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**DOI**
10.1016/j.scitotenv.2022.155936

**Publication date**
2022

**Document Version**
Final published version

**Published in**
Science of the Total Environment

Science of the Total Environment 838 (2022) 155936

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

# Effects of biological activated carbon filter running time on disinfection by-product precursor removal

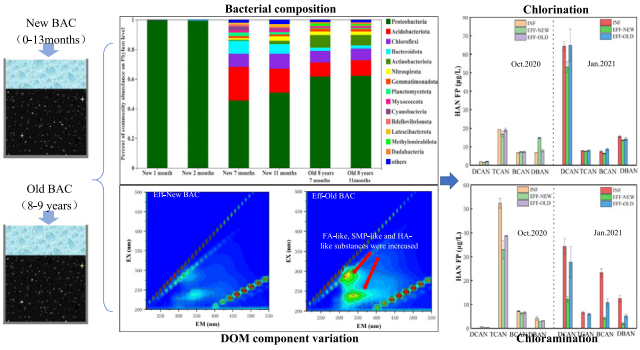
Feifei Wang [a], Jiazheng Pan [a,*], Yulin Hu [a], Jie Zhou [a], Haoqian Wang [a], Xin Huang [a], Wenhai Chu [b,*], Jan Peter van der Hoek [c,d]

[a] School of Environmental and Chemical Engineering, Shanghai University, 200444 Shanghai, PR China
[b] State Key Laboratory of Pollution Control and Resources Reuse, College of Environmental Science and Engineering, Tongji University, 200092 Shanghai, PR China
[c] Department of Water Management, Delft University of Technology, 2628 CN Delft, the Netherlands
[d] Research & Innovation Program, Waternet, 1096 AC Amsterdam, the Netherlands

## HIGHLIGHTS

- New BAC lowered DOM components while old BAC increased FA-, SMP- and HA-like substances.
- Formation potentials of C-DBPs ($THM_4$ and $HAA_7$) were lowered by both old and new BAC.
- Old BAC poorly reduced N-DBPs FPs, and even increased chlorinated BCAN and DCAN FPs.
- *Herminiimonas* may explain better DOC and $UV_{254}$ removal performance and lower DBP FPs.
- *Bradyrhizobium* might produce more EPS and result into increase of N-DBP FPs in old BAC filter.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

Editor: Qilin Wang

*Keywords:*
Biological activated carbon
Chlorination
Chloramination
Disinfection by-products
Formation potentials
High-throughput sequencing

## ABSTRACT

Biological activated carbon (BAC) filtration is usually considered to be able to decrease formation potentials (FPs) of disinfection by-products (DBPs) in drinking water treatment plant (DWTP). However, BAC filters with long running time may release microbial metabolites to effluents and therefore increase FPs of nitrogenous DBPs with high toxicity. To verify this hypothesis, this study continuously tracked BAC filters in a DWTP for one year, and assessed effects of old (running time 8–9 years) and new (running time 0–13 months) BAC filters on FPs of 15 regulated and unregulated DBPs. Results revealed that dissolved organic carbon (DOC) removal was slightly higher in the new BAC than the old one. All fluorescent components of dissolved organic matter evidently declined after new BAC filtration, but fulvic acid-like and soluble microbial product-like substances increased after old BAC filtration, which could be caused by microbial leakage. Correspondingly, new BAC filter generally removed more DBP FPs than the old one. 46.5% $HAA_7$ FPs from chlorination and 44.3% $THM_4$ FPs from chloramination were removed by new BAC filter. However, some DBP FPs, especially HAN FPs, were poorly removed or even increased by the old BAC filter. *Proteobacteria* could be a main contributor for DBP precursor removal in BAC filters. *Herminiimonas*, most abundant genera in new BAC filter, may explain its better DOC and $UV_{254}$ removal performance and lower DBP FPs, while *Bradyrhizobium*, most abundant genera in old BAC filter, might produce more extracellular polymeric substances and therefore increased N-DBP FPs in old BAC effluent. This study provided insight into variations of DBP FPs and microbial communities in the new and old BAC filters, and will be helpful for the optimization of DWTP design and operation for public health.

* Corresponding authors.
*E-mail addresses:* pjz19@shu.edu.cn (J. Pan), 1world1water@tongji.edu.cn (W. Chu).

http://dx.doi.org/10.1016/j.scitotenv.2022.155936
Received 9 March 2022; Received in revised form 25 April 2022; Accepted 10 May 2022
Available online 14 May 2022

## 1. Introduction

The disinfection of drinking water can effectively inactivate pathogenic microorganisms and eliminate water-borne diseases. It is considered to be one of the greatest achievements of public health in the 20th century (Li and Mitch, 2018). Chlorine and chloramine are two main chemical disinfectants used in drinking water treatment plants (DWTPs) to meet the microbial drinking water quality standards (Williams et al., 2005; Zhang et al., 2000). However, these disinfectants can react with dissolved organic matter (DOM) to produce disinfection by-products (DBPs) which are potentially carcinogenic and genotoxic to humans (Allard et al., 2015). So far, more than 700 DBPs have been recognized since trihalomethanes (THMs) were first discovered in 1970s, including haloacetic acids (HAAs) and haloacetonitriles (HANs) (Chen and Westerhoff, 2010; Richardson et al., 2007; Rook, 1974). Compared with chlorine disinfectant, chloramine disinfectant reduces the formation of regulated carbonaceous disinfection by-products (C-DBPs) and total organic halogens (Cuthbertson et al., 2020; Hua and Reckhow, 2007), but increases the formation of emerging unregulated nitrogenous DBPs (N-DBPs) (Chu et al., 2013; Cuthbertson et al., 2020; Plewa et al., 2004). HANs are typically nitrogenous DBPs with lower concentrations and greater cytotoxicity and genotoxicity than THMs and HAAs (Muellner et al., 2007; Plewa et al., 2008). And HANs and other emerging DBPs is still contribute, in many cases, the majority of the DBP-related toxicity risk in drinking water (Chuang et al., 2019; Cuthbertson et al., 2019; Komaki et al., 2014; Krasner et al., 2016).

Biological activated carbon (BAC) filters have been applied widely followed by chlorine/chloramine disinfection in DWTPs (Lu et al., 2021; Wan et al., 2021). BAC can remove DOM by adsorption and biodegradation. In the initial operation time, the removal rate of dissolved organic carbon (DOC) reached 40%–90% mainly by BAC adsorption (Dussert and Stone, 1994; Velten et al., 2011; Xing et al., 2008). Kim and Kang (2008 reported that over 99% $HAA_5$ formation potential (FP) and over 30% THM FP was removed by BAC filters in the first three months. Additionally, BAC can also decrease some N-DBP FPs. For instance, Krasner et al. (2012) found that 88 ± 6% ($n$ = 7) *N*-nitrosodimethylamine FP (NDMA FP), 39.6% dichloroacetonitrile (DCAN) FP, 40.2% dichloroacetamide (DCAcAm) FP, and 68.8% trichloronitromethane (TCNM) FP were removed by new BAC, respectively (Zheng et al., 2018). Kim and Kang (2008) reported that the removal efficiency of $HAA_5$ FP was increased to more than 99% after 6 months of operation and it may be attributed to biodegradation of DOM in BAC filters. Therefore, BAC filters are usually considered to be able to remove C-DBP and N-DBP FPs in the following disinfection process.

However, does a BAC filter always make chlor(*am*)inated water safer? Actually, in the past years, a few studies reported that some DBP FPs, especially the emerging unregulated DBPs with higher toxicity, increased due to BAC filtration (Cuthbertson et al., 2019; Zheng et al., 2018). For instance, Cuthbertson et al. (2020) reported that bromodichloronitromethane (BDCNM) increased from 800 to 1000 ng/L and dichloronitromethane (DCNM) increased from 200 to 300 ng/L after BAC filtration, and reported that dichloroacetonitrile precursors (>10 k Da) increased from 2.8 μg/L to 4.1 μg/L after BAC filtration. However, till now, the risk of DBP FPs increase caused by BAC filtration has not studied further, and the mechanisms behind the phenomenon were provided neither. To the authors' knowledge, a potential drawback of BAC, especially BAC with long running time, could be that the biofilm attaching on the activated carbon granules may be released into the effluent (Krasner et al., 2013; Simpson, 2008). The biofilm contains microbes, soluble microbial products (SMPs) and extracellular polymeric substances (EPS), which are considered as precursors of DBPs, especially N-DBPs (Cuthbertson et al., 2020; Zheng et al., 2018). Usually, the potentially increased N-DBPs is much more toxic than C-DBPs (Richardson et al., 2007). Therefore, the effect of BAC filters with a long running time on DBP FPs should be paid attention to, which is also a new crucial factor to be considered for the determination of activated carbon replacement cycle in DWTPs.

The biofilm attaching on BAC can biodegrade algae metabolites and DOM (Simpson, 2008). Previous studies reported that *Proteobacteria* was the dominant phylum, and *Sphingomonas*, *Bacillus*, *Hydrogenophaga* and *Pedomicrobium* accounted for 16%, 12.68%, 7.84%, and 7.32% at the genus level in up-flow BAC filter (Chen et al., 2019; Zheng et al., 2018). Some other studies further reported that Alphaproteobacteria was predominant in the BAC column filters, which might contribute to DOC and assimilable organic carbon removal (Liao et al., 2013; Zhang et al., 2019b) Till now, however, little has been known regarding the role of microorganisms in BAC filters on the removal of DBPs precursors. Investigation on microbial community evolution from new BAC to old BAC filter and their performance differences in DBP precursor removal is helpful to reveal the mechanism behind the risk of DBP formation caused by BAC filtration.

In this study, two BAC filters with different running times in a full scale DWTP located in the Yangtze River basin were selected as research objects and were continuously tracked for one year. The objectives of this study were to 1) assess the effect of old and new BAC filters on the formation potentials of 15 regulated and unregulated DBPs during the following chlorination/chloramination, and 2) explore the mechanism behind DBP FPs changes after BAC filtration, based on characteristic changes of effluent DOM as DBP precursor and microbial community structure differences between new and old BAC filters.

## 2. Methods and materials

### 2.1. Chemicals

DBPs standard solutions including $THM_4$ (trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM) and tribromomethane (TBM)), $HAA_7$ (dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), Bromochloroacetic acid (BCAA), dibromoacetic acid (DBAA), bromodichloroacetic acid (BDCAA), chlorodibromoacetic acid (CDBAA), and tribromoacetic acid (TBAA)) and $HAN_4$ (trichloroactetonitrile (TCAN), dichloroacetonitrile (DCAN), bromochloroacetonitrile (BCAN) and dibromoacetonitrile (DBAN)) as well as Methyl tert-butyl ether (MTBE, high performance liquid chromatography grade) were purchased from J&K Scientific Co., Ltd. (Shanghai, China). Sodium hypochlorite (NaOCl), ammonium chloride ($NH_4Cl$) and all other chemicals were obtained from Sinopharm Chemical Reagent Co. (Shanghai, China). All solutions were prepared with ultrapure water produced from a Milli-Q Gradient water purification system (18 MΩ·cm, Billerica, MA, USA). All the chemicals used in this study were of analytical grade.

### 2.2. Sampling

It has been reported that the activated carbon in many BAC filters can be used for 8–10 years (Andersson et al., 2001; Dong et al., 2021; Gerrity et al., 2018; Hu et al., 2019) . Some previous studies defined BAC filters into three phases (young, middle and old BAC filters) based on running time (Cuthbertson et al., 2019). In this study, the authors selected BAC filters with running time of 0–13 months and 8–9 years to represent new and old BAC filters. The old BAC filter has a potential risk in terms of N-DBP FPs, and therefore should be paid attention to. Since the water matrix could impact DBP FP removal efficiency by BAC, the new BAC filter in this study was considered as a reference to estimate old BAC performance.

To compare the differences in DBP FP removal as well as microbial diversity between old and new BAC filters, the water samples and BAC samples were collected from two BAC filters with different running times (0–13 months (the new BAC) and 8–9 years (the old BAC)) for seven times in November and December 2019, January, June, September and October 2020 as well as January 2021 for laboratory analysis. The samples originated from a DWTP with the source water from Taihu Lake, located in the Yangtze River basin, China. The main treatment units in this DWTP include pre-ozonation, coagulation, sedimentation, sand filtration, ozonation with a dosage of 0.4–0.5 mg $O_3$/L, BAC filtration and chlorine disinfection. After the ozonation, the effluent is subsequently led through parallel BAC

filters. In this study, the new BAC filter was filled with fresh activated carbon and the old filter had been running for approximately 8 years. More details of the raw water quality BAC filter can be found in Table S2 and Table S3. To avoid the interference of backwash, influent and effluent water samples as well as BAC samples were collected before regular backwash of every 6 days. Once collected, water samples were filtered through 0.45 μm filter membranes and were saved in the dark at 4 °C for less than 3 days before DBP FP tests. BAC samples were collected at 2–5 cm from the top of the two BAC filters bed and stored at −80 °C before DNA extraction. Process flow diagrams and sampling sites can be found in Fig. S1.

### 2.3. Chlorination and chloramination disinfection

DBP FP test, a well-established tool, has been widely used in previous studies to evaluate the total quantity of DBP precursors (He et al., 2020; Liu et al., 2016). In order to identify the change of DBP FPs caused by the old and the new BAC filters, DBP FP tests were conducted for the influent and the effluent waters. DBP FP tests were performed in triplicate in 40 mL amber glass volumetric bottles under headspace-free conditions in the dark at controlled room temperature (25.0 ± 0.5 °C). The detailed procedures can be found in previous studies (Chu et al., 2011b; Krasner et al., 2006). Briefly, NaClO stock solution with a concentration of 125 mg $Cl_2$/L was prepared as the chlorine disinfectant. Fresh monochloramine ($NH_2Cl$) solution was prepared in advance following the procedures of Mitch (Mitch and Sedlak, 2002). In DBP FP tests, the disinfection time of chlorine and chloramine were 24 h and 72 h, respectively. The chlorine and the chloramine demand were calculated according to Eqs. (1) and (2) respectively (Chu et al., 2011b; Krasner et al., 2006). The disinfectant dosages for influent and effluent were determined based on the actual DOC and $NH_4^+$-N concentration occurred in the influent and the effluent respectively. The additional chlorine dose of 10 mg/L was to guarantee sufficient chlorine during chlorination (Krasner et al., 2007). This chlorine dosage was also used in previous studies (Chu et al., 2013; Chu et al., 2010).

$$Cl_2 \text{ dosage} = 3 \times \text{DOC mg/L} + 7.6 \times NH_4{}^+ - \text{N mg/L} + 10 \text{ mg/L} \quad (1)$$

$$NH_2Cl \text{ dosage} = 3 \times \text{DOC mg/L} \quad (2)$$

Water samples were collected from the bottles at the end of the experiment. Immediately, DBPs were separated and extracted by MTBE via liquid-liquid extraction (LLE). Then, the extracted DBP samples were saved in the freezer for less than 24 h for the subsequent DBP analysis. Detailed information about the extraction procedure of DBPs is available in supplementary material.

### 2.4. Analytical methods

THMs, HANs and HAAs were measured using a gas chromatograph equipped with an electron capture detector (GC/ECD) (Clarus680, PerkinElmer, USA) based on the United States Environmental Protection Agency method 551.1 and 552.2. Other detailed information about the analytical method used for DBP analysis is available in Table S1.

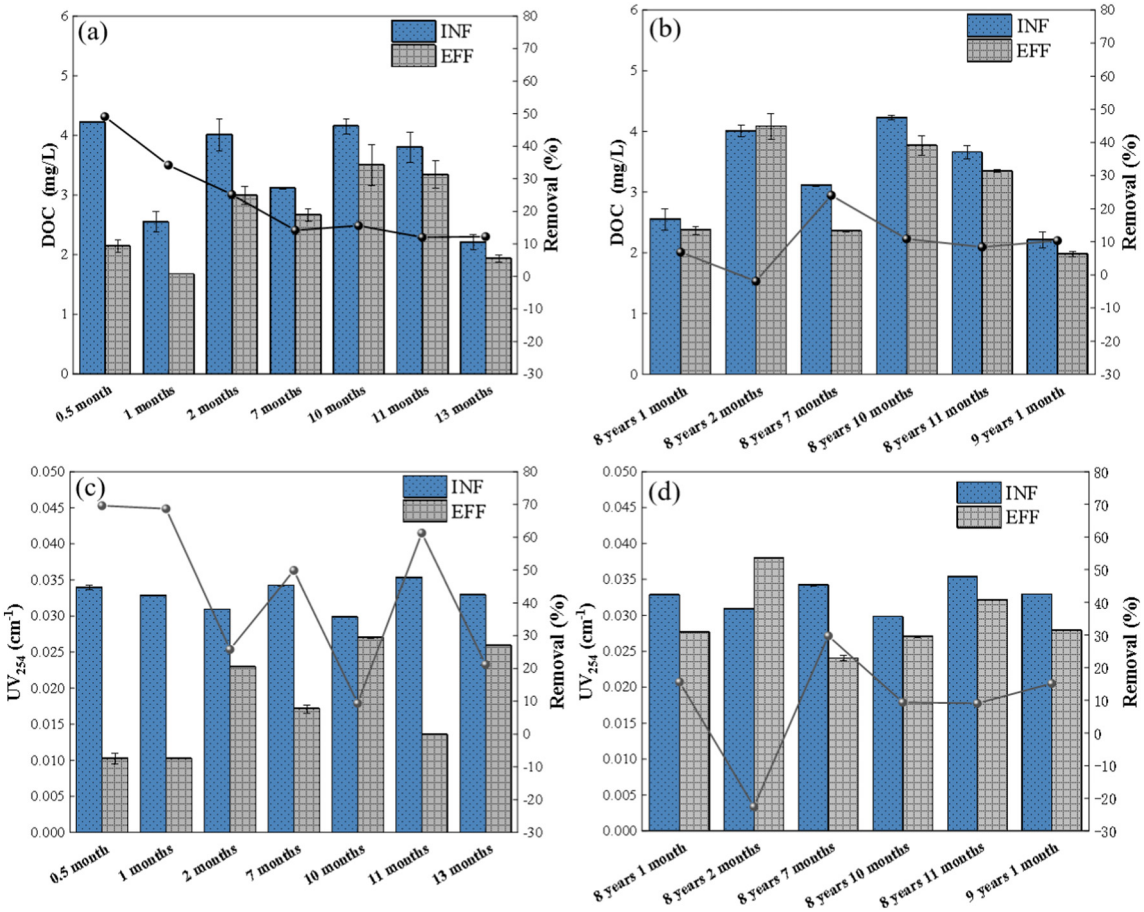



**Fig. 1.** Removal of DOC and $UV_{254}$ by the new BAC (a, c) and the old BAC (b, d). The column stands for the concentration of DOC or $UV_{254}$. The line represents the removal percentage of DOC or $UV_{254}$.

DOC in the influent and effluent of the BAC filters were measured by total organic carbon analyser (Jena multi C/N 2100, German) after a prefiltration through a 0.45 μm membrane. UV absorbance at 254 nm (UV254) was detected by a spectrometer (UV752, Shanghai Jingke Co. Ltd., China). The chlorine and chloramine concentrations were measured with a portable photometer (HACH Pocket Colorimeter TM, USA). ATP was used to characterize biological activity and biomass of the biofilm attached to GAC (Velten et al., 2011).Fluorescence spectrophotometer (HITACHI F-7000, Japan) was applied to analyze the variations of DOM in BAC filter influent and effluent samples. Milli-Q water was used as the blank spectra to neutralize the influence of Rayleigh and Raman scattering when the fluorescence regional integration (FRI) was calculated

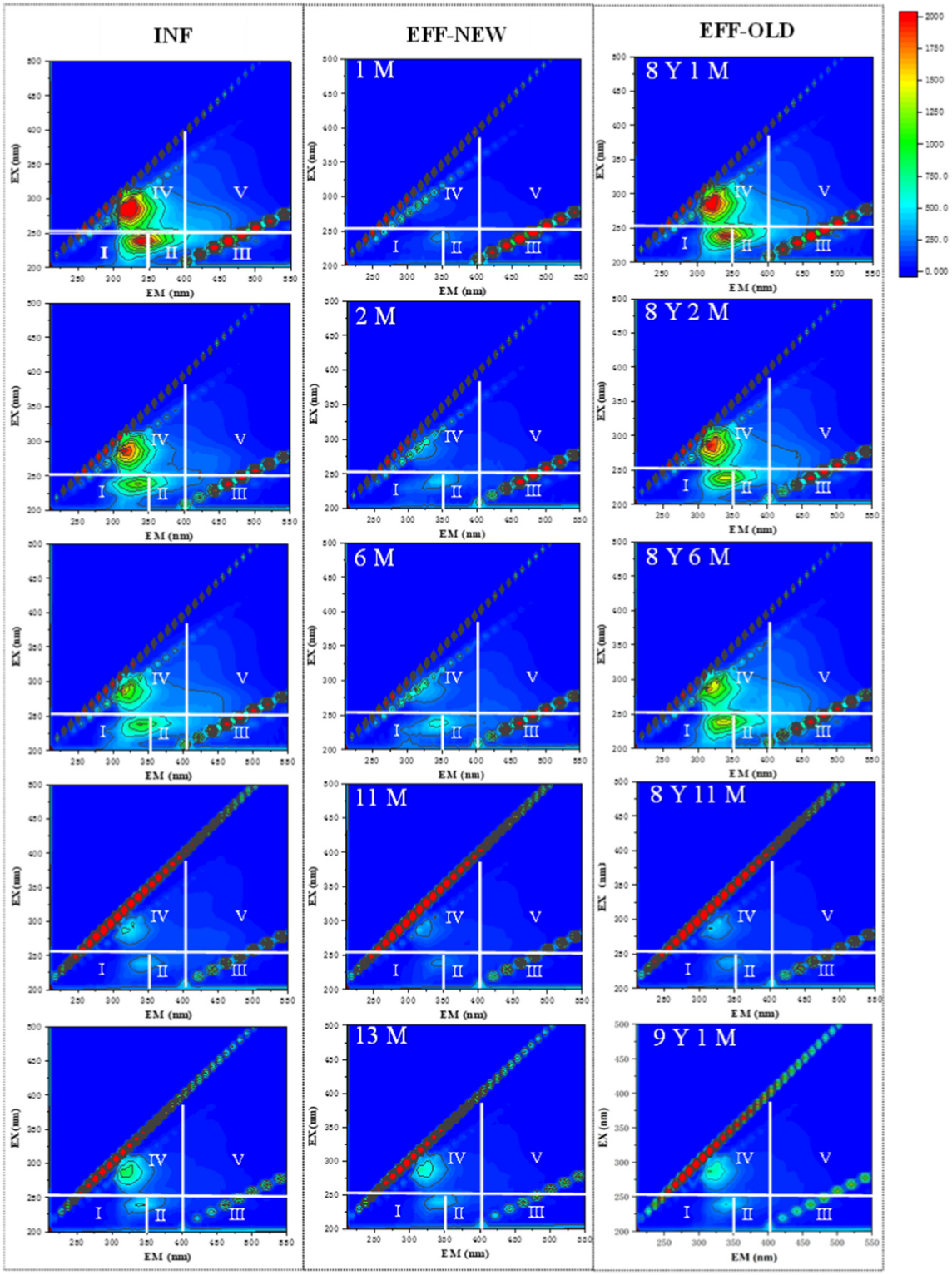



**Fig. 2.** Influent and effluent EEM spectra of the new BAC and the old BAC at different running time. Regions I-V represent aromatic protein-like type I, aromatic protein-like type II, fulvic acid-like, soluble microbial product-like, and humic acid-like substances, respectively. M and Y refer to month(s) and years, respectively.

(Chen et al., 2003). And all the BAC samples were examined by SEM (Sigma500, Zeiss, Germany) with accelerating voltage of 3 kV. All detailed detection and analytical methods were summarized in text 2 of supplementary material.

### 2.5. Microbial analysis

#### 2.5.1. DNA extraction, PCR amplification

DNA of the biofilm attached to GAC was extracted by Fast DNA® SPIN Kit for Soil (MP, USA). After DNA extraction, the concentration and purity of DNA were detected by Nano Drop 2000, and the integrity of DNA was detected by 1% agarose gel electrophoresis (voltage 5 V/cm, 20 min). The DNA samples, stored in a dry ice box, were sent to Majorbio Bio-pharm Technology Co., Ltd. (Shanghai, China) to determine the microbial 16S rRNA diversity.

The PCR amplification program was performed on a PCR instrument (ABIGeneAmp® 9700), and the PCR amplification was performed using primers 338F (ACTCCTACGGGAGGCAGCAG) and 806R (GAC TACHVGGTWTCTAAT). The PCR reaction system was 20 μL, including 4 μL fast pfu buffer, 2 μL dNTPs, 0.8 μL forward primer (5 μM), 0.8 μL Reverse Primer (5 μM), 0.4 μL Fast Pfu Polymerase, 0.2 μL BSA and 10 ng template DNA. The PCR reaction parameters were as follows: the pre-denaturation temperature was maintained at 95 °C for 3 min, and 27 cycles were performed (denaturation at 95 °C for 30 s, annealing at 55 °C for 30 s, extension at 72 °C for 45 s) and final extension at 72 °C for 10 min.

#### 2.5.2. High-throughput sequencing

All DNA date analysis was based on the second generation high-throughput sequencing platform Illumina Miseq PE300, the library was established by secondary PCR amplification. The sequencing results of each sample were spliced and quality controlled through the Meiji Biocloud platform, and the data were optimized. Then, the silva database was compared by operational taxonomic unit (OTU) clustering grouping.

## 3. Results and discussion

### 3.1. Removal of DOC and $UV_{254}$ by BAC filtration

To compare the difference in water quality purification between new and old BAC filters, Fig. 1 shows DOC and $UV_{254}$ values in the influent and effluent of these two BAC filters. The DOC and the $UV_{254}$ in the influent of new and old BAC filters were 2.4–4.2 mg/L and 0.031–0.035 $cm^{-1}$, respectively. As is shown in Fig. 1(a), the removal of DOC and $UV_{254}$ both exceeded 20% by the new BAC with running time of 2 months, mainly because of GAC adsorption (Korotta-Gamage and Sathasivan, 2017). After 7 months running, the removal of DOC and $UV_{254}$ by the new BAC were stable at around 15% and 20%, respectively. However, as shown in Fig. 1 (b), the old BAC filter showed relatively low DOC and $UV_{254}$ removal efficiencies, indicating that the new BAC filter could remove more DBP precursors than the old one.

### 3.2. EEM spectroscopic property after BAC filtration

To further identify which fractions of DOM as DBP precursors increased due to old BAC filtration, Fig. 2 shows the DOM EEM spectra of influent and effluent waters collected 5 times for 1 year from the new and the old BAC filters. Two peaks in region I and II and region IV appeared on the spectrum of BAC influent and effluent. This indicated the domination of SMP-like and AP-like substances in BAC influent and effluent DOM. The fluorescence intensities (FI) of SMP-like peak and AP-like peak was evidently weaker after the new BAC filtration, indicating that the new BAC filter acted as a sink of DOM. In contrast, the two peaks were hardly weakened, but instead enhanced after the old BAC filtration, indicating that the old BAC may acted as a source of DOM and leaked SMP-like and AP-like substance. Previous study has showed that SMP-like substances were the significant precursors of HANs (Chu et al., 2010). The intensities of two peaks were increased by old BAC filters. It was shown that old BAC process may leaked SMP-like and AP-like substance to increase the precursors of HANs.

FRI is a quantitative technique integrating the five regions volume of EEM spectra (Chen et al., 2003), to describe the content of each DOM fraction in water samples. The distribution of FRI in DOM was calculated according to the method described in a previous study (Chen et al., 2003) and the results are shown in Table 1. After new BAC filtration, $\Phi_{T,n}$values of I-V in the effluent were all observed to decline. With the new BAC filter, the removal of fluorescent component decreased. The highest removals of region I-V were 55.2%, 81.7%, 31.1%, 73.4% and 62.2% at running time of 0.5 months, respectively. Additionally, Zheng et al. (2018) also reported that 29.7% of AP-like and 36.9% of SMP-like substances were removed by a new BAC filter in a DWTP. These results further proved that new BAC filter is a sink of DBP precursors. However, $\Phi_{T,n}$values related to region III, IV and V increased in various degrees frequently after old BAC filtration (Table 1). On the average, 5.5% FA-like substance, 2.7% SMP-like substance and 1.8% HA-like substance increased in the effluent of the old BAC filter with a running time of 8–9 years. The increased $\Phi_{T,n}$values of region III indicated that more FA-like substances were leaked into the effluent, and Zhang et al. (2020) found that high concentrations of regulated carbonaceous DBPs (C-DBPs) were well correlated to the fluorescence intensities of FA-like and HA-like components. It indicated that the old BAC filtration may increase C-DBP FPs. The $\Phi_{T,n}$value of region IV was increased, indicating an increase of SMP-like substances in the effluent

**Table 1**
Distribution of FRI in DOM of the influents and effluents from BAC filtration.

| Operation time | Region I (aromatic protein I) | | | Region II (aromatic protein II) | | | Region III (fulvic acid–like) | | | Region IV (soluble microbial byproduct–like) | | | Region V (humic acid-like) | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $\Phi_{T,n}$ ($\times 10^6$ AU·nm$^2$) | | RE (%) | $\Phi_{T,n}$ ($\times 10^6$ AU·nm$^2$) | | RE (%) | $\Phi_{T,n}$ ($\times 10^6$ AU·nm$^2$) | | RE (%) | $\Phi_{T,n}$ $\times 10^6$ AU·nm$^2$) | | RE (%) | $\Phi_{T,n}$ ($\times 10^6$ AU·nm$^2$) | | RE (%) |
| | INF | EFF | | INF | EFF | | INF | EFF | | INF | EFF | | INF | EFF | |
| 0.5 M | 5.750 | 2.574 | 55.2 | 5.025 | 0.920 | 81.7 | 0.678 | 0.467 | 31.1 | 1.239 | 0.330 | 73.4 | 0.090 | 0.037 | 62.2 |
| 1 M | 0.897 | 0.141 | 84.2 | 1.235 | 0.206 | 83.3 | 0.195 | 0.173 | 11.4 | 0.312 | 0.116 | 62.8 | 0.022 | 0.012 | 45.3 |
| 2 M | 0.773 | 0.252 | 67.4 | 0.652 | 0.272 | 58.2 | 0.120 | 0.112 | 6.9 | 0.243 | 0.121 | 50.4 | 0.016 | 0.012 | 25.5 |
| 7 M | 0.581 | 0.346 | 40.4 | 0.659 | 0.398 | 39.5 | 0.144 | 0.118 | 17.9 | 0.212 | 0.135 | 36.2 | 0.021 | 0.014 | 30.4 |
| 11 M | 0.195 | 0.136 | 29.8 | 0.274 | 0.197 | 27.9 | 0.074 | 0.067 | 9.3 | 0.352 | 0.342 | 2.8 | 0.016 | 0.013 | 17.6 |
| 13 M | 0.228 | 0.199 | 12.5 | 0.299 | 0.260 | 13.3 | 0.046 | 0.046 | 1.6 | 0.2356 | 0.221 | 6.2 | 0.009 | 0.009 | 7.8 |
| 8 Y 1 M | 0.896 | 0.797 | 11.1 | 1.235 | 1.072 | 13.2 | 0.195 | 0.200 | −2.6 | 0.312 | 0.281 | 9.8 | 0.022 | 0.021 | 5.3 |
| 8 Y 2 M | 0.773 | 0.663 | 14.1 | 0.652 | 0.752 | −15.3 | 0.120 | 0.134 | −11.3 | 0.243 | 0.261 | −7.4 | 0.016 | 0.019 | −17.8 |
| 8 Y 7 M | 0.580 | 0.758 | −30.6 | 0.659 | 0.846 | −28.5 | 0.144 | 0.160 | −11.2 | 0.212 | 0.212 | −0.3 | 0.021 | 0.022 | −4.1 |
| 8 Y 11 M | 0.194 | 0.184 | 5.4 | 0.274 | 0.238 | 13.2 | 0.074 | 0.068 | 7.7 | 0.352 | 0.370 | −5.1 | 0.016 | 0.014 | 12.7 |
| 9 Y 1 M | 0.228 | 0.218 | 4.3 | 0.299 | 0.280 | 6.6 | 0.046 | 0.051 | −9.9 | 0.235 | 0.260 | −10.4 | 0.009 | 0.010 | −5.3 |

M means month (s), Y refers to years and RE is removal efficiency.

of the old BAC filter, and SMP-like substances have been proved to be important precursors of DBPs (Chu et al., 2011a). Therefore, may be more DBPs precursors were released into the old BAC effluent, resulting in high health risk.

### 3.3. Removal of $THM_4$ FPs, $HAA_7$ FPs and $HAN_4$ FPs after BAC filtration

The formation potentials of $THM_4$, $HAA_7$ and $HAN_4$ during the chlor(*am*)ination of influent and effluent waters from the two BAC filters are presented in Fig. 3. As is shown in Fig. 3(a), (b) and (c), in October, the concentrations of $THM_4$ FPs and $HAN_4$ FPs decreased from 32.9 ± 1.2 μg/L and 23.5 ± 6.2 μg/L in the influent to 25.2 ± 0.8 μg/L and 40.4 ± 1.6 μg/L in new BAC effluent, and 27.2 ± 1.3 μg/L and 35.5 ± 0.5 μg/L in old BAC effluent during chlorination. And the concentrations of $THM_4$ FPs, $HAA_7$ FPs and $HAN_4$ FPs decreased from 80.3 ± 9.4 μg/L, 420.1 ± 23.8 μg/L and 94.9 ± 2.6 μg/L in the influent to 71.8 ± 3.8 μg/L, 224.8 ± 46.1 μg/L and 80.3 ± 2.6 μg/L in new BAC effluent, and 81.7 ± 1.3 μg/L, 374.0 ± 4.2 μg/L and 97.6 ± 12.8 μg/L in old BAC effluent during chlorination in January. Similar trends were observed during chloramine disinfection. The concentrations of $THM_4$ FPs, $HAA_7$ FPs and $HAN_4$ FPs decreased by 44.3%, 12.3% and 56.9% by the new BAC filter, and by 21.1%, 5.41% and 30.1% by the old BAC filter. It was obviously found that the old BAC presented relatively poor removal ability in DBP FPs, especially DCAN, TCAA, DBCAA, TCAN, BDCM and TBAA of chlorination and TCAA, BDCAA, DBCAA and MBAA of chloramination. There were two possible reasons to explain why the old BAC performed poorly in DBP FPs removal. One is that the adsorption capacity of the old BAC was almost exhausted (Tan et al., 2017). The other one is that probably some DBP precursors leaked from old BAC to the water. Zheng et al. (2018) reported that the old BAC attached more microorganisms but produced more SMP, which could result in the increase of N-DBPs precursors in the effluent. In this study, total summed HANs FP significantly increased by 5.0 ± 3.0% after

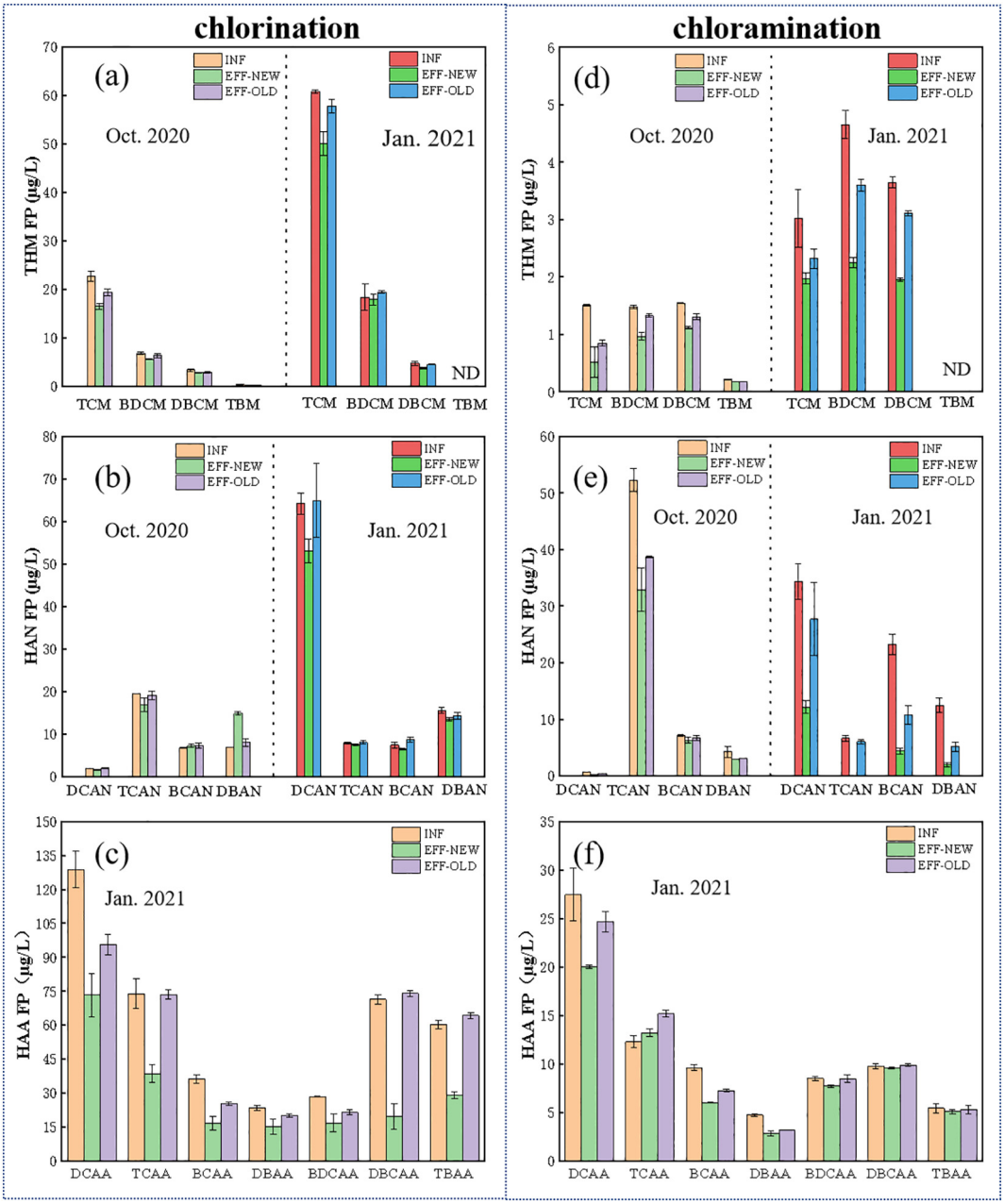



**Fig. 3.** Formation potential of $THM_4$, $HAA_7$ and $HAN_4$ of the influent (INF) and the effluent of old (EFF-OLD) and new (EFF-NEW) BAC filters. Water samples were collected in October 2020 (the left part of a, b, d and e), January 2021 (the right part of a, b, d and e, c and f). ND means no detection.

old BAC filtration for chlorinated DBPs ($p < 0.001$), indicating HANs precursors leakage from old BAC into the effluent, which is consistent with the increase of SMP-like substances in old BAC effluent (Table 1). Although the increase of total summed HANs FP by the old BAC was only 5%, but it cannot be ignored as HANs possess higher cytotoxicity and genotoxicity than C-DBPs. Considering the high toxicity of N-DBPs, the N-DBPs increase caused by old BAC filtration observed in this study reminds DWTP to consider the replacement cycle of GAC based on not only the regulated parameters but also the DBP FPs, especially N-DBP FPs, when chlorine disinfection follows BAC filtration.

### 3.4. BAC biofilm morphology and microbial activity

To compare the differences in biofilm structure between the new BAC filter and the old BAC filter, SEM micrographs of BAC at different running times are shown in Fig. 4. Many small pores were found on the surface of virgin GAC (Fig. 4(a)), but little was observed after 1 year (Fig. 4(c)) and 8 years running (Fig. 4(d)). Additionally, the BAC surface formed a wide range of interconnected biofilms after 1 year running (Fig. 4(c1)) and 8 years running (Fig. 4(d1)), respectively. The SEM results indicated that the BAC after 1 year running was covered by biofilm, and small pores were hardly observed. Therefore, the biodegradation was speculated to be dominant for water quality purification after 1 year running. In addition, the biofilm on the new BAC showed limited aggregation but exhibited larger cluster, and thicker biofilm was attached to the old BAC (Fig. 4(d1)).

Microbial activity of the biofilm attached to GAC was characterized by ATP (Hammes et al., 2010). ATP concentrations ranged from 161.21 to 802.28 ng/g GAC in the new BAC filters and from 303.51 to 543.64 ng/g GAC in the old BAC filters (Fig. 5), in the range of 300–1800 ng ATP /g GAC reported in previous studies (Di Tommaso et al., 2019; Velten et al., 2011; Velten et al., 2007). The ATP concentration in the new BAC filters was observed to be always higher than in the old BAC filters except for the half a month sample, indicating higher microbial activity in the new BAC and probably higher DBP precursors removal by microbial assimilation and respiration (Bishop et al., 2002; Noctor and Foyer, 1998).

It can be concluded that although thicker biofilm was observed on the old BAC, the new BAC exhibited higher microbial activity. The higher $UV_{254}$ removal rate (Fig. 1) and lower DBPs FP (Fig. 3) of the new BAC might be related to the relatively high microbial activity (Fig. 5) and adsorption capacity of the new BAC.

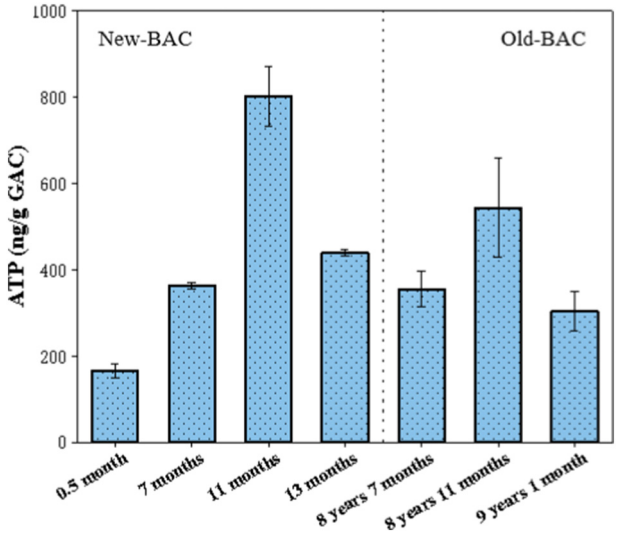



**Fig. 5.** ATP concentration of biofilm in new and old BAC filters.

### 3.5. Microbial community structure and composition in BAC filters

#### 3.5.1. Alpha diversity

Table 2 presents Chao index, Shannon index and Coverage index of bacteria attaching on the surface of activated carbon samples collected at different BAC running time. The Coverage index values for all the six samples were over 0.99 (Table 2), and the six rarefaction curves tended to increase with the increase of sequence number (Fig. S2), indicating that the sequence number was large enough to represent almost all microbial community in each BAC sample (Lemos et al., 2011; Zhang et al., 2019a). The results of Shannon and Chao index indicated that microbial community diversity increased with running time from 1 month to 7 months, and afterwards achieved a stable level. What is more, DOC removal by the new BAC filter became stable and achieved a similar level with the

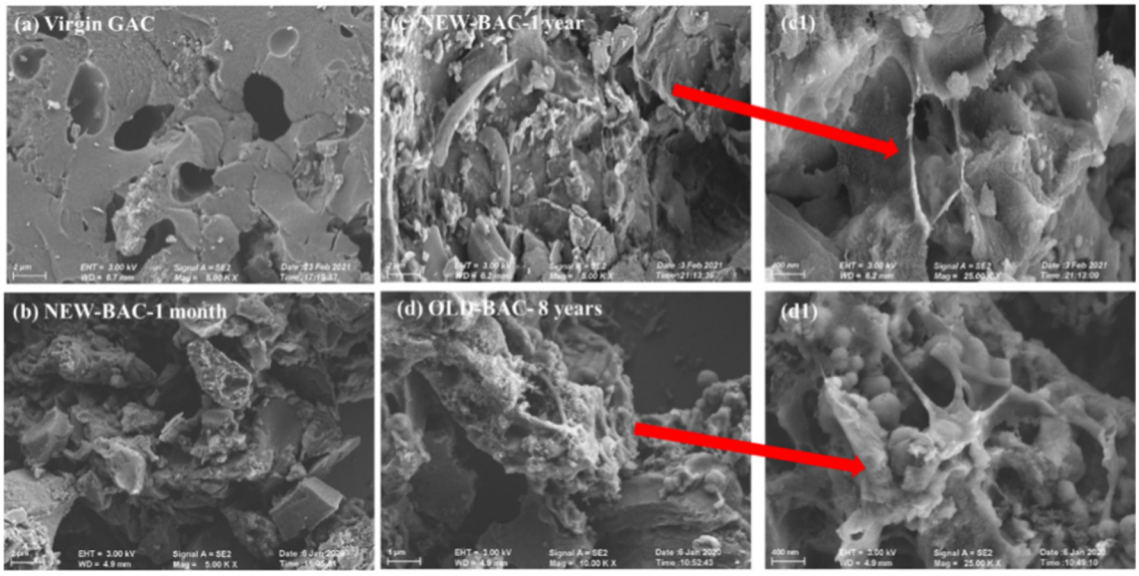



**Fig. 4.** SEM micrographs of BAC biofilm at different operating time (virgin, 1 month, 1 year, 8 years). c1 and d1 are partial enlarged perspective of new BAC and old BAC, respectively.

**Table 2**
Bacterial alpha diversity indexes at different running time ($n$ = 3).

| Sample | Chao index | Shannon index | Coverage index |
|---|---|---|---|
| New (1 month) | 186.9 ± 54.1 | 1.8 ± 0.1 | 0.99909 ± 0.00038 |
| New (2 months) | 204.5 ± 31.8 | 2.6 ± 0.2 | 0.99910 ± 0.00010 |
| New (7 months) | 922.5 ± 94.6 | 4.8 ± 0.2 | 0.99456 ± 0.00150 |
| New (11 months) | 916.9 ± 75.8 | 4.7 ± 0.1 | 0.99337 ± 0.00069 |
| Old (8 years 7 months) | 961.9 ± 55.8 | 4.5 ± 0.1 | 0.99633 ± 0.00113 |
| Old (8 years 11 months) | 1124.0 ± 69.7 | 5.2 ± 0.1 | 0.99339 ± 0.00099 |

old one after 7 months running as well (Fig. 3). These results implied that the mature biofilm had been formed after 7 months.

### 3.5.2. Beta diversity

The comparison of the similarity of the bacterial community was performed by principle coordinates analysis (PCoA) and Fig. 6(a) presents the results. It can be observed that for close running times the bacterial community time clustered together while differences in running times resulted into different clusters, suggesting that the running time greatly influenced the bacterial community. The clusters related to 7 months and 11 months showed an undeniable distance from the cluster related to initial phase samples, indicating that significant changes had took place in bacterial community during 7 months running. This result could explain why DOC and $UV_{254}$ removal increased to a stable level after a running time of 7 months (Figs. 3 and S3). Noticeably, the cluster containing old BAC samples also showed an undeniable distance from new BAC samples, which may have led to different biodegradation effects on DBP precursors. Fig. 6 (b) presents the bacterial community abundance on phylum level. It can be observed that bacterial diversity became more and more abundant with BAC running time. The results from Fig. 6(a) and (b) both clearly suggested that the running time greatly influenced the bacterial community. Significant evolution in microbial composition with BAC filer running time was observed. In the initial two months, *Proteobacteria is predominant*, accounting for >98%. Afterwards, the microbial community became diverse, and the *proteobacteria* proportion decreased to 57% in the 7th month, 42% in the 11st month, 58% and 57% in the 8th year. In the old and the new BAC filters, *Proteobacteria* was always predominant, followed by *Acidobacteria*, *Chloroflexi*, *Bacteroidota*, *Actinobacteriota* and *Nitrospirota*, which is in line with previous studies (Ma et al., 2013; Wang et al., 2018; Zhang et al., 2019a).

### 3.5.3. Microbial community structure

Illumina Miseq sequencing was conducted to investigate the microbial community structure of the biofilm attached on activated carbon samples collected at six different running times. The microbial community composition at phylum and genera levels is presented in Fig. 7 and Fig. S5.

The heatmap of the detailed correlation between running time and certain microorganism at phylum level is shown in Fig. 7(a). According to the results of Kruskal Wallis H test in Figs. S4 and S5, the most abundant species showed significant indigenous differences under different running times. Importantly, *Proteobacteria* was predominant in all running times, and *Proteobacteria* was also reported to be dominant bacteria in activated sludge systems for the removal of dissolved biodegradable organic compounds (Ma et al., 2013), which were the precursors of DBPs. Therefore, *Proteobacteria* may be the main contributor to the degradation of DBP precursors. The abundance of *Acidobacteriota*, *Chroloflexi* and *Bacteroidota* followed *Proteoba*cteria. But, the abundance of *Acidobacteriota* and *Bacteroidota* in the new BAC filter with running times of 7 and 11 months was 2–4 times higher than that in the old one. Previous studies indicated that *Acidobacteriota*, *Chroloflexi* and *Bacteroidota* can utilize various carbohydrates (Ren et al., 2014; Wei et al., 2016). Therefore, this may be another reason why the new BAC had better DOC removal performance than the old BAC.

In the genus level (Fig. 7(b)), *Herminiimonas* (0% - 46%), *Hydrogenophaga* (0% - 30%), *Phreatobacter* (0% -15%), *Beggiatoaceae* (0% - 10%) and *Bradyrhizobium* (0% - 8%) were mainly composed of detected OTUs (Fig. S5). *Herminiimonas* and *Hydrogenophaga* were most abundant

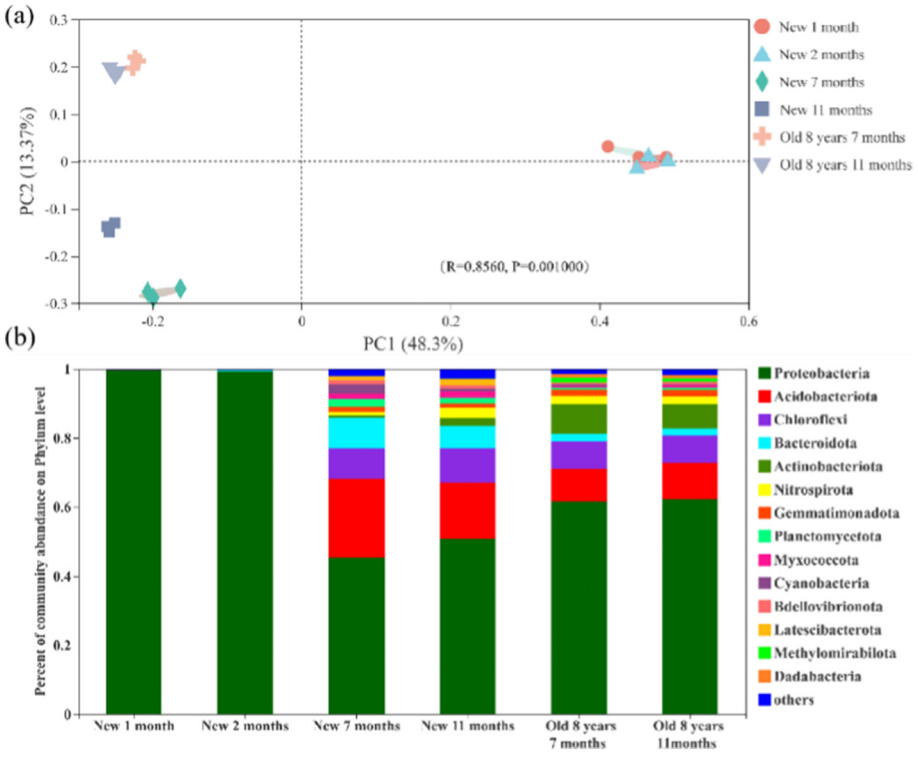



**Fig. 6.** Bacterial diversity based on (a) principal coordinate analysis of bacterial community similarity among different running time and (b) community composition at phylum level ($n$ = 3).

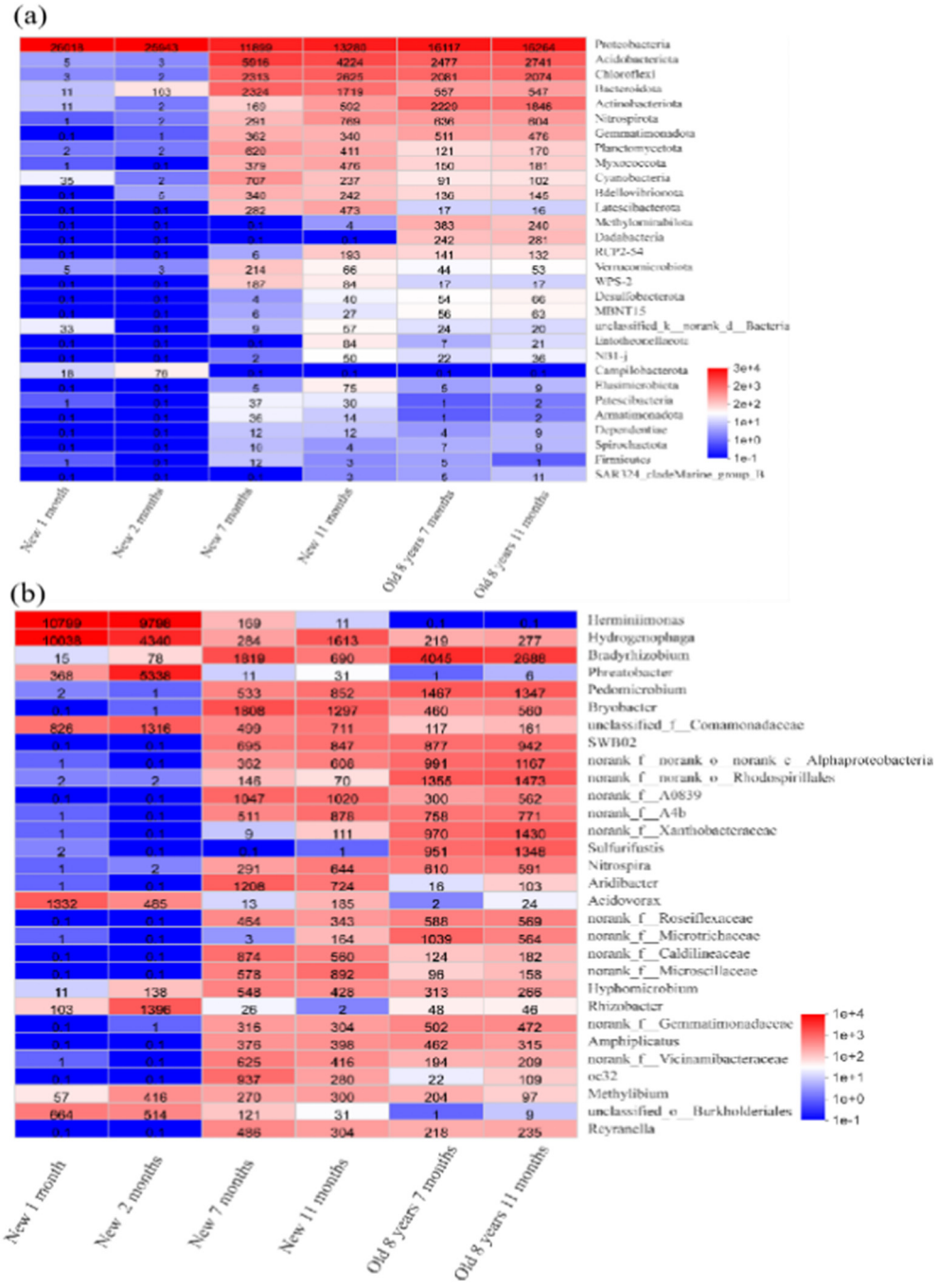



**Fig. 7.** Heatmap analysis of six different running time bacterial communities at a) phylum level and b) genera level. The y-axis is the clustering of the top 30 abundant species. The color intensity of scale indicates relative abundance of each specie. The number in each square is the average of OUT number for three parallel samples. Complete core genera (>1%) heatmap for all samples are shown in Fig. S3.

genus in the new BAC filter, which were both much higher than that in the old BAC filter (Fig. 7(b)). Previous studies showed that the *Herminiimonas* can degrade aromatic hydrocarbon compounds, such as toluene, p-cresol and phenylacetate (Kim et al., 2014; Washer and Edwards, 2007). This is a potential reason why the new BAC filter had better DOC and UV254 removal performance (Fig. 3) and lower DBP FPs than the old one (Fig. 5). Furthermore, *Bradyrhizobium*, belonging the class of *Rhizobiales*, was the most abundant genera in the old BAC filter, and previous studies reported that *Rhizobiales* could accelerate the secretion of EPS and facilitate the formation of biofilms (Cydzik-Kwiatkowska, 2015; Lin et al., 2014; Pang and Liu, 2007). What is more, as is shown in Table 1, SMP-like substances increased in the effluent of the old BAC filter but decreased in the effluent of new BAC filter. Therefore, probably *Rhizobiales* produced more EPS in the old BAC filter and EPS/SMP-like substances were released into the water, and further resulted into the increase of N-DBP FPs in the effluent of the old BAC filter (Fig. 5). The results imply that microbial/SMP-like substance leakage should be paid more attention when *Bradyrhizobium* is predominant in the bacterial community of BAC biofilm.

## 4. Conclusions

Although thicker biofilm was observed on the old BAC, the new BAC exhibited higher microbial activity. Correspondingly, the new BAC filter reached higher removal DOC efficiencies (2–7 month: 29.7 ± 6.4%; 7–13

month: 15.9 ± 7.9%) than the old one. In the effluent of new BAC filters all DOM fluorescent components were observed to decline. However, FA-like, SMP-like and HA-like substances were frequently observed to increase after old BAC filtration, indicating the leakage of microbial metabolites.

Both the old and new BAC filters had good removal efficiencies for chloraminated DBPs. Differently, $HAN_4$ FP increased by 5.0% after the filtration with old BAC for chlorinated DBPs. These results imply that drinking water managers should consider the replacement cycle of GAC based on not only the regulated parameters but also the DBP FPs, especially N-DBP FPs, when chlorine disinfection follows BAC filtration.

Microbial community diversity increased with running time, and close running times clustered together. *Proteobacteria* may be the main contributor to the degradation of DBP precursors. In the new BAC filter, *Herminiimonas* could be the reason for better DOC and $UV_{254}$ removal performance and lower DBP FPs in new filters. Furthermore, *Bradyrhizobium* was the most abundant genera in the old BAC filter, which may have produced more EPS and resulted into the increase of N-DBP FPs in the old BAC filter effluent.

## CRediT authorship contribution statement

**Feifei Wang**: Conceptualization, Writing-original draft, Writing - reviewing and Editing. **Jiazheng Pan**: Methodology, Investigation, Writing-original draft and Writing - reviewing and Supervision. **Yulin Hu**: Data Curation. **Jie Zhou** and **Haoqian Wang**: Methodology. **Wenhai Chu**: Resources, Supervision and Project administration. **Xin Huang** and **Jan Peter van der Hoek**: Resources and Writing - review and editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgements

This work was funded by State Key Laboratory of Pollution Control and Resource Reuse Foundation (NO. PCRRF19003), and Science and Technology Commission of Shanghai Municipality (NO. 20230714100).

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.155936.

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