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# Complex odor control based on ozonation/GAC advanced treatment: optimization and application in one full-scale water treatment plant

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## Abstract

**Background:** Taste and odor problem in drinking water is one major concern for consumers and water supply. Exploring the odor characteristics and the major odor causing compounds in the source water is the base for odor control in drinking water treatment plant (WTP). In this study, focusing on a newly constructed reservoir with Huangpu River as the source water, the occurrence of typical odorants and their variations were first identified. Correspondingly, the removal behavior in an ozone/GAC advanced treatment process was investigated.

**Results:** The results indicated that 2-methylisoborneol (2-MIB), geosmin (GSM), and bis (2-chloroisopropyl) ether (BCIE) have major contribution to the musty/earthy and chemical/septic odors in the source water, respectively. Pre-ozonation alone ( $1 \text{ mg L}^{-1}$ ) showed limited removal for 2-MIB and BCIE, at less than 30% and 20%, respectively, while combining with coagulation, sedimentation, and sand filtration, the removals were improved to higher than 50%. After post-ozonation, the desired removal was achieved at a  $1.5 \text{ mg L}^{-1}$  dosage with all the odorants decreased below the corresponding odor threshold concentrations (OTCs) in the effluents. Furthermore, at a  $1 \text{ mg L}^{-1}$  post-ozone addition, by combining with subsequent GAC process, the odor problem was solved as well.

**Conclusion:** To resolve the odor problem in the drinking water, the concentrations of the odorants at less than their OTCs need to be achieved. As 2-MIB and BCIE have low reactivity towards direct ozonation, a subsequent GAC is needed with a moderate dosage of post-ozonation ( $1 \text{ mg L}^{-1}$ ). Thus, for the odor problem in the source water, the suggested operation is:  $1 \text{ mg L}^{-1}$  of pre-ozonation in combination with coagulation, sedimentation, and sand filtration, followed by a  $1 \text{ mg L}^{-1}$  dosage of post-ozonation and finished by a GAC process.

**Keywords:** Odor, Ozonation, Granular activated carbon, 2-Methylisoborneol, Bis(2-chloroisopropyl) ether

## Background

Ensuring the esthetic satisfaction in drinking water is one major objective for water utilities. Usually, the occurrence of most odor issues was reported to associate with source water problems. For example, the bloom of algae

resulting from the excess of nutrients is one of the internal causes that leads to the production of the odorous metabolites [51]. Additionally, contaminant inputs such as tributaries, upstream industrial discharges, and oil spills from gas station lead to the external causes for the odor problem in the surface water [21, 25, 32]. Odor and taste compounds although have been reported to be non-toxic towards humans with the concentrations observed in the environment, their effects on public trust on drinking water safety is significant. In China, odor issue has become one growing concern for water safety, especially

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after the water crisis in Wuxi in 2007 [67], where serious complex odorants occurred in Lake Taihu water source. However, due to the limited information on odor characteristics and specific odor causing compounds, water treatment plants (WTPs) were usually not able to adopt applicable measures while encountering taste and odor problems.

Musty/earthy odor caused by the presence of 2-methylisoborneol (2-MIB) and geosmin (GSM) is the most encountered problem in drinking water, which is usually related with cyanobacteria proliferation [26, 33, 47], especially some benthic cyanobacteria such as *Oscillatoria* and *Phormidium* [56]. Huangpu River, the major source water in Shanghai, China, was reported to face musty/earthy odor problems seasonably, which was mainly correlated to 2-MIB produced by *Phormidium* [46]. Besides *Oscillatoria* and *Phormidium*, *Pseudanabaena* species have been reported to be a potential GSM producer [23].

Compared to musty/earthy odor compounds, the compounds that cause the chemical/swampy/septic smells are more complicated. A large number of compounds may result in an unpleasant odor in the surface water. Some sulfur-based odor may result from anaerobic bacteria [6, 9, 41]. For example, thiols (e.g., methanethiol) and thioether are the primary swampy/septic odor sources [16, 64], while thioether includes not only mono-sulfide thioether (e.g., dimethyl sulfide, diethyl sulfide, and dipropyl sulfide), but also multi-sulfide thioether (e.g., dimethyl disulfide (DMDS), dimethyl trisulfide (DMTS), and diethyl disulfide). Other odorous compounds (e.g., benzenes, phenols, esters, and heterocyclic compounds containing oxygen), arising from industrial activities such as meat rendering, rubber proofing, resin and plastic synthesis, and dye manufacturing, may cause odor problem at a very low concentration ( $\text{ng L}^{-1}$ ) [11, 12, 42, 43]. For example, bis(2-chloroisopropyl) ether (BCIE) is a chemical that is primarily used in industry as a solvent for fats, waxes and greases, as an extractant in paint and varnish removers, in spotting and cleaning solutions, and in textile processing [14]. Chemical/septic smelling compounds resulting from the industrial discharge and surface runoff [62], together with the natural odorous compounds (e.g., MIB and GSM) form the complex odor in the surface water. Complex odorants have been reported to be present in Huangpu River [17], causing a seasonal odor issue.

Conventional treatment processes showed a limited odorant removal. Pre-treatment, advanced treatment, or a combination of these techniques have been reported to improve the removal efficiency [39]. Compared to chlorination and potassium permanganate ( $\text{KMnO}_4$ ), ozonation revealed a greater removal efficiency [29, 30, 48]. Oxidation is effective to treat thiols and thioethers as well

as the musty/earthy and chemical odorous compounds [53]. However, the byproducts from ozonation such as bromide ions, precursor of aldehydes, ketones, and carboxylic acids cannot be neglected. Generally, granular activated carbon (GAC) is applied in combination with ozonation to remove the byproducts and the odorant residuals [28]. Studies have indicated that GAC was effective in removing musty/earthy compounds 2-MIB and GSM [13, 63]. A decreasing effectiveness was observed for the typical septic odorous compounds including thiols and thioethers through GAC [2]. By applying post-ozonation in combination with GAC, the odorants may be eliminated through oxidation and physiochemical adsorption. Adding pre-ozonation prior to post-ozonation and GAC improved the removal efficiency of recalcitrant compounds (e.g., emerging contaminants) [45, 65]. Such combination of treatment processes may apply to complex odor elimination when there is a significant input of the odorous compounds in the source water. Currently, the combined processes that are commonly involved include: (1) post-ozonation followed by GAC; and, (2) pre-ozonation, followed by post-ozonation, and GAC [17, 31].

A number of studies have focused on the odorant removal in water treatment processes. Ozonation and GAC were the most investigated unit processes [4, 8, 13, 20, 38, 40] as well as the  $\text{O}_3$ /GAC combined process [1, 39]. For the most part of these studies, great efforts have been made on the effectiveness of an individual process, while the additive and synergistic effects of a full treatment train have obtained less consideration. A few studies have investigated the effectiveness of a complete treatment train; however, the experiments were conducted in well controlled pilot systems. For example, Chen et al. [10] examined the removal of 2-MIB and GSM in a pilot-scale treatment train (including coagulation, sand filtration, ozonation, and GAC) where the odorants were spiked with pre-determined concentrations and the influent water quality was relatively stable. Moreover, the complex odor control has received less attention as well. For the above reasons, current study investigated and optimized an existing WTP treatment train reflecting real water conditions to achieve a simultaneous removal of a group of complex odorants.

Jinze Reservoir is a newly constructed reservoir, which is located at the upstream of Huangpu River, in Shanghai, China. The potential complex odor problem, especially during the startup period, needs to be evaluated; more importantly, an effective and practical control method needs to be developed. This study characterized the various odorants being present in the reservoir. The primary odor contributors were identified as well. The operation of the existing ozonation/GAC processes applied in one

downstream WTP was evaluated and optimized based on the practicable operational conditions. The application of the optimized treatment train was evaluated for 6 months to further verify its efficiency on complex odor control. This study provides a feasible odor elimination solution for the full-scale WTPs facing a potential complex odor issue.

## Materials and methods

### Chemicals and reagents

Odorant standards were ordered from Sigma-Aldrich (USA) with purity greater than 95%. A stock solution was prepared in methanol at a concentration of 10 mg L<sup>-1</sup> for each reference compound (Table 1). Analytical grade of sodium chloride (NaCl) and anhydrous sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>) were purchased from

Beijing Chemicals Ltd., China. Both chemicals (NaCl and Na<sub>2</sub>SO<sub>4</sub>) were pre-baked in a furnace at a temperature of 450 °C for 2 h to removal possible organics and moisture. A Milli-Q purification system was used to provide ultrapure water (18.2 MΩ cm) for chemical preparations.

### Sampling site and collection

Jinze Reservoir is an upstream water source on Huangpu River with a total volume of 9.1 million m<sup>3</sup> and a water supply capacity of 3.51 million m<sup>3</sup> day<sup>-1</sup>, serving 6.7 million residents from five districts in the southwest part of Shanghai, China. The reservoir started its operation in late 2016. The raw water had a chemical oxygen demand at 3.01 to 4.48 mg L<sup>-1</sup>. Serving as the source water of the downstream WTP, the occurrence of odorous compounds in the reservoir effluents were monitored monthly from January 2017 to December 2018 to identify their potential threats. In total, 13 compounds were selected for evaluation (Table 1), which was identified as the potential odorants in a previous study [18].

Xin Che Dun (XCD) WTP is a newly built WTP operating from March 2018. Taking Jinze Reservoir as its source water, XCD has a treatment capacity of 160,000 m<sup>3</sup> day<sup>-1</sup>. Detailed operating parameters are listed in Table 2. From April 2018 to July 2018, samples were collected weekly from the plant influents and the effluents after each unit process (pre-ozonation, sedimentation, filtration, post-ozonation, GAC, and clean water basin) to evaluate the removal efficiency.

All water samples were collected in 500 mL amber glass bottles that were pre-washed using nonphosphate laboratory-grade detergent solution (Liquinox™) and thoroughly rinsed with deionized (DI) water to remove detergent residue [3, 52]. 200 mL of the sample was used for flavor profile analysis (FPA), and 10 mL was prepared for determining the odorant concentrations. All analyses were completed within 24 h after sample collection.

**Table 1** Information of 13 investigated odorants

Odorants	Odor description	OTC (μg L <sup>-1</sup> ) <sup>a,b</sup>	Observed range (μg L <sup>-1</sup> ) <sup>c</sup>
DMDS	Swampy, septic	0.03	0.67–82.70
DMTS	Swampy, septic	0.01	0.23–1.59
Hexanal	Herbal/flower/almond	4.5	0.43–30.99
EB	Plastic/oily/chemical	150	0.05–56.60
BA	Herbal flavor	4.5	3.26–251.62
1,4-DCB	Almond/sweet	4.5	0.19–19.26
BCIE	Chemical/swampy/septic	0.10	6.93–900.29
2-MP	Medicinal odor	14.73	2.76–4.81
3-MP	Medicinal odor	12.89	3.82–20.94
2-MIB	Musty	0.005	0.14–35.48
2,4-Decadienal	Fishy/oily	0.029	2.76–21.04
Indole	Stink	0.10	6.88–30.24
GSM	Earthy	0.004	0.25–15.43

<sup>a</sup> [18]

<sup>b</sup> [55]

<sup>c</sup> Based on yearly monitoring of the odorant concentrations in Jinze Reservoir (January 2018 to December 2018)

**Table 2** Operating parameters in the water treatment plant

Process	Designed contact time (min)	Coagulant (aluminum sulfate) concentration (mg L <sup>-1</sup> ) <sup>a</sup>	Ozone dosage (mg L <sup>-1</sup> ) <sup>a</sup>	Actual contact time (min)	Backwash frequency
Pre-ozonation	3	–	0–1.0	3.4	–
Flocculation	20	20–30	–	23	–
Sedimentation	120	–	–	137	–
Sand filtration	30	–	–	34	24–48 h
Post-ozonation	12	–	0–1.5	14	–
GAC	12	–	–	14	5–10 days

<sup>a</sup> Coagulant concentration and ozone dosage may vary according to the actual influent qualities

### Sensory evaluation of odor

Flavor profile analysis (FPA) was adopted for odor characterization [61]. A panel with five panelists identified each odor attribute and its intensity based on a seven-point intensity scale (1: odor threshold; 2 and 4: very weak to weak odor; 6 and 8: moderate odor; 10 and 12: strong odor). Flasks (500 mL) containing 200 mL of water sample were heated to 45 °C in a water bath prior to the analysis. During the evaluation, each panelist held the bottom of the flask with one hand, opened the glass stopper with the other hand, smelled the samples; and recorded the description and intensity of each sample. The result of each sample was finalized after a consensus had been reached in the panel.

### Quantification of odorants

Analyses of samples were accomplished with solid phase microextraction (SPME) followed by gas chromatography tandem mass spectrometry (GC–MS/MS) with a triple quadrupole instrument (Shimadzu GCMS-TQ8050 NX) [27]. In total, 13 odorants were analyzed including the earthy/musty odor substances 2-MIB and GSM as well as the chemical and septic odorants (sulfides, BCIE, and other aromatics and aldehydes) (Table 1). Water sample (10 mL) was added to a 20 mL CTC vial that was preloaded with 2.5 g of NaCl. The SPME fiber was inserted to the vial for extraction. The extracted substances were injected to the GC column (Agilent VF-624MS; 0.32 mm × 1.80 μm × 60 m) in splitless mode for separation with a 1 mL min<sup>−1</sup> carrier gas flow. The inlet temperature was set to be 250 °C. With an initial temperature of 40 °C, the column temperature was ramped 8 °C min<sup>−1</sup> to 260 °C with a 10 min hold. The temperatures of the quadrupole and the ion source were 150 °C and 230 °C, respectively. The selective ion monitoring (SIM) mode was employed in the quantitative analyses [66]. The qualification of the targeted odorant was based on its retention time, ion fragmentation, and the corresponding abundance [15, 54]. The concentration of the odorant was calculated using the calibration curve generated by the workstation with a range of external standards and their corresponding peak areas.

### Odor activity value (OAV)

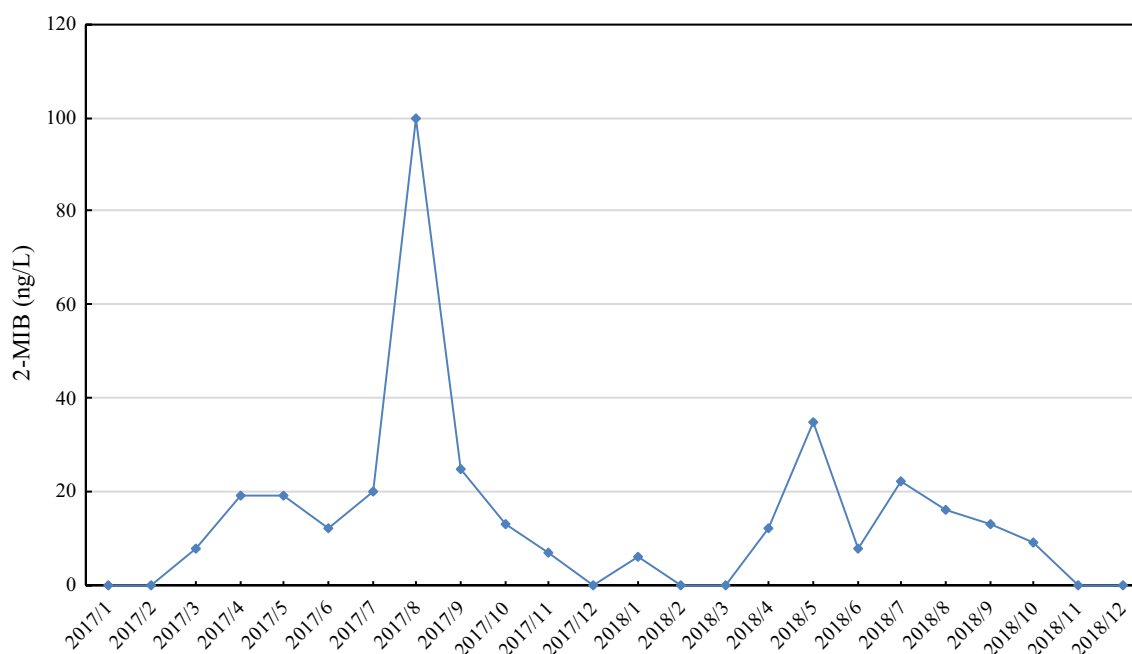
OAV was used to evaluate the contribution of the odor from each odorant. The value of OAV was calculated by dividing the measured concentration by its corresponding OTC. Odorants with OAVs at equal to or greater than one contribute greatly to the odor profile, while a value at less than one indicates a limited contribution [17].

## Results and discussion

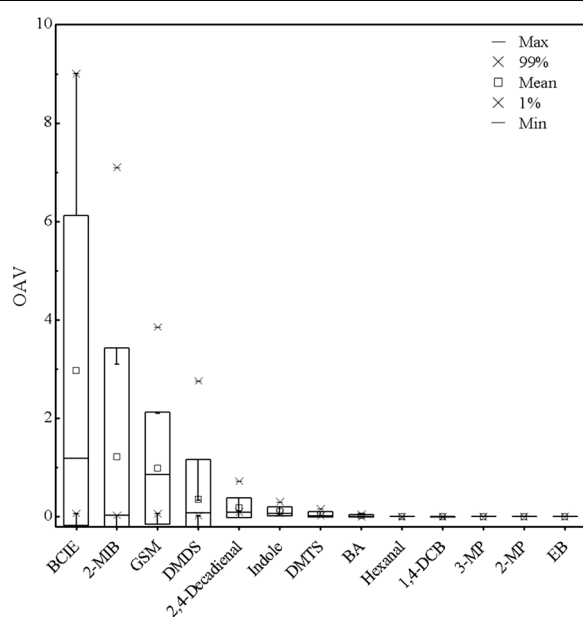
### Odor characterization and odorant identification in source water

Table 1 shows the results of the 13 targeted odorants in the WTP source water. As indicated, the concentration of 2-MIB increased in summer and autumn when algal blooms occurred relatively often. In this study, 2-MIB in Jinze Reservoir was approximately 100 ng L<sup>−1</sup> in August 2017 (Fig. 1), which is 20 times greater than its OTC (Table 1). GSM was detected in over 80% of the samples with approximately 40% exceeding the OTC (4 ng L<sup>−1</sup>). As an industrial chemical, BCIE was detected in all samples with relative high concentrations. Other odorants that were detected in more than 50% of the samples included thioethers (i.e., DMDS), aldehydes (i.e., hexanal and benzaldehyde (BA)), and benzenes (i.e., ethylbenzene (EB) and 1,4-dichloro-benzene (1,4-DCB)). Similarly to BCIE, the occurrence of these compounds may be related to industrial activities and discharges. For example, DMDS is a widely used soil fumigant [49] and can be produced in industrial process such as the wood-pulp industry and oil refineries [22]. DMDS was observed in swine facilities [35] as well. BA is related to the production of food preservatives, pharmaceuticals, and cosmetics [50]. EB is mostly used in producing styrene [5]. The presence of the various odorants in Jinze Reservoir is not surprising, as Huangpu River has long been reported to have issues regarding complex odorants [18, 46, 60]. Guo et al. [18] indicated that DMDS and BCIE were observed to be the primary contributors for chemical/septic smell, while 2-MIB and GSM contribute the most for musty/earthy odor. The complex odor issue in Huangpu River was also observed by Yang et al. [60] and Guo et al. [19], where nine odorous compounds belonging to benzenes, ethers, pyrazines, thioethers, and heterocyclic compounds were detected.

All targeted odorants were detected in the reservoir to some extent (0.05 to 900.29 ng L<sup>−1</sup>) (Table 1); their corresponding OAVs were calculated to further identify the compounds that contributed the most to the odor profile (Fig. 2). GSM and 2-MIB were observed to be the major earthy/musty odor contributor in Jinze, while BCIE and sulfide compounds such as DMDS were responsible for the chemical/septic odor. BCIE obtained an average OAV at as high as 2.97, which may result from its greater concentration (averaged at 297.18 ng L<sup>−1</sup>) compared to other odorous compounds. With an OTC at 5 ng L<sup>−1</sup>, 2-MIB, on the other hand, still showed an OAV at greater than one despite a relative low concentration (approximately averaged at 6 ng L<sup>−1</sup>). Compounds with low OTCs, though may have low detected concentration, cannot be neglected from odor concern. Moreover, seasonal effect needs



**Fig. 1** Concentration of 2-MIB in Jinze Reservoir from January 2017 to December 2018. A spike in 2-MIB concentration in August 2017 may relate to an algal bloom event



**Fig. 2** Odor activity values (OAVs) of targeted odorants in the reservoir arranging in a descending order. BCIE and 2-MIB were the two odorants with an average OAV at great than one indicating their strong contribution to the septic and the earthy/musty odor, respectively

to be taken into account as well. For example, greater 2-MIB concentrations in summer (Fig. 1) reflected a potential off-flavor event. Other odorous compounds

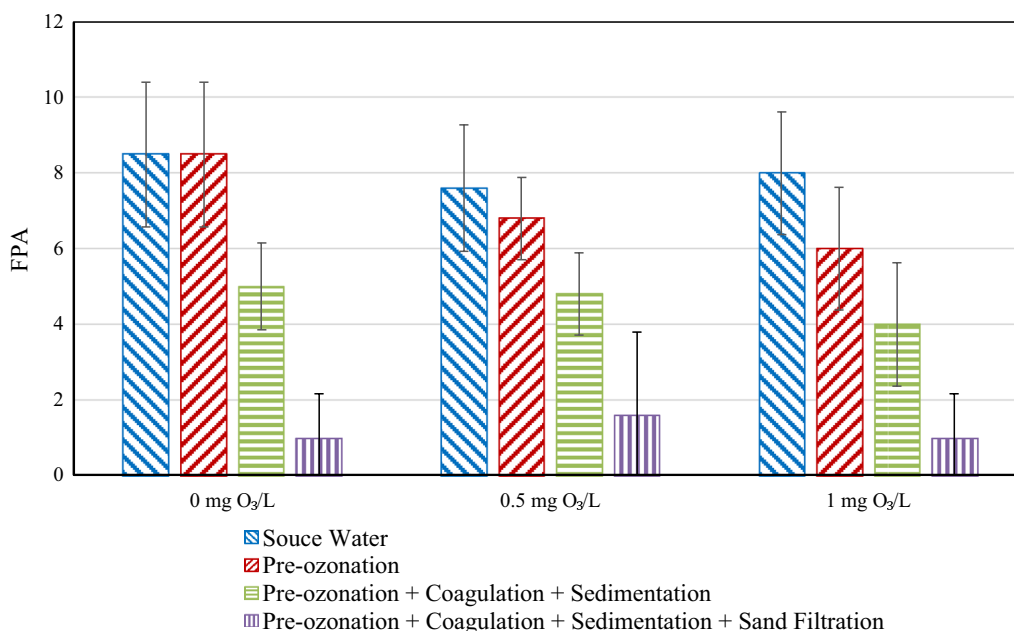
including DMDS and DMTS, 2,4-decadienal, indole, and BA, contributed to septic, fishy, stinky, and herbal flavor odors, respectively (Table 1 and Fig. 2); these compounds together formed a very complex odor in the source water.

#### Removal of odorous compounds in XCD WTP

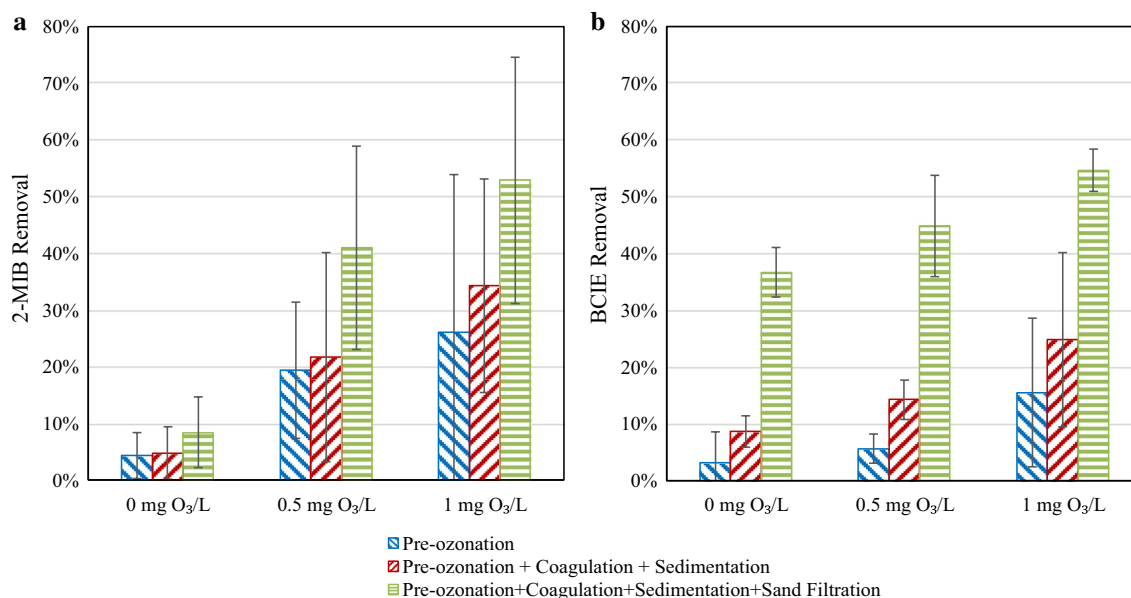
##### Preliminary evaluation of odor elimination

FPA was conducted to preliminarily evaluate the complex odor control efficiency in the WTP (Fig. 3). Source water showed an average odor intensity of eight indicating a moderate odor and the need for odor control. Compared to no pre-oxidation condition, application of pre-ozonation improved the odor reduction. Specifically, with a dosage of  $0.5 \text{ mg O}_3 \text{ L}^{-1}$ , FPA intensity of the source water was reduced to seven. Further increasing the dosage to  $1 \text{ mg O}_3 \text{ L}^{-1}$ , the intensity decreased to six. By combining subsequent coagulation and sedimentation process, the FPA intensity decreased to four, indicating a weak odor (Fig. 3). Greater performance was achieved after sand filtration with the intensity reduced to the threshold value (i.e., 1). Although pre-ozonation alone revealed some reduction in odor intensity, to reduce the intensity to an odor-free level, a combined process that involves additional advanced treatment such as post-ozonation and GAC needs to be considered. As 2-MIB and BCIE were the two major odor contributors, their removal efficiencies were investigated through the WTP





**Fig. 3** Evaluation of the FPA in the source water and after each unit treatment process in XCD WTP. The dosage of pre-ozonation was demonstrated on the X axis. FPA odor intensity at 0 indicates odor-free samples; at 1 indicates the odor threshold; at 2 to 4 indicates a very weak to weak odor; at 6 to 8 indicates a moderate odor; and at 10 to 12 indicates a strong odor

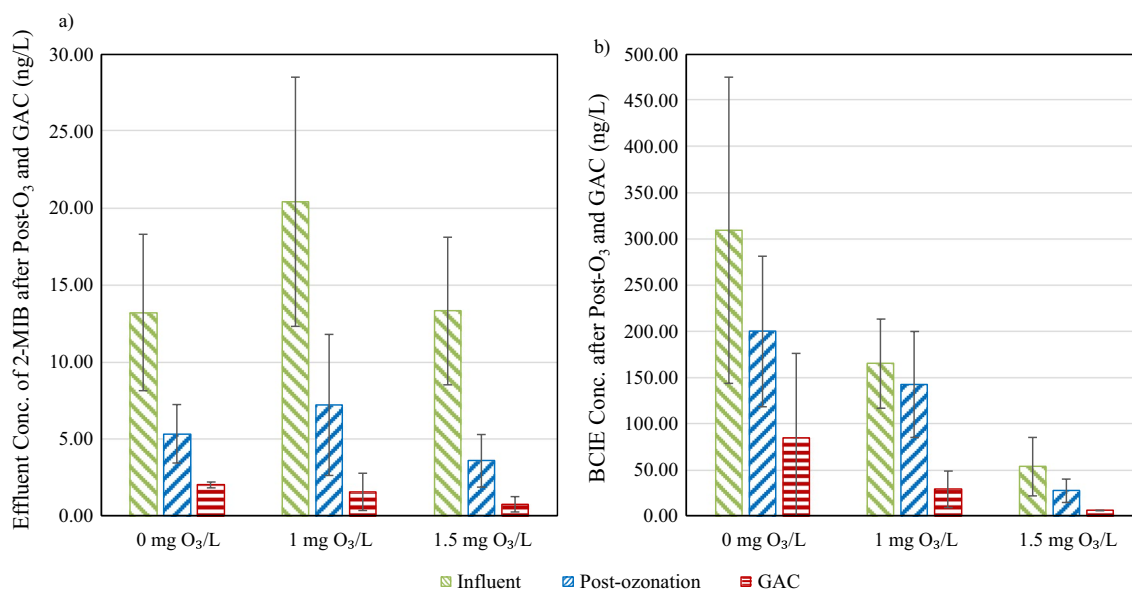


**Fig. 4** Removal of **a** 2-MIB and **b** BCIE shown as a function of pre-ozonation dosages. The removal efficiency of pre-ozonation alone was compared with a combined treatment process (i.e., pre-ozonation followed by coagulation, sedimentation, and sand filtration). Odorant removal in pre-ozonation alone with a zero dosage was due to the application of powdered activated carbon (10 mg L<sup>-1</sup>) at the intake of the source water (i.e., a pre-treatment process for the WTP)

to evaluate the odor reduction and optimize the applicable operational conditions in the existing water treatment facilities (Figs. 4 and 5).

#### Evaluation of pre-ozonation for odor removal

Removal of 2-MIB was less than 5% without the pre-ozonation (Fig. 4a). With subsequent treatment processes



**Fig. 5** Effluent concentration of **a** 2-MIB and **b** BCIE from post-ozone and GAC in XCD WTP; their average influent concentrations were shown as well for comparison

(i.e., coagulation, sedimentation, and sand filtration), the overall removal was still less than 10%, indicating the need to apply pre-ozone. Pre-ozone was investigated with dosages of 0.5 mg L<sup>-1</sup> and 1 mg L<sup>-1</sup>, which are the typical dosages applied in XCD WTP. Generally, increasing the dosage of pre-ozone demonstrated a greater removal. Specifically, removal increased to 19% with a 0.5 mg L<sup>-1</sup> pre-ozone dosage, and further improved to 26% with a dosage of 1 mg L<sup>-1</sup>. Limited removals were also observed in a pilot plant study conducted by Yang et al. [60], where 2-MIB removal was approximately 20% and 30% with a pre-ozone dosage of 0.5 mg L<sup>-1</sup> and 1 mg L<sup>-1</sup>, respectively. Natural organic matters (NOMs) in water consumed the ozone and OH<sup>•</sup> and inhibited the oxidation of the odorants [59]. Pre-ozone targeted mostly on the NOMs and revealed a competing effect on 2-MIB removal during degradation [65]. Xie et al. [59] observed a significant effect on the odorant reduction when NOM exceeded 1 mg L<sup>-1</sup>. With an influent total organic carbon (TOC) ranging from 3.03 to 4.00 mg L<sup>-1</sup> and an average turbidity at 19.35 ± 4.20 NTU in current study, 2-MIB was not effectively removed.

Coagulation and sedimentation have been reported to be ineffective in removing odorants [44]. However, adding a pre-oxidation and a subsequent filtration may improve the overall removals [10]. In this study, pre-ozone in combination with coagulation, sedimentation, and sand filtration achieved an average reduction of 41% with a pre-ozone dosage of 0.5 mg L<sup>-1</sup> and an average

removal at 52% with a dosage of 1 mg L<sup>-1</sup> (Fig. 4a). Pre-ozone broke down the large organic molecules (e.g., odorants such as 2-MIB) which may enhance their electrostatic interactions with coagulant flocs [36]. Though the combined treatment revealed an improvement, the removal was still insufficient as the effluent concentration of 2-MIB from the sand filtration was 5.60 ± 2.09 ng L<sup>-1</sup>, which is greater than its OTC.

Similar trends were observed in BCIE removals. Consistent with other studies [60], even though an improved reduction of BCIE was observed with an increasing pre-ozone dosage, the removal was still limited (Fig. 4b). Specifically, removal increased from 3 to 6% with a 0.5 mg L<sup>-1</sup> dosage, and further improved to 15% with a dosage of 1 mg L<sup>-1</sup>. BCIE was relatively resistant to pre-ozone as the removal was less than 20% with the greatest dosage; again the presence of NOM in the source water decreased the potential of ozonation to target on the odorants. Similarly, the combined treatment, i.e., ozonation followed by coagulation, sedimentation, and sand filtration, showed limited improvement. Greatest removal (i.e., 55%) was observed at the highest ozone dosage. However, with an effluent concentration of 152.40 ng L<sup>-1</sup> in the sand filtration, BCIE still exceeded the OTC after a series of treatment units. As both concentrations of 2-MIB and BCIE remained above their olfactory perception thresholds, the optimal dosage of pre-ozone was set to be 1 mg L<sup>-1</sup> (i.e., the greatest dosage in the applicable range of the WTP) and

the optimization of the post-ozonation dosage was performed based on this dosage.

#### **Evaluation of post-ozonation for odor removal**

The dosages of post-ozonation again were selected based on the typical operational range in XCD (i.e., 0.8 mg L<sup>-1</sup> to 1.5 mg L<sup>-1</sup>) used in the full-scale WTP. 2-MIB concentration was reduced to below the OTC with a post-ozonation dosage of 1.5 mg L<sup>-1</sup> (Fig. 5a).

Compared to 1.5 mg L<sup>-1</sup>, a dosage of 1 mg L<sup>-1</sup> did not show an acceptable effluent concentration (>5 ng L<sup>-1</sup>), because the 2-MIB in the influent was two times greater (up to approximately 30 ng L<sup>-1</sup>) on the day of sampling. Influent concentration affected the treatment efficiency significantly (Fig. 5a) [24, 39]. With a dosage of 1 mg O<sub>3</sub> L<sup>-1</sup>, Park et al. [39] observed a removal of less than 80%, when the influent 2-MIB concentration was 50 ng L<sup>-1</sup>; this removal increased to greater than 95% when a 1.5 mg L<sup>-1</sup> of ozonation was applied. 2-MIB is an aliphatic alcohol with a bicyclic structure. The hydroxyl group (–OH) attaching on the alkyl ring acts as an electron-withdrawing group. As compounds with no aromatic moieties but containing electron-withdrawing functional group are resistant to direct ozonation [58], 2-MIB was observed to be recalcitrant. The low reactivity was indicated by the low ozone rate constant ( $k_{O_3}=0.35$  to  $10\text{ M}^{-1}\text{ s}^{-1}$ ) as well [40, 57]. Ozone is a selective oxidant with rate constant ranging from  $10^{-1}$  to  $10^6\text{ M}^{-1}\text{ s}^{-1}$  [65, 68] while hydroxyl radical (OH·) is less selective with rate constant ( $k_{OH}$ ) differing by less than one order of magnitude [58]. An indirect reaction with OH· may dominate over the ozone during 2-MIB oxidation. Westerhoff et al. [57] reported the  $k_{OH}$  ranging from  $1 \times 10^9$  to  $6 \times 10^9\text{ M}^{-1}\text{ s}^{-1}$  for 2-MIB in bulk water samples that were collected from five different surface water locations; similar results were observed by Ma et al. [34] as well. OH· acted as the most significant reactive species during the degradation of the odorants in the ozonation process. OH· is generated from the decay of ozone in water. As a result, when the influent concentration of 2-MIB was high, a moderate dosage of post-ozonation (1 mg L<sup>-1</sup>) did not produce an adequate amount of OH· to oxidize the recalcitrant compound. In such case, a greater dosage (i.e., 1.5 mg L<sup>-1</sup>) or a subsequent advanced process (i.e., GAC) is needed, as the concentration of OH· increases with a greater ozone dosage [57].

Similarly, the greatest BCIE removal efficiency was obtained with a 1.5 mg L<sup>-1</sup> dosage. After adding the post-ozonation, the effluent concentration was reduced to 26.74 ng L<sup>-1</sup>, below the OTC (Fig. 5b). Compared to the results without post-ozonation, adding 1 mg L<sup>-1</sup> ozone though revealed a greater reduction in BCIE, the 142.21 ng L<sup>-1</sup> effluent concentration was still above the

OTC (100 ng L<sup>-1</sup>). Again, when the upstream has a sudden spike in odorant concentration, the moderate post-ozonation dosage in the WTP would not satisfy the odor requirement. BCIE is an aliphatic compound with two chlorine functional groups; these two polar groups are electron-withdrawing and leads to a low reactivity towards ozone [7]. With the source water concentration at up to approximately 500 ng L<sup>-1</sup>, a 1.5 mg L<sup>-1</sup> dosage of post-ozonation is recommended for treating BCIE. Otherwise, with a moderate post-ozonation dosage, a subsequent process for the removal of the remaining BCIE is necessary.

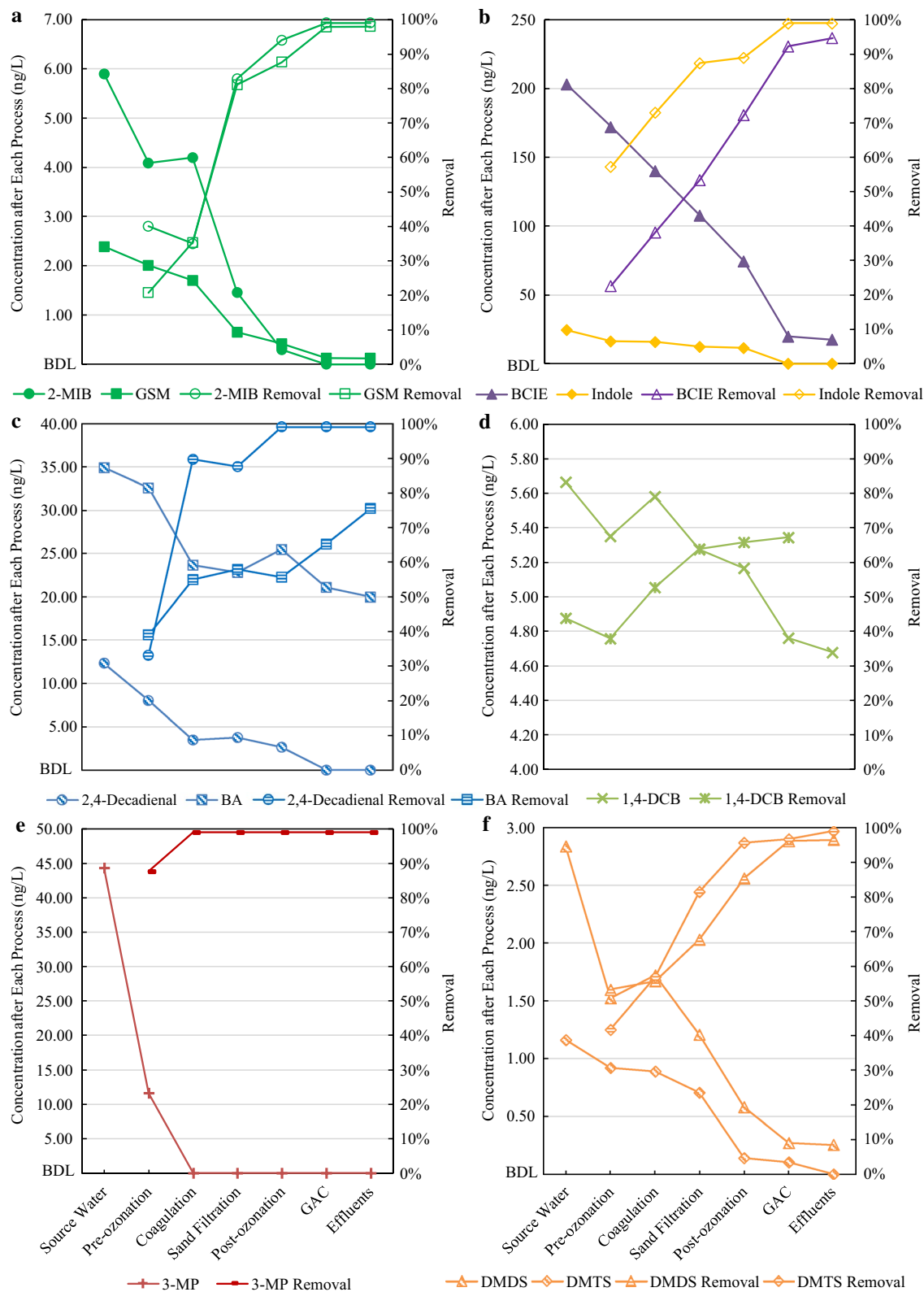
#### **Suggestions for complex odor control**

To remove the complex odors simultaneously, all the odorants need to be removed below their OTCs. As discussed above, a post-ozonation dosage of 1 mg L<sup>-1</sup> is insufficient to remove the complex odorants when a peak influent concentration is observed. The effectiveness of ozonation was significantly affected by the influent conditions. However, after GAC process (size: 8 × 30 Mesh; empty bed contact time (EBCT): 14 min; filter velocity: 10 m h<sup>-1</sup>), all the odorant compounds were removed to below the OTCs regardless of the source water conditions (Fig. 5). Consistent with other studies [17, 39, 60, 63], GAC was applicable for removing odorants with a wide range of influent concentration ( $10^0$  to  $10^2\text{ ng L}^{-1}$ ). But breakthrough of the GAC needs to be considered. Post-ozonation prior to GAC was observed to improve the breakthrough time as well as increase the removal efficiency in GAC [37]. Therefore, using GAC as the final polishing step, a post-ozonation (1 mg O<sub>3</sub> L<sup>-1</sup>) was selected for the optimal treatment train to save the cost and minimize the ozonation byproducts (i.e., pre-ozonation (1 mg O<sub>3</sub> L<sup>-1</sup>) + coagulation + sedimentation + sand filtration + post-ozonation (1 mg O<sub>3</sub> L<sup>-1</sup>) + GAC (EBCT: 14 min)), while at the same time a desirable performance of complex odor control was also achieved.

#### **Continuous operation for verification**

The optimal treatment train was operated continuously for 6 months to verify the performance of the complex odor control in XCD (Fig. 6). The results indicated that the natural odorants (2-MIB and GSM) were removed at greater than 99% and 98%, respectively, both below their odor thresholds (Fig. 6a). The optimal treatment train demonstrated a good removal for BCIE (ether), the major chemical/septic odor contributor as well. A removal of 23% and 72% was achieved in pre- and post-ozonation, respectively, the GAC further ensured a 92% reduction, achieving a desirable effluent concentration for BCIE (Fig. 6b). After the treatment, indole was not detected in the effluent (Fig. 6b). Aldehydes (2,4-decadienal and





**Fig. 6** Concentration after each process and average removal shown with corresponding groups. Samples were collected monthly from July 2018 to December 2018 for verification of treatment efficiency. Since 2-MP (phenol) was only detected in two samples and hexanal (aldehyde) and EB (benzene) were not detected in all source water samples, they were not shown in this figure

BA) were removed at greater than 99% and 75%, respectively, with the effluent concentrations at below their corresponding OTCs (Fig. 6c). Benzene (1,4-DCB) was removed at 67%, however, the effluent concentration was 4.68 ng L<sup>-1</sup>, which is significantly less than its threshold value (4500 ng L<sup>-1</sup>) (Fig. 6d). Pre-ozonation in combination with coagulation showed an excellent removal efficiency for 3-MP (phenol). A greater than 99% reduction was observed with 3-MP removed to below the detection limit (Fig. 6e). Removal of DMDS and DMTS (thioethers) was approximately 60% after pre-ozonation while after post-ozonation this removal increased to greater than 85% and further improved to greater than 95% after GAC (Fig. 6f); their concentrations in the effluents were 0.25 ng L<sup>-1</sup> and below detection limit, respectively. The investigated odorants were effectively removed to below their OTCs in the effluents. Overall, the treatment train after optimization is a great solution for complex odor control.

## Conclusions

This study investigated the fate of the odorants in an existing WTP. The effectiveness of the current treatment units was evaluated and the optimal strategy to resolve the complex odor issue was explored. 2-MIB and BCIE were observed to be the major contributors of the musty/earthy and chemical/septic odors, respectively. The application of post-ozonation was the key for improving the odorant removals. With a 1.5 mg L<sup>-1</sup> dosage of post-ozonation, all odorants were removed below the OTCs. When applying a 1 mg L<sup>-1</sup> post-ozonation dosage, by combining GAC, the complex odor issue was well controlled regardless of the odorant concentrations in the influents. The optimal and cost-effective treatment train for a promising complex odor control involved a 1 mg O<sub>3</sub> L<sup>-1</sup> pre-ozonation in combination with coagulation, sedimentation, and sand filtration, followed by a post-ozonation with a dosage of 1 mg O<sub>3</sub> L<sup>-1</sup>, and a GAC process (EBCT: 14 min); this solution was verified in an extended 6-month study where all the odorants were removed simultaneously.

## Abbreviations

–OH: Hydroxyl group; 1,4-DCB: 1,4-Dichloro-benzene; 2-MIB: 2-Methylisoborneol; 2-MP: 2-Methyl-phenol; 3-MP: 3-Methyl-phenol; BA: Benzaldehyde; BCIE: Bis(2-chloroisopropyl) ether; BDL: Blow detection limit; DI: Deionized; DMDS: Dimethyl disulfide; DMTS: Dimethyl trisulfide; EB: Ethylbenzene; EBCT: Empty bed contact time; FPA: Flavor profile analysis; GAC: Granular activated carbon; GC–MS/MS: Gas chromatography tandem mass spectrometry; GSM: Geosmin; KMnO<sub>4</sub>: Potassium permanganate; *k*<sub>O<sub>3</sub></sub>: O<sub>3</sub> rate constant; NaCl: Sodium chloride; Na<sub>2</sub>SO<sub>4</sub>: Anhydrous sodium sulfate; NOM: Natural organic matter; OAV: Odor activity value; OH·: Hydroxyl radical; OTC: Odor threshold concentration; SPME: Solid phase microextraction; TOC: Total organic carbon; WTP: Water treatment plant; XCD: Xin Che Dun.

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## Authors' contributions

PX designed the experiments, analyzed the data, and wrote the paper. SYZ, HY, LJ and ZW mainly performed the FPA of typical odorants. DQY, JWY and DZ reviewed and edited the manuscript. All authors read and approved the final manuscript.

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Not applicable.

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Not applicable.

## Competing interests

The authors declare that they have no competing interests.

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