



Effect of disinfection method on odor and disinfection byproduct control in drinking water treatment

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Received 30 December 2014; Accepted 7 August 2015

ABSTRACT

Odor problems in drinking water have drawn worldwide concern. Chlorine oxidation of odor in water can produce disinfection byproducts (DBPs). In this study, effect of disinfection method (common disinfections (sodium hypochlorite disinfection, chloramine disinfection, chlorine dioxide disinfection), synergistic disinfection of chlorine dioxide and sodium hypochlorite, sequential chlorine disinfection, and UV synergistic disinfection) on odor and DBP control in drinking water treatments were investigated. The results showed that chloramine and chlorine dioxide could better control odor and DBP. The best ratio of sodium hypochlorite to chlorine dioxide (synergistic disinfection) for odor and DBP control was 1:1. UV synergistic disinfection (compared with chloramine and chlorine dioxide) had little effect on odor and DBP control. Disinfection methods showed little effect on odor and DBP control during pre-chlorination + conventional water treatment process. Odor and DBP could be efficiently controlled by chloramine and chlorine dioxide disinfection during pre-ozonation followed by biological activated carbon + O₃ process.

Keywords: Disinfection method; Odor; DBP; Drinking water treatment

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Presented at the 7th International Conference on Challenges in Environmental Science and Engineering (CESE 2014) 12–16 October 2014, Johor Bahru, Malaysia

1. Introduction

Odor in drinking water not only affects its potability, but also does harm to human health, which raises worldwide concerns. World Health Organization and United States Environmental Protection Agency (USEPA) both have strict standards for the value of odor. The source of odor in the water body is mainly derived from two ways: naturally occurring odor (mainly caused by aquatic organisms such as algae and fungi) and anthropogenic odor (organic matter in industrial wastewater and domestic sewage, which are directly discharged into the water body, or chemical products made of natural organic matters and chemical reagent during the water treatment process).

There are many sensory and analytical methods available for detecting odor in drinking water. Sensory detecting methods include threshold odor number analysis [1] and flavor profile analysis (FPA) [2]. Instrumental detecting methods include closed-loop stripping analysis [3], purge & trap [4], liquid–liquid extraction [5], solid-phase extraction [6], solid-phase microextraction [4], liquid-phase microextraction [7], stir bar sorptive extraction [8], gas chromatography–mass spectrometry [9], etc. Furthermore, there are combined sensory and instrumental methods [10] and enzyme-linked immunosorbent assay [11]. All these methods have their own advantages and disadvantages. The sensory methods are superior in the practical application, because the human nose is more sensitive than most available instrumentations and rapid results can guide the corresponding utilities to take timely action [2]. Considering the accuracy among the sensory methods, FPA is the best method.

Concentration and threshold of odorous compounds in the water body are extremely low, and traditional water treatments exhibit limited the removal efficiency. Thus, appropriate technologies should be considered according to the practical characteristics of water resources. Powered activated carbon (PAC) for its economy and excellent adsorbability has been widely used as the adsorbent for odorous compounds [12,13]. Sometimes, odor cannot be effectively removed by PAC alone, and other technologies combined with PAC may be employed, such as $O_3 + PAC$ [1] and $KMnO_4 + PAC$ [14]. Oxidation (by O_3 , Cl_2 , ClO_2 , etc.) [15] and advanced oxidation process (such as $UV + H_2O_2$, $UV + O_3$, $O_3 + H_2O_2$, and vacuum ultraviolet light) [16–18] are also used to treat odorous compounds such as 2-methylisoborneol (2-MIB) and geosmin (GSM) in the water. Furthermore, 2-MIB and GSM can be removed by the biological process (biological sand filter, biological aerated filter, granular activated carbon [GAC] filter, etc.) [19–21].

However, few technologies could remove odor and odorous compounds completely, an extremely low concentration of the residual odorous compounds (2-MIB, GSM, etc.) could still produce smelly odor in drinking water. Furthermore, DBPs were formed when disinfectants (chlorine, ozone, chlorine dioxide, or chloramines) react with naturally occurring organic matter, anthropogenic contaminants, bromide, and iodide during the production of drinking water [22]. Emmert et al. [23] found sodium hypochlorite feedstocks might be a significant source of three haloacetic acids (HAAs) species in finished water for utilities that used hypochlorite ion feedstocks. SUVA correlated strongly with trihaloacetic acids and unknown total organic halogen yields, whereas weak correlations were observed between SUVA and trihalomethane (THM) and dihaloacetic acid yields during chlorination [24]. The degradation of phenylurea herbicides by chlorine dioxide and the formation of DBPs from subsequent chlorination or chloramination were investigated by Tian et al. [25]. Zhang et al. also found that the UV/chlorine process produced more haloacetonitriles than chlorination [26]. Furthermore, Zhai et al. [27,28] found brominated disinfection byproducts (Br-DBPs) were generally more cytotoxic and genotoxic than their chlorinated analogs. The quantity of odorous compounds might not be reduced significantly by disinfection process. However, it is worth well to investigate whether the pungent smell could be concealed by disinfection process.

In this study, common disinfections (sodium hypochlorite disinfection, chloramine disinfection, chlorine dioxide disinfection), synergistic disinfection of chlorine dioxide and sodium hypochlorite, sequential chlorine disinfection, and UV synergistic disinfection were selected as representative disinfection processes, which were frequently used in water treatment plants. After disinfected by these different disinfection methods, odor of finished water (measured by FPA method) and DBP formation were investigated to study the effect of disinfection processes on odor and DBP control. Moreover, in practical water treatment plants with different treatment technologies, odor and DBP control by different disinfection methods were also studied to improve the potability.

2. Materials and methods

2.1. Materials

All chemical reagents were supplied by Sinopharm Chemical Reagent Co. Ltd (Shanghai, China), and were analytical grade. Deionized water was obtained from a Milli-Q ultrapure water purification system.

Chloramine solution: sodium hypochlorite solution and ammonium sulfate solution in accordance with Cl_2 : N = 4:1 (mass ratio) reacted by stirring for 10 min at pH 9 [29], and monochloramine was the main product. Chlorine dioxide was prepared by the reaction of 25 mL hydrochloride (3 mol/L) and sodium chlorite solution (10 g sodium chlorite dissolved in 750 mL water).

2.2. Flavor profile analysis (FPA)

FPA was used to quantify the odor intensity of water and required a minimum of four trained panelists [30] as described in Standard Method 2170B [30]. Panelists defined an intensity rating to the odor of each water sample by a 7-point category scale: 0 = not detectable odor, 2 = very weak, 4 = weak, 6 = weak-to-moderate, 8 = moderate, 10 = moderate-to-strong, 12 = strong. Ten panelists were selected from students and faculties from the water treatment plants. They had an age range of 18–60 and consisted of 5 males and 5 females. After systemic FPA training [2,31], these 10 panelists began to test the odor of water samples.

FPA was conducted using 200 mL of odorant solutions in 500-mL wide-mouth Erlenmeyer glass flasks. Samples were warmed inside a water bath ($45 \pm 1^\circ\text{C}$) for 30 min and then FPA odor rating value to each water sample was assigned by panelists, ranging from 0 to 12 (even numbers). In the end, a mean and standard deviation was calculated for each water sample.

2.3. Analysis methods

THMs (including trichloromethane) and HAAs (including dichloroacetic acid, trichloroacetic acid, haloacetic acids, choral hydrate) as DBPs were analyzed by a gas chromatograph (Agilent 6890) with an electron capture detector, based on USEPA methods 551.1 and 552.4 [32,33].

2.4. Characteristics of water

All raw water of batch experiments was derived from the effluent of biological activated carbon (BAC) + O_3 process (principal process of a certain drinking water treatment plant). Characteristics of the water were $\text{TOC} = 2.978 \text{ mg/L}$, UV absorbance = 0.018 cm^{-1} , $\text{NH}_3\text{-N} = 0.24 \text{ mg/L}$, turbidity = 0.1 NTU, pH = 7.018 and $T = 25^\circ\text{C}$.

There were two water treatment processes (old process: coagulation + sedimentation + filtration + disinfection, new process: O_3 + BAC) in drinking water treatment plant E for treating river D. The raw water from the effluent of filters (old process) with $\text{TOC} = 4.11 \text{ mg/L}$, UV absorbance = 0.068 cm^{-1} , $\text{NH}_3\text{-}$

$\text{N} = 0.58 \text{ mg/L}$, turbidity = 1.8 NTU, pH 6.813 and $T = 25^\circ\text{C}$. O_3 + BAC process was the new and principal process and characteristics of the raw water were the same as in batch experiments.

2.5. Experimental process

2.5.1. Common disinfection

Referred to the traditional disinfection dosage in the water plants, the dosage of sodium hypochlorite and chloramine were selected as 1–2 mg/L, the dosage of chlorine dioxide was 0.5–1.0 mg/L.

Five-hundred milliliter water samples were input into some beakers (1,000 ml). Then, 1.0, 1.5, and 2.0 mg/L sodium hypochlorite, 1.0, 1.5, and 2.0 mg/L chloramine, and 0.5, 0.75, and 1.0 mg/L chlorine dioxide were added into the corresponding beakers. All beakers were sealed by sealing films and were kept in dark for 30 min reaction. In the end, 200 ml of finished water was chosen for FPA and disinfection analysis.

Chloramine was measured using an HACH Pocket Colorimeter II (Model 46700-001, USA). Chlorine dioxide was measured using an HACH Pocket Colorimeter II (Model 5953051, USA).

2.5.2. Sequential chlorine disinfection

Five-hundred milliliter water samples were input into some beakers (1,000 ml). Then, 2.5 mg/L sodium hypochlorite, 2.5 mg/L sodium hypochlorite with ammonium sulfate solution (in accordance with Cl_2 : N = 4:1 (mass ratio)) reacted 15 min for chloramine, and 2.5 mg/L chloramine were added into the corresponding beakers. The following steps were the same as 2.5.1.

2.5.3. Synergistic disinfection of chlorine dioxide and sodium hypochlorite

Five-hundred milliliter water samples were input into some beakers (1,000 ml). Then, 0.4 mg/L sodium hypochlorite + 1.2 mg/L chlorine dioxide (1:3), 0.8 mg/L sodium hypochlorite + 0.8 mg/L chlorine dioxide (1:1), 1.2 mg/L sodium hypochlorite + 0.4 mg/L chlorine dioxide (3:1), 1.6 mg/L sodium hypochlorite, and 1.6 mg/L chlorine dioxide were added into the corresponding beakers. The following steps were the same as 2.5.1.

2.5.4. UV synergistic disinfection

Five-hundred milliliter water samples were input into some beakers (1,000 ml). Two beakers were irradiated by UV for 90 s and then 2.0 mg/L

chloramine and 1.5 mg/L chlorine dioxide were added into the corresponding beakers. The other two beakers without UV irradiation were added with 2.0 mg/L chloramine and 1.5 mg/L chlorine dioxide, respectively. The following steps were the same as 2.5.1.

The UV irradiation system consisted of a 9-L stainless steel reactor (i.d. = 20 cm, height = 32 cm) with a 400-mL quartz tube (i.d. = 4.5 cm) fixed in the center and was operated at room temperature (25°C). Four low-pressure Hg UV lamps (TUV 11 W T5 4P-SE, Philips, Netherlands) with quartz sleeves were symmetrically fixed in the center of the reactor. The UV intensity was changed by turning on different numbers of UV lamps and detected using a light intensity meter (UV-C luxometer, Photoelectric Instrument Factory of Beijing Normal University, Beijing, China) at 254 nm.

3. Results and discussions

3.1. Effect of common disinfection on odor and DBP control

As shown in Fig. 1(a), the dosage of sodium hypochlorite increased from 1 to 2 mg/L, the average FPA intensities were 6, 8, and 8, respectively. Weak chlorine odor could be noticed at 1 mg/L sodium hypochlorite, and chlorine odor intensity was enhanced with the increased dosage of sodium hypochlorite. The similar tendency was also found when chloramine was added. However, FPA intensities were not influenced by the dosage of chlorine dioxide, all of which were 6 (obvious earthy odor) at different dosages (0.5, 0.75, and 1 mg/L). So odor control by common disinfection followed the sequence: chloramine > chlorine dioxide > sodium hypochlorite.

THMs, which were produced by disinfection with chloramine and chlorine dioxide, were all less than 4 µg/L. All corresponding HAAs were no more than 3 µg/L. The DBP production was little affected by the dosages of chloramine and chlorine dioxide. However, THMs and HAAs were more than 10 and 20 µg/L, which were produced by disinfection of sodium hypochlorite. More amounts of HAAs were found with the increased dosage of sodium hypochlorite, but THMs remained unchanged in Fig. 1(b). So DBP control by common disinfection followed the sequence: chloramine ≈ chlorine dioxide > sodium hypochlorite. Bond et al. [34] found DBP formation was much more reduced when considering pre-formed monochloramine compared to chlorine. Zhang et al. [35] found chlorine dioxide could effectively control DBP, especially trihalomethanes (I-THMs). Meanwhile, Tian

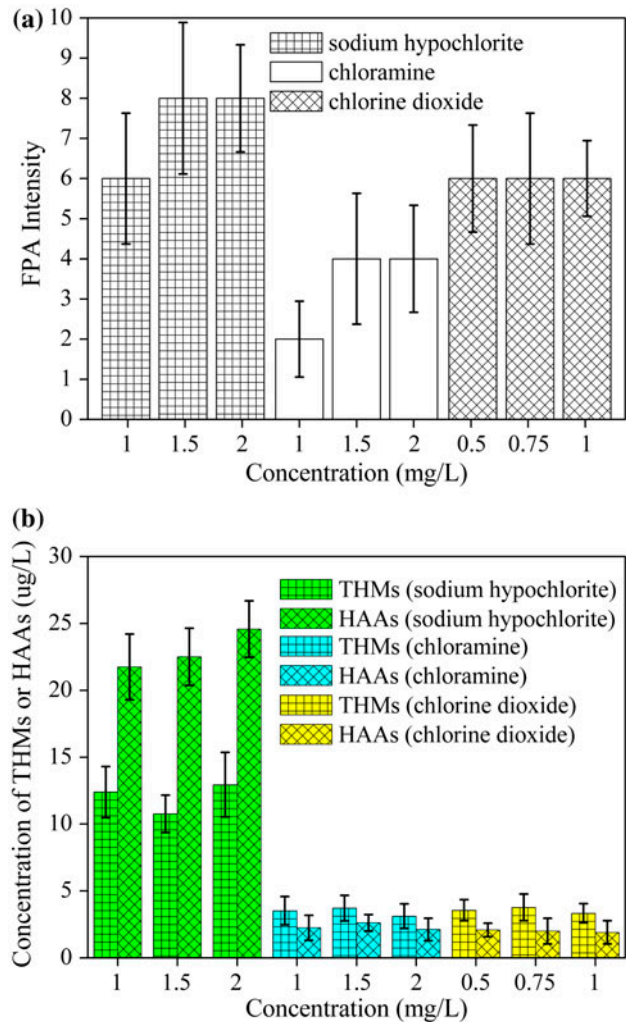


Fig. 1. Effect of common disinfection on odor (a) and DBP control (b).

et al. [36] also found DBP control by chlorine dioxide was much better than that of chlorine.

3.2. Effect of sequential chlorine disinfection on odor and DBP control

Based on the previous batch experiments, 15 min transformation time of sodium hypochlorite to chloramine was best for odor control. Meanwhile, the initial dosage of sodium hypochlorite (1.5, 2.0, 2.5, and 3.0 mg/L) had little effect on odor control in sequential chlorine disinfection (data not shown).

It is shown in Fig. 2(a), that sequential chlorine disinfection (15 min transformation time by 2.5 mg/L sodium hypochlorite) and 2.5 mg/L chloramine could effectively control odor, with the lowest average FPA

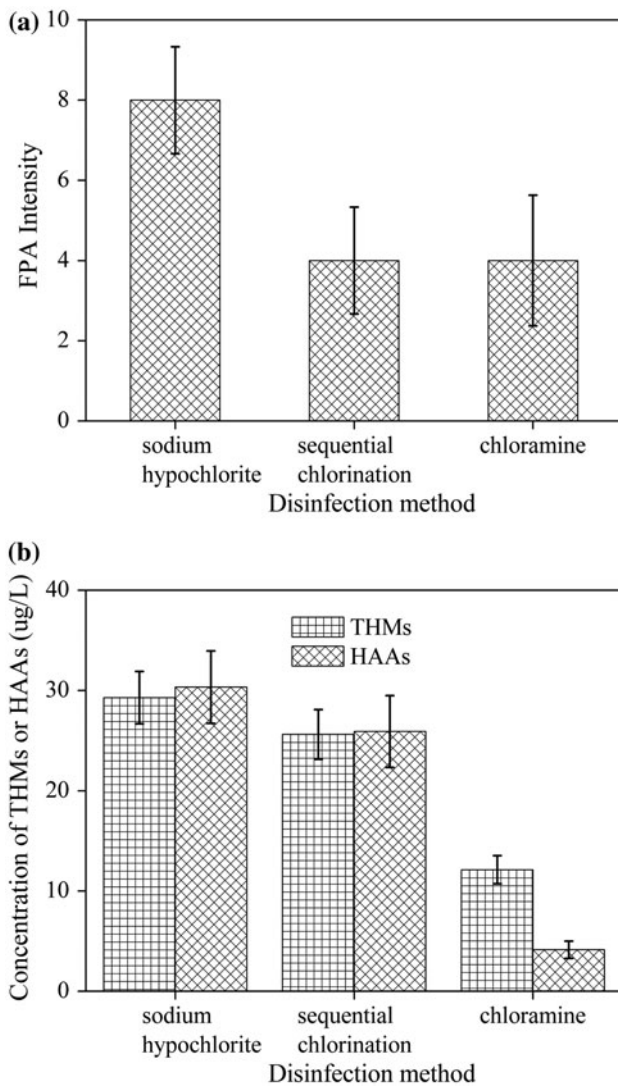


Fig. 2. Effect of sequential chlorine disinfection on odor (a) and DBP (b) control.

intensities of 4 and 4 compared with the average FPA intensity of 8 by 2.5 mg/L sodium hypochlorite disinfection. The maximum level of DBPs (THMs: $29.3 \pm 2.6 \mu\text{g/L}$ and HAAs: $30.3 \pm 3.6 \mu\text{g/L}$) was formed after 2.5 mg/L sodium hypochlorite disinfection, and minimum THMs ($12.1 \pm 1.4 \mu\text{g/L}$) and HAAs ($4.1 \pm 0.9 \mu\text{g/L}$) were produced by 2.5 mg/L chloramine disinfection. Sequential chlorine disinfection led to medium DBP formation (THMs: $25.6 \pm 2.5 \mu\text{g/L}$ and HAAs: $25.9 \pm 3.6 \mu\text{g/L}$), which was slightly lower than that of DBP formation caused by 2.5 mg/L sodium hypochlorite disinfection. It was consistent with the research of Bond et al. [34], who showed chloramination was effective at reducing the concentrations of THMs and haloacetonitriles in drinking water compared with chlorination.

3.3. Effect of synergistic disinfection of chlorine dioxide and sodium hypochlorite on odor and DBP control

The ratios of sodium hypochlorite to chlorine dioxide (1:1 and 3:1) were best for odor control in Fig. 3(a), and the corresponding average FPA intensities were 2 and 2, respectively. The average FPA intensities of sodium hypochlorite to chlorine dioxide (1:3) and 1.6 mg/L chlorine dioxide were 4 and 4, respectively. The maximum FPA intensity (6) was found at 1.6 mg/L sodium hypochlorite disinfection. So the odor (the FPA intensity) could be controlled by adjusting the ratio of sodium hypochlorite to chlorine dioxide.

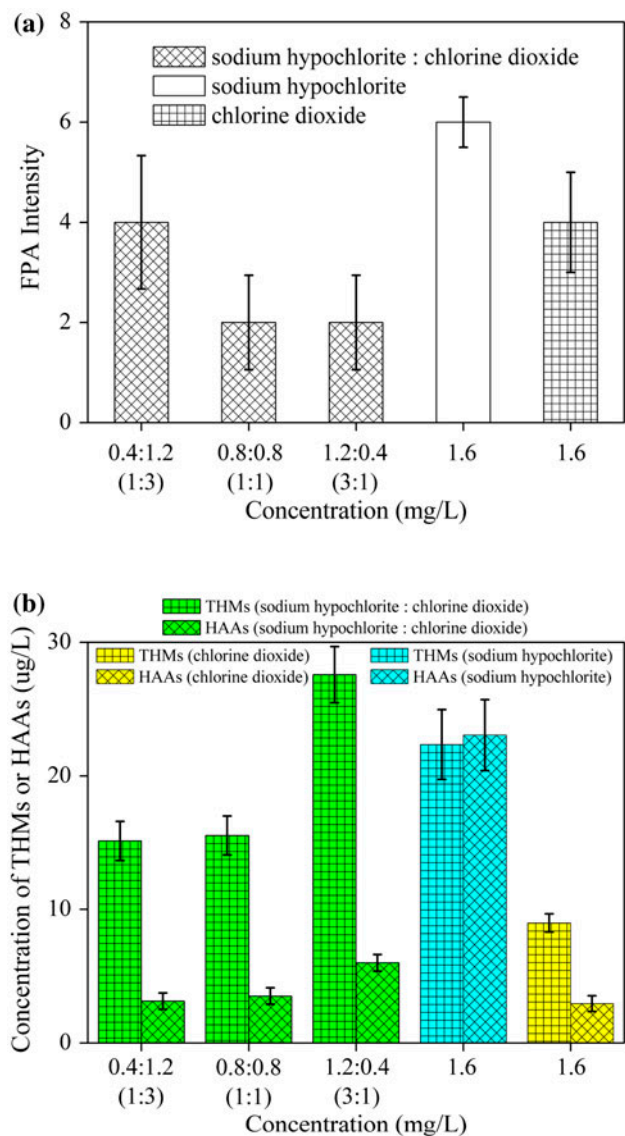


Fig. 3. Effect of synergistic disinfection of chlorine dioxide and sodium hypochlorite on odor (a) and DBP (b) control.

As shown in Fig. 3(b), DBP formation (THMs: $9.0 \pm 0.7 \mu\text{g/L}$, HAAs: $2.9 \pm 0.6 \mu\text{g/L}$) was the least, when the water was disinfected by 1.6 mg/L chlorine dioxide. Disinfection by sodium hypochlorite to chlorine dioxide (1:3 and 1:1) generated 68% THMs, 7% HAAs and 72% THMs, 21% HAAs (THMs: $15.1 \pm 1.5 \mu\text{g/L}$, HAAs: $3.1 \pm 0.6 \mu\text{g/L}$ and THMs: $15.5 \pm 1.5 \mu\text{g/L}$, HAAs: $3.5 \pm 0.6 \mu\text{g/L}$) more than that of 1.6 mg/L chlorine dioxide. The most amounts of DBP formation were produced by 1.6 mg/L sodium hypochlorite and sodium hypochlorite to chlorine dioxide (3:1), which were THMs: $22.4 \pm 2.6 \mu\text{g/L}$, HAAs: $23.1 \pm 2.7 \mu\text{g/L}$ and THMs: $26.6 \pm 2.1 \mu\text{g/L}$, HAAs: $6 \pm 0.6 \mu\text{g/L}$. Due to the residual chlorine in the water, the chlorite was oxidized to form chlorine dioxide in synergistic disinfection of chlorine dioxide and sodium hypochlorite. Because sodium hypochlorite converted chlorite to chlorine dioxide, and became chloride itself. The concentration of sodium hypochlorite decreased, and the concentration of chlorine dioxide increased. Compared with sodium hypochlorite, chlorine dioxide (at the same concentration) generated less THMs and HAAs. Thus, the DBP formation decreased after the conversion. With the increased dosage of sodium hypochlorite, THMs increased much more than HAAs. THMs and HAAs, which were produced by synergistic disinfection of sodium hypochlorite and chlorine dioxide (1:1), reduced by 31 and 85% compared with that of 1.6 mg/L sodium hypochlorite. Therefore, odor and DBP formation could be controlled by adjusting the ratio of sodium hypochlorite to chlorine dioxide. This result was consistent with Wang et al. [37], who showed the combination of chlorine dioxide and sodium hypochlorite for disinfection of drinking water could achieve better disinfection efficiency and decrease disinfection byproducts.

3.4. Effect of UV synergistic disinfection on odor and DBP control

It can be seen in Fig. 4, odor was reduced by UV pretreatment. However, DBP control was not obviously affected by UV synergistic disinfection compared with chloramine and chlorine dioxide disinfection. The average FPA intensities for disinfection of UV + 2 mg/L chloramines and UV + 2 mg/L chlorine dioxide were both 2, comparing with 4 for either 2 mg/L chloramine or 2 mg/L chlorine dioxide. All these disinfection methods (UV + 2 mg/L chloramine, 2 mg/L chloramine, UV + 2 mg/L chlorine dioxide and 2 mg/L chlorine dioxide) had similar DBP formation control, with corresponding THMs: 14.4 ± 1.3 , 14.9 ± 1.3 , 14.4 ± 1.0 , $14.4 \pm 1.1 \mu\text{g/L}$ and HAAs: 2.4 ± 0.5 , 2.8 ± 0.6 , 3.0 ± 0.6 , $2.8 \pm 0.6 \mu\text{g/L}$.

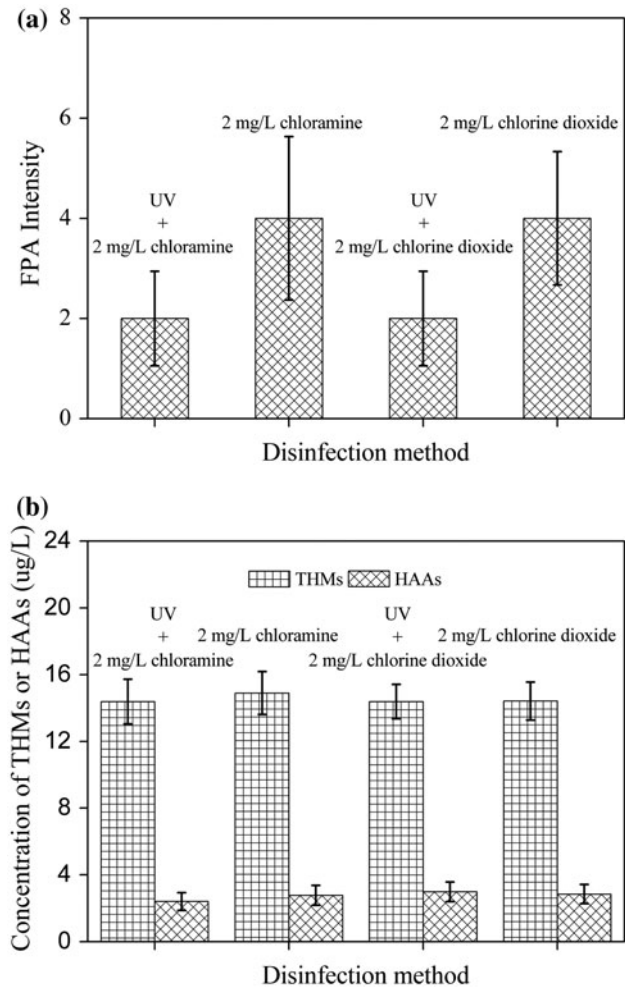


Fig. 4. Effect of UV synergistic disinfection on odor (a) and DBP (b) control.

Unlike the common disinfection, UV disinfection inactivated the bacterium via the power of irradiation, by changing the structure of the cell and impacting its normal function. However, the reaction between organic matters in water did not occur under this frequency UV irradiation. Therefore, compared with chloramine and chlorine dioxide disinfection, UV synergistic disinfection had certain effect on odor control, but little effect on DBP control.

3.5. Effect of the water treatment process on odor and DBP control

3.5.1. Old water treatment process (conventional water treatment process)

Pre-chlorination (2.5–3.5 mg/L) was operated in the old conventional water treatment process. $\text{NH}_3\text{-N}$ and chlorine reacted and chloramine was formed

considering high concentration of $\text{NH}_3\text{-N}$ in the upstream during pre-chlorination process. Thus, pre-chlorination process could be referred to the pre-chloramination process actually, which could enhance the efficiency of coagulation for turbidity, COD, and $\text{NH}_3\text{-N}$ removal. The result was consistent with Wu et al., they also found pre-chlorination enhanced the removal efficiencies of color, turbidity, ammonia nitrogen, COD, and UV_{254} by the coagulation sedimentation and microfiltration system, and reduced membrane fouling to a certain extent [38].

However, the main problems were DBP formation (THMs and HAAs), which was produced by chemical reaction between the precursor of DBP and chlorine. The practical measured DBP formation was THMs: $13.2 \pm 1.3 \mu\text{g/L}$ and HAAs: $18.6 \pm 0.8 \mu\text{g/L}$, due to the limited removal efficiency of DBP during coagulation + sedimentation of conventional water treatment process [39].

As shown in Fig. 5, the average FPA intensities increased with the dosage of sodium hypochlorite and chlorine dioxide, which increased from 6 to 10 and from 4 to 10, respectively. However, the increased dosage of chloramine showed no improvement on odor control. More DBP formation was found after disinfection compared with that in the effluent of filtration and produced HAAs was much more than that of THMs (as shown in Fig. 5(b)). However, the difference between these DBP (THMs and HAAs) was unobvious. Because pre-chlorination process was operated in the old water treatment process, and then high concentration of chlorine was remained in the effluent of filtration water.

The dosage of pre-chlorination was high, and it was the main problem in the old water treatment process, showing little effect of disinfection methods on odor and DBP control in the effluent of filtration. The dosage of pre-chlorination should be controlled strictly to avoid too much DBPs production. Thus, it was suggested that pre-chlorination needed to be replaced by other pre-processes.

3.5.2. New water treatment process (BAC + O_3 process)

Pre-ozonation was operated in the new water treatment process, providing better removal efficiency of pollutants (turbidity, organic matters), more efficiently DBP formation control, and much more safety drinking water compared with pre-chloramination.

Pre-ozonation combined with BAC + O_3 process showed better removal efficiencies for organic matters, $\text{NH}_3\text{-N}$, manganese, and precursors of DBP [40–42]. Moreover, GAC would transfer into BAC during long-term operation. Geosmin (GSM) and 2-methylisoborneol

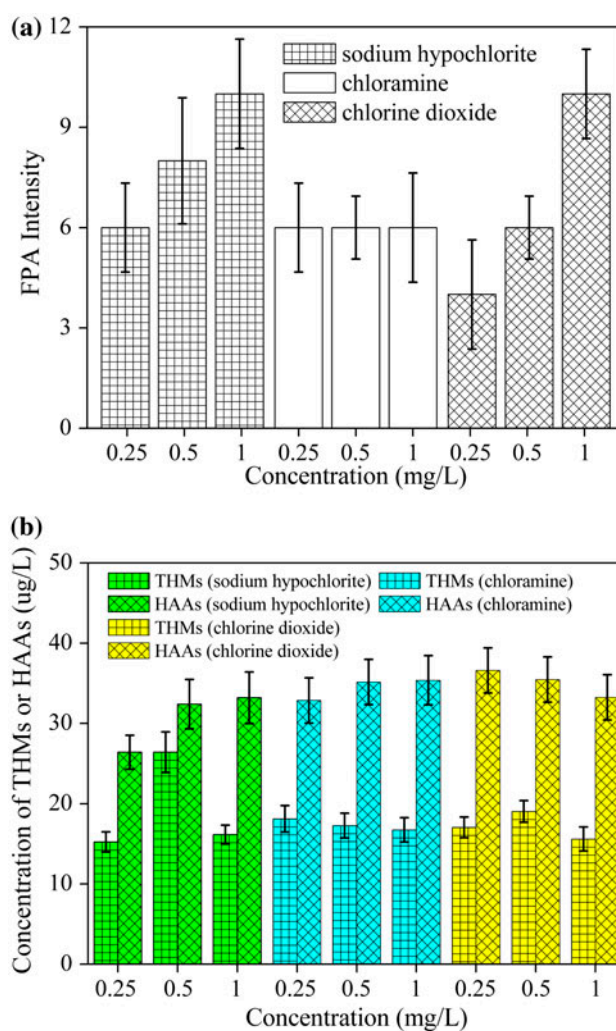


Fig. 5. Effect of conventional water treatment process on odor (a) and DBP (b) control.

(2-MIB) would be efficiently removed by oxidation of O_3 , adsorption and biodegradation by BAC [43].

As shown in Fig. 6(a), there was chlorine odor in the effluent using sodium hypochlorite and the average FPA intensities increased (6, 8, and 8) with more sodium hypochlorite. Chlorine odor and earthy odor were also found in the effluent when chloramine and chlorine dioxide were used for disinfection. For excellent quality of effluent treated by BAC + O_3 process, the amount of disinfectant was much more reduced as the obvious advantage.

Disinfection by chloramine and chlorine dioxide could better control DBP formation, and THMs and HAAs all kept as constant at about $2.5 \mu\text{g/L}$. Whereas, THMs: $12.9 \pm 1.4 \mu\text{g/L}$ and HAAs: $24.5 \pm 2.2 \mu\text{g/L}$ were detected when using sodium hypochlorite as disinfectant.

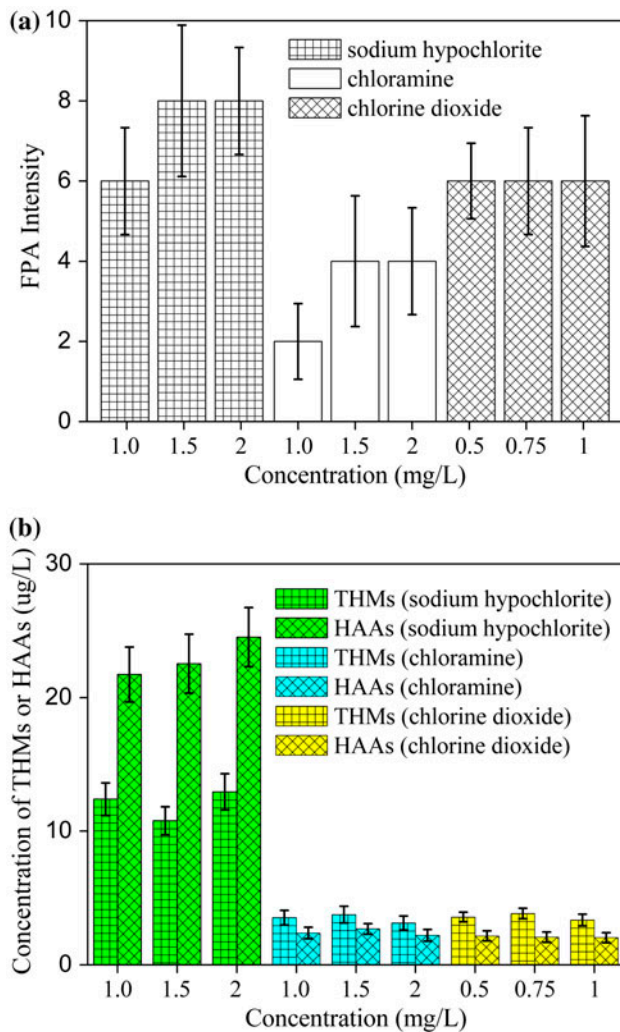


Fig. 6. Effect of O_3 + BAC process on odor (a) and DBP (b) control.

4. Conclusions

In this study, effect of disinfection method on odor and DBP control in drinking water treatment were investigated. The results showed that:

- (1) Odor control by common disinfection followed the sequence: chloramine > chlorine dioxide > sodium hypochlorite. DBP control by common disinfection followed the sequence: chloramine \approx chlorine dioxide > sodium hypochlorite.
- (2) The best ratio of sodium hypochlorite to chlorine dioxide (synergistic disinfection) for odor and DBP control was 1:1. The corresponding THMs and HAAs reduced by 31 and 85% compared with that of single sodium hypochlorite disinfection.
- (3) UV synergistic disinfection compared with chloramine and chlorine dioxide disinfection had little effect on odor and DBP control.
- (4) Disinfection methods showed little effect on odor and DBP control during pre-chlorination + conventional water treatment process. Odor and DBP could be efficiently controlled by chloramine and chlorine dioxide disinfection during pre-ozonation followed by BAC + O_3 process.

Acknowledgements

The authors gratefully acknowledge the financial support provided by the State Key Laboratory of Urban Water Resource and Environment (HIT, grant number 2013DX12), Major Science and Technology Program for Water Pollution Control and Treatment (grant number 2012ZX07408001), and National Science Foundation (grant numbers 51208140, 21206125, and 51478314).

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