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Investigating the impact of temperature and organic matter on the removal of selected organic micropollutants during bank filtration: A batch study

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ABSTRACT

Riverbank filtration (RBF) represents a low-cost and sustainable alternative to advanced treatment technologies to pre-treat or remove several organic micropollutants (OMPs) from surface water. The objective of this research was to investigate the efficacy of biodegradation and adsorption processes in the removal of OMPs at high temperatures (20–30 ± 2 °C) during RBF. Laboratory-scale batch studies were conducted using silica sand at different temperatures (20, 25 and 30 °C) to study the removal of 19 OMPs (6 polyaromatic hydrocarbons (PAHs), 8 herbicides and 5 insecticides) from various water sources with different organic matter characteristics. Simazine, atrazine, metolachlor, and isoproturon exhibited partial persistent characters ($16\% <$ removal $< 59\%$), which apparently decreased with increase in temperature. DDT, pyriproxyfen, pendimethalin, β-BHC, endosulfan sulfate and PAHs with high hydrophobicity (solubility in terms of $\log S < -4$) tend to be well adsorbed onto sand grains (removal $> 80\%$), regardless of temperature, redox conditions or type of organic carbon fraction fed to the batch reactors. These findings indicate that these hydrophobic compounds are effectively removed during RBF regardless of the environmental conditions. Hydrophilic compounds (molinate, dimethoate, and propanil) showed temperature-dependent characteristics for influent water with low organic matter; their attenuation increased at higher temperature (removal $> 95\%$) due to the high microbial activity. This study revealed that temperature is an important parameter affecting the removal of OMPs with hydrophilic and low-hydrophobicity characters. However, temperature has less influence on the removal of highly hydrophobic OMPs during RBF process and thus should be considered during RBF system design.

1. Introduction

The need for high-quality drinking water is increasing rapidly worldwide as a result of increasing urbanization and population growth. However, contamination of surface water resources by domestic and industrial wastewater increase the costs of treatment. Most developing countries employ conventional water treatment techniques, including coagulation, flocculation, clarification, and filtration processes, and these methods are usually followed by disinfection with chlorine. However, convectional water treatment is not effective enough in removing persistent compounds, such as organic micro-pollutants (OMPs) [1]. Riverbank filtration (RBF) could be a viable and cost-effective treatment option for water supply schemes to reduce the concentration of these persistent pollutants, as well as removing pathogens, algal toxins, and organic matter [2,3].

RBF is achieved when a hydraulically connected well located near a surface water system is pumped continuously, causing induced filtration flow towards the interception aquifer [3]. Tufenkji [4] reported that well-operated RBF facilities positioned in proper hydrogeological conditions can provide relatively inexpensive high-quality water that may require little further treatment. This technique has been employed for hundreds of years in Europe and North America. According to Schmidt [5], bank filtration contributes to 75% of the drinking water production in Berlin (Germany). Recently, this technique was applied in countries (such as Egypt and India) with different hydrological and climate conditions [6,7]. The water temperature of RBF wells located along the Nile River (Egypt), the Ganga River (India) and the Yamuna River (India) were determined to be 26.4, 28.6, and 30 °C, respectively [8,9]. However, the maximum recorded temperature for a bank filtration field along the west coast of the Netherlands in the period of 2012–2014 was 22 °C [10]. The effectiveness of RBF under these extreme climate conditions is still not fully understood. Thus, the main

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objective of this research was to study the efficacy of RBF in removing chemical pollutants (with a focus on OMPs) under these conditions.

OMPs removal during RBF is mainly attributed to adsorption and biodegradation, which are highly influenced by the surrounding environmental conditions including temperature, redox and organic composition of the source water [11]. Various studies were conducted using laboratory-scale experiments to investigate the effect of organic matter composition of influent water on the removal of OMPs during RBF. According to Lim [12] and Maeng [13], there is a positive relationship between biodegradable dissolved organic carbon (BDOC) concentration in the feed water and OMP removal, while other studies expound a negative relationship [14,15]. This contradiction can be explained in terms of different OMP classes and characteristics, influent water characteristics and biological activity that is largely affected by the surrounding climate and environmental conditions. In the same regard, temperature may also directly affect the performance of RBF by altering the soil sorption characteristics and exert indirect effects through alternating the redox conditions in the infiltration area, thereby impacting the removal of OMPs [16]. During RBF, the initial infiltration zone is always characterized by oxic conditions followed by anoxic (NO_3^- as electron acceptor) and anaerobic conditions (Fe^{3+}/ Mn^{4+} as electron acceptor). The redox zone size is largely based on the aquifer characteristics and the raw water quality. Few studies have been developed to determine the effect of redox conditions on OMP removal during RBF processes. Maeng [13] illustrated that 17α-ethinylestradiol (EE2) and 17β-estradiol removals were not significantly affected by the redox conditions. Bertelkamp [17] reported that some OMPs including dimethoate, diuron, and metoprolol showed redox-dependent removal behaviour in favour of oxic conditions. However, these studies were undertaken at room temperature. Thus, it is important to assess the effect of redox conditions on the removal mechanisms of OMPs at high temperature.

During this research, laboratory-scale batch studies were conducted to understand the behaviour of different OMP classes (PAHs and pesticides) during RBF and to assess the influence of different environmental conditions (temperature, redox, and influent organic matter composition) on their removal as well as to propose guidelines for OMPs removal prediction during RBF processes at arid climate conditions.

2. Materials and methods

2.1. Experimental set-up

To assess the effect of temperature, redox conditions and organic matter composition on the removal of different classes of OMPs, 34 batch reactors were set up. 100 g of silica sand (grain size, 0.8–1.25 mm) were placed in 0.5-L glass reactors bottles. Initially, sand in the reactors was bio-acclimated with 400 mL of Nile River water mixed with secondary treated wastewater (1:1) under their respective environmental conditions of temperature (20, 25 and 30 ± 2 °C) and redox conditions (oxic, anoxic and anaerobic) (Table 1).The temperature levels were set based on field feedback from previous studies [9,18]. The acclimation period lasted for 60 days with a hydraulic residence time of 5 days until the reactors biologically stabilized (i.e., acclimated) with respect to dissolved organic carbon (DOC) removal.

2.2. Effect of organic composition and temperature on OMPs removal

After the acclimation period, the batch reactors were fed with different types of water of different organic matter composition (humics “humic and fulvic”, and protein): Water from the Nile River at Gabal Taqoq (Aswan, Egypt) (NR) was used to simulate influent water with low organic content. Other water types were developed by mixing the Nile River water with a) secondary treated wastewater from Kima wastewater treatment plant at Aswan, Egypt (ST), b) treated wastewater from oxidation ponds at Balana, Egypt (OP), and c) water extractable soil organic matter (WEOM). WEOM was prepared following the method of Guigue [19], and described in Supplemental Information (S1). The main purpose of using this type of influent water was to simulate the effect of influent water with a high humic concentration on OMP removal efficiency. All influents were filtered through a micro-sieve (38 μm) before application to the batch reactors. Each reactor was injected with the selected OMPs at a concentration of 5 μg/L each, this concentration is corresponded to the OMPs concentrations detected in the Egyptian surface water systems [20,21–23] and other surface water systems worldwide [24,25]. Then, the batch reactors were rotated on a shaker at 100 rpm and incubated at the desired temperature for 30 days. A blank experiment was undertaken by injecting the same concentration of the selected OMPs in Milli-Q water, without sand and incubated under similar conditions, to investigate the loss of OMPs.

2.3. Abiotic experiments

To assess the role of adsorption in the OMP removal process, 6 abiotic batch reactors were employed using NR influent water injected with 20 mM $HgCl_2$ to suppress biological activity [26]. The experiment was conducted at three different temperatures (20, 25, 30 ± 2 °C).

2.4. Redox experiments

To investigate the effect of different redox conditions on the OMPs removal efficiency, 2 reactors were developed under anaerobic condition by degassing with a nitrogen stream to dissipate dissolved oxygen (DO) from the reactors (i.e., DO < 0.2 mg/L) [27], while another 2 reactors were run under anoxic conditions by degassing with a nitrogen stream and injecting 10 mg/L NO_3^-.

2.5. Organic micropollutants (OMPs)

The selected OMPs consisted of 6 polyaromatic hydrocarbons, 8 herbicides and 5 insecticides covering a wide range of physico-chemical properties (Table 2). All OMPs were of analytical grade standard solutions and purchased from Accustandard, USA. Pesticides were analysed with LC–MS-MS following EPA Method 536. PAHs were measured using a GC–MS-MS instrument and following the EPA-625 method. Limits of quantification (LOQ) for pesticides and PAHs is presented in Table S1.

Table 1
Process conditions of batch experiments.

Influents	Temperature	Redox conditions	Biotic/abiotic
Nile River (NR)	20, 25, 30 °C	Oxic	biotic
Nile River: Secondary treated wastewater (ST)	20, 25, 30 °C	Oxic	biotic
Nile River: oxidation ponds treated wastewater (OP)	20, 25, 30 °C	Oxic	biotic
Nile River : Water extractable organic matter (WEOM)	20, 25, 30 °C	Oxic	biotic
Nile River (NR)	20, 25, 30 °C	Oxic	abiotic
Nile River (NR)	25 °C	anoxic/ anaerobic	biotic

Table 2
List of OMPs studied and their properties.[1, 2]

	Chemical structure	pKa Charge at pH 8	Water solubility mg/mL	LogS at pH 8	LogD at pH 8	LogP
Molinate	$C_9H_{17}NOS$	n.a	9.70E-01	−2.1	2.34	2.34
Simazine	$C_7H_{12}ClN_5$	3.23	1.00E-02	−3.45	1.78	1.78
Isoproturon	$C_{12}H_{18}N_2O$	n.a	7.20E-02	−3.08	2.57	2.57
Atrazine	$C_8H_{14}ClN_5$	3.2	3.47E-02	−3.8	2.2	2.2
Propanil	$C_9H_9C_{l2}NO$	1.21	2.25E-01	0	0.56	3.85
Dimethoate	$C_5H_{12}NO_3PS_2$	−4.5	2.50E+01	−2	0.34	0.34
Pendimethalin	$C_{13}H_{19}N_3O_4$	−1	3.00E-04	−4.32	4.82	4.82
Metolachlor	$C_{15}H_{22}ClNO_2$	n.a	5.30E-01	−3.68	3.45	3.45
Pyriproxyfen	$C_{20}H_{19}NO_3$	2.86	1.63E-02	−5.11	4.75	4.75
Picloram	$C_6H_3C_{l3}N_2O_2$	−0.21	2.41E+02	0	−1.43	2
DDT	$C_{14}H_9Cl_5$	n.a	8.50E-05	−6.55	6.46	6.46
Endosulfan sulfate	$C_9H_6C_{l6}O_4S$	n.a	4.80E-04	−4.9	n.a	3.16
β-BHC	$C_6H_6Cl_6$	n.a	2.40E-04	−4.7	4.35	4.35
Naphthalene	$C_{10}H_8$	n.a	3.10E-02	−3.3	2.96	2.96
Fluorene	$C_{13}H_{10}$	n.a	1.69E-03	−4.8	3.74	3.74
Anthracene	$C_{14}H_{10}$	n.a	4.34E-05	−5.6	3.95	3.95
Pyrene	$C_{16}H_{10}$	n.a	1.35E-04	−6.9	4.28	4.28
Chrysene	$C_{18}H_{12}$	n.a	2.00E-06	−7.7	4.94	4.94
Benzo (b) fluoranthene	$C_{20}H_{12}$	n.a	1.50E-06	−7.7	5.27	5.27

[1] https://chemicalize.com.

[2] http://www.t3db.ca/.

2.6. *Characterization of influents and effluents*

The collected samples were stored at 4 °C after 0.45-μm filtration (Whatman, Dassel, Germany) and analysed within 3 days of collection to minimize biodegradation of organic matter. Dissolved organic matter (DOC in mg C L^{-1}) was measured using a total organic carbon analyser (TOC-VCPN (TN), Shimadzu, Japan). UV absorbance [cm^{-1}] was monitored at 254 nm by a UV/Vis spectrophotometer (UV-2501PC Shimadzu). Specific ultraviolet absorbance ($SUVA_{254}$) [L mg^{-1} m^{-1}]), an indicator for the relative aromaticity and humic content of the bulk organic matter, was determined after dividing UV_{254} absorbance by DOC concentration. Characterization of organic matter fractions (humic, fulvic, and protein) was performed using a fluorescence excitation-emission spectrophotometer (F-EEM) as described in [28]. The measurement method is presented in Supplemental Information (S2). Peak picking technique was used to characterize the fluorescence excitation and emission matrices data of the influents and effluents water [29,30]. Three regions of peaks were identified at distinct excitation and emission wavelengths: primary humic-like OM peak (P1) was assigned at excitation = 250 nm and emission = 440 nm; (2) secondary humic-like OM peak (P2) was measured at excitation = 320 nm and emission = 440 nm; (3) protein-like OM peak (P3) was determined at excitation = 240 nm and emission = 330 nm. Furthermore, two fluorescence indices were used to characterize the organic composition of the feed water. (1) Humification index has used an indicator for the humic content of the feed water, it is calculated by dividing the peak area under the emission spectra 435–480 nm by the peak area 300–345 nm 435–480 nm, at excitation 254 nm [29,31]. (2) Fluorescence index (FI) has used to identify the relative contribution of terrestrial (low values) and microbial (high values) dissolved organic matter to the full fluorescence spectrum. It is estimated as a ratio of the fluorescence intensity at emission wavelength 450 nm to that at 500 nm for a definite excitation wavelength (370 nm) [29,32]. FI typically ranges between 1.2–1.7 for surface water systems, where high value is indicative for higher biodegradable content [33]. Nitrate and phosphate concentrations (mg/L) of the influent and effluent samples were quantified using ion chromatography (881 Compact IC pro, Metrohm anions, Swiss), whereas ammonia (mg/L) was determined using an ion chromatograph (881 Compact IC pro, Metrohm cations, Swiss). Heterotrophic plate counts (HPC) were used as an indicator of sand biomass activity following the method described in [34]. At the end of the experiment, duplicate wet sand samples (2–2.5 g) were suspended in 50 mL of autoclaved tap water and sonicated at a power of 40 W for 2 min to suspend the biomass into the solution. 0.1 mL of microbial suspensions were spread in triplicate over a surface of R2A agar plates and incubated for 48 h at 35 °C.

2.7. *Data analysis*

Two-way ANOVA was used to determine the significance impact of temperature, influent organic matter and redox conditions on the removal of OMPs in batch reactors. The criterion level of significance (α) was 0.05.

3. Results and discussion

3.1. *Characterization of influents and effluents water*

Relevant water quality parameters of the feed water added to batch reactors and the average changes in their quality characteristics during the incubation period at different temperatures are summarized in Table 3. At all temperature set-points, OP water exhibited the highest DOC removal (51–54%) followed by ST water (46–48%), WEOM water (28–33%), and NR water (9–18%). The higher DOC reduction for OP and ST are mainly attributed to their high biodegradable organic matter concentrations, which support the biological activity associated with the sand. The average concentrations of HPC originating from the biomass associated with the OP, ST and WEOM sand reactors at 25 °C were 1411 ± 33, 1343 ± 96 and 1153 ± 81 CFU/mL, respectively. However, the average HPC for NR water bio-acclimated sand was 866 ± 62 CFU/mL at the same temperature, which suggests that the contribution of biodegradation to the overall DOC removal is reduced at low DOC concentration. Furthermore, ANOVA test results revealed that temperature has a significant effect on the biological activity of the sand ($p = 0.02$). It was found that the biological activity apparently increased with temperature (Table S2). The results indicated that the HPC values associated with the bio-acclimated sand of NR batch reactors increased by 16 and 17% at 25 and 30 °C compared to the value at 20 °C (723 ± 98 CFU/mL); this illustrates that the role of biodegradation in DOC removal is increased with rising temperature. This was also confirmed with SUVA results, which exhibited as well an increase with temperature, indicating that there is preferred degradation of non-aromatic compounds at higher temperature. SUVA increased by 14, 16 and 21% for effluent NR waters at 20, 25 and 30 °C, respectively, compared to the influent value. The same trend was observed for ST and OP waters, where SUVA increased by (34, 42 and 47%) and (12, 29 and 33%), respectively, at the same temperatures. However, WEOM water did not exhibit the same trend, its highest increase (11%) of SUVA was recorded at 20 °C, and was attributed to its high humic content which is preferentially removed by adsorption [34].

To characterize the organic matter characteristics of influents and effluents, samples were taken at the beginning and end of the batch experiments to measure fluorescence intensity. Three dominant peaks at definite wavelengths representing the bulk organic matter fractions (humic, fulvic and protein) were discriminated [13]. Table 4 and table S3 show that change in fluorescence intensity for each of the identified peaks is highly dependent on the organic matter composition and temperature. Humics compounds (humic and fulvic) bear a negative charge and thus tend to be removed by adsorption during the infiltration process [13,35]. In this research, abiotic batch reactors exhibited a relatively larger decrease of humics compound fluorescence intensity at lower temperatures. Therefore, it can be concluded that humics reduction (by adsorption) exhibits an inverse relationship with temperature rise, this was also reported by Abdelrady [35]. A similar trend of better attenuation at lower temperatures (20 °C) was observed for NR

Table 3
Characteristics of the influent and effluent water.

Influent			DOC (mg L^{-1})	SUVA (L mg^{-1} m)	NO_3^{-1} (mg L^{-1})	NH_3^{+1} (mg L^{-1})	PO_4^{-3} (mg L^{-1})	pH
NR		**Influent**	4.05 ± 0.69	1.82	N.D.	N.D.	N.D.	7.42
	20 °C	**Effluent**	3.31 ± 0.39	2.11	0.09	N.D.	N.D.	7.53
	25 °C	**Effluent**	3.35 ± 0.69	2.16	0.17	N.D.	N.D.	7.78
	30 °C	**Effluent**	3.68 ± 0.47	2.3	0.14	N.D.	N.D.	7.85
ST								
		Influent	10.62 ± 0.59	1.49	0.15	2.34	1.64	7.96
	20 °C	**Effluent**	5.72 ± 0.34	2.27	0.64	0.25	0.71	8.11
	25 °C	**Effluent**	5.51 ± 0.59	2.58	0.89	N.D.	0.69	8.18
	30 °C	**Effluent**	5.63 ± 0.41	2.79	0.59	N.D.	0.68	8.17
OP		**Influent**	10.31 ± 0.68	1.56	0.14	2.61	1.84	8.08
	20 °C	**Effluent**	5.05 ± 0.47	1.78	0.19	0.17	1.09	8.13
	25 °C	**Effluent**	4.83 ± 0.68	2.19	0.56	N.D.	1.09	8.16
	30 °C	**Effluent**	4.65 ± 0.39	2.34	0.56	N.D.	1.02	8.28
WEOM		**Influent**	9.41 ± 0.79	2.44	N.D.	1.04	0.24	8.11
	20 °C	**Effluent**	6.34 ± 0.71	2.73	0.12	N.D.	0.08	8.17
	25 °C	**Effluent**	6.75 ± 0.21	2.29	0.36	N.D.	0.08	8.29
	30 °C	**Effluent**	6.78 ± 0.54	2.51	0.36	N.D.	0.07	8.24

N.D.: not detected.
Limit of quantitation (LOQ) for NO_3^{-1}(0.05 mg/L), NH_3^{+1}(0.2 mg/L) and PO_4^{-3}(0.05 mg/L).

effluent water in which the biodegradable matter was limited. However, OP, WEOM and ST effluents showed an increase of humics fluorescence intensity. This increase displays a positive relationship with temperature rise for ST effluent and a negative relationship for OP effluents (Fig. 1). Previous studies [34,35] suggested that bacteria present in the soil are able to form new fluorescent organic compounds associated with existing dissolved organic matter during the long-term infiltration process.

Apparently, fluorescence intensity of protein-like components (peak 3) decreased with increasing temperature for all organic water matrices. An exception was observed for WEOM water, which exhibited higher protein compound removal at low temperature that may be attributed to adsorption. Additionally, the results of abiotic conditions verified that the adsorption process is more influential at low temperature, even for biodegradable materials.

The fluorescence indices were used to get insight information about the organic composition of the feed water. The results refer that ST and OP have the highest concentration of biodegradable organic matter. The fluorescence index (FI) were 1.67, 1.5, 1.36 and 1.25 for ST, OP, NR and WEOM feed waters, respectively. By contrast, WEOM feed water possessed the highest humic content, its humification index (HIX) was 0.75, while it was 0.56, 0.55 and 0.5 for ST, OP and NR, respectively.

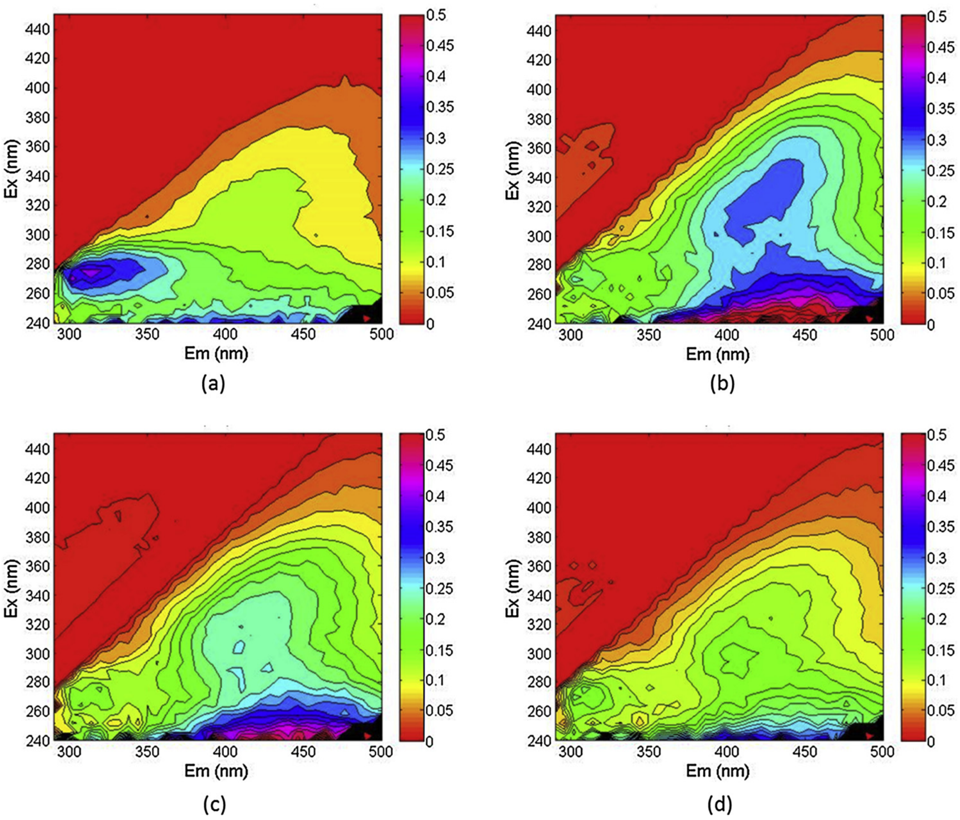
3.2. Effect of feed water organic matter composition on OMPs removal

Laboratory-scale batch reactors were conducted at high temperatures (20–30 °C) to assess the impact of feed water organic composition on the removal of selected OMPs. Four different water types (NR, ST, OP, and WEOM) were introduced into the batch. Biodegradation and adsorption are the main mechanisms for removal OMPs during the bank filtration processes, and they are considerably impacted by the raw water characteristics [11]. In this research, a statistically significant effect of the influent organic matter composition on OMP removal was observed at 20 °C (*p-value = 0.02*). However, no statistically significant effect was observed at higher temperatures of 25 °C (*p-value = 0.06*) and 30 °C (*p-value = 0.09*), indicating that the influence of feed water organic matrix on OMP removal is lower at higher temperatures. Among the OMPs studied, removal of DDT, pyriproxyfen, pendimethalin, and picloram in NR blank reactors was 78.3 ± 7.3%, 69.7 ± 4.5%, 90.4 ± 3.6%, 90.1 ± 5.6%, and 94%, respectively. These removal efficiencies were 5–7% higher for the other water types tested. This implies that abiotic removal mechanisms such as volatilization and hydrolysis may play a major role in reducing the concentration of these OMPs during the batch study. Furthermore, the removal efficiency of these OMPs was higher than 99% in the batch reactors regardless of the water organic matrix, a result mainly attributed to adsorption (Fig. 2). Similarly, β-BHC and endosulfan sulfate exhibited removal between 80–94% which was mainly attributed to adsorption (Fig. 2). Thus, it can be concluded that high hydrophobic compounds ($\log S > 4$) tend to be highly removed (> 80%) by adsorption during the long filtration process (> 30 days). These results are in agreement with Rodríguez-Liébana [36], who asserted that neutral hydrophobic pesticides, such as pendimethalin, are mainly removed by abiotic processes in soil irrigated with treated wastewater.

Simazine, atrazine, metolachlor and isoproturon ($-2.5 > \log S > -4$) demonstrated more persistence during the batch study (removal efficiencies varying between 16–59%) (Fig. 2). They exhibited a relatively

Table 4
Fluorescence peaks intensities (RU) in oxic batch reactors operated at different temperatures using different feed water types.

Feed water type	Fluorescence peaks (RU)											
	P1 (λex/λem = 245/440 nm)				P2 (λex/λem = 345-/440 nm)				P3 (λex/λem = 240/340 nm)			
	NR	ST	OP	WEOM	NR	ST	OP	WEOM	NR	ST	OP	WEOM
Influent	0.32	0.32	0.30	1.18	0.20	0.12	0.25	0.45	0.34	0.45	0.44	0.43
Effluent-20 °C	0.28	0.35	0.63	1.62	0.13	0.18	0.33	0.68	0.19	0.25	0.18	0.28
Effluent-25 °C	0.34	0.55	0.48	1.83	0.24	0.28	0.26	0.58	0.16	0.20	0.16	0.31
Effluent-30 °C	0.37	0.61	0.35	2.18	0.24	0.34	0.2	0.54	0.12	0.18	0.16	0.34



(a) (b) (c) (d)

Fig. 1. F-EEM spectra: (a) influent OP, (b) effluent at 20 °C, (c) effluent at 25 °C, (d) effluent at 30 °C.

higher removal efficiency from ST and OP influent waters containing higher concentrations of biodegradable organic compounds. This finding implies that co-metabolism may play a role in the removal of these compounds. This is consistent with the findings of Bertelkamp [15], who studied the effect of three feed waters with different organic fractions (hydrophilic, hydrophobic, transphilic) on the removal of OMPs (include atrazine and simazine) in three similar sand columns, and found that the biodegradability of the feed water significantly affects the removal efficiency of OMPs during the filtration process. Orlandini [37] reported that atrazine removal is increased in the presence of high concentrations of biodegradable materials which enhance the biological activity associated to the sand and enables the existing bacteria to use atrazine as a source of carbon or nitrogen. On the other hand, these moderate hydrophobic OMPs exhibited lower removal efficiencies for NR feed water that mainly ascribed to its low organic content and thereby the low diversity of microorganisms in the sand. Likewise, lower removal efficiencies were observed for WEOM feed water that might attribute to its high humic content and the competition between OMPs and humic compounds for adsorption sites that should be considered during the bank filtration installing processes. Molinate, dimethoate and propanil compounds with relatively high solubility (logS > -2.5) exhibited more biodegradable character during the batch study (Fig. 2). According to Singh [38], highly soluble pesticides are more subject to degradation. The high biodegradability characteristics of these compounds are mainly ascribed to the presence of electron donating functional groups (amine groups) in their structures [39]. Therefore, the removal efficiencies of these OMPs decreased significantly under abiotic conditions (Fig. 3, Figure S1, and Figure S2).

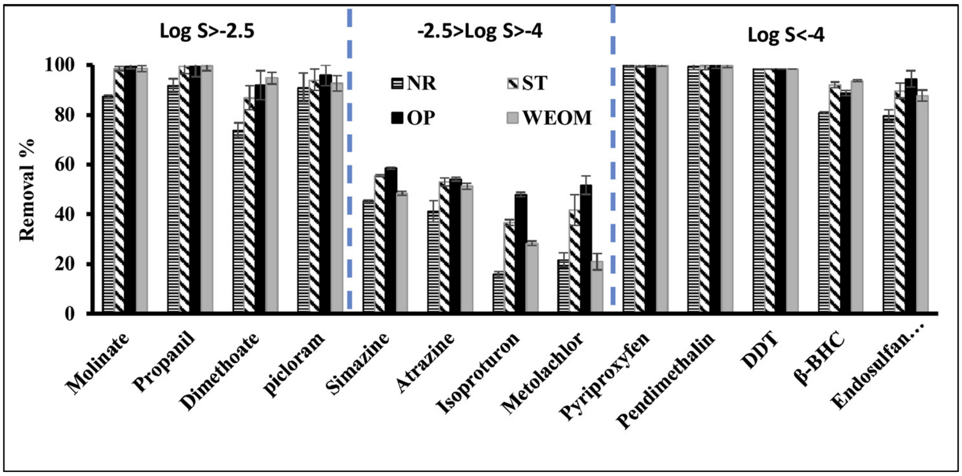


Fig. 2. Impact of influent organic matter type on the removal of OMPs in batch reactors (20 °C, oxic).

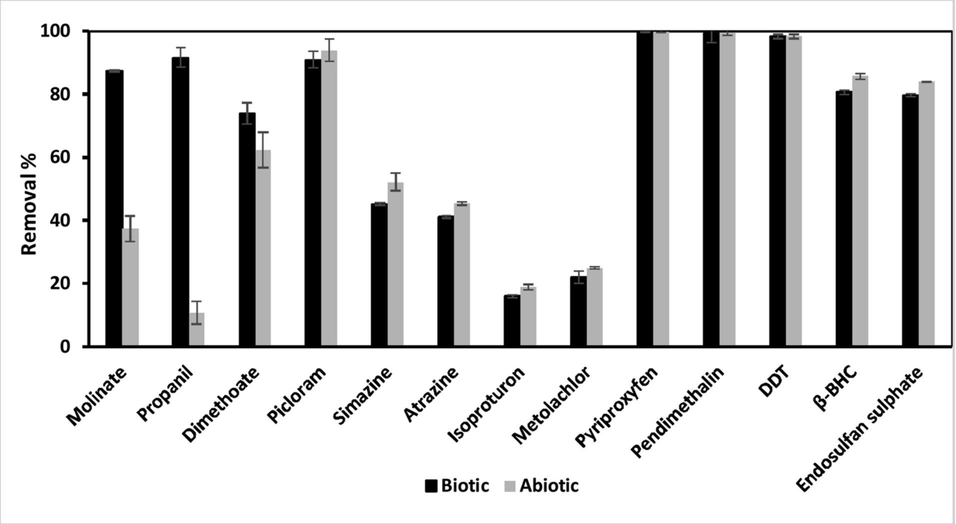


Fig. 3. Removal of OMPs under biotic and abiotic conditions at 20 °C for NR influent water.

These results emphasize the importance of the biodegradation process in the removal of these hydrophilic compounds during the RBF process. This is in agreement with other researchers [15,40] who discovered that adsorption of these compounds to the soil is very weak. With respect to the effect of influent organic matter, the removal efficiencies of these compounds decreased slightly under conditions of limited organic content at a temperature of 20 °C (Fig. 2). The removal of molinate, propanil and dimethoate were decreased by 12 ± 0.65%, 9 ± 0.52% and 13–18%, respectively, for NR during a residence time of 30 days in the batch reactors compared to their removal in water of higher influent biodegradable matter (OP, ST). Nunes [41] illustrated that co-metabolic processes mainly dominated their degradation; thus, the presence of an additional carbon source could enhance degradation.

PAH compounds are highly toxic and classified as probable human carcinogens. Blank batch reactors exhibited limited removal of these hydrophobic compounds (less than 5%) at all tested temperatures. Furthermore, no significant effect of influent organic matrix was observed on the removal of these compounds. In this research, the removal of these compounds exceeded 80% under different influent organic conditions. Lamichhane [42] demonstrated that biodegradation and hydrolysis of PAH compounds are very limited. Accordingly, it can be concluded that the high removal observed in this study is mainly attributed to sorption and prolonged residence time (30 days). It was observed that PAH compounds with more than three aromatic rings were thoroughly removed (> 99%) (Figure S3).

3.3. *Effect of temperature on OMP removal*

To assess the effect of temperature on the removal of OMPs during the RBF process, batch experiments were conducted at three different temperatures, 20, 25, and 30 ± 2 °C, using different water types in oxic conditions. Attenuation of 5 of the 13 selected pesticides investigated in this study showed temperature dependency (Figs. 4 and S4). The removals of pendimethalin, pyriproxyfen and DDT exceeded 95% at all three temperatures tested, although previous studies [43–45] illustrated the temperature-dependent attenuation behaviour of these OMPs during the infiltration process. The high attenuation of these compounds is mainly attributed to their high adsorption efficiencies and prolonged residence times, as previously described. In the same regard, Maeng [13] conducted laboratory-scale batch studies at 16–17 °C to assess the removal of selected OMPs (e.g., pharmaceuticals) and reported as well the high removal (> 80%) of the hydrophobic compounds (e.g., bezafibratem ibuprofen, naproxen, logS < -4).

The same trend was observed for PAH compounds. However, naphthalene, fluorene, and anthracene exhibited lower removal values at higher temperatures (30 °C), likely due to their lower adsorption efficiency at higher temperature or the desorption process which may take place during the prolonged residence time. Hiller [46] illustrated that less hydrophobic PAHs (e.g., naphthalene) demonstrate lower adsorption efficiency and higher desorption characteristics at high temperatures. Thus, post-treatment of bank filtrate may be required to remove these compounds at high temperature. Likewise, picloram exhibited a high removal efficiency (> 90%) at all processed temperature, an effect that may be ascribed to abiotic process (e.g., hydrolysis and volatilization). The temperature-dependent removal of molinate, dimethoate, and propanil was observed when the influent organic content was limited (NR), as their removal efficiencies were increased considerably at high temperature. Removal of molinate and dimethoate in abiotic batch reactors significantly increased with increase in temperature, indicating that the hydrophobicity of these compounds increased at high temperature; this was also previously reported by Rani [47]. However, propanil exhibited temperature-independent removal under abiotic conditions. This implies that the biotic process plays a positive role in its removal at higher temperature. Attenuation of atrazine, simazine, metolachlor, isoproturon, endosulfan sulfate and β-BHC were markedly increased at high temperatures for all influent waters under both biotic and abiotic conditions. The high attenuation of these hydrophobic compounds with increasing temperature is mainly attributed to their increasing adsorption capacity at higher temperature.

3.4. *Effect of redox on removal of OMPs*

The effect of three redox conditions including oxic, anoxic (NO_3^- as electron acceptor) and anaerobic (Fe^{+2}, Mn^{+2} as electron acceptors) on the removal of 19 OMPs was examined for different water characteristics at high temperature (25 °C.). Effluent dissolved oxygen in the suboxic batch reactors was less than 0.2 mg/L, and the average removal of nitrate in anoxic reactors was 92 ± 5.8%. Fig. 5 shows the average removal of the selected pesticides at 25 °C under different redox conditions.

Various studies [15,48] asserted that the removal efficiencies of DDT, pyriproxyfen and, pendimethalin decreased under sub-oxic conditions. However, it was obvious in this research that the removals of these OMPs are redox-independent and exceed 99% in all batch reactors operating at 25 °C. This is mainly attributed to their high adsorption efficiency at higher temperature as well as the prolonged residence time in the soil. In the same manner, PAHs also exhibited high

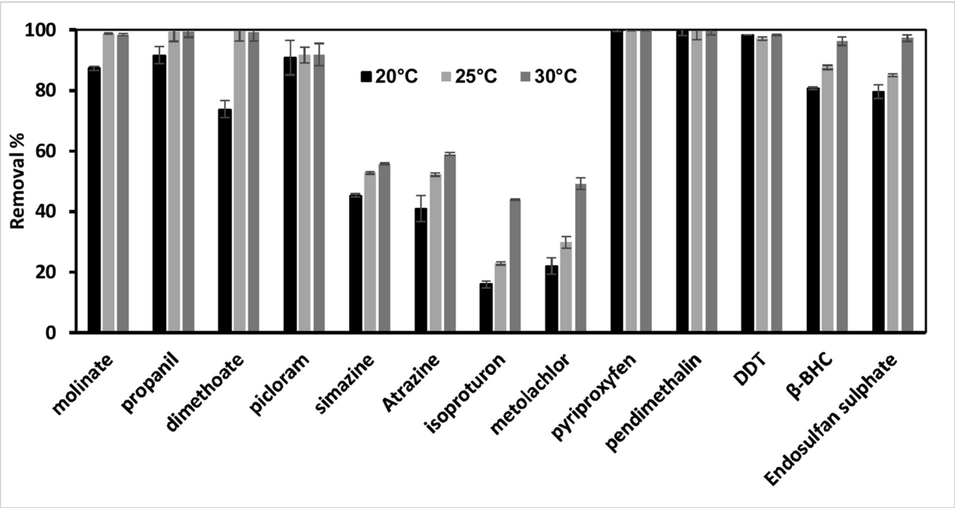


Fig. 4. Removal of OMPs at different temperatures (20, 25, 30 °C) under oxic condition for NR water.

removal efficiencies (> 99%) at the same temperature regardless of the redox conditions, as adsorption was the dominant mechanism. Apparently, if the hydrophobic compound exhibits logP > 4, it tends to be highly removed (> 80%) during the filtration process (residence time: 30 days) regardless of the redox, organic composition, and temperature environment, at least for the OMPs tested during this research. Likewise, dimethoate displayed high removal efficiency (> 90%) independent of the redox condition. Inversely, Bertelkamp [17] reported that dimethoate removal is highly reduced under sub-oxic conditions: its biodegradation rate is 72% lower under anaerobic conditions and reduced by 85% under anoxic conditions when compared to oxic conditions. This is probably because the experiments were conducted at low temperature (12 °C). This implies that dimethoate is preferentially removed due to an increase in microbial activity with increasing temperature regardless of the environment of redox conditions. Similarly, abiotic removal mechanisms (i.e., hydrolysis) may also have played a role in the removal efficiency at high temperature [49]. On the other hand, triazines (atrazine and simazine) demonstrated the same persistence under the redox conditions tested. In contrast, Gimbel [50] and Shahgholi [51] revealed that the suboxic condition resulted in higher biodegradation removal of triazines than did the oxic condition during the RBF process. This contradiction can be explained in terms of the absence of organisms that are able to consume triazine compounds in the batch reactors [17]. This trend was also observed for metolachlor, which showed high persistence independent of the redox conditions. Low biodegradation of metolachlor under different redox conditions was reported in other studies [52]. Seybold [53] studied metolachlor degradation in wetland soil and water microcosms and stated that metolachlor is degraded under anaerobic condition with a half-life of 67 days. Therefore, it can be concluded that RBF does not effectively remove all of OMPs in an acceptable manner in temperate regions and that post-treatment may be required for drinking water purposes.

Molinate and propanil exhibited a high dependency on the redox conditions, since their biodegradation was highly increased under oxic conditions compared to anoxic and anaerobic conditions. The removal of these pesticides under different redox conditions during RBF has not yet been studied. However, these results are consistent with Lopes [40] and Nunes [41] who tracked the environmental fate of these hydrophilic compounds in wet soils. Therefore, it can be concluded that the compounds require longer residence time to be removed during the suboxic infiltration process. However, some molinate transformation products such as molinate sulfoxide are also potential toxicants and have higher solubility compared to the original compound; therefore, they have high potential to be leached during bank filtration, which requires further investigation.

Endosulfan sulfate and β-BHC exhibited relatively higher removal under sub-oxic conditions. The removal efficiencies for endosulfan sulfate and β-BHC were increased by 8.7 and 11.8% for the anoxic condition and 6.7 and 13% under anaerobic conditions, respectively. Baczynski [54] illustrated that organochlorine pesticides are degraded effectively under anaerobic conditions and that this process is temperature-dependent, increasing slightly at higher temperature.

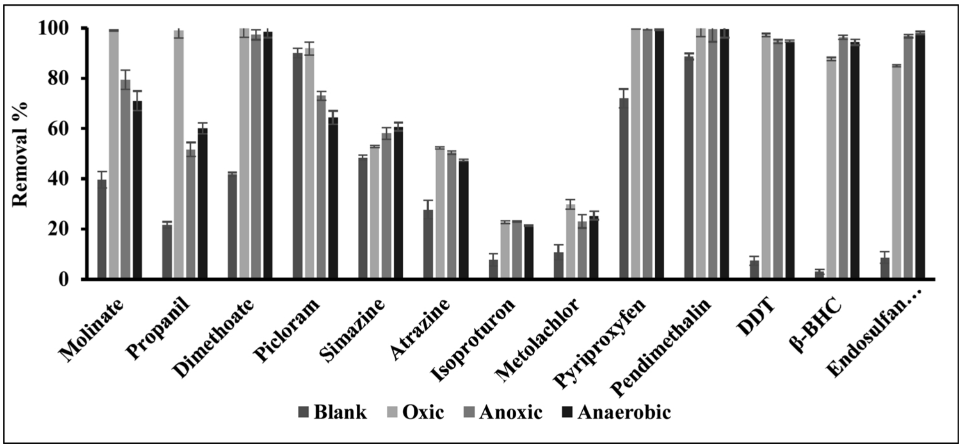


Fig. 5. Removal of OMPs under different redox conditions at 25 °C for NR influent water.

4. Conclusions

Batch studies were conducted to assess the removal efficiency of some micro-organics pollutants during the RBF process (travel time = 30 days) under different environmental conditions. Based on the results of this research, the following conclusions can be drawn:

- Labile compounds (i.e., protein-like) is the most amenable DOM fraction to be removed during RBF at all the tested temperatures.
- Atrazine, simazine, isoproturon, and metolachlor (-2.5 > logS > -4) were the most persistent compounds and were mainly removed by adsorption process (< 45%, residence time = 30 days). The removal of these compounds was enhanced at higher temperatures and in the presence of organic matter. This finding indicates that co-adsorption, as well as co-metabolism, may play significant roles in the removal of these compounds.
- Poorly soluble OMPs with logs < -4 (i.e., DDT, pyriproxyfen, pendimethalin, β-BHC, and endosulfan sulfate) were highly removed (> 80%) by adsorption (residence time = 30 days) regardless of the environmental conditions (i.e., temperature, redox and influent organic characteristics). However, these slightly soluble compounds may degrade in the soil and produce more toxic and more soluble compounds which may leach into the bank filtrate. Further investigation is required in this phenomenon.
- The removal efficiencies of soluble compounds with logS > -2.5 (molinate, propanil, and dimethoate) were below 40% under abiotic conditions. Removal efficiencies of these hydrophilic compounds increased to > 70% under biotic conditions. This implies that biodegradation has a key role in the attenuation of these compounds during RBF. Thus, influent temperature and biodegradable organic matter concentration may affect the removal of these OMPs due to their influence on microbial activity and the co-metabolism processes taking place in the filtration area. The removal of these compounds was found to be higher than 95% at 25 °C.
- DDT, pyriproxyfen, and pendimethalin exhibited redox-independent behaviour at high temperatures (25 °C), as their removal efficiencies exceeded 95%.
- Molinate and propanil showed redox-dependent removal behaviour with high attenuation under oxic conditions (> 87%). In contrast, β-BHC, and endosulfan sulfate showed slightly higher biodegradation under sub-oxic conditions (> 94%).
- Atrazine, simazine, isoproturon, and metolachlor, characterized by persistence properties under oxic conditions, were also not removed under sub-oxic conditions. Thus, post-treatment may be required to remove these OMPs from bank filtrate.
- Polyaromatic hydrocarbons (PAHs) compounds exhibited high removal (> 80%) during the batch study regardless of the organic composition of the feed water and the redox environment. However, a significant effect of temperature was observed for lower hydrophobic PAHs (e.g., naphthalene, fluorene, and anthracene) at high temperature that may attribute to lower adsorption efficiency or desorption process, which needs to be investigated
- In summary, the results indicated that higher temperature enhanced the removal of OMPs during the RBF process, although it decreased the attenuation of bulk organic matter.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jece.2019.102904.

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