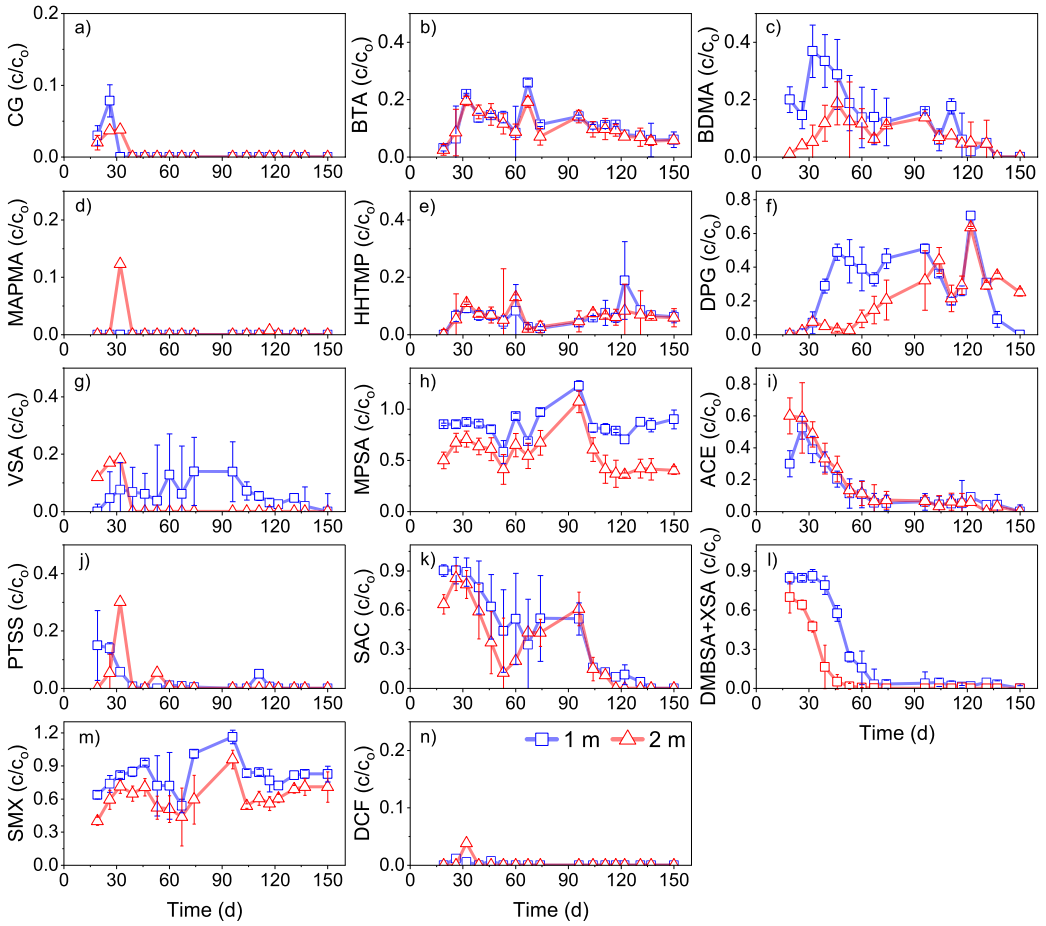

Fig. 6. Relative concentrations of (a) CG, (b) BTA, (c) BDMA, (d) MAPMA, (e) HHTMP, (f) DPG, (g) VSA, (h) MPSA, (i) ACE, (j) PTSS, (k) SAC, (l) DMBSA + XSA, (m) SMX and (n) DCF showing bio-transformation in the sand columns after 1 and 2 m passage (relative concentrations were calculated using averaged values of the replicates, error bars indicate standard deviations for replicates).

correlated with time, which means that with adaptation (or operational time), improved elimination was observed for these OMP. Elimination of BTA was noted in the first sand column (Fig. 6b). However, insignificant elimination was observed in the second column likely due to low biological activities. Regression analysis between time and degradation of BTA (R^2 = 0.75) revealed improved elimination with adaptation. Previous studies reported that BTA is sometimes well removed [62,63] and sometimes partially removed [21] through BF. Contradictory to this, it was reported that BTA is poorly removed by the BF process and remains persistent under all conditions [52,64].

BDMA indicated sorption in both of the sand columns at the start of the experiment (Fig. 6c); however, after 3 BV in the first and 4 BV in the second columns BDMA showed bio-transformation. A significantly higher percentage of BDMA was eliminated in the first column compared to the second column. The bio-transformation rate improved over time as the regression analysis showed a strong

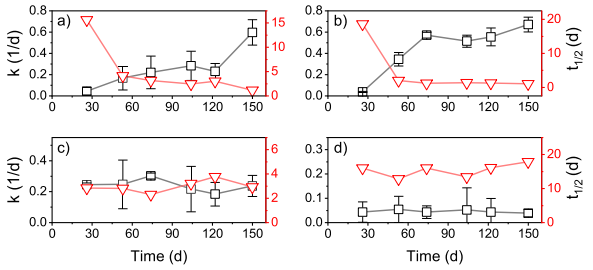

Fig. 7. First order rate constants (*k;* left y-axis) and half-lives ($t_{1/2}$; right y-axis) of (a) ACE, (b) DMBSA and XSA, (c) HHTMP and (d) SMX in two sand columns at a different times during the experiment (data evaluation (first-order kinetics) is shown in Fig. S5).

relationship between time and degradation ($R^2 = 0.71$ (first column) and 0.60 (second coulum)). The behavior of BDMA in a simulated BF is reported in this study for the first time.

ACE showed improved elimination with time ($R^2 = 0.96$ (first column) and 0.91 (second coulum)) and biodegradation was only observed in the first columns due to high microbial activities (Fig. 6i). However, biodegradation was insignificant in the second sand column. Likewise, the rate constant increased over time, indicating enhanced degradation with adaptation (Fig. 7a). Improved elimination of ACE was observed for up to 70 days (ca. 6.5 BV); however, no further increase in elimination was noted after that. It might be because about 95% of ACE was biologically degraded and microorganisms were not capable of degradating very low concentrations. It was also reported that ACE was attenuated under oxic conditions [52,65]. Contradictory, it was mentioned that ACE is not removed by BF [66].

SAC showed improved elimination with adaptation (Fig. 6k). Moreover, the elimination was higher in the first columns compared to the second columns due to the high biological activities in the first columns. Similarly, Di Marcantonio et al. [16] reported the degradation of SAC through BF.

The behavior of DMBSA and XSA was not reported in previous studies. However, this study revealed that degradation of DMBSA and XSA improved with adaptation and were completely eliminated after 70 days (ca. 6.5 BV). In addition, a significant increase in the rate constant of DMBSA and XSA was observed (Fig. 7b).

VSA, HHTMP, DPG, MPSA and SMX were eliminated time-independently during the experiment (Fig. 6). The rate constant for these compounds were constant over time (Fig. S4), for instance, HHTMP (Fig. 7c) and SMX (Fig. 7d) showed insignificant change in rate constant values. The behavior of HHTMP, DPG and MPSA in a simulated BF is reported first time in this study. However, these OMP were frequently detected in the surface water and groundwater and reported as PM compounds [5,7].

DPG showed sorption up to 47 days (ca. 5 BV) in the first columns and 105 days (ca. 9 BV) in the second columns (Fig. 6f). After 47 days, DPG was well degraded in the first columns, but no elimination was observed in the second columns. In contrast to this, Zahn et al. [7] reported that DPG photolyzes but does not biodegrade and is stable to hydrolysis.

VSA was fully eliminated in the first columns (Fig. 6g); VSA is known to be efficiently removed under oxic conditions [15,22,48]. SMX showed partial elimination in the 12 days HRT and degradation was slightly higher in the first columns (Fig. 6m). Other studies confirmed redox-dependent elimination of SMX; van Driezum et al. [63] found slow and only partial degradation under oxic conditions. On the contrary, based on field data, it was suggested that SMX is better degradable under strictly anaerobic [67] and under anoxic [29,67–69] conditions, than under aerobic ones.

The rate constants for all the OMP were high in the first sand columns. While most OMP (16 out of 24) were insignificantly eliminated in the second sand columns. BDMA, VSA, DPG, SAC, MPSA, DMBSA, XSA and SMX showed significant (ANOVA, $p > 0.05$) elimination in the second columns. Hence, these eight compounds were biodegraded in the investigated sand column setup mimicking a BF process, even under low biological activities. However, BTA, HHTMP and ACE were persistent in low biological active sands.

3.4. Correlations between OMP concentration profiles and quality parameters

Pearson's correlations were calculated between the reduction of different parameters and OMP eliminations in columns along the flow-path for days 26, 60, 111 and 150. Results in Fig. 8 (day 150) and Fig. S7 (days 26, 60 and 111) revealed that the biodegradability of the organic carbon could significantly affect the eliminations of OMP.

For the eliminations of BDMA, VSA, ACE, PTSS, SAC, DMBSA and XSA, significant correlations (Pearson's $r > 0.90$) were observed

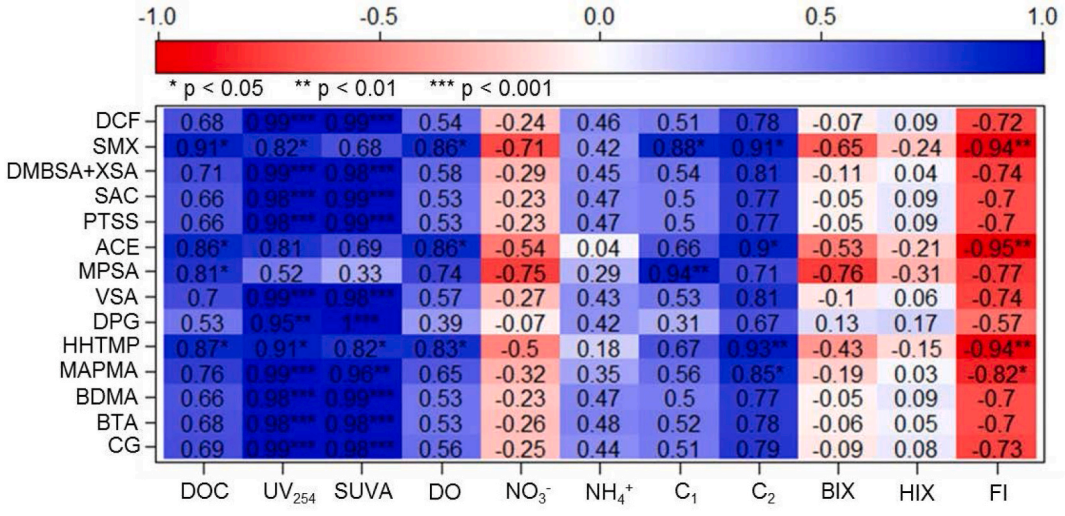

Fig. 8. Heat map of Pearson's correlations between reductions of different quality parameters (with one row per OMP) along the flow-path for day 150 (the profiles of all quality parameters shown above (Figs. 2 and 3) and profiles for OMP are presented in Figure S8).

with the reductions of SUVA and UV_{254} on the selected days (26, 60, 111 and 150). These OMP displayed improved eliminations with adaptation in the columns (Fig. 6), but the reductions for DOC, SUVA and UV_{254} were constant throughout the experimental period (Fig. 2). Therefore, the observed eliminations of these OMP is not directly connected with the DOC removals, abatements of UV_{254} or SUVA reductions in the sand columns. The eliminations for these OMP were presumably due to biological activities as described above (section 3.3.2). Similarly, previous studies reported that varying DOC concentrations [70] and availability of easily degradable organic carbon [71] did not significantly affect OMP biodegradation.

CG, BTA, MAPMA and DCF eliminations correlated with the reductions in SUVA and UV_{254} on the selected days. These OMP showed complete elimination within 0.5 m in the first columns (Fig. S8). Therefore, the influence of the quality parameters on the eliminations of these OMP cannot be explained based on available datasets. Further research is needed, e.g. with increased influent concentrations of these OMP.

The eliminations of SMX, MPSA and HHTMP were constant in the sand columns throughout the study (Fig. 6). Significant correlations (Pearson's $r > 0.80$, $p < 0.05$) were observed for the eliminations of HHTMP and the reductions in quality parameters (SUVA and UV_{254} and C_2) on the selected days. The eliminations of SMX (Pearson's $r > 0.80$) and MPSA (Pearson's $r > 0.70$) were parallel to the removals of humic substances (C_1 and C_2). Thus, humic substances in parallel with the eliminations of SMX, MPSA and HHTMP. Similarly, previous studies reported the influence of dissolved humic substances on the elimination of SMX and other OMP [72,73]. SMX was presumably removed by sorption onto soils. This study found potential correlations between the eliminations of SMX, MPSA and HHTMP and the reductions of humic substances (C_1 and C_2) in sand columns for the first time. However, subsequent in-depth research with varying DOM concentrations and compositions is needed to confirm this hypothesis.

4. Conclusion

The study evaluated the fate (persistency or bio-transformation) of 24 emerging contaminants using saturated oxic sand columns simulating BF processes under controlled boundary conditions. Additionally, the influence of DOM on the removals of OMP was analyzed. Two humic-like fDOM fractions were identified (using EEM-PARAFAC) in the influent, and they were eliminated similarly to the overall DOC in the sand column systems.

OMP such as MEL, ATA, PRI, DCA, TFMSA, AAMPS and CBZ were persistent under oxic conditions and mobile throughout the study. Hence, no effects of biological activity were observed in the columns and no sorption onto the sand was noted. Even the maximum HRT of 12 days was not enough for the biodegradation of these compounds. BETMAC and DIOTOG were also persistent but showed sorption. Sorption phenomena for DIOTOG were higher in the first columns than in the second columns because of the higher organic content in the first columns.

Four OMP (CG, PTSS, MAPMA and DCF) were completely eliminated and six (BTA, BDMA, ACE, SAC, DMBSA and XSA) showed improved elimination with ongoing adaptation. The elimination of these OMP were presumably due to biological activities in the sand columns. Five OMP (VSA, DPG, HHTMP, MPSA and SMX) showed constant eliminations, independent of adaptation times. The eliminations of HHTMP (Pearson's $r > 0.80$, $p < 0.05$), MPSA (Pearson's $r > 0.70$) and SMX (Pearson's $r > 0.80$) correlated with the removals of humic substances (C_1 and C_2).

The attenuation of the investigated OMP were correlated with the HRT and biological activities in the columns. HRT, adaptation time and bioactive sands were critical parameters for the biodegradation of the OMP in the experimental column setup and therefore are likely to play an important role in the remediation of these OMP through BF. The experiments were conducted with constant DOM concentrations and compositions. Hence, the eliminations of OMP at varying DOM concentrations and compositions remain to be investigated.

Author contribution statement

Muhammad Zeeshan: Conceptualization, Methodology, Formal analysis, Data curation, Investigation, Visualization, Modeling, Writing - original draft preparation, Writing - review & editing, **Pia Schumann:** Writing - reviewing and editing, **Silke Pabst:** Formal analysis, Writing - reviewing and editing, **Aki Sebastian Ruhl:** Supervision, Writing - reviewing and editing Supervision, Writing - reviewing and editing.

Data availability statement

Data will be made available on request.

Additional information

Supplementary content related to this article has been published online at [URL].

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.heliyon.2023.e15822.

References

[1] T. Reemtsma, et al., Mind the Gap: Persistent and Mobile Organic Compounds Water Contaminants that Slip through, ACS Publications, 2016.
[2] M. Neumann, I. Schliebner, Protecting the Sources of Our Drinking Water–A Revised Proposal for Implementing Criteria and an Assessment Procedure to Identify Persistent, Mobile and Toxic (PMT) and Very Persistent, Very Mobile (vPvM) Substances Registered under REACH. Mobile and Toxic (PMT) and Very Persistent, Very Mobile (vPvM) Substances Registered under REACH, German Environment Agency, Dessau-Rosslau, 2017, p. 16.
[3] S.E. Hale, et al., What's in a name: persistent, mobile, and toxic (PMT) and very persistent and very mobile (vPvM) substances, Environ. Sci. Technol. 54 (23) (2020) 14790–14792.
[4] H. Arp, S. Hale, REACH: improvement of guidance and methods for the identification and assessment of PMT/vPvM substances, Texte 126 (2019) 2019.
[5] S. Schulze, et al., Occurrence of emerging persistent and mobile organic contaminants in European water samples, Water Res. 153 (2019) 80–90.
[6] C. Huang, et al., The distribution of persistent, mobile and toxic (PMT) pharmaceuticals and personal care products monitored across Chinese water resources, J. Hazard. Mater. Lett. 2 (2021), 100026.
[7] D. Zahn, et al., Identification of potentially mobile and persistent transformation products of REACH-registered chemicals and their occurrence in surface waters, Water Res. 150 (2019) 86–96.
[8] M. Scheurer, et al., Persistent and Mobile Organic Chemicals in Water Resources: Occurrence and Removal Options for Water Utilities, Water Supply, 2021.
[9] S.-S. Liu, et al., In situ measurement of an emerging persistent, mobile and toxic (PMT) substance-Melamine and related triazines in waters by diffusive gradient in thin-films, Water Res. 206 (2021), 117752.
[10] I.J. Neuwald, et al., Occurrence, distribution, and environmental behavior of persistent, mobile, and toxic (PMT) and very persistent and very mobile (vPvM) substances in the sources of German drinking water, Environ. Sci. Technol. 56 (15) (2022) 10857–10867.
[11] M. Collard, et al., Persistence and mobility (defined as organic-carbon partitioning) do not correlate to the detection of substances found in surface and groundwater: criticism of the regulatory concept of persistent and mobile substances, Sci. Total Environ. (2022), 161228.
[12] B. Hu, et al., Riverbank filtration in China: a review and perspective, J. Hydrol. 541 (2016) 914–927.
[13] M. Zeeshan, et al., Seasonal variations in dissolved organic matter concentration and composition in an outdoor system for bank filtration simulation, J. Environ. Sci. (2023).
[14] A.C. Kondor, et al., Occurrence of pharmaceuticals in the Danube and drinking water wells: efficiency of riverbank filtration, Environ. Pollut. 265 (2020), 114893.
[15] J. Filter, et al., Varying attenuation of trace organic chemicals in natural treatment systems–a review of key influential factors, Chemosphere (2021), 129774.
[16] C. Di Marcantonio, et al., Organic micropollutant removal in full-scale rapid sand filters used for drinking water treatment in The Netherlands and Belgium, Chemosphere 260 (2020), 127630.
[17] U. Bayarsaikhan, et al., Fate of leaf litter deposits and impacts on oxygen availability in bank filtration column studies, Environ. Res. 164 (2018) 495–500.
[18] M. Zeeshan, et al., The fate of nitrification and urease inhibitors in simulated bank filtration, J. Environ. Manag. 335 (2023), 117485.
[19] A.F. Henzler, J. Greskowiak, G. Massmann, Modeling the fate of organic micropollutants during river bank filtration (Berlin, Germany), J. Contam. Hydrol. 156 (2014) 78–92.
[20] I.S. Barkow, et al., Seasonal dynamics modifies fate of oxygen, nitrate, and organic micropollutants during bank filtration—temperature-dependent reactive transport modeling of field data, Environ. Sci. Pollut. Res. 28 (8) (2021) 9682–9700.
[21] J. Filter, M. Jekel, A.S. Ruhl, Impacts of accumulated particulate organic matter on oxygen consumption and organic micro-pollutant elimination in bank filtration and soil aquifer treatment, Water 9 (5) (2017) 349.
[22] K. Hellauer, et al., Advancing sequential managed aquifer recharge technology (SMART) using different intermediate oxidation processes, Water 9 (3) (2017) 221.
[23] C. Bertelkamp, et al., Sorption and biodegradation of organic micropollutants during river bank filtration: a laboratory column study, Water Res. 52 (2014) 231–241.
[24] U. Berger, et al., Assessment of Persistence, Mobility and Toxicity (PMT) of 167 REACH Registered Substances, 9, Umweltbundesamt (UBA), 2018.
[25] T. Scheytt, et al., Determination of sorption coefficients of pharmaceutically active substances carbamazepine, diclofenac, and ibuprofen, in sandy sediments, Chemosphere 60 (2) (2005) 245–253.
[26] M. Pucher, et al., staRdom: versatile software for analyzing spectroscopic data of dissolved organic matter in R. Water 11 (11) (2019) 2366.
[27] Y. Zhang, et al., The contribution of phytoplankton degradation to chromophoric dissolved organic matter (CDOM) in eutrophic shallow lakes: field and experimental evidence, Water Res. 43 (18) (2009) 4685–4697.
[28] J.E. Birdwell, A.S. Engel, Characterization of dissolved organic matter in cave and spring waters using UV–Vis absorbance and fluorescence spectroscopy, Org. Geochem. 41 (3) (2010) 270–280.
[29] S. Grünheid, G. Amy, M. Jekel, Removal of bulk dissolved organic carbon (DOC) and trace organic compounds by bank filtration and artificial recharge, Water Res. 39 (14) (2005) 3219–3228.
[30] K.R. Murphy, et al., OpenFluor–an online spectral library of auto-fluorescence by organic compounds in the environment, Anal. Methods 6 (3) (2014) 658–661.
[31] T.B. Bittar, et al., Seasonal dynamics of dissolved, particulate and microbial components of a tidal saltmarsh-dominated estuary under contrasting levels of freshwater discharge, Estuar. Coast Shelf Sci. 182 (2016) 72–85.
[32] M. Chen, et al., Dynamics of dissolved organic matter in riverine sediments affected by weir impoundments: production, benthic flux, and environmental implications, Water Res. 121 (2017) 150–161.
[33] P.G. Dainard, C. Guéguen, Distribution of PARAFAC modeled CDOM components in the north pacific ocean, bering, chukchi and beaufort seas, Mar. Chem. 157 (2013) 216–223.
[34] M. Panettieri, et al., Grassland-cropland rotation cycles in crop-livestock farming systems regulate priming effect potential in soils through modulation of microbial communities, composition of soil organic matter and abiotic soil properties, Agric. Ecosyst. Environ. 299 (2020), 106973.
[35] K. Wheeler, D. Levia, J. Hudson, Tracking senescence-induced patterns in leaf litter leachate using parallel factor analysis (PARAFAC) modeling and self-organizing maps, J. Geophys. Res.: Biogeosciences 122 (9) (2017) 2233–2250.
[36] M. Chen, et al., Production of fluorescent dissolved organic matter in Arctic Ocean sediments, Sci. Rep. 6 (1) (2016) 1–10.
[37] D.N. Kothawala, et al., Selective loss and preservation of lake water dissolved organic matter fluorescence during long-term dark incubations, Sci. Total Environ. 433 (2012) 238–246.
[38] X. He, et al., Spectroscopic characterization of water extractable organic matter during composting of municipal solid waste, Chemosphere 82 (4) (2011) 541–548.

[39] Q. Li, et al., Investigating the spectral characteristic and humification degree of dissolved organic matter in saline-alkali soil using spectroscopic techniques, Front. Earth Sci. 11 (1) (2017) 76–84.
[40] S.R. Brogi, et al., Optical and molecular characterization of dissolved organic matter (DOM) in the Arctic ice core and the underlying seawater (Cambridge Bay, Canada): implication for increased autochthonous DOM during ice melting, Sci. Total Environ. 627 (2018) 802–811.
[41] K.L. Ziegelgruber, et al., Sources and composition of sediment pore-water dissolved organic matter in prairie pothole lakes, Limnol. Oceanogr. 58 (3) (2013) 1136–1146.
[42] J. Gluge, et al., Information requirements under the essential-use concept: PFAS case studies, Environ. Sci. Technol. (2021).
[43] B.J. Sieira, et al., Chlorination and bromination of 1, 3-diphenylguanidine and 1, 3-di-o-tolylguanidine: kinetics, transformation products and toxicity assessment, J. Hazard Mater. 385 (2020), 121590.
[44] R. Alhelou, B. Seiwert, T. Reemtsma, Hexamethoxymethylmelamine–a precursor of persistent and mobile contaminants in municipal wastewater and the water cycle, Water Res. 165 (2019), 114973.
[45] S.E. Hale, et al., Persistent, mobile and toxic (PMT) and very persistent and very mobile (vPvM) substances pose an equivalent level of concern to persistent, bioaccumulative and toxic (PBT) and very persistent and very bioaccumulative (vPvB) substances under REACH, Environ. Sci. Eur. 32 (1) (2020) 1–15.
[46] S. Schulze, et al., Using REACH registration data to rank the environmental emission potential of persistent and mobile organic chemicals, Sci. Total Environ. 625 (2018) 1122–1128.
[47] H. An, et al., The behavior of melamine in biological wastewater treatment system, J. Hazard Mater. 322 (2017) 445–453.
[48] M. Munz, et al., Temperature-dependent redox zonation, nitrate removal and attenuation of organic micropollutants during bank filtration, Water Res. 162 (2019) 225–235.
[49] J. Regnery, et al., Start-up performance of a full-scale riverbank filtration site regarding removal of DOC, nutrients, and trace organic chemicals, Chemosphere 127 (2015) 136–142.
[50] E.Y. Zeng, Persistent Organic Pollutants (POPs): Analytical Techniques, Environmental Fate and Biological Effects, Elsevier, 2015.
[51] C. Bertelkamp, et al., A predictive multi-linear regression model for organic micropollutants, based on a laboratory-scale column study simulating the river bank filtration process, J. Hazard Mater. 304 (2016) 502–511.
[52] V. Burke, et al., Temperature dependent redox zonation and attenuation of wastewater-derived organic micropollutants in the hyporheic zone, Sci. Total Environ. 482 (2014) 53–61.
[53] C. Bertelkamp, et al., The effect of redox conditions and adaptation time on organic micropollutant removal during river bank filtration: a laboratory-scale column study, Sci. Total Environ. 544 (2016) 309–318.
[54] S. Huntscha, et al., Degradation of polar organic micropollutants during riverbank filtration: complementary results from spatiotemporal sampling and push-pull tests, Environ. Sci. Technol. 47 (20) (2013) 11512–11521.
[55] M. Garcia, et al., Adsorption of cationic surfactants on biosolids, Jornadas del Comite Espanol de la Detergencia 27 (1997) 343.
[56] M. Garcia, et al., Fate and effect of monoalkyl quaternary ammonium surfactants in the aquatic environment, Environ. Pollut. 111 (1) (2001) 169–175.
[57] C. Schwarzer, et al., Physiological and electron microscopical investigations on syntrophic dicyandiamide degradation by soil bacteria, Soil Biol. Biochem. 30 (3) (1998) 385–391.
[58] R. Geerts, et al., Microorganisms hydrolyse amide bonds; knowledge enabling read-across of biodegradability of fatty acid amides, Biodegradation 25 (4) (2014) 605–614.
[59] B. Wiese, et al., Removal kinetics of organic compounds and sum parameters under field conditions for managed aquifer recharge, Water Res. 45 (16) (2011) 4939–4950.
[60] K. Urano, Z. Kato, Evaluation of biodegradation ranks of priority organic compounds, J. Hazard Mater. 13 (2) (1986) 147–159.
[61] D. Focht, F. Williams, The degradation of p-toluenesulfonate by a Pseudomonas, Can. J. Microbiol. 16 (5) (1970) 309–316.
[62] J. Müller, J.E. Drewes, U. Hübner, Sequential biofiltration–A novel approach for enhanced biological removal of trace organic chemicals from wastewater treatment plant effluent, Water Res. 127 (2017) 127–138.
[63] I.H. van Driezum, et al., Spatiotemporal resolved sampling for the interpretation of micropollutant removal during riverbank filtration, Sci. Total Environ. 649 (2019) 212–223.
[64] T. Reemtsma, et al., Polar pollutants in municipal wastewater and the water cycle: occurrence and removal of benzotriazoles, Water Res. 44 (2) (2010) 596–604.
[65] A. Sanz-Prat, et al., A model-based analysis of the reactive transport behaviour of 37 trace organic compounds during field-scale bank filtration, Water Res. 173 (2020), 115523.
[66] M. Scheurer, et al., Performance of conventional multi-barrier drinking water treatment plants for the removal of four artificial sweeteners, Water Res. 44 (12) (2010) 3573–3584.
[67] C.K. Schmidt, F.T. Lange, H.-J. Brauch, Assessing the impact of different redox conditions and residence times on the fate of organic micropollutants during riverbank filtration, in: 4th International Conference on Pharmaceuticals and Endocrine Disrupting Chemicals in Water, 2004.
[68] M. Jekel, S. Grunheid, Is Bank filtration an effective barrier against organic substances and pharmaceutical residues? GAS UND WASSERFACH WASSER ABWASSER 148 (10) (2007) 698.
[69] C. Schmidt, F. Lange, H.-J. Brauch, Characteristics and evaluation of natural attenuation processes for organic micropollutant removal during riverbank filtration, Water Sci. Technol. Water Supply 7 (3) (2007) 1–7.
[70] C. Bertelkamp, et al., The effect of feed water dissolved organic carbon concentration and composition on organic micropollutant removal and microbial diversity in soil columns simulating river bank filtration, Chemosphere 144 (2016) 932–939.
[71] J. Filter, et al., Impacts of autochthonous particulate organic matter on redox-conditions and elimination of trace organic chemicals in managed aquifer recharge, Environ. Sci. Pollut. Res. (2023).
[72] Y. Liu, et al., Influences of humic acid on the bioavailability of phenanthrene and alkyl phenanthrenes to early life stages of marine medaka (Oryzias melastigma), Environ. Pollut. 210 (2016) 211–216.
[73] B. Wang, et al., Effect of straw-derived dissolved organic matter on the adsorption of sulfamethoxazole to purple paddy soils, Ecotoxicol. Environ. Saf. 203 (2020), 110990.