

Semivolatile organic compounds removal and health risk reduction in drinking water treatment biofilters applying different backwashing strategies

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Abstract This study aimed to investigate the removal of 24 semivolatile organic compounds in Yangtze River (China) source water treated by six biofilters using different backwashing methods. Health risks induced by the pollutants in the influent and effluent water were also assessed based on the chemical detections. Comparatively, the biofilter backwashed with both air (15 m/h, 3 min) and water (8 m/h, 5 min) was most efficient in removing semivolatile organic compounds and reducing health risk. PCR-denatured gradient gel electrophoresis showed that backwashing alterations posed considerable influences on microbial community structure in the six biofilters. About 72.4 % of di-*n*-butyl phthalate and 81.8 % of bis(2-ethylhexyl)phthalate (two main semivolatile organic compounds in the river water) were removed under the optimal backwashing conditions. However, in the effluent of each biofilter, non-carcinogenetic risks of 2,6-dinitrotoluene and bis[2-ethylhexyl]phthalate and carcinogenetic risks of dibenz[a,h]anthracene and benzo[a]pyrene did not reach safety levels, revealing that these pollutants in the source water deserve more public health concerns. This study might serve as a basis for biofiltration process optimization and also as a benchmark for the authorities to reduce the health risk induced by exposure to the hazardous pollutants.

Keywords Biological filtration · Drinking water contamination · Operational optimization · Public health

Introduction

Yangtze River is the longest river in Asia and drains one-fifth of China's land area. The river basin containing one-third of China's population is one of the fast developing regions of China. Yangtze River is being used as drinking water source for most of the people living in the basin. However, the region is suffering from enormous environmental deterioration along with rapid industrialization. Recently, various semivolatile organic compounds (SVOCs), including polycyclic aromatic hydrocarbons (Xu et al. 2001), polychlorinated biphenyls (Sun et al. 2002) and environmental endocrine disruptors (Wu et al. 2009b), have been detected in the water of Yangtze River, which is possibly threatening the safety of the drinking water supplied for the people living along the river. Therefore, these chemicals have to be effectively removed from drinking water in this region to reduce potential public health risk.

Biofiltration has been widely used in drinking water treatment for the removal of organic pollutants (Ifelebuegu 2011; Kim 2009). This technique enhances biological substrate removal by providing a surface for indigenous water microorganisms to attach and grow. Compared to conventional sand filtration, granular activated carbon (GAC) biofilters often shows higher efficiencies (Chien et al. 2006) due to effective bioadsorption and biodegradation in these biofilters, since dissolved oxygen is sufficient for the microorganisms (Zhang et al. 2010). However, biofilter performance is affected by various factors including temperature (Fazaelipour 2010), substrate (Rene et al. 2008; Yu et al. 2009), hydraulic loading (Hassan and Sorial 2010;

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Zhang et al. 2010) and filter media (Liu et al. 2001). Recent studies have focused on the regulation and optimization of operational biofiltration conditions so as to improve removal efficiency of pollutants in wastewater (Spagni et al. 2010) or drinking water treatment (Devi et al. 2008).

Frequent water or air backwashing can remove loosely attached large microbial aggregates in favor of dead cells removal and microbial community stabilization (Bai et al. 2005; Choi et al. 2007). However, limited information is available about the influence of backwashing strategy selection on the removal of SVOCs in drinking water treatment biofilters and reduction of health risks induced by the hazardous substance.

This study investigated the removal of 24 SVOCs in Yangtze River source water treated by six biofilters applying different backwashing strategies. The biofilters were continuously operated at Beihekou Tap Water treatment Plant (Nanjing, China) during April to November of 2009 and the water and GAC samples were collected and transported to Nanjing University (China) for chemical analyses and biodiversity investigations. The induced non-carcinogenic and carcinogenic risks were assessed for the SVOCs in influent and effluent water of the biofilters. This study might serve as a basis for regulation and optimization of biofiltration process and also as a benchmark for the authorities to control health risk induced by exposure to the hazardous pollutants in drinking water.

Materials and methods

Biofilters

For effective removal of SVOCs in source water, six biofilters, numbered from BF1 to BF6, were continuously run for more than 180 days at Beihekou Tap Water Plant. The main part of each biofilter was a transparent polyvinyl chloride column with an inner diameter of 10 cm and an effective height of 100 cm. Detailed information about configuration and size of the biofilters was previously described (Zhang et al. 2009). Each filter was packed with GAC (2.3 L) supported by sand (1.1 L). The biofilters were fed with effluent water from a pilot-scale settling tank in Beihekou Tap Water Plant. During the start-up period, the filters were continuously and stably operated for more than 30 days with influent hydraulic loading rate at 1.5 m/h. In each biofilter, dissolved oxygen was kept at 2.7–2.9 mg/L, and water temperature ranged from 22.5 to 26.9 °C. During this period, the six biofilters had a similar performance and showed no significant difference in the removal of chemical oxygen demand (COD_{Mn}), assimilable organic carbon (AOC), ammonia nitrogen (NH₃-N), total nitrogen (TN) and total phosphorus (TP). In order to investigate the effects

Table 1 Backwashing strategies used in this study (applying different air or water flow rates and durations)

Biofilter	Air backwashing		Water backwashing	
	Flow (m/h)	Duration (min)	Flow (m/h)	Duration (min)
BF1	15	3	–	–
BF2	35	5	–	–
BF3	–	–	8	5
BF4	–	–	15	8
BF5	15	3	8	5
BF6	35	5	15	8

of backwashing on biofilter performance and SVOCs removal, different backwashing strategies were applied in the six biofilters (Table 1). Air backwashing (in BF1 and BF2) and water backwashing (in BF3 and BF4) were used to prevent clog of the biofilters. A combined backwashing method using air followed by water was employed in BF5 and BF6. After the backwashing alterations, the treatment systems ran stably for more than 5 weeks.

Water quality assessment

During the experimental periods, water samples were collected every 24 h from both influent and effluent of the six biofilters for the measurement of COD_{Mn}, AOC, NH₃-N, TN and TP. COD_{Mn} was measured using the acid–potassium permanganate method (NEPAC 2002). NH₃-N, TN and TP were determined using the methods recommended by APHA (APHA 2005), and AOC was measured according to Li et al. (2004). GAC samples were also collected every 24 h at the sampling site of each column (30 cm above bed bottom) and biomass was measured according to Yu et al. (2002).

SVOC measurement

During the post-regulation period (5 weeks), influent and effluent water were sampled for SVOCs measurement at day 7, 14, 21, 28 and 35 (at the end of each backwashing interval). Water samples (10 L) were collected in brown glass bottles previously washed with detergent, followed by deionized water, 2 M nitric acid, deionized water again and finally influent or effluent water. The water samples were then transferred to lab in an ice bath within 3 h. The water samples were filtered onto cellulose ester filters (0.45-μm pore size) and kept at 4 °C until analyses of the 24 SVOCs listed in Table 2. Extraction of the SVOCs was conducted using the method recommended by EMC (2011). Guide DSQ II Single Quadrupole GC/MS (ThermoQuest, San Jose, USA) with selected ion monitoring mode was used to

Table 2 Components in source water fed to the biofilters

Component	Concentration	Component	Concentration
pH	7.8–8.0	Bis[2-ethylhexyl]phthalate (ng/L)	535–1,098
Ca (mg/L)	25.8–33.7	Butyl benzyl phthalate (ng/L)	10.3–20.8
Mg (mg/L)	4.7–6.2	Chrysene (ng/L)	7.2–9.7
Na (mg/L)	8.1–12.9	Dibenz[a,h]anthracene (ng/L)	8.6–17.0
K (mg/L)	2.2–3.4	Diethyl phthalate (ng/L)	7.0–25.9
COD _{Mn} (mg/L)	2.55–3.61	Dimethyl phthalate (ng/L)	55.3–150.9
AOC (mg/L)	0.38–0.62	Di- <i>n</i> -butyl phthalate (ng/L)	1,316–2,575
NH ₃ -N (mg/L)	0.15–0.28	2,6-Dinitrotoluene (ng/L)	201.3–541.1
TN (mg/L)	0.66–1.08	2,4-Dinitrotoluene (ng/L)	13.5–22.5
TP (mg/L)	0.10–0.18	Fluorene (ng/L)	9.1–18.0
Acenaphthylene (ng/L)	5.3–12.2	Hexachlorobenzene (ng/L)	3.0–5.7
Anthracene (ng/L)	26.2–46.1	Hexachlorocyclopentadiene (ng/L)	8.9–13.4
Benzo[a]anthracene (ng/L)	11.8–20.9	Indeno[1,2,3- <i>cd</i>]pyrene (ng/L)	5.9–17.4
Benzo[a]pyrene (ng/L)	7.4–14.2	Isophorone (ng/L)	5.2–15.6
Benzo[b]fluoranthene (ng/L)	9.6–19.9	Phenanthrene (ng/L)	12.6–30.8
Benzo[k]fluoranthene (ng/L)	12.8–19.1	Pentachlorophenol (ng/L)	9.8–18.8
Bis[2-ethylhexyl]adipate (ng/L)	102.0–199.4	Pyrene (ng/L)	7.8–11.2

determine the concentrations according to Wu et al. (2009b). Quality assurances were preformed according to Wu et al. (2009a, b). Detection limits of the compounds ranged from 0.4 to 2 ng/L (a signal-to-noise ratio of 3).

DNA extraction and PCR

For microbial community structure analysis, total DNA was extracted from each GAC sample collected from the six biofilters at the 35th day after the backwashing regulation. GAC was vortexed for 10 min to separate bacterial cells. The liquid phase (50 mL) was centrifuged and the pellet was used for DNA extraction with Fast Soil Genome DNA Isolation Kit (Biotake, China). Integrity of DNA was confirmed after electrophoresis on 0.8 % agarose gels by comparison with DNA ladder (λ HindIII, Takara, Japan). Purity of the DNA was spectrophotometrically determined by the ratio of absorbance at 260 and 280 nm. A 472 base-pair hypervariable region of the 16S rRNA gene was amplified by polymerase chain reaction (PCR). PCR primers and amplification conditions were previously described in detail (Zhang et al. 2010). Duplicate PCRs were carried out for each sample with sterile water as negative control, and PCR products were checked by electrophoresis on 1 % agarose gel with ethidium bromide in 1× TAE buffer at 120 V for 15 min.

DGGE and phylogenetic analysis

After 16S rRNA PCRs, denatured gradient gel electrophoresis (DGGE) was used to investigate microbial

diversity in the biofilters. The DGGE assay was performed using Bio-Rad DcodeTM Gene Mutation Detection System (Bio-Rad, USA) following the method of Zhang et al. (2010). After electrophoresis, the desired bands were cut from the DGGE gel and DNA was recovered using Poly-Gel DNA Extraction Kit (Omega Co., USA). PCR product of each DNA fragment was purified using PCRquick spinTM PCR Product Purification Kit (iNtRon Biotechnology Co., South Korea). The DNA fragments were cloned directly into pUCm-T vector using a TA cloning kit (Promega Co., USA). DNA sequencing was performed by automated means at Sangon (Shanghai, China) using M13 primer set. The 16S rRNA homology searches were conducted using the BLAST program at the National Centers for Biotechnology Information (National Institutes of Health, Bethesda, USA). Nucleotide sequences were aligned using the multiple sequence alignment program CLUSTAL W (Thompson et al. 1994). The GenBank sequences having the highest identity to those obtained in this study were retrieved for phylogenetic tree construction. Neighbor-joining trees were constructed using MEGA version 4.1 and bootstrap analysis with 1,000 replicates was performed to evaluate the significance of the nodes.

Health risk assessment

In order to further compare the performance of the biofilters, the induced health risks were assessed for the SVOCs in influent and effluent water. Health risk assessment was carried out on a basis of reliable exposure pathways of contaminants (USEPA 2011c). Exposure to the organic



pollutants in drinking water was considered mainly via the following pathways: (a) direct ingestion of water consumption, and (b) dermal absorption of contaminants in water adhered to exposed skin. Carcinogenic and non-carcinogenic risks were assessed based on the two exposure pathways in this study. The assessment included exposure dose determination (ingestion and dermal absorption pathway), hazard quotient calculation, non-carcinogenic hazard index (HI) and carcinogenic target risk (TR) calculations, and uncertainty analysis. The assessment procedures and methods, as well as the mathematical models, were previously described in detail (Wu et al. 2010).

Statistical analysis

Experimental results were statistically analyzed using SPSS 11.0 (SPSS Inc., Chicago, USA). The least significant differences test was used to measure the differences of each water quality parameter, SVOC level and health risk among the six biofilters. An analysis of variance (ANOVA) test was carried out to assess the homogeneity of variance and $p < 0.05$ was considered statistically significant.

Nucleotide sequence accession numbers

The sequences of 16S rRNA genes obtained in this study have been deposited in GenBank under accession No. HQ843996 to HQ844001.

Results and discussion

Pollutant removal performance of the six biofilters

Table 2 shows the concentrations of the components in the inlet water of the biofilters. During the start-up period (>30 days), the filters were operated continuously and stably with a water backwashing (20 m/h for 3 min) interval at 7 days. Within this period, operational performance showed no significant difference in the removal of COD_{Mn} ($80.3 \pm 1.0\%$), AOC ($88.1 \pm 1.2\%$), $\text{NH}_3\text{-N}$ ($66.6 \pm 0.8\%$), TN ($58.8 \pm 0.9\%$) and TP ($78.2 \pm 1.6\%$). At the end of this period, backwashing conditions were altered in each biofilter, except for BF3 (Table 1). After water backwashing, intensity was increased to 15 m/h for 8 min in BF4; each removal of COD_{Mn} , AOC, $\text{NH}_3\text{-N}$ and TN showed a significant decrease ($p < 0.05$) (Fig. 1a), revealing an evident effect of water backwashing intensity on biofilter performance.

Air backwashing also had a considerable influence on the removal of the pollutants. With weekly air backwashing at 15 m/h for 3 min, BF1 had high removal in COD_{Mn} , AOC, $\text{NH}_3\text{-N}$, TN and TP, demonstrating that appropriate

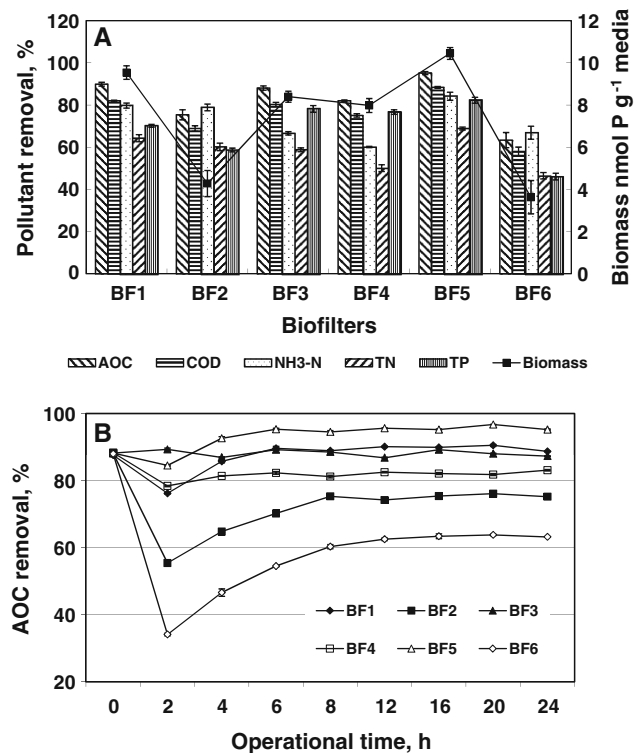


Fig. 1 Removal percentages of AOC (assimilable organic carbon), COD_{Mn} (chemical oxygen demand [Mn]), $\text{NH}_3\text{-N}$ (ammonia nitrogen), TN (total nitrogen) and TP (total phosphorus) in the six biofilters during the operation period (35 d) after the backwashing alterations

air backwashing conditions contribute to microbial activity improvement in the biofilters (Fig. 1a). However, too high backwashing air flow in BF2 resulted in the decrease in microbial biomass and the pollutant removal (each $p < 0.05$). A combined backwashing using both water and air was applied for BF5 and BF6. Among the six biofilters, BF5 had the highest removal in each of COD_{Mn} (88.4 %), AOC (95.2 %), $\text{NH}_3\text{-N}$ (84.2 %), TN (68.9 %) and TP (82.3 %) (each $p < 0.05$), indicating that the combined backwashing method is more effective in pollutant removal compared with separate use of air or water. However, BF6 showed a very poor performance in pollutant removal, since too high backwashing intensity in the biofilter resulted in the low biomass (3.6 nmol P/g media) (Fig. 1).

These results have shown that GAC biofilter can serve as a cost-effective and environmentally friendly process for advanced drinking water treatment, which is supported by previous studies applying the process to remove organic carbon (Tian et al. 2009) and nitrogen (Zhang et al. 2010) in source water. Compared with other biological processes used for drinking water treatment, biofiltration (especially using GAC as media) enhances substrate removal by providing a surface for indigenous water microorganisms to attach and grow (Moll et al. 1998). Previous studies showed that over

80 % of AOC could be removed in a pilot-scale GAC filtration process (Chien et al. 2006); while only 26.5 % of total organic carbon was removed by sand filtration in 30 conventional drinking water treatment plants (Kim 2009).

Combined use of air (15 m/h for 3 min) and water (8 m/h for 5 min) was considered as the optimal backwashing method in this study. GAC biofilters accumulate both biological and non-biological particles on the filter bed, and intermittent backwashing is necessary for the biofilters to prevent clog. Some previous studies investigated the effects of backwashing alterations on biofilter performance (Liu et al. 2001; Zheng and Long 2008). Liu et al. (2001) has indicated that chlorine in backwashing water affects biofilter performance and GAC filters are more resistance to chlorinated backwashing water than anthracite filters. Zheng and Long (2008) revealed that a long backwashing interval (>20 days) was practicable in an intermittent aerated biofilter developed for phosphorus removal in sewage. In this study, AOC removal (Fig. 1b) and microbial mass (Fig. 1c) in each biofilter were recovered within 24 h after backwashing, indicating that the backwashing intensities were acceptable for the biofilters. However, the backwashing alterations affected biomass increase and pollutant removal. AOC, COD and TP removal had positive correlations with biomass increase (each $p < 0.05$), but nitrogen removal showed no correlation to the bacterial growth ($p > 0.05$). Although hydrophobic bacteria are more strongly attached to the GAC surface than hydrophilic particles (Ahmad and Amirtharajah 1998), air or water backwashing with too high intensity can detach active cells from the media surface to cause biomass decrease and microbial community damage.

Effects of backwashing on removal of SVOCs

This study investigated the changes of concentrations of 24 individual SVOCs in the biofiltration systems. Total concentration of the SVOCs in the inlet of the biofilters was determined to be 2,391–4,923 ng/L. These 24 SVOCs are divided into three groups, i.e., 12 polycyclic aromatic hydrocarbons (PAHs), six phthalates (PAEs), and six benzene derivatives or heterocycle compounds (BDHCs). The total influent concentrations of PAHs, PAEs and BDHCs were about 180, 3,028 and 419 ng/L, respectively (Fig. 2). PAEs in Yangtze River source water had the similar level to those in Taiwan River (Yuan et al. 2002), while PAHs concentrations were lower than those in several of other Chinese rivers, including Minjiang River (Zhang et al. 2004), Qiantang River (Zhu et al. 2008) and Yellow River (Li et al. 2006). Among the 24 SVOCs, di-*n*-butyl phthalate (1,946 ng/L), bis(2-ethylhexyl)phthalate (817 ng/L) and 2,6-dinitrotoluene (371 ng/L) contributed most to the total concentration (Table 2).

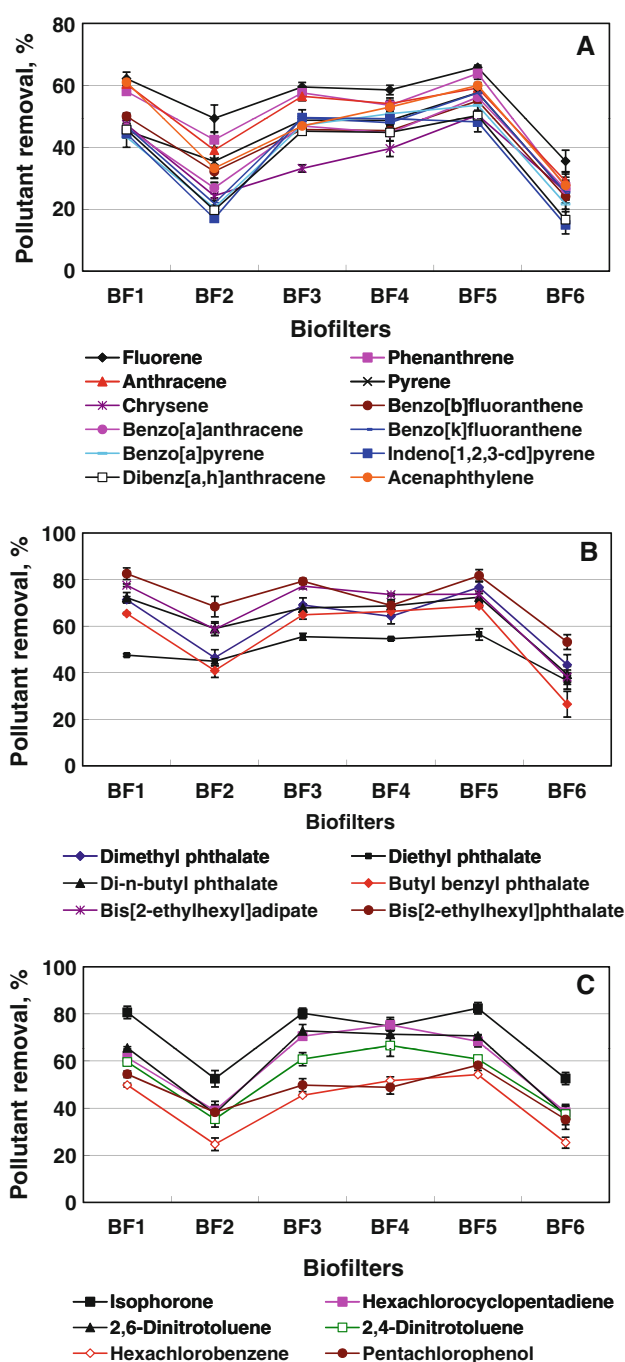


Fig. 2 Removal percentages of 24 semivolatile organic compounds, including 12 polycyclic aromatic hydrocarbons (a), six phthalates (b), and six benzene derivatives or heterocycle compounds (c), in the six biofilters applying different backwashing strategies

Backwashing alterations significantly influenced the removal of 24 SVOCs in the biofilters. Comparatively, BF5 had the highest removal in PAHs (56.4 %), PAEs (71.6 %) and BDHCs (65.8 %). BF2 and BF6 showed poor performance in SVOCs removal, indicating that high air flow hindered metabolism of the SVOCs by microorganisms in the biofilters (Fig. 2). However, BF3 and BF4 shared a



similar removal performance, demonstrating that the water backwashing alteration posed a relatively slighter effect.

This study showed that PAHs were more redundant to ready microbial utilization than the other two groups (Fig. 2a), since these chemicals are composed of fused aromatic rings whose biochemical persistence arises from dense clouds of π -electrons on both sides of the ring structures making them resistant to nucleophilic attack (Zhang et al. 2006). However, biofilter cultivated with functional microorganisms can remove over 70 % of benzo(a)pyrene in drinking water (Zhang et al. 2009). Among the PAHs, fluorene showed to be the most readily degraded (Fig. 2a). Comparatively, fluorene comprises fewer benzene rings and the carbon atom linking the two benzene rings in fluorene molecule is often attacked by the oxygen free radical to form 9-fluorenone under the catalysis of naphthalene 1,2-dioxygenase, with a result that the two rings are easily divided under subsequent oxidation reactions (Zhang et al. 2006).

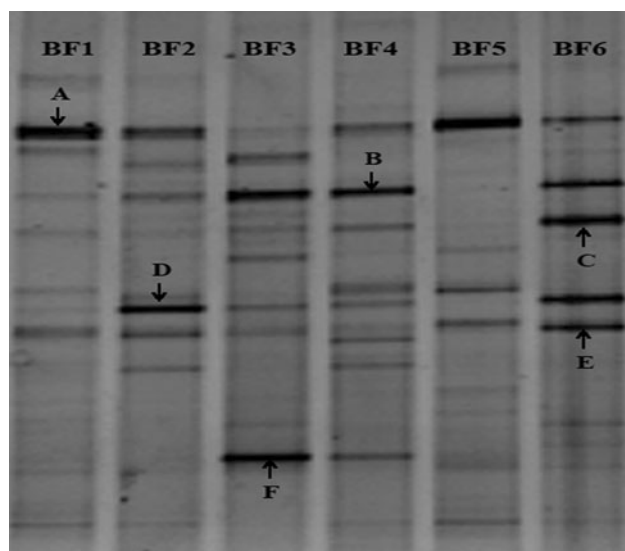
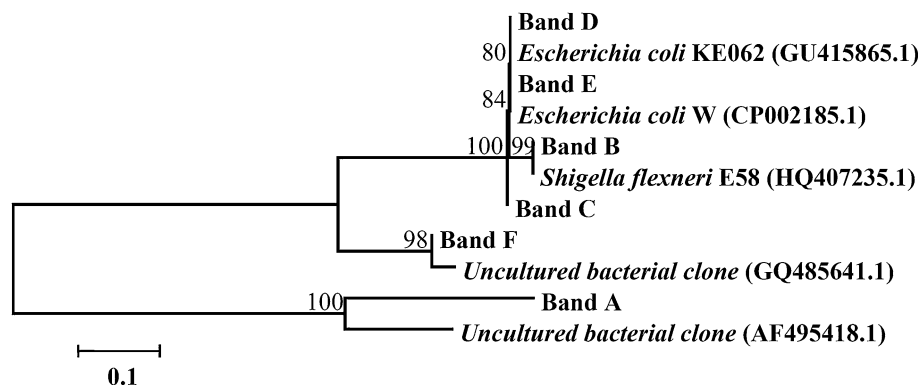


Fig. 3 Denaturing gradient gel electrophoresis (DGGE) analysis of PCR amplified 16S rDNA extracted from the media in the six biofilters (BF1–BF6)

Fig. 4 16S rDNA phylogenetic analysis of the dominant microbial species in the biofilters using the neighbor-joining method (MEGA version 4.1). Bootstrap analysis with 1,000 replicates was used to evaluate the significance of the nodes. GenBank accession numbers of the sequences are given in parenthesis



Among the PAEs, di-*n*-butyl phthalate and bis(2-ethyl-hexyl)phthalate, the two main SVOCs in the influent water, showed the highest removal of 72.4 and 81.6 % in BF5, respectively (Fig. 2b). Compared to PAHs and BDHCs, PAEs had relatively higher removal in the biofilters, because the linear structure of these molecules is more easily attacked by free radicals. Isophorone was metabolized more readily because a carbonyl group is located on the ring of the compound, which makes ring-opening to take place more easily (UM-BBD 2011). More than 70 % of 2,6-dinitrotoluene was removed in each of BF3, BF4 and BF5, but the removal efficiency was lower than 37 % in BF2 and BF6, further confirming the hypothesis that the biofilters are more sensitive to the air backwashing alteration. Information is limited about the removal of the SVOCs in biofilters, but it is known that biofilters are effective in removing volatile organic compounds (Lebrero et al. 2010; Rattanapan et al. 2010). Hassan and Sorial (2010) showed that the removal efficiency of benzene was 90 % in a controlled trickle bed air biofilter at a loading rate of 64 g/m³/h.

Effects of backwashing on microbial community structure

In this study, 16S rDNA PCR–DGGE was used to differentiate the microbial community structures in the six biofilters with different backwashing strategies. A total of 12 bands were excised from the DGGE gel, out of which six yielded unanimous sequences (Fig. 3). By BLAST search algorithm, high similarity values were observed after DNA sequencing, and these results were confirmed by phylogenetic analysis (Fig. 4). Band A had 99 % sequence similarity to an uncultured bacterial clone from activated sludge of wastewater treatment plants (GenBank accession No. AF495418.1) and band B was identified to be *Shigella flexneri*. Bands C, D and E were nicely grouped together and belonged to *Escherichia coli*. DNA sequencing showed that band F was closely related to an uncultured bacterial clone (GenBank accession No. GQ485641.1) previously

detected in drinking water treatment biofilters (Zhang et al. 2010).

Backwashing regulations posed obvious impacts on the microbial community structure in the biofilters. Band A was the dominant species in BF1 and BF2 (using air backwashing), while bands B and F appeared in the lanes of BF3 and BF4 (using water backwashing) and were very weak in the lanes of BF1 and BF2. *E. coli* represented by band D grew well in the biofilter with high air backwashing flow. The bacterial pattern in biofilter BF5 showed fewer intense bands, of which band A was sequenced and had a high identity of an uncultured bacterial strain from wastewater treatment plants (GenBank accession No. AF495418.1), suggesting that the species might be involved in the high pollutant removal efficiency in BF5. *Shigella* sp. (band B) was frequently detected in drinking water and is closely related to public health surveillance for various infectious diseases (Koutsotoli et al. 2006). *E. coli* is well known to have high detection frequency in drinking water (Juhna et al. 2007) and is capable of utilizing a variety of organic compounds including PAHs, phthalate, trinitrotoluene and heterocycle compounds (UM-BBD 2011). These dominant bacteria can secrete various biochemical enzymes with biodegradation function (Gonzalez-Perez et al. 2007), which might contribute to the removal of the SVOCs.

Previous studies showed that microbial community structures in biofilters could adapt to changing operational conditions, e.g., inlet loadings (Zhang et al. 2010) and substrates starvation (Moll et al. 1998). However, few investigations have been documented about the effect of backwashing on microbial community structure in biofilters. The alterations in air or water backwashing intensity control the biofilm formation, as well as microbial composition and activity in the biofilters (Liu et al. 2001). In turn, the microbial activity governs the substrate utilization performance of the biofilters (Moll et al. 1998).

Health risk induced by the SVOCs in influent and effluent

Non-carcinogenic and carcinogenic risk quotients of the SVOCs in the influent and effluent water were calculated to evaluate the potential health risk induced by these contaminants. Given that health risk analysis was performed only for the pollutants with available health guidelines, this study calculated the non-carcinogenic HI of 12 SVOCs (Table 3) and carcinogenic risk TR of other 12 SVOCs (Table 4). The SVOCs in the influent and effluent posed a slight adverse effect on public health because HI was lower than 1.0 (non-carcinogenic risk threshold) and TR was below 1.0×10^{-6} (carcinogenic risk threshold) for most of the individual SVOCs via

Table 3 Non-carcinogenic hazard index (HI) (via ingestion and dermal) of the SVOCs in influent and effluent water of the biofilters

Pollutant	Ingestion						Dermal					
	Influent			Effluent			Influent			Effluent		
	BF1	BF2	BF3	BF4	BF5	BF6	BF1	BF2	BF3	BF4	BF5	BF6
Anthracene	4.42E-03	1.75E-03	2.70E-03	1.93E-03	1.81E-03	3.14E-03	1.35E-03	8.25E-04	5.90E-04	6.22E-04	5.53E-04	9.61E-04
Bis[2-ethylhexyl]phthalate	1.50E+00	2.62E-01	4.73E-01	3.11E-01	2.75E-01	7.01E-01	4.58E-02	1.45E-02	9.51E-03	1.43E-02	8.41E-03	2.15E-02
Diethyl phthalate	7.55E-04	3.97E-04	4.17E-04	3.37E-04	3.29E-04	4.80E-04	4.62E-05	2.43E-05	2.06E-05	2.10E-05	2.02E-05	2.94E-05
Dimethyl phthalate	3.78E-02	1.08E-02	2.02E-02	1.17E-02	8.82E-03	2.14E-02	1.16E-02	3.32E-03	3.58E-03	4.14E-03	2.70E-03	6.55E-03
Di- <i>n</i> -butyl phthalate	7.14E-01	1.98E-01	2.93E-01	2.30E-01	1.97E-01	4.33E-01	1.09E-02	3.03E-03	3.52E-03	3.42E-03	3.01E-03	6.63E-03
2,4-Dinitrotoluene	3.30E-01	1.33E-01	2.13E-01	1.29E-01	1.29E-01	2.06E-01	3.37E-02	1.36E-02	1.32E-02	1.13E-02	1.32E-02	2.11E-02
2,6-Dinitrotoluene	1.36E+01	3.03E+00	5.49E+00	2.40E+00	2.58E+00	5.54E+00	1.39E+00	3.09E-01	2.45E-01	2.57E-01	2.64E-01	5.66E-01
Fluorene	1.24E-02	4.72E-03	6.32E-03	5.05E-03	4.27E-03	8.04E-03	1.52E-03	5.78E-04	6.18E-04	6.33E-04	5.22E-04	9.84E-04
Hexachlorobenzene	2.00E-01	9.20E-02	1.38E-01	1.00E-01	8.39E-02	1.37E-01	2.45E-02	1.13E-02	1.69E-02	1.09E-02	1.03E-02	1.68E-02
Hexachlorocyclopentadiene	6.79E-02	2.62E-02	4.14E-02	2.00E-02	2.15E-02	4.19E-02	4.16E-03	1.60E-03	1.22E-03	1.02E-03	1.31E-03	2.56E-03
Isophorone	1.91E-03	3.69E-04	9.06E-04	3.77E-04	3.35E-04	9.04E-04	2.92E-04	5.65E-05	1.39E-04	7.37E-05	5.13E-05	1.38E-04
Pentachlorophenol	1.05E-01	4.78E-02	6.48E-02	5.27E-02	4.38E-02	6.80E-02	3.20E-02	1.46E-02	1.98E-02	1.64E-02	1.34E-02	2.08E-02
Pyrene	1.16E-02	6.34E-03	7.49E-03	5.96E-03	4.92E-03	8.67E-03	2.67E-03	1.46E-03	1.37E-03	1.37E-03	1.13E-03	1.99E-03
Total HI	1.66E+01	3.82E+00	6.75E+00	3.27E+00	3.36E+00	7.17E+00	1.56E+00	3.67E-01	6.50E-01	3.22E-01	3.18E-01	6.66E-01



Table 4 Carcinogenic target risk (TR) (via ingestion and dermal) induced by the SVOCs in influent and effluent water of the biofilters

Pollutant	Ingestion						Dermal					
	Influent			Effluent			Influent			Effluent		
	BF1	BF2	BF3	BF4	BF5	BF6	BF1	BF2	BF3	BF4	BF5	BF6
Benzol[a]anthracene	4.38E-07	2.36E-07	2.32E-07	2.43E-07	1.92E-07	2.23E-07	8.38E-08	6.14E-08	4.46E-08	4.65E-08	3.68E-08	6.18E-08
Benzol[a]pyrene	2.88E-06	1.64E-06	1.53E-06	1.42E-06	1.34E-06	2.26E-06	4.41E-07	3.52E-07	2.34E-07	2.18E-07	2.06E-07	3.47E-07
Benzol[b]fluoranthene	3.94E-07	1.97E-07	2.14E-07	2.15E-07	1.77E-07	2.99E-07	6.04E-08	4.08E-08	3.27E-08	3.29E-08	2.71E-08	4.57E-08
Benzol[k]fluoranthene	4.27E-08	2.28E-08	3.35E-08	2.16E-08	1.82E-08	3.19E-08	3.27E-08	2.56E-08	1.66E-08	1.71E-08	1.39E-08	2.44E-08
Bis[2-ethylhexyl]phthalate	4.19E-07	7.34E-08	1.33E-07	1.31E-07	7.70E-07	1.96E-07	1.28E-08	4.06E-09	2.66E-09	3.99E-09	2.36E-09	6.01E-09
Chrysene	2.26E-09	1.18E-09	1.70E-09	1.36E-09	1.12E-09	1.67E-09	1.73E-10	1.30E-10	1.15E-10	1.04E-10	8.54E-11	1.28E-10
Dibenz[a,h]anthracene	3.43E-06	1.86E-06	2.75E-06	1.89E-06	1.71E-06	2.86E-06	5.25E-07	4.21E-07	2.87E-07	2.90E-07	2.61E-07	4.37E-07
2,4-Dinitrotoluene	2.05E-07	8.27E-08	1.32E-07	8.02E-08	8.02E-08	1.28E-07	2.09E-08	1.35E-08	8.19E-09	6.98E-09	8.18E-09	1.31E-08
Hexachlorobenzene	2.56E-07	1.18E-07	1.77E-07	1.13E-07	1.07E-07	1.75E-07	3.13E-08	2.16E-08	1.57E-08	1.39E-08	1.31E-08	2.14E-08
Indeno[1,2,3-c]pyrene	3.12E-07	1.74E-07	2.60E-07	1.59E-07	1.62E-07	2.67E-07	9.56E-08	7.95E-08	4.83E-08	4.87E-08	4.97E-08	8.16E-08
Isophorone	3.62E-10	7.01E-11	1.72E-10	9.14E-11	6.37E-11	1.72E-10	5.54E-11	2.63E-11	1.10E-11	1.40E-11	9.75E-12	2.63E-11
Pentachlorophenol	2.09E-07	9.56E-08	1.30E-07	1.07E-07	8.77E-08	1.36E-07	6.41E-08	3.96E-08	3.22E-08	3.29E-08	2.68E-08	4.16E-08
Total TR	8.59E-06	4.50E-06	6.51E-06	4.37E-06	3.95E-06	6.68E-06	1.37E-06	1.06E-06	7.23E-07	7.10E-07	6.45E-07	1.08E-06

ingestion or dermal exposure (Table 3). However, exposure to influent 2,6-dinitrotoluene or bis[2-ethylhexyl]phthalate via ingestion can induce hazardous risk since the HI values were more than 1.0. Dermal exposure to 2,6-dinitrotoluene might also cause damages to human health. High concentrations of the two chemicals in influent water may account for the higher non-carcinogenetic risks. Additionally, total HI and TR of the influent SVOCs were both higher than the threshold values, suggesting that the integrated risks induced by these chemicals deserve more concerns (Table 3).

Dibenz[a,h]anthracene and benzo[a]pyrene contributed most to the total carcinogenetic risk (Table 4), although concentrations of these two compounds were lower than most of the other SVOCs. Among PAHs, dibenz[a,h]anthracene and benzo[a]pyrene are well known to have the highest carcinogenetic risk (USEPA 2011b), and the carcinogenetic risk threshold is recommended as only 2.9 ng/L for both the chemicals in drinking water (USEPA 2011a). Wu et al. (2010) indicated that the two contaminants in Yangtze River source water could induce potential carcinogenic effects under a worst-case scenario.

Biofilter backwashing alterations greatly influenced the reduction of the health risk induced by SVOCs. Comparison of total HI or TR showed that effluent from BF3, BF4 and BF5 induced relatively lower health risks, which is in accordance with the chemical detections (each $p < 0.05$). Total dermal HI was evidently reduced to below 1.0 in each of the six biofilters, but the risk via ingestion was still more than the criteria for each filter effluent. Among the SVOCs, 2,6-dinitrotoluene contributed most to the total ingestion HI. The effluent water from BF1, BF3, BF4 and BF5 had no dermal carcinogenetic risk, but each can cause potential carcinogenetic risk via ingestion, which mainly arose from dibenz[a,h]anthracene and benzo[a]pyrene in the water. This study showed that too high air backwashing flow posed a negative effect on the reduction of health risk induced by the SVOCs, evidenced by the relatively less risk reduction in BF2 and BF6. Water backwashing, or combined backwashing with water and air at an appropriate flow, was relatively more efficient in minimizing the health risk.

Conclusion

This study revealed that backwashing significantly influenced biomass and microbial community structure and subsequently affected SVOCs removal and health risk reduction in biofilters. Compared with separate use, combined backwashing with air and water at an appropriate intensity is more efficient in both pollutant removal and risk reduction. This study is expected to provide some



useful baseline information for drinking water biofiltration process optimization. Future studies may include in-depth and comprehensive investigations of the optimal operational conditions in biofilters, so as to remove hazardous pollutants and reduce potential public health risk in a more efficient manner.

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