Efficiency and Mechanisms of Biochar Aerogel-Assisted Biodegradation of Taste and Odor Compounds in a One-Step Membrane Bioreactor for Rural Drinking Water Production

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Efficiency and mechanisms of biochar aerogels-assisted biodegradation of taste & odor compounds in a one-step membrane bioreactor for rural drinking water production

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Abstract: In many rural areas, lakes and reservoirs represent common sources for drinking water, but these water bodies are more likely affected by taste & odor problems that cause discomfort to consumers. Taste & odor compounds, especially 2-methylisobornol (2-MIB) and geosmin (GSM), are not easily removed using conventional water treatment processes, and this problem is exacerbated in rural areas where treatment systems are often sparser, modest, and dated compared to typical urban water sources. Herein, a combined process deploying biochar aerogels-supported biofilms and performed in an ultrafiltration reactor (BAB-UF) was evaluated and investigated to treat rural water polluted with 2-MIB and GSM. During a 40-day experiment, the system performance was analyzed at different values of the empty bed contact time, while the microbial communities in different BAB-UF reactors were examined extensively. The process proved to be effective in removing 2-MIB and GSM, predominantly through biodegradation. Specifically, using biochar aerogels as suspended fillers in the reactor and an EBCT of roughly 1 h, the removal rate of 2-MIB/GSM was higher than 95%, and the effluent satisfied the requirements for domestic drinking water. Microorganisms with specific functions were enriched in different BAB-UF reactors and governed the transformation process, highlighting the importance of system tuning for achieving the desired biological function, hence product water quality.

Keywords: Biochar aerogel; Geosmin (GSM); 2-methylisobornol (2-MIB); Microbial community; Rural drinking water treatment.

Synopsis: This study proposes and analyzes a combined process of biotransformation and selective filtration to produce safe drinking water from rural reservoir water polluted with taste & odor compounds.

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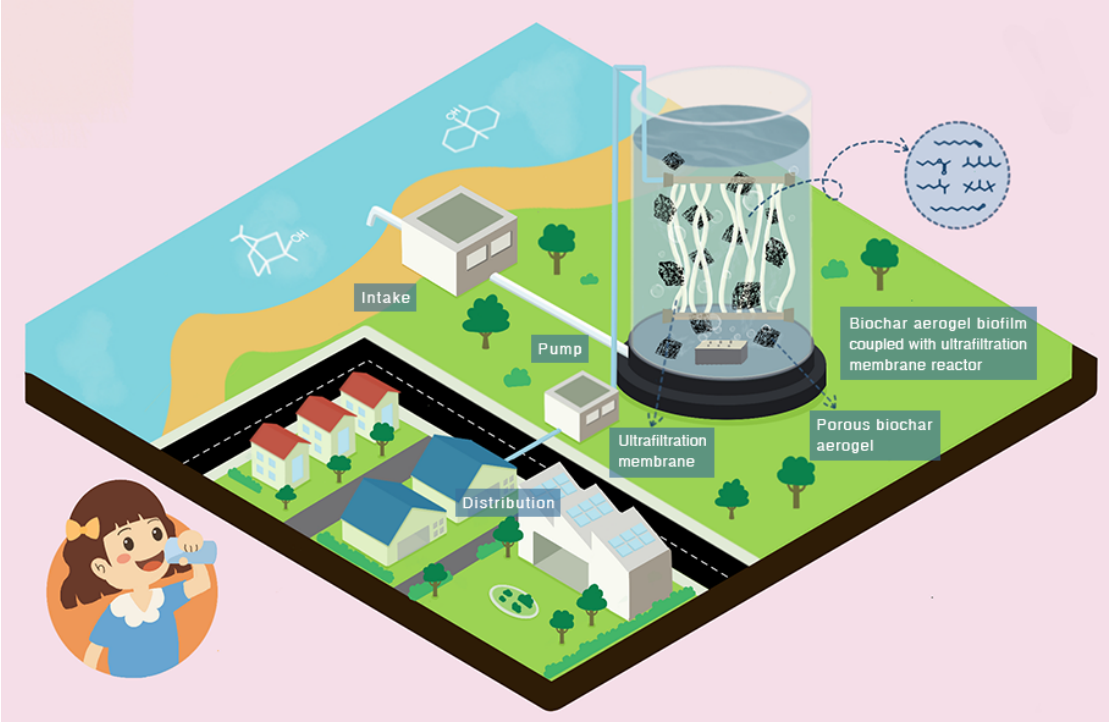


1. Introduction

Drinking water quality in rural areas is a global concern. Currently, approximately 2.2 billion people, particularly those living in remote rural areas, e.g., mountainous, pastoral, and forest areas, do not have access to safely managed or high-quality drinking water.[1, 2] For example, according to a national Chinese survey, about 80% of water bodies in China have taste & odor issues, particularly, lakes and reservoirs that serve as main water sources in rural areas.[3] Human life depends on access to clean, safe drinking water, and consumers instinctively avoid water that tastes or smells unpleasant because this characteristic may be an indicator of a harmful supply.[4-6] The terpenes, *2-methylisobornyl* (2-MIB, $C_{11}H_{20}O$) and geosmin (GSM, *trans-1,10-dimethyl-trans-9 decanol*-$C_{12}H_{22}O$), originate from the metabolism and biodegradation activity of certain types of cyanobacteria [7, 8] and they have been identified in water supplies as key odorants of the musty/earthy group.[9] The concentrations of 2-MIB and GSM in water can be perceived by the human nose at concentrations as low as 6 ng/L[4] and 4 ng/L[10], respectively.

Studies have shown that 2-MIB/GSM are not removed to concentration values below their odor threshold concentrations by conventional drinking water treatment processes consisting of coagulation-flocculation, sedimentation, and filtration.[8, 11, 12] Treatment methods that have been successfully employed by centralized water treatment plants to remove 2-MIB/GSM include granular activated carbon (GAC) adsorption and advanced oxidation processes (AOP)[13]. Several researchers have

66 demonstrated the use of oxidants, such as ozone and hydrogen peroxide, and the use of
67 ultraviolet light (UV) for the removal of 2-MIB/GSM[14-16]. However, these methods
68 have limitations for rural water treatment. For instance, activated carbon has a limited
69 adsorption capacity and needs to be replaced frequently for capacity regeneration. In
70 addition, adsorption is strongly influenced by the presence of natural organic material
71 (NOM) in natural water .[17] NOM is generally present in much higher concentrations
72 (mg/L) compared to T&O compounds (ng/L). The various fractions of NOM occupy
73 some adsorption sites and cause inhibition of adsorption of target odor substances. Also,
74 large NOM molecules may block the diffusion pathway of odorants and thwart
75 transport toward the adsorption sites[13]. As a result, high doses of activated carbon are
76 required for successful removal of the T&O compounds. Most rural drinking water
77 supply systems have lower capacity than urban systems, are managed by operators with
78 typically less professional experience compared to those operating in urban areas,[1] and
79 the addition of costly chemicals or complicated operating procedures associated with
80 adsorption and advanced oxidation processes would require sustained management and
81 maintenance that are not compatible with such rural systems. A simpler approach,
82 characterized by less complexity and requiring minimum skilled oversight, would
83 benefit water supply in rural areas.

84 Biological processes play an important role in the future of drinking water
85 treatment, especially for developing countries, due to their environmental friendliness.
86 Biological drinking water treatment relies on the activity of non-pathogenic bacteria for

biochemical degradation, and to the ability of these microbes to produce a biologically stable water that prevents the growth of other pathogenic microorganisms in the water distribution system.[18] These processes remove pollutants with minimal technological complexity, require the addition of little or no chemicals, and limit the creation of by-products that may be toxic or harmful for humans or the environment. Common biological technologies include slow and rapid sand biofiltration (SSF/RSF), biological activated carbon (BAC), and biological aerated filter (BAF).[7, 19] For example, BAC consists of microbes attaching and growing on the GAC surface to form a biofilm, which removes contaminants as water filters through the media, thus exploiting adsorption and biodegradation simultaneously, and extending the GAC service life.[20, 21] Biosand and BAC depth filtration, as well as their aerated counterpart (BAF), are effective methods for removing low molecular weight organic compounds and are becoming widespread processes for advanced water treatment in large urban water supply systems also in developed countries (such as Japan, the Netherlands, the United States).[22, 23]

A possible biological treatment alternative consists of a combined process whereby biological activated carbon is used within a membrane bioreactor. In such a configuration, the biologically-active media are suspended as fillers in the aerated raw water and the product water is extracted through membrane filtration. It has been suggested that such system would improve the quality of treated water and also help alleviating membrane fouling.[24, 25] Since membrane filtration can intercept most of the

suspended solids, microorganisms, and macromolecules in water,[26, 27] and since it can be applied in rugged mountain areas to meet drinking water treatment needs of remote areas,[28] the hypothesis of this study is that the combination of biological treatment and ultrafiltration should provide a simple and easy-to-operate way to solve odor problems associated with 2-MIB and GSM in rural drinking water systems. To date, little or no research has been conducted on these processes, resulting in a substantial lack of knowledge related both to the engineering and to the underlying mechanisms characterizing the related systems.

This study investigates the use of activated carbon-assisted biological degradation combined with an ultrafiltration bioreactor to address T&O issues in rural drinking water. To reduce the adsorption of NOM on the support carbon material, biochar aerogels were used as suspended biological fillers to form an aerogel-supported biofilm (BAB). A schematic diagram of the combined one-step process (BAB-UF reactor) is shown in **Figure 1.** Specifically, this study examines: (1) the treatment performance of rural reservoir water containing 2-MIB and GSM via four different BAB-UF reactors; (2) the biodegradation rates of 2-MIB and GSM; (3) the possible pathways of biodegradation of 2-MIB and GSM; (4) the efficiency and potential of the system for drinking water treatment; (5) the microbial communities developing in the reactor and their role on treatment performance.

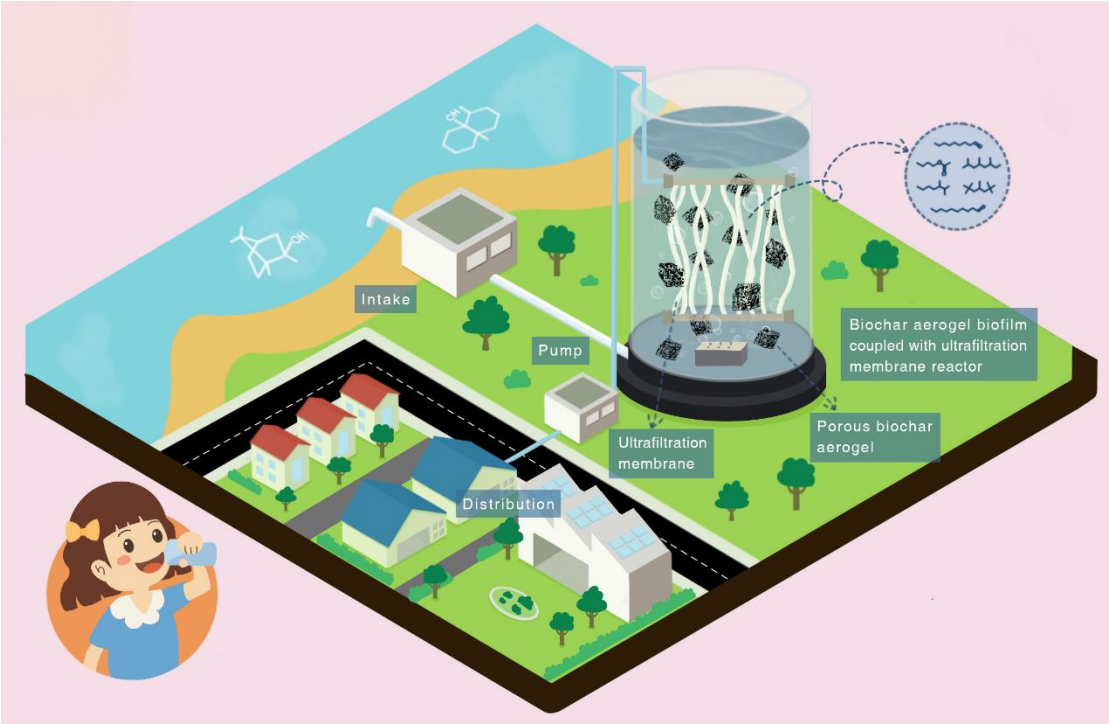


Figure 1. The schematic diagram of the combined one-step process (BAB-UF reactor)

2. Material and Methods

2.1. Materials.

Water from a reservoir in Zhong county (Chongqing, China) was used as experimental water. Wahaha brand bottled water was used for the establishment of 2-MIB/GSM standard curves and for blank control experiments. A standard mixture of 2-MIB (trace CERT®) and GSM (trace CERT®) was purchased from Sigma-Aldrich [13]. Sodium chloride (NaCl, 98% purity), potassium hydroxide (KOH, 90%), acetic acid (CH_3COOH, 99.5% purity), and sodium azide (NaN_3, 99%) were provided by Agilent Technologies (China) Co., Ltd. Chitosan was purchased from Jinan Haidebe Marine Biological Engineering Co., Ltd. (China), and glucose was purchased from Chengdu Kelong Chemical Reagent Factory (China). The ultrafiltration hollow fiber membranes

used in this study were provided by Litree Purifying Technology Co., Ltd (Hainan, China). Peristaltic pumps (BT-300-2 J, Longer Pump, China) were used to feed water from the tanks to the BAB-UF reactors, as well as to extract effluents from the ultrafiltration hollow fiber membrane modules.

2.2. Methods

2.2.1. BAB-UF reactors set-up.

The shallow sediment of the reservoir was collected for microbial enrichment, and a 28-day acclimation process was required to allow microbial aggregation and biofilm formation on the suspended fillers. Specific details regarding the acclimation process and associated data can be found in the Supporting Information (**SI**, **Text S1 and Figure S1-2**). Following the 28-day acclimation period, degradation experiments were initiated. Actual water from the reservoir mixed with 2-MIB and GSM at a concentration of 100 ng/L was used as the raw water (RW), which was supplied to all reactors simultaneously. Because odor problems are seasonal[29], with outbreaks typically lasting about 1 to 2 months during the warm weather season, the study was conducted in the laboratory for 40 days. Basic information of the different BAB-UF reactors is shown in **Table 1**.[4] Concentrations of 2-MIB and GSM were measured every three days for 33 days in the influent and effluent streams from the BAB-UF reactors. After the systems achieved stability, the concentration of odorant was gradually increased from 100 ng/L to 200 ng/L.

Table 1. Operating parameters of the BAB-UF reactors in this work.

Operators ID	Suspended Fillers	Flow rate (ml/min)	EBCT (min)	Gas-water ratio	Running Time (d)
UF	-	0.524	-	3:1	40
BAB-20-UF	BA	0.524	18	3:1	40
BAB-40-UF	BA	0.524	36	3:1	40
BAB-60-UF	BA	0.524	54	3:1	40

Note: The running time of the reactors only includes the degradation experiments period.

2.2.2. Experimental setup comprising biodegradation, adsorption and aeration in BAB-UF

The following three control experiments were conducted on the 40th day of the experiment when the influent 2-MIB/GSM concentration was 200 ng/L. In one control experiment, only aeration was applied while the fillers and ultrafiltration membranes were not present, to understand the rate of volatilization. In a second control experiment, Wahaha purified water containing 300 mg/L sodium azide was used to soak the filler in the BAB-UF device for 2 h, which inhibited the biological activity of the biofilm without affecting other functions (e.g., adsorption).[20] A third control test was conducted in which the membrane reactors operated simultaneously under the same conditions but without aeration and without the presence of the ultrafiltration membrane. The four full BAB-UF reactors were instead run at different empty bed contact time (EBCT) of 18, 36, 54 min and contained 20%, 40%, and 60% fillers, respectively. In all cases, we measured the influent 2-MIB/GSM concentrations and the effluent concentrations three

178 times, and for each setup we calculated the average effluent concentration as a
179 percentage of the influent concentration to obtain the removal rate of 2-MIB and GSM.
180 The biodegradation contribution in the full BAB-UF reactors was obtained by
181 subtracting the aeration and adsorption rates from the total BAB-UF removal rate.
182
183 **2.2.3. Preparation of biochar aerogel fillers for biofilm support**
184 The biochar adsorbent powder was prepared using the sol-gel method;[30-32] refer to
185 the SI (**Text S2**) for a detailed description of the method. The method of BAB aerogel
186 production was adapted from previous research[33] and comprised with the following
187 steps: chitosan powder with a mass concentration of 4.0 wt% was dissolved in 1.0 wt%
188 aqueous glacial acetic acid solution by stirring, resulting in a yellow transparent
189 homogeneous solution. The previously prepared 0.1wt% biochar adsorbent powder was
190 added to the chitosan solution and stirred for 30 min to ensure even dispersion. The
191 mixture was then injected quickly with a syringe into a cylindrical mold about 7 mm
192 height and about 5 mm in diameter, and then rapidly frozen at −20°C before being
193 freeze-dried for 24 h. The resulting dried granular aerogel was removed from the mold
194 and placed in a 4 wt% aqueous NaOH solution, where it was shaken for 2 h at room
195 temperature to fully react and neutralize the acid in the aerogel. Finally, the pellet
196 aerogels were washed with pure water until neutral pH to obtain the final BAB supports.

2.3. Analytical methods

2.3.1. 2-MIB and GSM analysis

The concentrations of 2-MIB and GSM were analyzed using a gas chromatography-mass spectrometry (GC-MS) system, which was coupled with a solid phase micro-extraction (SPME) fiber holder and SPME fiber assembly-divinylbenzene/carboxen/polydimethylsiloxane (DVB/CAR/PDMS). An Agilent 8890 Series gas chromatograph equipped with an HP-5 MS capillary column (30 m × 0.25 mm × 0.25 mm film thickness) interfaced to an Agilent 7010B mass selective detector was used. To quantify the 2-MIB/GSM, 35 mL of water sample and 3.5 g of NaCl baked at 300 °C in a muffle for 2 h were mixed evenly into a 40 mL brown headspace bottle. The bottle was placed into a 65 °C water bath for preheating, and extraction was carried out for 15 min. The extraction head was then inserted into a GC injection port for 5 min for quantitative analysis of 2-MIB/GSM. The carrier gas was high purity He (99.999%, Rising Corp., China) at a column flow rate of 1 mL/min. The injection port operated in a separation mode with split ratio of 20:1 while the temperature maintained at 250 °C. The column oven initial temperature was set at 60 °C for 2.5 min, increased to 250 °C at a rate of 10 °C/min, and then maintained for 5 min. The MS ion source temperature, the four-stage rod temperature, and the auxiliary heating zone temperature were maintained at 230 °C, 150 °C, and 280 °C respectively. The solvent delay was set to 7.5 min. The MS ionization energy was set to 70 eV, and the scanning mode was set at multiple reaction monitoring (MRM). The quantitative ion pair parameters are 95/67

for 2-MIB,112/97 for GSM. Standard chromatograms and 1-200 ng/L 2-MIB/GSM standard curves are presented in the SI (**Figure S4**).

2.3.2. Effluent quality analysis

A portable multifunctional Ultrameter II 6PFC (Myron L Company, Carlsbad, USA) was utilized to determine the total dissolved solids (TDS). COD_{Mn} was determined based on the permanganate index (GB 11892-89). DOC and UV_{254} values were obtained, respectively, with a TOC analyzer (TOC-L CPH, Shimadzu, Japan) and with a UV–vis spectrophotometer (Orion AquaMate 8000, Thermo Fisher, USA). The pH was measured using a pH meter (PB-10, Sartorius Scientific Instruments Co, Ltd., Gottingen, Germany), while turbidity was measured using a turbidimeter (2100Q, Hach Company, Loveland, USA). A three-dimensional excitation-emission matrix (3D EEM) fluorescence spectrum (F-7000, Hitachi, Japan) was used to characterize the DOM components in the effluent and on the membrane contaminated layers. For detailed information on the related methods, please refer to our previous studies. [34, 35]

2.3.3. 16S-rRNA analysis

The microbial diversity of the biofilm formed on the suspended fillers in the continuous mode BAB-UF reactor at different times was sequenced using 16S-rRNA to analyze changes in microbial communities and dominant functional microorganisms over time. In the four BAB-UF reactors, a total of six BAB filler samples and two sections of ultrafiltration membrane samples were collected on days 10 and 40. For

each BAB filler sample, 2 g of packing material from approximately the middle depth of the reactor was taken and placed into a sterilized 10 mL polyethylene bottle. The sample was then transported to the analytical laboratory on ice packs, while an equal volume of packing was backfilled immediately after removal. The detailed operation method of 16S-rRNA can be found in the SI (**Text S4**).

2.3.4. Membrane fouling analysis

The membrane fouling layer morphology was examined with scanning electron microscopy (SEM, Regulus 8230, Hitachi, Japan) on membrane samples after their use in the reactor. Before SEM analysis, the dried sample surfaces were coated with Au by a model MS-2S gold coating instrument (IXRF Systems Inc., USA), reaching a thickness of approximately 2 nm.

3. Results and Discussion

3.1. Biodegradation is the dominant removal mechanism of 2-MIB and GSM

In this study, a fraction of 2-MIB/GSM was lost due to natural volatilization since the experimental device could not be kept sealed (due to continuous aeration, the pressure in the reactor would increase if it was completely sealed, resulting in increased water flow resistance and reduced water flow velocity). The results of the control experiments suggested that the fractions of volatilized 2-MIB and GSM were 32.4% and 42.3%, respectively, at a starting concentration of 100 ng/L, while 56.25% and

61.01%, were lost at a starting concentration of 200 ng/L. These losses were accounted for when computing raw water concentrations and in the following calculations.

By observing the influent and effluent concentrations 2-MIB/GSM from the four BAB-UF reactors throughout 40 day of testing, it is clear that the removal efficiency increased with EBCT; see **Fig. 2a, b**. For example, the effluent 2-MIB/GSM concentrations were 6 and 4 ng/L, respectively, from BAB-40-UF and BAB-60-UF, hence lower than the human nose perception threshold and attaining values that respect the local standards for drinking water quality (GB 5749-2022). Note that the removal rate obtained in the control experiments comprising the UF membrane only was around or lower than 30%, due to the small molecular size of the two T&O compounds that cannot be excluded from entering and moving across the membrane pores. Such removal rates recorded in the control experiment are attributed to the presence and activity of bacteria on the ultrafiltration membrane. These bacteria were remnants from the initial acclimation phase conducted prior to the commencement of the control experiment.

The contribution of aeration, adsorption, biodegradation to the overall 2-MIB/GSM removal rate is presented in **Fig. 2c**. Aeration was responsible for 19.5% and 13.4% of the total removal of the two substances. The adsorption removal rates were instead 2.7%, 9.5%, and 18.2% for 2-MIB and, quite similarly, 3.0%, 10.4%, and 18.8% for GSM, from the three reactors characterized by EBCT 18, 36, 54, respectively. Indeed, the contribution of biodegradation was dominant and estimated as

follows: UF only (11.8%, 8.7%), BAB-20-UF (46.3%, 63.9%), BAB-40-UF (56.1%, 69.6%), BAB-60-UF (58.5%, 66.0%). The findings indicate that the process of adsorption did not provide substantial removal, possibly due to the chemical nature and to the low concentrations of 2-MIB and GSM. On the other hand, biodegradation contributed to around 60% of the overall removal rate, which aligns with the conclusions drawn by earlier research[7], and represented the dominant removal mechanism in the examined system.

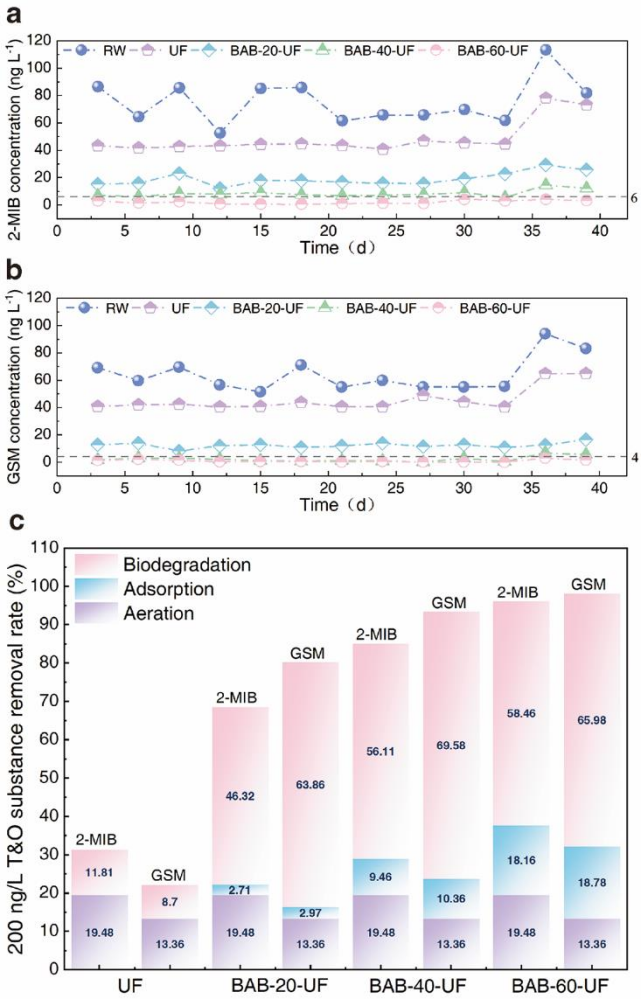


Figure 2. Removal rates of T&O compounds: (a) 2-MIB concentrations and corresponding removal rates in different reactors: the grey line indicates the 2-MIB

odor threshold in water at 6 ng/L; (b) GSM concentrations and corresponding removal rates in different reactors: the grey line indicates the GSM odor threshold in water at 4 ng/L; (c) contributions to removal rates from different mechanisms. RW refers to the reservoir water containing 2-MIB/GSM.

3.2. The effluent from the BAB-UF systems met local drinking water standards

The water quality parameters of influent and effluent streams from different BAB-UF reactors are displayed in **Fig. 3**. The COD_{Mn} of effluents from BAB-60-UF and BAB-40-UF were below 3 mg/L, thus meeting local drinking water standards (GB5749-2022); see **Fig. 3a, b**. Specifically, the COD_{Mn} removal efficiency was higher than that of the UF device alone, suggesting that BAB adsorbed and/or biodegraded part of organic compounds. Considering DOC, the average removal rate increased by roughly 12% with each doubling of EBCT, achieving approximately 44% in BAB-60-UF (**Fig. 3c, d**). Other than removal rate itself, the stability of removal was apparently higher for reactors characterized by longer EBCT. A similar trend was also observed for the UV254 parameter, which can be used to quantify the amount of dissolved organic pollutants with unsaturated bonds in the raw water. As shown in **Fig. 3e**, the removal of UV_{254} was between approximately 19.0 (BAB-20-UF) and 46% (BAB-60-UF). The fact that the removal rate of UV_{254} was higher than that of DOC indicates that the BAB-UF reactor more efficiently degraded unsaturated organic and aromatic compounds.

Turbidity and TDS were used as additional indicators related to the quality of drinking water. As shown in **Fig. 3f** and consistent with the removal behavior of organics, the removal rates generally increased with EBCT and the average turbidity of the effluent of BAB-UF reactors was lower than 0.5 NTU, indicating higher than 90% removal rate of turbidity in all cases. Finally, the TDS did not change considerably, converting from 120-140 ppm in the RW to 100-130 ppm in the treated effluents, the minimal removal likely due to uptake by microorganisms for growth.

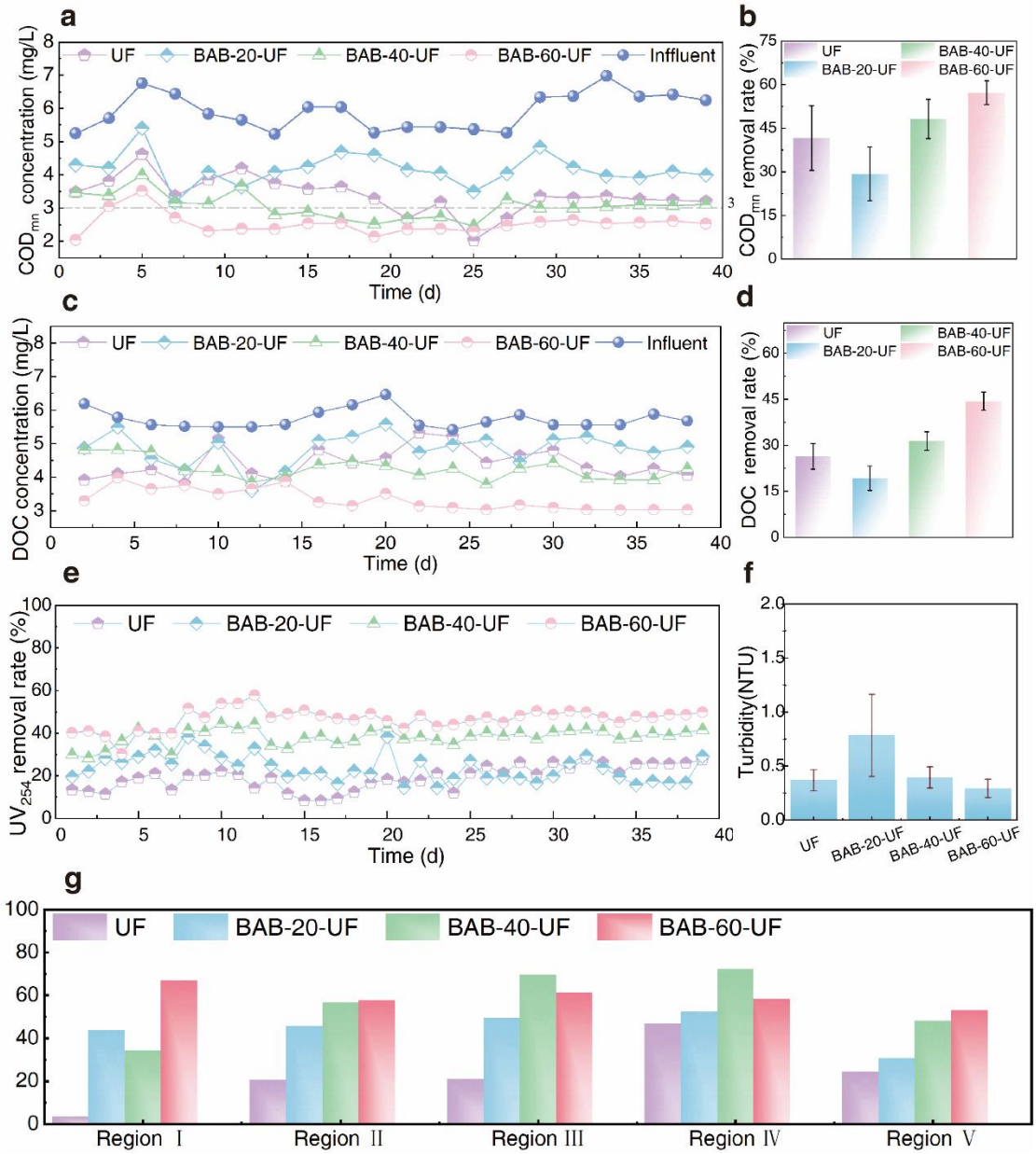


Figure 3. Water quality parameters of influent and effluent streams from different BAB-UF reactors during 40-days. (a) COD_{Mn} concentrations; (b) corresponding average removal rates of COD_{Mn}: here, the grey dotted line marks the local drinking water standard limits for COD_{Mn} in the GB5749-2022 legislation; (c) DOC concentrations; (d) corresponding average removal rates of DOC; (e) UV_{254} removal rates; (f) average turbidity concentrations in the effluents. (g) Removal rates of Fluorescence EEM spectra computed for different BAB-UF reactors.

Beside overall removal rates, the changes in the composition of DOC were examined with fluorescence EEM spectroscopy. The EEM spectrum may be classified into five regions: Region Ⅰ: (Ex/Em = 220-250/280-330 nm, tyrosine protein-like substances), region Ⅱ: (Ex/Em=220-250/330-380 nm, tryptophan protein-like substances), region III: (Ex/Em = 220-250/380-480 nm, fulvic acid-like matters), region Ⅳ: (Ex/Em = 250-440/280-380 nm, soluble microbial by-product-like matters), and region V: (Ex/Em = 250-400/380-540 nm, humic acid-like components).[34, 36] As shown in **Fig. S5e**, fulvic acid-like matters (region III), soluble microbial by-product-like matters (region IV), and humic acid-like components (region V) were the main organic constituents in the raw water, far more than tyrosine protein-like substances (regions I and II). This observation is attributed to the fact that, similar to previous studies, fulvic acid-like matters (region III) and humic acid-like components (region V) are the major components of NOM in surface waters.[37] The removal efficiency of BAB-

UF for regions III and IV were the highest and most consistent for the different systems, with removal rates higher than 50% observed for the two higher EBCT values for regions II, III, IV, and V, consistent with the removal efficiency of COD_{Mn} and UV_{254}; see **Fig. 3g**. This result implies that the BAB-UF reactors are especially apt to the purification of hydrophilic low and medium molecular weight organic components, which actually represent the largest fraction of organics in rural surface water sources, such as lakes and reservoirs.

3.3. Adaptation of the microbial community in BAB-UF systems

All analyzed samples had coverage values greater than 0.99 (see **Text S4.2**) , indicating that the sequencing depth was sufficient to cover most microorganisms, even some rare species.[35, 38] The flattening of the rarefaction curve in **Fig. 4a** also indicates sufficient sequencing depth. The combined result imply that the community richness and diversity of the 40th day samples was higher than that of the 10th day samples.

Fig. 4b and **c** show the microbial community composition at the genus level (relative abundance > 0.05% in all samples) and at the phylum level (relative abundance > 0.1% in all samples), respectively. Similar to previous studies using BAF or other methods for drinking water treatment,[28,39] *Proteobacteria* were the most abundant and broad in all samples (in this study, 40%-88% of the samples on the 10th day, and 51%-93% of the samples on the 40th day). Another dominant phylum was *Bacteroidetes*, which are well-known degraders of organic matter.[40] The above two phyla, together

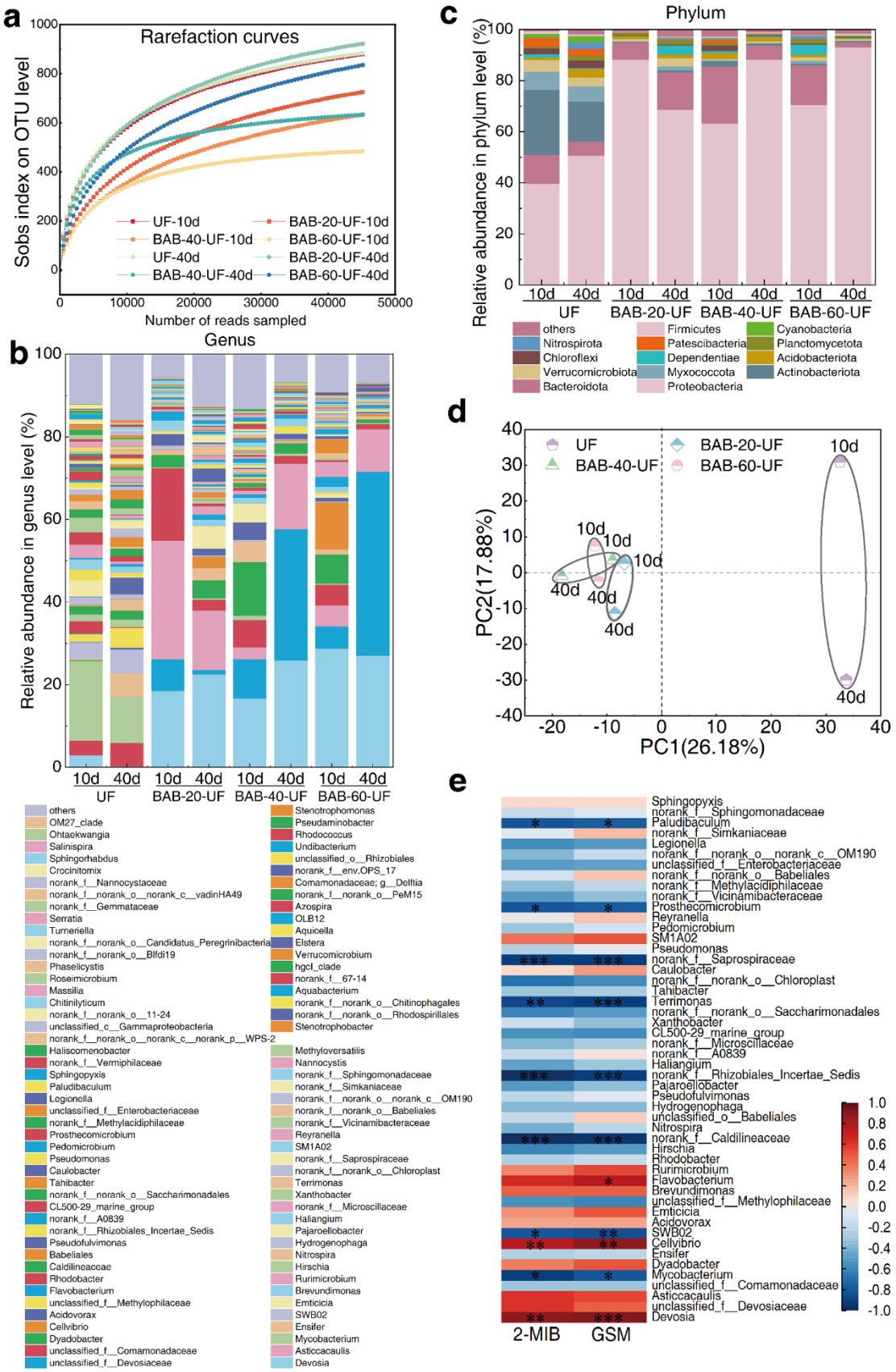
with *Actinobacteriota*, *Myxococcota* and *Verrucomicrobiota*, accounted for more than 81% of the community.

Principal component analysis (PCA) of OTUs levels of microbial communities in different BAB-UF reactors (**Fig. 4d**) provides information on the similarity of the bacterial community structure between the four reactor samples and the effect of different operating times on the bacterial community structure. The bacterial community structures of BAB-20-UF, BAB-40-UF, and BAB-60-UF were similar, whereas the bacterial community structures of the reactors using ultrafiltration alone showed substantial differences. This result may be mainly attributed to the fact that the ultrafiltration membrane is not suitable for microbial attachment and growth compared to fillers and that it has a relatively poor ability to absorb substances from the influent water. The results also imply that materials with strong bioattachment capabilities may play an important role in acclimating stable microbial communities. While there were some differences in the bacterial community structure across the three BAB-UF reactors, the essential microorganisms were comparable. According to the data plotted in Fig. 4c, these core microbes were *Devosia*, *unclassified_f_Devosiaceae*, *Asticcacaulis*, *Unclassified_f_Comamonadaceae*, *Dyadobacter*, *Ensifer* and *Cellvibrio*, all belonging to *Proteobacteria* and *Bacteroidetes*, indicating that a large number of microorganisms potentially capable of degrading organic matter were adapted and retained in the biofilm-rich systems.

To further investigate microorganisms activity for 2-MIB/GSM removal, correlation analysis (**Fig. 4f**) was performed between the 2-MIB/GSM removal rate and the genus-level microbial community (top 50 genus in abundance). The results suggest that *Devosia* with abundance of 0.2% to 26% was positively correlated with substance removal ($p<0.01$ and $p<0.001$ for 2-MIB and GSM, respectively). It has been reported that *Devosia* isolated from enriched bacterial cultures showed effective degradation of alcohols, e.g., deoxynivalenol, whose molecular structure is similar to that of GSM.[41] Degradation enzymes in *Devosia* may belong to the aldo-keto reductase (AKR) family.[42] *Cellvibrio* (0.9%-13%) were also positively correlated with the removal rates of 2-MIB and GSM ($p<0.05$ and $p<0.01$, respectively), but its mechanistic contributions requires additional research. Finally, *Flavobacterium* (0.2%-2.4%) had a significant correlation with the GSM removal rate ($p<0.05$). Yuan *et al.* reported that the ability of *Flavobacterium* to remove 2-MIB with an initial concentration of 515 ng/L reached 96.3%.[7, 19] According to the correlation analysis results of this study, *Flavobacterium* may also contribute to the degradation of GSM, but further studies are needed to prove this hypothesis. It is worth noting that *Pseudomonas*, *Sphingomonas sp.* were found in the four groups of BAB-UF reactors, and it has been pointed out that these two bacteria are likely to participate in the biodegradation of 2-MIB and GSM.[7, 19] Indeed, the abundance of *Pseudomonas* and *Sphingomonas* in the BAB-UF reactors at 40 days was an order of magnitude higher than that measured at 10 days, and also it increased with BAB filler amount,

corroborating previous reports. Note that the correlation analysis discussed above is hypothetical and based on data obtained from the 16s rRNA assay. Additional investigations will be conducted in future studies to test this hypothesis and the relationships with other water elements will also be taken into consideration. Specific microbiological tests will be conducted for each distinct bacterium in the water at the appropriate 2-MIB and GSM concentrations.

In conclusion, after the completion of the acclimation phase, the microbial communities in the different BAB-UF reactors were adapted to the raw water. After long-term operation, *Proteobacteria* was dominant in all communities. It appears that suspended fillers with robust biological affinity were able to acclimate stable microbial communities, which in turn effectively broke down contaminants in water. In addition, several microorganisms were found have a strong correlation with 2-MIB/GSM removal, including *Devosia*, *Cellvibrio*, *Flavobacterium*, *Pseudomonas*, as well as *Sphingomonas*. These correlations shall be further evaluated and quantified by future studies.



417

418 **Figure 4.** Microbial community analysis in four BAB-UF reactors: a) OTUs-level

419 rarefaction curves at different operation times. Bacterial community compositions of
420 (b) genus (> 0.05%), (c) the phylum (> 0.1%) and (d) principal component analysis. (e)
421 are the correlation analysis of the above genus-level (top 50) microbial community and
422 2-MIB/GSM removal rate variables ("*" represents a value of $p < 0.05$, "**"
423 represents a value of $p < 0.01$ and "**" represents a value of $p < 0.001$).
424
425 **3.4 Biochar aerogel fillers slightly improved the membrane fouling behavior**
426 The morphology of the fouling layer on the membrane surface in various BAB-
427 UF reactors was analyzed with SEM; see representative micrographs and results in **Fig.**
428 **S8**. Surfaces were covered with a thick fouling layer. Interestingly, the fouling layer of
429 BAB-60-UF membrnes was the thinnest among the aerogel-assisted reactors,
430 suggesting that EBCT ought to be no less than roughly 54 min to control fouling in the
431 investigated system. Therefore, the impact of aerogel was minor but not negligible, and
432 may deserve further study to understand how the presence of fillers might change the
433 interaction of microorganisms and organic matter with the membrane.
434
435 **ASSOCIATED CONTENT**
436 **Supporting Information**
437 Additional experimental materials, methods, and procedures for data processing;
438 discussion on DTPs and comparative analysis; and a number of other supporting
439 materials are provided in Texts S1−S5, Tables S1−S4, and Figures S1−S8.

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