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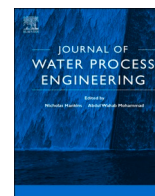To cite this publication, please use the final published version (if applicable).
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The fate of heavy metals during bank filtration: Effect of dissolved organic matter

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ABSTRACT

The effectiveness of bank filtration (BF) is highly dependent on the source water quality (e.g., organic matter composition, pH, and concentration of heavy metals (HMs)). In this study, the impact of dissolved organic matter (DOM) on the removal of selected metals (Cu, Zn, Pb, Se, and Ni) during BF was investigated. Column studies were conducted at 30 °C with feed water sources of different organic matter composition. Excitation–emission matrix fluorescence coupled with parallel factor analysis (PARAFAC–EEM) was used to characterise the organic composition of the feed waters. Moreover, another series of column studies was conducted to assess the impact of natural organic matter type (humic, protein) and concentration on the HMs removals. The experimental results revealed a high Pb(II) removal efficiency during filtration, which depends only slightly on the organic matter content of the feed water. In comparison, the removals of Cu, Zn and Ni ranged between 65 and 95 %; and relied significantly on the organic concentration and composition in the raw waters. Humic compounds (terrestrial or microbial) demonstrated adequate ability to reduce the removal efficiencies of these HMs during the infiltration. Conversely, biodegradable matter was found to be effective in enhancing the sorption of HMs onto the sand grains. The Se-removal was enhanced when the feed water contained a higher concentration of biodegradable matter. In general, it can be concluded that the organic composition of the source water affects profoundly the removal of HMs during the BF, and should be considered in the design of BF systems.

1. Introduction

Bank filtration (BF) is a natural and economical water treatment technology which can be applied worldwide, provided that the hydrogeological conditions and quality of the regarding river or lake are satisfactory [1,2]. The technique involves induced infiltration of the river/lake water towards the production well, situated at some distance from the river/lake. The subsurface sediments act as natural filters that are capable of removing different types of contaminants from the river/lake water [3]. This technology has been implemented in Europe for more than 100 years, and recently, the application of BF has extended into many developing countries worldwide. Compared to surface water treatment, BF is considered one of the most cost-effective, known technology that can be effectively applied due to its simplified design, lower operation and maintenance and costs. BF is known as an efficient

technique to remove the pollutants from source water and supplying high-drinking water quality [4,5]. BF can remove different contaminants including organic micro-pollutants, microbial pathogens, nutrients, and heavy metals (HMs) [6,7].

HMs are widely distributed in the earth's crust and are naturally non-biodegradable. Metals such as mercury (Hg), cadmium (Cd), arsenic (As), chromium (Cr), thallium (Tl), zinc (Zn), nickel (Ni), copper (Cu), selenium (Se), and lead (Pb) are commonly found in surface waters. A small number of HMs (in trace amounts) have an essential role in the metabolism of humans and animals, however, higher concentrations may be toxic. HMs constitute a significant public health concern because of their bio-accumulative nature in biotic systems and must therefore be removed from drinking water [8]. Excess exposure to Pb causes irreversible brain damage and encephalopathy symptoms. Drinking water with high levels of Cu may cause nausea, vomiting, stomach cramps, and

sometimes diarrhoea [9]. In addition, Se amounts above the maximal contaminant level can cause numbness in the extremities, circulation problems, neurological impairment, and fingernail or hair loss. High levels of Zn in drinking water may cause stomach cramps, vomiting, and sometimes nausea [8]. Moreover, a high concentration of Ni can damage the DNA of the hands [10].

The removal of HMs during BF is highly dependent on the site conditions, source water quality matrix, and HM type. Various studies have been conducted to investigate the biogeochemical processes taking place during the infiltration of river water into alluvial aquifers and its impacts on the removal of HMs [11,12]. These studies highlight that the adsorption of HMs during the filtration process is considerably impacted by the water quality matrix of the raw water, more specifically, the pH, dissolved oxygen, and dissolved organic matter (DOM). DOM contains multiple functional groups that have an affinity for HMs and hence may affect the mobility of HMs in aquatic and soil systems during the BF process [13].

The interaction of DOM and HMs in surface water occurs in different ways [13]. DOM in surface water forms complexes after reaction with HMs, which reduces the bioavailability and toxicity of HMs to aquatic organisms [10]. The type of DOM such as humic substances (fulvic acid and humic acid) influences the geochemical mobility of metal ions during the infiltration processes [13]. According to Reuter et al. [14], metal–humic complexes in natural water systems have a higher stability than inorganic–metal compounds. Hence, the presence of DOM affects the variation in the behaviour of metal pollutants in natural water systems.

The risk of contaminated drinking water due to HMs is increasing, in particular in developing countries where industrial and human activities result in the discharge of waste with high organic concentration into water bodies [10]. In a previous study [15], the authors discussed the behaviour of DOM fractions during the BF process. However, the effect of DOM on heavy metal removal during BF processes is not well understood. In addition, hardly any studies to elucidate the impact of DOM fractions (humic, fulvic, and protein) on the behaviour of HMs during the BF process had been carried out. Hence, the main objective of this research was to study the behaviours of heavy metals during infiltration and extent to which these behaviours could be affected by the concentration and composition of organic matter in the source water, especially in hot climate regions. This is a prerequisite step towards the successful application of BF in arid climate countries. Therefore, laboratory-scale column studies were conducted at a controlled room temperature (30 °C) to investigate the effect of the feed water source and natural organic matter (NOM) type (humic-like, protein-like tyrosine, the combination of protein-like and humic acid) on the removal of HMs (Cu, Zn, Ni, Pb, and Se) during the infiltration process.

2. Materials and methods

2.1. Filter media characteristics

The silica sand (grain size 0.8–1.25 mm) used in this study was bought from Wildkamp company (Netherlands). The characteristics of the sand is presented in Table S1. The media was washed with non-chlorinated tap water to discard the debris before introducing into the columns. The average bulk density and porosity of sand were 1500 kg/m^3 and 0.42, respectively. The organic content and the HMs concentrations of the sand were determined using the acid-extraction method described in [16]. The biological activity associated with the saturated sand was estimated in terms of Adenosine triphosphate (ATP) following the technique described in previous studies [15,17].

2.2. Column experiment

2.2.1. Experimental set-up

Laboratory-scale soil column setup was used to assess the impact of

organic matter on the removal of HMs during the filtration process. The PVC pipes with an internal diameter of 4.2 cm and a total height of 50 cm were used for all experiment. All pipes were fitted with a stopper leaving a space of 5 cm at the top and 5 cm for supporting materials (saturated gravels) at the bottom. Natural silica sand with a grain size ranging between 0.7 and 1.25 mm was filled up on top of the gravel for about 40 cm in all columns. Then the column was fitted with connectors from both ends to fix the influent and effluent tube. The tube was connected to the influent tank of about 10 L by using a polyethylene tube. The influents and effluents tubes were replaced after every five days and also covered with black materials and foils to reduce biofilm formation during the experimental process. The influent container was cleaned with diluted HCl and washed several times with distilled water before introducing the feedwater into the tank. Aeration was executed by inducing air tubes connected from the oxygen gas diffuser into the feed water tanks. The dissolved oxygen (DO) was monitored continuously to ensure the oxygen supplied is enough to support the experimental condition (DO > 7 mg/L). To introduce the feedwater from the tank into the column at the desirable hydraulic load rate, a variable speed peristaltic pump was attached to the bottom of the column. The experiment was implemented at a controlled temperature room (30 °C) to assure the possibility of removing HMs during BF in hot climate areas. Different parameters were monitored periodically to ensure that the experiments were conducted under the desired conditions. Such parameters include the dissolved oxygen, pH of the influent, infiltration rate and the concentration of HMs in the feed water.

2.2.2. Effect of DOM composition of raw-water on HM removal during BF

Four PVC columns were fed with different types of source waters to investigate the effect of the feed-water organic matter composition on the removal of HMs. Therefore, four types of feed water were prepared from different sources including (I) non-chlorinated tap water (NCTW) with a low organic content, (II) Delft canal (DC) water collected from a canal, (III) secondary treated wastewater obtained from the Harnaschpolder WWTP and mixed with Delft canal water to simulate contaminated surface water systems (DCWW), and (IV) water containing extractable organic matter (WEOM) which contains higher concentrations of humic compounds, which was prepared following the method proposed by Guigue et al. [18]. The physical–chemical water quality parameters of all feed waters were analysed before the start of the experiment (Table 1). The pH of the feed waters was adjusted to 7.8 ± 1 using NaOH solution. The column was run in the up-flow mode and the empty bed contact time was 12 h.

2.2.3. Effect of NOM type on HM removal during BF

This experiment was principally conducted to investigate the impact of the NOM type and concentration on the HM removal efficiency during a BF. Therefore, four columns were developed and ripened at 30 °C, and hydraulic rate of 0.3 m/day. The first column was fed with NCTW and used as a control sample. The other columns were fed with mixtures of

Table 1
Characteristics of the feed waters used for the laboratory- columns experiment.

Parameters	DC	DCWW	WEOM	NCTW
pH	8.3 ± 0.20	8.0 ± 0.10	8.5 ± 0.10	7.8 ± 0.20
DO (mg/L)	8.4 ± 0.20	7.9 ± 0.20	7.9 ± 0.10	8.5 ± 0.30
EC (μS/cm)	881 ± 0.58	838 ± 0.80	700 ± 0.50	551 ± 0.58
Total Nitrogen TN (mg/L)	1.1 ± 0.06	6.7 ± 0.14	1.8 ± 0.01	1.1 ± 0.06
NH$_4^-$ -N (mg/L)	0.03 ± 0.00	1.2 ± 0.03	0.7 ± 0.02	0.03 ± 0.00
PO$_4$-P	< 0.3	1.6 ± 0.00	< 0.3	< 0.3
Se(IV) (μg/L)	< 2	< 2	< 2	< 2
Cu(II) (μg/L)	16.7 ± 0.28	8.2 ± 2.30	138.5 ± 2.12	5.2 ± 2.30
Pb(II) (μg/L)	< 5	< 5	< 5	< 5
Ni(II) (μg/L)	< 5	< 5	< 5	< 5
Zn(II) (μg/L)	29.5 ± 2.10	27.5 ± 0.71	47.5 ± 0.50	7.1 ± 1.30

All values were presented as Mean value ± standard deviation, n = 7.

NCTW and different NOM types: (i) humic (HA), (ii) TY, and (iii) a mixture of 50 % HA and 50 % TY (HA:TY). Each type of NOM was injected at four different concentrations (5, 10, 15, and 20 mg-C/L). The experiments were conducted for each concentration and samples from the influent and effluent were taken daily.

2.2.4. Effect of NOM type on specific HM removal efficiency

The main target of this experiment was to analyse the impact of NOM type on the removal efficiency of specific HM's during saturated infiltration and to assess whether the presence of multiple metals in the feed water can affect the adsorption characteristics of other HM's and their behaviours toward the organic matter fractions. Therefore, eight columns were prepared and ripened for two months. Subsequently, the columns were fed with NCTW containing different NOM types. Cu(II) and Se(IV) were spiked individually into the feed water of the four columns, and the experiment took seven days at each hydraulic rate.

The Thomas model was used to estimate the breakthrough of the metals and impact of the NOM type on the adsorption parameters (adsorption capacity and rate) of the tested metals during the filtration process. This kinetic model assumes that adsorption is governed by the mass transfer and chemical reaction processes, and this reaction is a reversible second-order reaction, which obeys the Langmuir adsorption kinetics. Thomas model in the linear form is described as follows (Eq. 1) [19]:

$$\ln\left(\frac{C_0}{C_t} - 1\right) = \frac{K_{TH}\, q_0\, X}{Q} - K_{TH} C_0\, t \tag{1}$$

K_{TH} is Thomas adsorption rate constant (L/hr.μg), q_0 is the adsorption capacity (μg/g), X is the mass of adsorbent (g), Q is the flow rate of the feed water (L/hr), $C_{0 \text{ and }} C_t$ are the initial and breakthrough concentration (μg/L).

2.3. Analytical methods

Samples were collected from both influent and effluent of the columns and filtered using 0.45 μm filtration (Whatman, Dassel, Germany). The samples collected were acidified with 0.5 mL of concentrated HCl and stored at a controlled temperature room (4 °C). The metal (Cu, Zn, Ni, and Pb) concentrations were determined by inductively coupled plasma–mass spectrometry (Xseries II Thermo Scientific). The limit of detection (LOD) of metals was 10 μg/L. The Se concentration was quantified with a graphite furnace atomic absorption spectrophotometer (Solaar MQZe GF95, Thermo Electron Co.), where the LOD was 5 μg/L.

The combustion technique was employed to determine the organic content (DOC in mg-C/L) of the influent and effluent samples using a total organic carbon analyser (TOC-VCPN (TN), Shimadzu, Japan) (LOD = 20 mg/L). Specific ultraviolet absorbance (SUVA$_{254}$) [L mg^{-1} m^{-1}] was estimated to define the aromaticity of the influent and effluent. SUVA$_{254}$ was calculated as a ratio between the DOM of the water sample and its ultraviolet absorbance (UV) at 254 nm [m^{-1}]. The UV absorbance was determined using a UV/Vis spectrophotometer (UV-2501PC Shimadzu).

The organic constituents of the samples were determined with the fluorescence excitation–emission matrix (EEM) technique (Fluoromax-3 spectrofluorometer, HORIBA Jobin Yvon, Edison). The fluorescence intensity of the samples was determined at the excitation wavelengths (λex) 240–452 nm with 4 nm intervals and at the emission wavelengths (λem) 290–500 nm with 2 nm intervals.

The fluorescence indices, including the humification index (HIX), fluorescence index (FIX), and biological index (BIX), were used to characterise the organic fluorescence characteristics of the feed water. These indices were described in details by Gabor et al. [20]. HIX is an indicator of the humification degree of the feed water, and estimated as follows [21]:

$$HIX = \frac{\sum FI_{\lambda em\,(434-480\ nm)}}{\sum FI_{\lambda em\,(434-480\ nm)} + \sum FI_{\lambda em\,(300-344\ nm)}} \quad at\ \lambda ex\ (254\ nm) \tag{2}$$

FIX is used to detect the source of DOM in water (terrestrial or microbial). The value of FIX for each sample was estimated following Eq. 3 [20], and it varies between 1.2–1.8 with lower values refer to DOM of terrestrial origin.

$$FIX = \frac{FI_{\lambda em\,(450)}}{FI_{\lambda em\,(500)}} \quad at\ \lambda ex\ (370\ nm) \tag{3}$$

BIX was used to estimate the contribution of microbial DOM to the total organic content of the feed water. This parameter was determined using Eq. 4 [22].

$$BIX = \frac{FI_{\lambda em\,(380)}}{FI_{\lambda em\,(430)}} \quad at\ \lambda ex\ (310\ nm) \tag{4}$$

2.4. Fluorescence modelling

The fluorescence EEM spectroscopy was combined with the parallel factor analysis (PARAFAC) algorithm to discretise the fluorescence dataset into independent fluorescence components. This technique has been widely used to identify organic matter characteristics of surface and subsurface water systems [15,23]. The PARAFAC technique uses an alternating least-squares algorithm to separate the fluorescence dataset into independent fluorescence components with different dissolved organic fractions and a residual error term (Eq. 5) [24].

$$X_{ijk} = \sum_{f=1}^{f} a_{if} b_{jf} c_{kf} + \varepsilon_{ijk}$$
$$, i = 1,\ldots\ldots,I;\ j = 1,\ldots\ldots,J;\ k = 1,\ldots\ldots,k;\ f = 1,\ldots\ldots,F \tag{5}$$

where X_{ijk} is the fluorescence intensity of the ith sample at the kth excitation and jth emission wavelength; f represents the number of fluorescence components; a_{if} is the score of each f th component and relates to the fluorophore f concentration in sample i; b_{jf} is the scaled estimates (loading) of the emission spectrum of the f th fluorescence component; c_{kf} links to the excitation spectrum of the f th fluorescence component and expressed as loading; and ε_{ijk} is the residual term representing the unaccounted variation of the model.

A fluorescence dataset of 80 samples collected from the feed water was used to develop and validate the PARAFAC models following the steps proposed by Murphy et al. [24]. Briefly, an initial exploratory examination was first implemented to identify the poor quality measured data (outliers) and removed them from the dataset. The outliers (samples or variables) are commonly produced due to sampling or measuring errors and are determined by conducting the leverage analysis. The leverage value ranges between zero and one and expresses the deviation of measurement from the average data distribution. Then, models with different components (3–7) proceeded and tested using different diagnostics techniques, including: split-half validation, the randomness of residuals and core consistency [24].

2.5. Data analysis

The two-way analysis of variance (ANOVA) and post-hoc Tukey test were used to assess the significance of the impact of organic composition of the feed water and NOM type on the removal of HMs during the column filtration process; the impact was considered significant if the level of significance (p) was below or equal to 0.05.

In addition, a Spearman rank correlation (ρ) analysis was conducted to determine the relationship between the fluorescence characteristics of the feed water and the HM removal during the filtration process. The strength of the relationship was considered high if the correlation value $\rho > 0.7$, moderate if $0.4 < \rho > 0.7$, and low if $\rho < 0.4$ [25].

3. Results

3.1. Impact of feed water source on HM removal efficiency

The effect of the organic composition of the feed water on the HM removal efficiency was studied with laboratory-scale columns and different water sources spiked with various concentrations of HMs. The experiments were conducted at different hydraulic rates (0.3, 0.6, and 1.0 m/day).

3.1.1. Organic characteristics of feed water

The results (Table 2) infer that NCTW had the lowest concentration of organic matter; its value ranged between 3.47 and 3.67 mg-C/L. This feed water also showed had low SUVA (1.76 ± 0.14 L/mg-g) and HIX (0.49) values, which indicates its aliphatic organic composition. The DOC of the DC water (11.25 ± 0.81 mg/L) and DCWW (11.41 ± 0.59 mg/L) were significantly higher than that of the WEOM feed water (9.7 ± 0.24 mg/L). However, the WEOM contained a higher concentration of aromatic compounds than the DC water and DCWW; the average SUVA values of the DC water, DCWW, and WEOM were 2.99 ± 0.11, 3.72 ± 0.39, and 3.82 ± 0.27 L/mg-g, respectively. These results were confirmed by the fluorescence indices, which revealed that the WEOM had a higher humic concentration (HIX = 0.84) and fewer microbial-derived compounds (BIX = 0.54) than the DC water (HIX = 0.8, BIX = 0.72) and DCWW (HIX = 0.74, BIX = 0.79). Moreover, the FIX of the WEOM was evidently lower than that of the other feed waters, suggesting that its humic content originated mainly from terrestrial sources.

3.1.2. Effect of organic composition of feed water on HM removal efficiency

The results showed that Pb(II) had the highest capability to be removed during the filtration process; the effluent concentration of Pb (II) in all columns was below the detection limit (5 μg/L) of the instrument under all experimental conditions. However, Cu(II), Zn(II), and Ni (II) exhibited lower removal efficiencies (65 %–95 %). These removal efficiencies were highly dependent ($p < 0.05$) on the organic concentration and composition of the feed water, with a preferential removal at a low organic concentration (Fig. 1). The removal efficiencies of Cu(II), Ni(II), and Zn(II) for DC feed water were 88 % ± 1%, 88 % ± 6%, 90 % ± 2%, for the DCWW 66 % ± 4%, 75 % ± 9%, 70 % ± 3%, for the WEOM 79 % ± 1%, 85 % ± 3%, 87 % ± 4%, and for the NCTW 98 % ± 1%, 93 % ± 3%, 97 % ± 1% at a hydraulic loading rate of 1.0 m/day. These removal efficiencies changed by ± (1–16 %) at lower hydraulic rates (0.6 and 0.3 m/day) (Figs. S1 and S2). Therefore, the hydraulic loading rate had no significant effect ($p = 0.2–0.47$) on the HM removal efficiencies (Cu(II), Zn(II), Ni(II)) during the filtration process. Nevertheless, Se(IV) was the most persistent metal to be removed during the filtration process. The removal efficiency of Se(IV) depended highly on the feed-water organic composition ($p < 0.001$). The average removal efficiencies of Se(IV) for the columns fed with DC water, DCWW, WEOM, and NCTW were 23 % ± 5%, 25 % ± 5%, 7% ± 4%, and 4% ± 2% at a hydraulic loading rate of 1.0 m/day, respectively. Moreover, the

statistical analysis revealed that the infiltration rate had no significant effect ($p = 0.2$) on the Se(IV) removal efficiency. The Se(IV) removal efficiency in this study was linked to the biological activity associated with the sand. The ATP concentrations of the sand media of the columns fed with DCWW, DC water, WEOM, and NCTW were 9.3 ± 1.28, 9.2 ± 1.04, 5.1 ± 0.33, and 3.3 ± 0.47 μg/g, respectively. Furthermore, the correlation between Se(IV) removal efficiency and ATP concentration was 0.96.

3.2. Impact of fluorescence organic compounds on HM removal

3.2.1. Fluorescence modelling

The PARAFAC–EEM model decomposed successfully the fluorescence data collected from the influents of the columns into three main components representing different organic composition groups. The contour plots and loadings of the identified fluorescence components are presented in Fig. 2. The Openfluor database, which employs Tucker's congruence coefficient to determine the similarity degree between the components, was used to identify the separated PARAFAC components and matched them to those recognised at the same excitation and emission wavelengths in the previous studies (Table S2). The first component (PC1) exhibited maximal excitation wavelengths (λex) at 240 and 344 nm and maximal emission wavelengths (λem) at 474 nm. Hence, this component probably corresponded to humic compounds derived from terrestrial sources [26]. Component 2 (PC2) exhibited two maxima (λex) at 240 and 300 nm and a single maximum (λem) at 396 nm. This component matched well with humic compounds originating from microbial sources [23]. Component 3 (PC3) exhibited two maxima (λex) at ≤240 and 272 nm and a maximum (λem) at 316 nm. This component resembled protein-like fluorophores originating from microbial sources [15]. The results (Table 2) refers that WEOM and DCWW feed water has the highest content of terrestrial and microbial humic compounds, respectively. Whereas, NCTW retained with the lowest concentration of the three organic fluorescence compounds.

3.2.2. Relationship between DOM fluorescence composition and HM removal efficiency

The correlations between HM removal efficiency and organic fluorescence characteristics of the feed waters are presented in Fig. 3. The results demonstrate that the organic matter concentration and fluorescence composition of the feed water had a significant impact on the HM behaviour during the filtration process. Pb(II) was impacted least by the variations in the organic concentration of the feed water; the correlation between the Pb(II) removal efficiency and organic matter concentration was (ρ) = -0.21. Conversely, DOM suppressed the removal efficiencies of Cu(II), Ni(II), and Zn(II) in the sand surface, and negative relationships between the removal efficiencies of these metals and organic concentration of the feed water were observed. The fluorescence indices results revealed that the humic content of the feed water affected negatively the adsorption of Cu(II), Zn(II), and Ni(II), and negative correlations between the removal efficiency of the previously mentioned metals and fluorescence characteristics (HIX, BIX, and FIX) of the feed water were

Table 2
Organic characteristics of the feed waters used for the laboratory- columns experiments.

| Parameters	Units	DC	DCWW	WEOM	NCTW
pH	——	8.3 ± 0.20	8.0 ± 0.10	8.5 ± 0.10	7.8 ± 0.20
DOC	mg-C/L	11.25 ± 0.81	11.41 ± 0.59	9.7 ± 0.24	3.6 ± 0.11
UVA$_{254}$	(cm^{-1})	0.34 ± 0.04	0.45 ± 0.04	0.37 ± 0.02	0.06 ± 0.01
SUVA	(L/mg-g)	2.99 ± 0.11	3.72 ± 0.39	3.82 ± 0.27	1.76 ± 0.14
HIX	——	0.8 ± 0.08	0.74 ± 0.05	0.84 ± 0.07	0.49 ± 0.03
FIX	——	1.23 ± 0.27	1.37 ± 0.19	1.06 ± 0.23	1.04 ± 0.33
BIX	——	0.72 ± 0.11	0.79 ± 0.21	0.54 ± 0.06	0.39 ± 0.05
PC1	RU	0.91 ± 0.15	1.02 ± 0.28	1.6 ± 0.21	0.09 ± 0.02
PC2	RU	1.06 ± 0.33	1.2 ± 0.25	1.05 ± 0.28	0.14 ± 0.08
PC3	RU	0.76 ± 0.12	0.85 ± 0.19	0.54 ± 0.3	0.15 ± 0.05
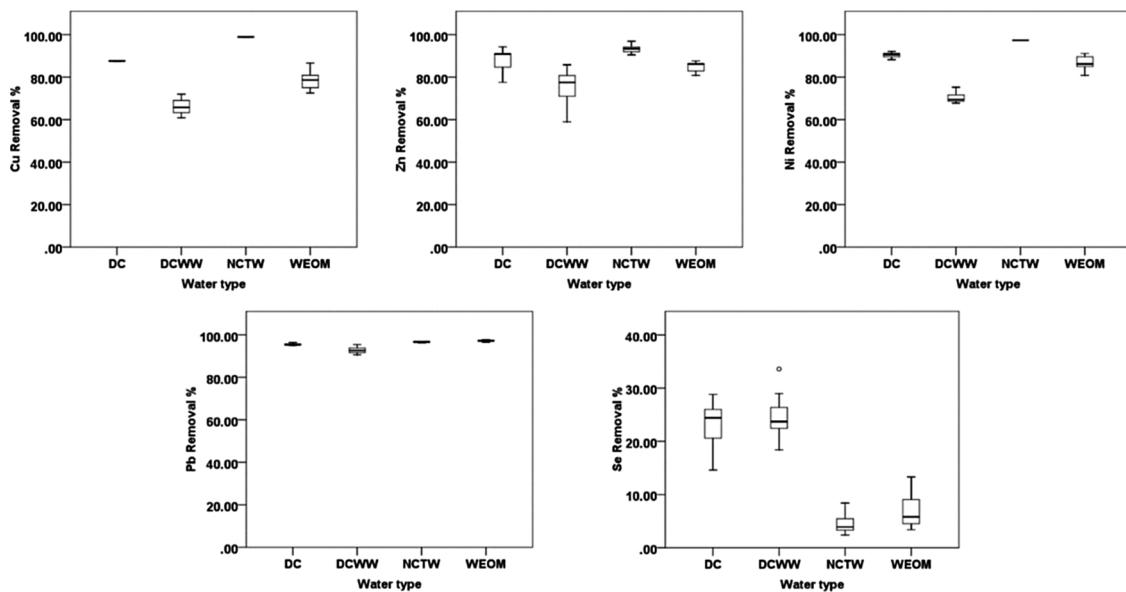
Fig. 1. Removal of Cu, Zn, Ni, Pb (initial concentration = 150 μg/L) and Se (10 μg/L) during aerobic column infiltration at HLR = 1.0 m/day (temperature = 30 °C).
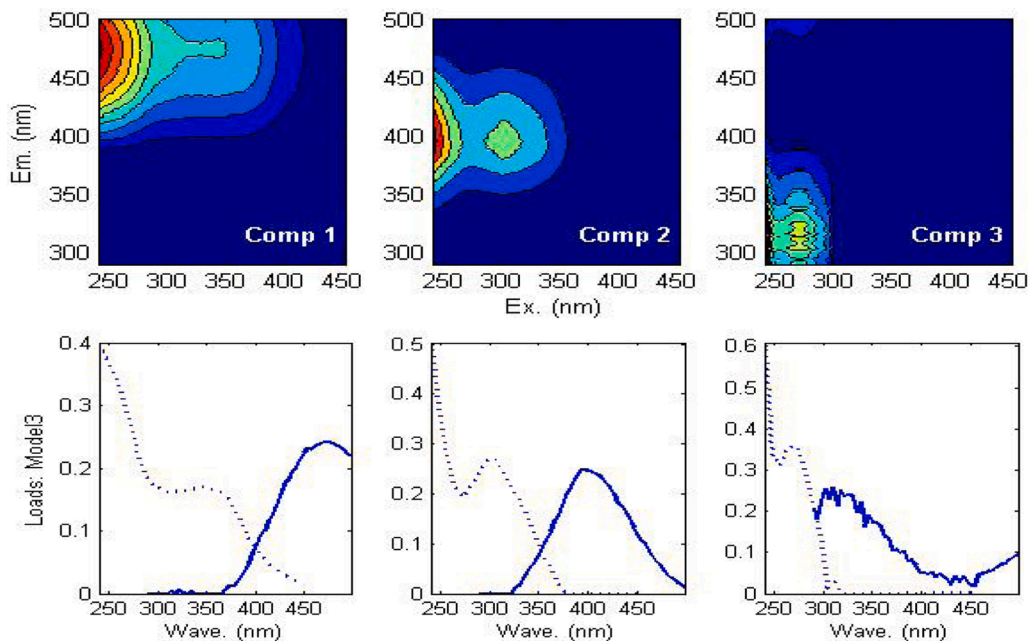

Fig. 2. Contour plots of the three PARAFAC components separated from the complete measured F-EEMs dataset for the influent and effluent of the columns experiment.

observed. Cu(II) was more affected by the changes in the humic content of the feed water than Ni(II) and Zn(II); the correlations between Cu(II), Ni(II), and Zn(II) removal efficiencies and HIX of the feed water were -0.63, -0.53, and -0.36, respectively. The PARAFAC data revealed that the terrestrial humic and microbial humic substances were highly correlated with the removal efficiencies of Cu(II), Ni(II), and Zn(II). Thus, the humic compounds, regardless of their structures, can react with the three metals and reduce their adsorption efficiencies.

In contrast, organic matter enhanced the removal efficiency of Se(IV) during the filtration process; a strong positive correlation was detected between the Se(IV) removal efficiency and the organic concentration of the feed water. The fluorescence data revealed that the Se(IV) removal efficiency depended weakly on the level of humic compounds in the feed water. In addition, a weak correlation ($\rho = 0.33$) was detected between

the Se(IV) removal efficiency and HIX of the feed water. However, the BIX ($\rho = 0.69$) and FIX ($\rho = 0.57$) exhibited a good correlation. The Se (IV) removal efficiency correlated well with the fluorescence intensity of the protein-like ($\rho = 0.72$) and microbial humic compounds ($\rho = 0.58$). In conclusion, the Se(IV) removal efficiency showed a stronger correlation to the biodegradable matter concentration of the feed water during the filtration process.

3.3. Impact of NOM on HM removal efficiency

The laboratory-scale column experiments were conducted to investigate thoroughly the effect of organic matter on HMs removal efficiency during filtration with different NOMs (HA, TY, and a mixture of HA:TY) at different concentrations (5, 10, 15, 20 mg/L) in the feed water. The
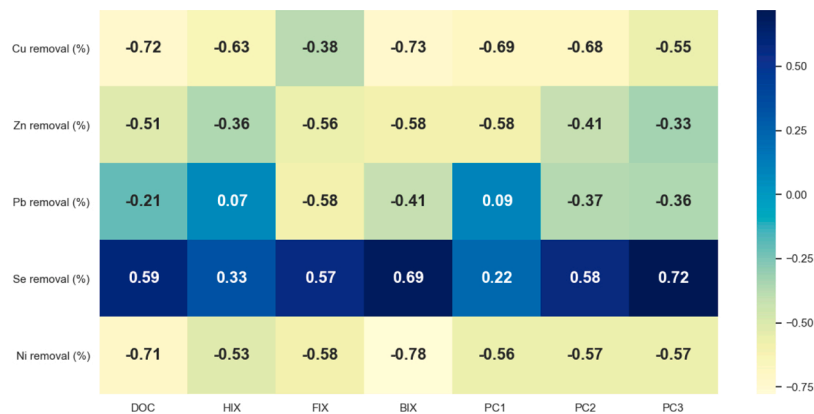
Fig. 3. Correlations between HM removal efficiency (%) and fluorescence characteristics of feed water.
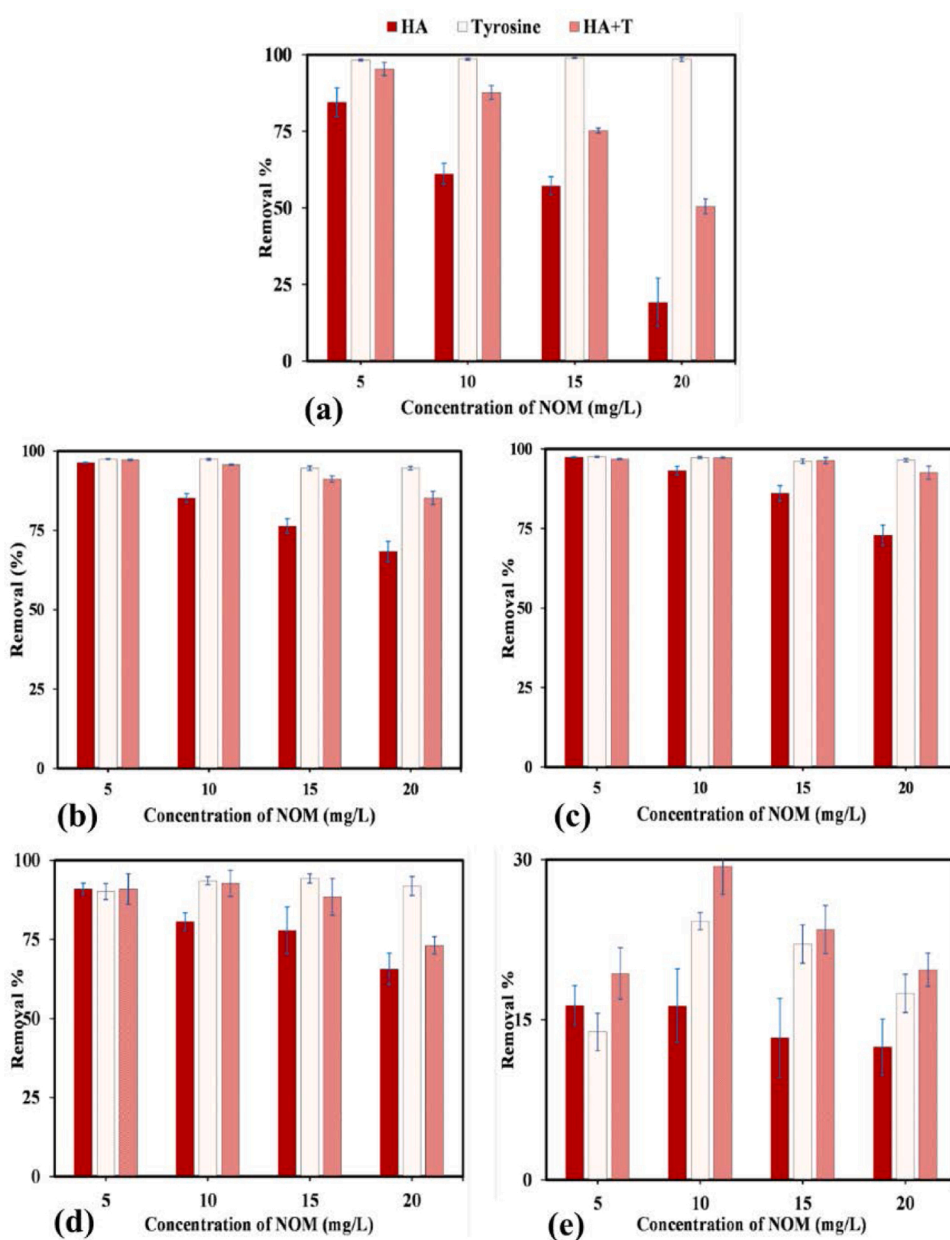

Fig. 4. Removal of (a) Copper, (b) Zinc, (c) Lead, (d) Nickel, and (e) Selenium during column infiltration with NCTW spiked with different NOM concentrations (5, 10, 15, 20 mg-C/L) at HLR = 0.3 m/day.

fluorescence characteristics of the feed water are presented in Fig. S3.

3.3.1. Effect of NOM type and concentration on HM removal efficiency

The type and concentration of the NOM had a significant influence on the HM removal efficiency during the filtration process (Fig. 4). The removal efficiency of Pb(II) exceeded 90 % in the presence of TY. The post-hoc tests (Tukey method) showed that there were no statistically significant differences in the removal efficiencies of Pb(II) in feed water containing different concentrations of TY or a mixture of HA and TY. This indicates the dominate role of labile organic compounds (TY) in enhancing the removal efficiency of Pb(II) during the filtration process. However, regarding the column fed with HA, the Pb(II) removal efficiency exhibited a gradual decrease with increasing HA concentration in the feed water. The Pb(II) removal efficiencies were 97 % $\pm$ 0.12 %, 93 % $\pm$ 1.5 %, 86 % $\pm$ 2.5 %, and 73 % $\pm$ 3% for humic concentrations of 5, 10, 15, and 20 mg-C/L in the feed water, respectively. This implies that the highly concentrated HA compounds (>5 mg-C/L) possessed sufficient capacity to suppress the adsorption of Pb(II) onto the sand surface.

The NOM type affected significantly the removal efficiencies of Cu (II), Ni(II), and Zn(II) during the filtration process, with a preferential removal in the presence of TY. However, no significant difference in the removal efficiencies of these metals was observed for the columns fed with different concentrations of TY because their removal efficiencies all exceeded 90 %. By contrast, the HA compounds deteriorated the adsorption efficiencies of these metals during the filtration process. The removal efficiencies of Cu(II), Zn(II), and Ni(II) were 85 % $\pm$ 5%, 91 % $\pm$ 2%, and 96 % $\pm$ 0.6 % for the column fed with 5 mg-C/L HA, respectively. As these removal efficiencies decreased to 19 % $\pm$ 8%, 66 % $\pm$ 5%, and 68 % $\pm$ 5% for a HA concentration of 20 mg-C/L in the feed water, respectively, Cu(II) was most influenced by the increasing HA concentration. The same trend was observed for the column which was fed with NCTW spiked with an HA:TY mixture, whereby the removal efficiencies of Cu(II), Ni(II), and Zn(II) decreased by 45 %, 18 %, and 12 % when the feed mixture concentration increased by 15 mg/L, respectively. Thus, humic compounds had a higher capacity than the TY compounds to suppress the adsorption efficiencies of Cu(II), Ni(II), and Zn(II) on a sand surface.

Se(IV) removal efficiency depended highly on the type and concentration of NOM in the feed water during filtration process. The column fed with a mixture of HA and TY exhibited the highest Se(IV) removal capacity during the filtration. However, the post-hoc results revealed that there were no significant differences in the removal efficiencies of Se(IV) between the columns fed with TY and HA:TY. On the other hand, Se(IV) exhibited a higher removal persistence in the absence of TY; the removal efficiency of Se(IV) was 16 % $\pm$ 3% for the column fed with 10 mg-C/L HA. This removal efficiency decreased to 13 % $\pm$ 2% and 12 % $\pm$ 1% in the feed water with 15 and 20 mg-C/L HA, respectively. Therefore, it can be deduced that biodegradable compounds are feasible for the Se(IV) removal in filtration processes.

3.3.2. Kinetic study of Cu(II) and Se(IV) in infiltration process

Among the tested metals, Cu(II) and Se(IV) were most impacted by the varying NOM types and concentrations during the filtration process. Therefore, laboratory-scale column studies were conducted to determine the adsorption kinetics of these two metals under different experimental conditions and the impact of the presence of other metals. This set of experiments was conducted under the same experimental conditions; however, a fixed concentration of each metal was individually injected into the NCTW feed water spiked with different NOM types (HA, TY, HA:TY). Furthermore, the Thomas model was used to describe the breakthrough curves (Fig. S4) and estimate the adsorption parameters (adsorption capacity and rate) of Cu(II) and Se(IV) during the filtration process (Tables 3 and 4). The results show that the experimental data and model agreed well (high R^2 and low X^2).

According to Table 3, the adsorption behaviour of Cu(II) in the columns fed with different NOM compositions exhibited evident

differences. TY had a positive effect on the adsorption characteristics of Cu(II) during the filtration process. In addition, the breakthrough time of Cu(II) was longer for the column fed with NCTW and TY than for the control column fed with only NCTW. Based on the Thomas model, the adsorption capacity of Cu(II) increased from 2.01–2.89 µg/g when 10 mg-C/L TY was added to the feed water. By contrast, HA compounds had a negative effect on the adsorption capacity of Cu(II). Its value decreased to 0.53 and 1.01 µg/g when HA and HA:TY were added to the NCTW feed water, respectively. Furthermore, the kinetic results revealed that the presence of other metals in the raw water reduced the effect of humic compounds on the adsorption of Cu and increased the adsorption capacity of the sand; the adsorption capacities were 2.83 and 1.9 µg/g for the columns fed with HA and HA:TY in the presence of other metals, respectively. However, this effect was negligible for the columns supplied with NCTW and TY.

Regarding Se(IV) (Table 4), the adsorption capacity of the sand was lower for the column fed with HA (0.003 µg/g) and higher for the column fed with HA:TY (0.016 µg/g) and TY (0.024 µg/g). Thus, the biodegradable compounds (TY) appeared to promote the removal of Se (IV) during the filtration process. However, the presence of other metals in the raw water had a negative effect on the adsorption characteristics of Se(IV) during the filtration process. The adsorption capacities of the sand decreased to 0.002, 0.019, and 0.012 µg/g for the columns fed with HA, TY, and HA:TY, respectively.

4. Discussion

4.1. Removal of HMs during bank filtration

The Pb(II) removal efficiency was higher than other metals during filtration, regardless of the surrounding environmental conditions. More specifically, the Pb removal efficiency was above 90 % under all experimental conditions (different hydraulic rates and feed-water organic compositions). According to Kalakodio et al. [27], precipitation and adsorption are the main mechanisms which remove Pb(II) during the sand filtration process. Furthermore, a PHREEQC analysis was conducted in this study to determine the precipitation characteristics of the added Pb(II). Based on the results, its concentration in the feed water was insufficient for precipitation (saturation index SI < 1), which implies that the removal of Pb(II) during the column experiment was mainly caused by adsorption. Several studies [28–30] highlighted that Pb has a higher affinity to the organic matter and silicates groups at the surface of the soil than other metals. Sontheimer [31] reported an average Pb(II) removal efficiency of 75 % at BF sites along the Rhine River (Germany). In this research, the experiment was conducted at high temperature (30 $\pm$ 2 °C), which considerably enhanced the adsorption characteristics of the metal during filtration. Awan et al. [32] reported that Pb(II) has a high probability to become hydrolysed in water. Thus, it can be readily chemisorbed on the sand. The hydrolysis and adsorption processes were strongly enhanced by the increased temperature. Furthermore, Guanxing et al. [33] highlighted that the adsorption of Pb (II) into the soil is an endothermic reaction, and its efficiency is positively related to temperature. The high Pb(II) removal capacity of a sand filtration process was also observed in column experiment conducted at 25 °C [34]. Thus, it can be concluded that the BF technique is capable of removing Pb(II) from source water, in particular under hot-climate conditions.

The removal efficiencies of Cu(II), Zn(II), and Ni(II) were lower (65 %–95 %) than that of Pb(II) during filtration and statistically independent of the flow rate of the feed water. Consequently, the retention time did not play a significant role in the removal of these metals during the filtration process. The efficiency of the BF technique in the removal of these metals under different hydrological and climate conditions has been investigated. Nagy-Kovács et al. [3] reported removal efficiencies of 59 %–99 % of these metals at BF sites along the Danube River (Hungary). Awan et al. [32] highlighted that the removal efficiencies of

Table 3
Thomas adsorption parameters of Cu(II) of columns fed with different NOM types in the presence/absence of metals.

	Cu(II) [individual]				Cu(II) [mixed]			
	K_{TH} (L/hr.µg)	q_0 (µg/g)	R^2	X^2	K_{TH} (L/hr.µg)	q_0 (µg/g)	R^2	X^2
HA	5.00×10^{-05}	0.53	0.96	0.80	1.33×10^{-05}	1.63	0.95	0.36
TY	2.20×10^{-05}	2.89	0.94	0.02	2.53×10^{-05}	2.83	0.87	0.14
HA:TY	4.80×10^{-05}	1.01	0.96	0.31	2.27×10^{-05}	1.90	0.98	0.12
NCTW	3.15×10^{-05}	2.01	0.94	0.10	3.87×10^{-05}	2.40	0.91	0.22

Table 4
Thomas adsorption parameters of Se(IV) for the columns fed with different NOM types in the presence/absence of metals.

	Se(IV) [individual]				Se(IV) [mixed]			
	K_{TH} (L/hr.µg)	q_0 (µg/g)	R^2	X^2	K_{TH} (L/hr.µg)	q_0 (µg/g)	R^2	X^2
HA	1.41×10^{-03}	0.003	0.91	0.07	2.14×10^{-03}	0.002	0.89	0.34
TY	4.64×10^{-04}	0.024	0.91	0.03	4.64×10^{-04}	0.019	0.87	2.1
HA:TY	5.93×10^{-04}	0.016	0.95	0.03	5.29×10^{-04}	0.012	0.94	2.2
NCTW	2.14×10^{-03}	0.004	0.96	1.99	1.47×10^{-03}	0.0017	0.92	0.12

these metals are highly dependent on their affinity to negatively charge groups (e.g., OH^-) at the soil surface, which depends strongly on the environmental conditions (e.g. temperature, pH, and organic content of the raw water).

A high persistence of Se(IV) was observed during the column filtration process; the removal efficiency of Se(IV) did not exceed 40 % under all experimental conditions. This removal efficiency was statistically independent of the infiltration rate of the raw water. Schmidt et al. [35] reported a low Se(IV) removal efficiency (15 %) at a BF site along the Rhine River (Germany). The lower Se(IV) removal efficiency during the filtration process was mainly ascribed to its natural characteristics of a high solubility and low adsorption characteristics, which was also reported by Li et al. [36].

4.2. Impact of organic matter on HM removal efficiency

The organic composition of the feed water had a minor effect on the Pb(II) removal efficiency during filtration. According to Ahmed et al. [37], organic compounds augment the Pb(II) removal efficiency by forming adsorbable complexes. However, this research study revealed that feed water with a higher HA concentration (>15 mg-C/L) might suppress the removal efficiency of Pb(II) by 20 %. Likewise, HA had a negative impact on the removal efficiencies of Cu(II), Zn(II), and Ni(II) during filtration. The highest reduction in removal efficiency was observed when the WEOM (higher terrestrial humic content) and DCWW (higher microbial humic content) feed water were used. The experimental results showed that the addition of 20 mg-C/L HA compounds to the feed water reduced the removal efficiencies of Cu(II), Zn(II), and Ni(II) by 40 %–75 %. Similarly, the Se(IV) removal efficiency decreased with increasing HA concentration in the feed water. The lower removal efficiency of the former metals in the presence of a higher HA concentration was mainly caused by (I) the ability of organic matter to react with metals and to form aqueous complex compounds, which increases the mobility of the metals and reduces their adsorption efficiencies in the sand surface [38]. (II) HA compounds have a high potential to accumulate on the sand surface, which reduces its adsorption capacity [39]. (III) HA compounds contain active functional groups (e.g. carboxylic, phenol, and catechol OH) which may associate to the minerals on the sand surface and compete with metal for the adsorption sites [40]. (IV) The HA compounds can change the soil structure by improving its aggregation and thereby reducing the number of adsorption sites [41]. Interestingly, the fluorescence data demonstrated that the HA compounds, regardless of their source (microbial or terrestrial), reacted with these metals and reduced their adsorption efficiency. Moreover, HA compounds enhanced the desorption rates of the metals

via chelating and exchange reaction processes. Tian et al. [42] illustrated that the desorption rate of Se(IV) increases by approximately 50 % in the presence of hydrophobic HA compounds; the hydrophilic compounds could release a low amount (below 3.5 %) of the adsorbed Se [43].

The impact of HA compounds on the metal removal depends strongly on the properties of the metal and its adsorption mechanisms on the sand surface. In this research study, Cu(II) was most impacted by the variation in the HA concentration in the feed water. However, Cu(II) exhibited a higher removal efficiency than Zn(II) and Ni(II) in the column fed with low-organic content feed water (NCTW). The sorption of Cu(II) is predominantly based on the formation of strong electrostatic and covalent bonds with negative functional groups (e.g. organic matter) on sand surface grains [39]. By contrast, Zn(II) and Ni(II) tend to remain on the surface through an electrostatic attraction process [39]. Therefore, Cu(II) is expected to have a higher affinity than Zn(II) and Ni(II) for the functional groups on the sand grains during the filtration process. However, this affinity decreases with increasing humic concentration in the feed water. Hence, Cu(II) prefers to react with soluble HA substances and to form aqueous complexes than to be adsorbed onto active organic sorption sites of the soil. The Zn(II) and Ni(II) adsorption rates were also affected (to a lower extent) by the humic content of the feed water. Zhao et al. [38] pointed out that Cu(II) binds stronger to the active groups (e.g. phenols [−OH], amines [−NH₂], and carboxyl [−COOH]) of microbial and terrestrial humic compounds than Zn(II) and Ni(II).

By contrast, the biodegradable (protein-like) compounds enhanced the HM removal efficiency during filtration. According to Abdelrady et al. [44], LMW organic compounds tend to be adsorbed faster onto the sand surface than high-molecular weight (HMW) compounds during a filtration process. Therefore, a metal with a higher affinity towards LMW compounds is more likely to be removed during filtration. Moreover, biodegradable matter promotes biological activity associated with the sand and thus enhances the biosorption and accumulation of metal on its surface [45]. In this study, the sand column with a higher microbial activity (higher ATP concentration) had a higher HM removal efficiency during the infiltration process. Moreover, 5 mg-C/L TY was sufficient for optimising removal of HM (Cu, Zn, Ni, and Pb) during the filtration. However, it worth to note that it might enhance the development of an anaerobic environment within the infiltration zone, and consequently, promote the reduction and releasing of metals into the bank filtrate.

4.3. Impact of metals on sorption of Cu and Se during bank filtration

The conducted kinetic experiments revealed that the adsorption rate

of Cu(II) increased significantly when the feed water contained a mixture of metals. Hence, the existence of other metals in the feed water retarded presumably the formation rate of the aqueous Cu(II)–HA complex. By contrast, the presence of other metals in the raw water had an adverse effect on the adsorption capacity of the sand for Se(IV) during filtration; a lower Se(IV) removal efficiency impact, even when the raw water contained LMW (TY) or HMW (humic) organic compounds. Dinh al. [46] pointed out that LMW organic compounds tend to react with metals and form stable ring-complexes that compete with Se(IV) for adsorption sites. However, HMW compounds can reduce the adsorption capacity of sand by accumulating and blocking adsorption sites or changing the sand structure, as previously discussed. Therefore, it can be concluded that good removal of Se(IV) can be achieved with BF when the raw water has a low concentration of metals and high content of biodegradable organic matter.

5. Conclusions

Laboratory-scale column experiments were conducted to assess the impact of the feed water and organic composition on the removal of HMs (Cu, Zn, Ni, Pb, and Se) during the BF process, and the following conclusions are drawn:

- Pb is the most probable metal to be adsorbed into the sand surface during the filtration process. However, humic compounds at concentrations above 15 mg-C/L reduced the adsorption efficiency by approximately 20 % in pure water.
- Cu, Zn, and Ni exhibited lower removal efficiency during the filtration process (70 %–95 %) for the column fed with different water sources. Furthermore, the experimental results showed that 10 mg-C/L humic compounds could reduce the adsorption efficiencies of these metals by 40 %–75 %.
- Se was the most persistent metal during the column filtration process among the tested metals; its removal was less than 40 % under all the experimental conditions.
- Biodegradable matter enhanced the adsorption rates of the aforementioned metals, whereas humic compounds negatively affected their removal efficiencies.
- PARAFAC fluorescence data revealed that presence of both terrestrial and/or microbial humic compounds reduced the adsorption efficiency of the metals.
- The kinetic study revealed that the presence of other metals in the raw water reduced the influence of the humic compounds on the adsorption of Cu and thus enhanced its removal efficiency. Conversely, the presence of HMs exhibited a negative effect on the removal of Se during the filtration process.

Therefore, this study highlighted the impact of dissolved organic matter composition on the removal efficiencies of HMs, which should be considered in the design and installation of bank filtration wells.

Declaration of Competing Interest

None

Acknowledgments

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.jwpe.2020.101563.

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