



ISSN 1001-0742
CN 11-2629/X

2012

Volume **24**
Number **7**

JOURNAL OF
**ENVIRONMENTAL
SCIENCES**



Sponsored by
Research Center for Eco-Environmental Sciences
Chinese Academy of Sciences

JOURNAL OF ENVIRONMENTAL SCIENCES

(<http://www.jesc.ac.cn>)

Aims and scope

Journal of Environmental Sciences is an international academic journal supervised by Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. The journal publishes original, peer-reviewed innovative research and valuable findings in environmental sciences. The types of articles published are research article, critical review, rapid communications, and special issues.

The scope of the journal embraces the treatment processes for natural groundwater, municipal, agricultural and industrial water and wastewaters; physical and chemical methods for limitation of pollutants emission into the atmospheric environment; chemical and biological and phytoremediation of contaminated soil; fate and transport of pollutants in environments; toxicological effects of terrorist chemical release on the natural environment and human health; development of environmental catalysts and materials.

For subscription to electronic edition

Elsevier is responsible for subscription of the journal. Please subscribe to the journal via <http://www.elsevier.com/locate/jes>.

For subscription to print edition

China: Please contact the customer service, Science Press, 16 Donghuangchenggen North Street, Beijing 100717, China. Tel: +86-10-64017032; E-mail: journal@mail.sciencep.com, or the local post office throughout China (domestic postcode: 2-580).

Outside China: Please order the journal from the Elsevier Customer Service Department at the Regional Sales Office nearest you.

Submission declaration

Submission of an article implies that the work described has not been published previously (except in the form of an abstract or as part of a published lecture or academic thesis), that it is not under consideration for publication elsewhere. The submission should be approved by all authors and tacitly or explicitly by the responsible authorities where the work was carried out. If the manuscript accepted, it will not be published elsewhere in the same form, in English or in any other language, including electronically without the written consent of the copyright-holder.

Submission declaration

Submission of the work described has not been published previously (except in the form of an abstract or as part of a published lecture or academic thesis), that it is not under consideration for publication elsewhere. The publication should be approved by all authors and tacitly or explicitly by the responsible authorities where the work was carried out. If the manuscript accepted, it will not be published elsewhere in the same form, in English or in any other language, including electronically without the written consent of the copyright-holder.

Editorial

Authors should submit manuscript online at <http://www.jesc.ac.cn>. In case of queries, please contact editorial office, Tel: +86-10-62920553, E-mail: jesc@263.net, jesc@rcees.ac.cn. Instruction to authors is available at <http://www.jesc.ac.cn>.

Copyright

© Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. Published by Elsevier B.V. and Science Press. All rights reserved.

CONTENTS

Aquatic environment

Investigation of the hydrodynamic behavior of diatom aggregates using particle image velocimetry Feng Xiao, Xiaoyan Li, Kitming Lam, Dongsheng Wang.....	1157
Shellac-coated iron oxide nanoparticles for removal of cadmium(II) ions from aqueous solution Jilai Gong, Long Chen, Guangming Zeng, Fei Long, Jiuhua Deng, Qiuya Niu, Xun He.....	1165
Prediction of DOM removal of low specific UV absorbance surface waters using HPSEC combined with peak fitting Linan Xing, Rolando Fabris, Christopher W. K. Chow, John van Leeuwen, Mary Drikas, Dongsheng Wang.....	1174
Photo-production of dissolved inorganic carbon from dissolved organic matter in contrasting coastal waters in the southwestern Taiwan Strait, China Weidong Guo, Liyang Yang, Xiangxiang Yu, Weidong Zhai, Huasheng Hong.....	1181
One century sedimentary record of lead and zinc pollution in Yangzong Lake, a highland lake in southwestern China Enlou Zhang, Enfeng Liu, Ji Shen, Yanmin Cao, Yanling Li.....	1189
Antimony(V) removal from water by iron-zirconium bimetal oxide: Performance and mechanism Xuehua Li, Xiaomin Dou, Junqing Li.....	1197
Carbonaceous and nitrogenous disinfection by-product formation in the surface and ground water treatment plants using Yellow River as water source Yukun Hou, Wenhai Chu, Meng Ma.....	1204
Water quality evaluation based on improved fuzzy matter-element method Dongjun Liu, Zhihong Zou.....	1210
Formation and cytotoxicity of a new disinfection by-product (DBP) phenazine by chloramination of water containing diphenylamine Wenjun Zhou, Linjie Lou, Lifang Zhu, Zhimin Li, Lizhong Zhu.....	1217

Atmospheric environment

Chemical compositions of PM _{2.5} aerosol during haze periods in the mountainous city of Yong'an, China Liqian Yin, Zhenchuan Niu, Xiaoqiu Chen, Jinsheng Chen, Lingling Xu, Fuwang Zhang.....	1225
Decomposition of trifluoromethane in a dielectric barrier discharge non-thermal plasma reactor M. Sanjeeva Gandhi, Y. S. Mok.....	1234
Transverse approach between real world concentrations of SO ₂ , NO ₂ , BTEX, aldehyde emissions and corrosion in the Grand Mare tunnel I. Ameur-Bouddabbous, J. Kasperek, A. Barbier, F. Harel, B. Hannoyer.....	1240
A land use regression model incorporating data on industrial point source pollution Li Chen, Yuming Wang, Peiwu Li, Yaqin Ji, Shaofei Kong, Zhiyong Li, Zhipeng Bai.....	1251

Terrestrial environment

Effect of vegetation of transgenic Bt rice lines and their straw amendment on soil enzymes, respiration, functional diversity and community structure of soil microorganisms under field conditions Hua Fang, Bin Dong, Hu Yan, Feifan Tang, Baichuan Wang, Yunlong Yu.....	1259
Enhanced flushing of polychlorinated biphenyls contaminated sands using surfactant foam: Effect of partition coefficient and sweep efficiency Hao Wang, Jiajun Chen.....	1270
Transpiration rates of urban trees, <i>Aesculus chinensis</i> Hua Wang, Xiaoke Wang, Ping Zhao, Hua Zheng, Yufen Ren, Fuyuan Gao, Zhiyun Ouyang.....	1278

Environmental biology

Methanogenic community dynamics in anaerobic co-digestion of fruit and vegetable waste and food waste Jia Lin, Jiane Zuo, Ruofan Ji, Xiaojie Chen, Fenglin Liu, Kaijun Wang, Yunfeng Yang.....	1288
Differential fate of metabolism of a disperse dye by microorganisms <i>Galactomyces geotrichum</i> and <i>Brevibacillus laterosporus</i> and their consortium GG-BL Tatoba R. Waghmode, Mayur B. Kurade, Anuradha N. Kagalkar, Sanjay P. Govindwar.....	1295

Environmental catalysis and materials

Effects of WO _x modification on the activity, adsorption and redox properties of CeO ₂ catalyst for NO _x reduction with ammonia Ziran Ma, Duan Weng, Xiaodong Wu, Zhichun Si.....	1305
Photocatalytic degradation of bisphenol A using an integrated system of a new gas-liquid-solid circulating fluidized bed reactor and micrometer Gd-doped TiO ₂ particles Zhiliang Cheng, Xuejun Quan, Jinxin Xiang, Yuming Huang, Yunlan Xu.....	1317
Effect of CeO ₂ and Al ₂ O ₃ on the activity of Pd/Co ₃ O ₄ /cordierite catalyst in the three-way catalysis reactions (CO/NO/C _n H _m) Sergiy O. Soloviev, Pavlo I. Kyriienko, Nataliia O. Popovych.....	1327

Environmental analytical methods

Development of indirect competitive fluorescence immunoassay for 2,2',4,4'-tetrabromodiphenyl ether using DNA/dye conjugate as antibody multiple labels Zi-Yan Fan, Young Soo Keum, Qing-Xiao Li, Weilin L. Shelver, Liang-Hong Guo.....	1334
A novel colorimetric method for field arsenic speciation analysis Shan Hu, Jinsuo Lu, Chuanyong Jing.....	1341
Aminobenzenesulfonamide functionalized SBA-15 nanoporous molecular sieve: A new and promising adsorbent for preconcentration of lead and copper ions Leila Hajiaghatabaei, Babak Ghasemi, Alireza Badieli, Hassan Goldoos, Mohammad Reza Ganjali, Ghodsi Mohammadi Ziarani.....	1347



Carbonaceous and nitrogenous disinfection by-product formation in the surface and ground water treatment plants using Yellow River as water source

Yukun Hou^{1,2,*}, Wenhai Chu^{1,*}, Meng Ma²

1. College of Environmental Science and Engineering, Tongji University, Shanghai 200092, China

2. Zhengzhou Water Supply Corporation, Zhengzhou 450013, China

Received 27 December 2012; revised 16 January 2012; accepted 01 March 2012

Abstract

This work investigated the formation of carbonaceous and nitrogenous disinfection by-products (C-DBPs, N-DBPs) upon chlorination of water samples collected from a surface water and a ground water treatment plant (SWTP and GWTP) where the conventional treatment processes, i.e., coagulation, sedimentation, and filtration were employed. Twenty DBPs, including four trihalomethanes, nine haloacetic acids, seven N-DBPs (dichloroacetamide, trichloroacetamide, dichloroacetonitrile, trichloroacetonitrile, bromochloroacetonitrile, dibromoacetonitrile and trichloronitromethane), and eight volatile chlorinated compounds (dichloromethane (DCM), 1,2-dichloroethane, tetrachloroethylene, chlorobenzene, 1,2-dichlorobenzene, 1,4-dichlorobenzene, 1,2,3-trichlorobenzene and 1,2,4-trichlorobenzene) were detected in the two WTPs. The concentrations of these contaminants were all below their corresponding maximum contamination levels (MCLs) regulated by the Standards for Drinking Water Quality of China (GB5749-2006) except for DCM (17.1 µg/L detected vs. 20 µg/L MCL). The SWTP had much higher concentrations of DBPs detected in the treated water as well as the DBP formation potentials tested in the filtered water than the GWTP, probably because more precursors (e.g., dissolved organic carbon, dissolved organic nitrogen) were present in the water source of the SWTP.

Key words: carbonaceous disinfection by-products; nitrogenous disinfection by-products; formation potential; free chlorine; water treatment plant

DOI: 10.1016/S1001-0742(11)61006-1

Introduction

Drinking water disinfection (esp. chlorination) results in the formation of disinfection by-products (DBPs) which may cause adverse health effects on human beings. DBPs are mainly formed in the reactions between their precursors present in source water and the disinfectant (esp. chlorine) added during water treatment. Over the past three decades, much more DBP studies have focused on the currently regulated carbonaceous DBPs (C-DBPs) (e.g., trihalomethanes (THMs) and haloacetic acids (HAAs)) (Bellar et al., 1974; Krasner et al., 2006), creating a better understanding of the formation and control of THMs and HAAs in water treatment plants (WTPs) than those of emerging unregulated DBPs, e.g., nitrogenous DBPs (N-DBPs) (Muellner et al., 2007). The concentrations of N-DBPs, including haloacetonitriles (HANs), halonitromethanes (HNMs), and haloacetamides (HAcAms), are generally at a lower level (ng/L to µg/L) than those of C-DBPs (Krasner et al., 2006; Chu et al., 2011a), but are more toxic (Plewa et al., 2004, 2008; Muellner et al., 2007).

The levels of DBPs and their precursors in WTPs

provide basic information for health risk assessment, regulation and water treatment process optimization. Since the 1980s, the surveys for DBPs in the treated water of WTPs have been carried out in many countries. Halogenated N-DBPs were detected in some WTPs of the United States, such as HANs, HNMs and HAcAms (Krasner et al., 1989, 2006, 2007a; McGuire et al., 2002; Mitch et al., 2009). Similar surveys were also carried out in Canada (Williams et al., 1995), Australia (Simpson and Hayes, 1998) and Scotland (Goslan et al., 2009). However, these studies mainly focused on HANs, and much less data were reported on HAcAms and HNMs.

Toxicological studies have reported that N-DBPs (e.g., HAcAms and HNMs) show much higher cytotoxicity and genotoxicity than the regulated THMs and HAAs to mammalian cells (Plewa et al., 2004, 2008). Investigating the formation of N-DBPs in WTPs is therefore an urgent task. Chu et al. (2011b) reported the formation of N-DBPs, including HANs, HNMs and HAcAms, in a typical surface water treatment plant (SWTP) located in southern China. However, the surface water quality varies significantly in different regions of China due to significantly different climatic and geographical characteristics. Zhang et al. (2011) investigated the formation potential of 4 THMs and 7 HAAs in 13 source water samples collected from the

* Corresponding author. E-mail: hou.yukun@126.com (Yukun Hou); 1world1water@tongji.edu.cn (Wenhai Chu)

northeast to south China covering four of the seven major water basin areas (Songhua River Basin, Hai River Basin, Yangtze River Basin and Huai River Basin). It was found that the organic compounds in different source waters exhibited different reactivities with chlorine, which probably caused the different characterization for the formation of N-DBPs. However, comprehensive information on the formation of C-DBPs and N-DBPs from source water in Yellow River Basin (e.g., Zhengzhou City) is little known. The Yellow River, with a total length of 5464 km, is the second longest river in China. With artificial pollution (e.g., factory discharges and sewage from fast-expanding cities), the Yellow River is being polluted, which brings great challenges to WTPs along Yellow River. The conventional treatment process could not effectively remove DBP precursors, probably causing the extensive formation of DBPs in drinking water, including C-DBPs and N-DBPs, threatening people's health.

The aim of this study was to evaluate the concentrations of DBPs and the removal efficiency of the precursors (denoted by the DBP formation potentials (FPs)) in two typical WTPs located in Zhengzhou City that use the surface and ground waters from the Yellow River as source water, respectively. For N-DBPs, two HAcAms (dichloroacetamide (DCAcAm) and trichloroacetamide (TCAcAm)), four HANs (dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), bromochloroacetonitrile (BCAN) and dibromoacetonitrile (DBAN)) and one HNM (trichloronitromethane (TCNM)) were determined. For C-DBPs, four regulated THMs (chloroform (CF), bromodichloromethane (BDCM), dibromochloromethane (DBCM) and bromoform (BF)), nine HAAs (chloroacetic acid (CAA), dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), bromoacetic acid (BAA), bromochloroacetic acid (BCAA), bromodichloroacetic acid (BDCAA), dibromoacetic acid (DBAA), dibromochloroacetic acid (DBCAA), tribromoacetic acid (TBAA)), and some other volatile chlorinated compounds (e.g., dichloromethane (DCM)) were also measured. It is expected that the survey data would be helpful for DBPs health risk assessment, regulation and water treatment process optimization.

1 Materials and methods

1.1 Chemicals

The standard solutions of the studied DBPs were purchased from Sigma-Aldrich (USA), except DCAcAm (98.5%) and TCAcAm (99%) which were supplied from Alfa Aesar (Karlsruhe, Germany). Ethyl acetate, obtained from Fisher Scientific (Waltham, USA), was used to extract DCAcAm and TCAcAm. All other chemicals and reagents were supplied by Sinopharm Chemical Reagent (Shanghai, China) with at least analytical grade purity.

1.2 Sample collection and preparation

Water samples were collected from a SWTP and a ground water treatment plant (GWTP) located in Zhengzhou

City, China, in November 2011. The treatment processes adopted in the SWTP included coagulation (34 mg/L polyaluminium chloride), sedimentation, filtration, and chlorination (3.1 mg/L chlorine), which are prevalent in Chinese WTPs. In contrast, only two treatment processes (i.e., filtration and chlorination) were adopted in the GWTP where about 0.95 mg/L free chlorine was dosed as disinfectant. Although the two plants both use the Yellow River as their water source, the source water of the GWTP (bank-infiltration groundwater) had much better quality than that of the SWTP (surface water). Once collected, the water samples were immediately quenched of disinfectant residuals through addition of about 10 mg/L ascorbic acid. Glacial acetic acid was added to lower the sample pH to 4.8–5.5 for HAN analysis. With regards to HAcAm analysis, the sample pH was adjusted to 5.0 ± 0.2 with glacial acetic acid to inhibit the potential hydrolysis of HAcAms under neutral or basic conditions (Chu et al., 2012). Subsequently, the samples were filtered through a pre-rinsed 0.7- μm glass filter membrane (Whatman, UK), and then stored in the refrigerator at 4°C until used. The water quality parameters of influent, and before and after filtration in the two WTPs are compared in Table 1.

1.3 Analysis

The sample pH was measured onsite using a solid selective electrode connected to a multimeter (WTW3410, German). Turbidity was also measured onsite by a turbidimeter (HACH 2100N, USA). Dissolved organic carbon (DOC) and total dissolved nitrogen (TDN) were analyzed by a TOC analyzer coupled with a total nitrogen measurement unit (TOC-VCPH, Shimadzu, Japan). The DON concentration was calculated as the difference between TDN and dissolved inorganic nitrogen (DIN). DIN including ammonia, nitrate, and nitrite, and free and total chlorine were measured with a portable spectrophotometer (HACH DR2800, USA). UV_{254} was measured by a UV/Vis double beam spectrophotometer (Unico4802, USA). The specific UV (SUVA) is the UV absorbance at the 254 nm wavelength divided by the DOC of a water sample, which normalizes the aromatic-biased UV_{254} measurement over the overall organic load in the water according to the US EPA method 415.3. SUVA is a good surrogate for aromatic organics (Edzwald and Tobiasson, 1999).

THMs, HANs (DCAN, TCAN, BCAN and DBAN), HNM (TCNM) and some other volatile chlorinated DBPs (e.g., DCM) were all analyzed with a purge & trap sample concentrator (eclipse4660, OI, USA) and gas chromatograph/mass spectrometry (GC/MS) (QP2010, Shimadzu, Japan) according to US EPA method 524.2. Nine HAAs were analyzed by a GC (Shimadzu-QP2010, Japan) coupled with an electron capture detector according to US EPA method 552.2. Two HAcAms were analyzed by liquid-liquid extraction and GC/MS detection (Chu et al., 2010). The DBP FP test was conducted with free chlorine following the method of Krasner et al. (2007b). The method detection limits (unit $\mu\text{g/L}$) were as follows: 0.067 (CF), 0.071 (BDCM), 0.087 (DBCM), 0.085 (BF), 0.11 (DCAN), 0.091 (TCAN), 0.15 (BCAN,

Table 1 Water quality parameters of influent before and after filtration in the studied SWTP and GWTP

Parameters	SWTP			GWTP		
	Raw water	Before filtration	After filtration	Raw water	Before filtration	After filtration
Turbidity (NTU)	30.2	1.76	0.14	3.29	0.11	0.12
pH	8.22	8.12	7.98	7.60	7.69	7.64
UV ₂₅₄ (cm ⁻¹)	0.035	0.005	0.004	0.005	0.001	0.001
NH ₄ ⁺ -N (mg/L)	0.27	< 0.02	< 0.02	0.20	< 0.02	< 0.02
NO ₃ ⁻ -N (mg/L)	2.30	2.81	2.84	0.08	0.16	0.15
NO ₂ ⁻ -N (mg/L)	0.018	< 0.001	< 0.001	0.001	< 0.001	< 0.001
DOC (mg/L)	15.6	3.28	2.67	1.76	0.55	0.42
DON (mg/L)	1.00	0.93	0.84	0.19	0.17	0.15
SUVA (L/(mg·m))	2.24	1.52	1.50	2.84	1.81	2.38

DOC: dissolved organic carbon; DON: dissolved organic nitrogen; SUVA: specific UV.

TCNM), 0.26 (DBAN), 0.10 (DCAcAm, TCACAm), 0.70 (CAA, TBAA), 0.50 (DCAA, TCAA, BAA, BCAA, BDCAA, DBAA, DBCAA), and < 0.10 for eight volatile chlorinated compounds (DCM, 1,2-dichloroethane, tetrachloroethylene, chlorobenzene, 1,2-dichlorobenzene, 1,4-dichlorobenzene, 1,2,3-trichlorobenzene and 1,2,4-trichlorobenzene).

2 Results and discussion

2.1 Formation of THMs

Four THMs (i.e., CF, BDCM, DBCM and BF) have recently been listed in the National Standards for Drinking Water Quality of China (GB5749-2006), with maximum contamination levels (MCLs) at 60, 100, 60, and 100 µg/L, respectively. Figure 1a shows that the concentrations of the above four THMs and total THM (TTHM) in the finished water of the SWTP were all much higher than those of the GWTP. This can be mainly ascribed to the better source water quality of the GWTP than that of the SWTP. For example, the concentrations of DOC (representing the primary precursors of C-DBPs) were 15.6 and 1.76 mg/L in the raw water of the SWTP and GWTP, respectively (Table 1). Similarly, it was found the THM FPs (representing the precursor levels) in the filtered water (right before chlorination) of the SWTP were also substantially higher than those in the GWTP (Fig. 1b),

which is in agreement with the previous inference. As indicated in Fig. 1b, the concentrations of four THMs in the finished water of both the SWTP and GWTP were lower than their MCLs. However, it should be pointed out that the concentrations of CF and TTHM of the SWTP might exceed their MCLs during the long time delivery of water in the pipelines, as cautioned by their FPs in Fig. 1b.

2.2 Formation of HAAs

The concentrations of nine HAAs in the finished water of the SWTP and GWTP are shown in Fig. 2a. The results indicated that BAA, DBAA, DBCAA, and TBAA were undetectable in the finished water of the SWTP and GWTP, and CAA (0.85 µg/L) was only detected in the SWTP. Similar to THMs, the concentrations of all detected HAA were significantly higher in the SWTP than those in the GWTP. In addition, Fig. 2b shows that the SWTP had notably higher HAA FPs than the GWTP. The MCLs of DCAA and TCAA were regulated as 50 and 100 µg/L, respectively, which were both higher than their concentrations detected in the finished water of the two WTPs (Fig. 1a). It is noted that the concentration of THAA might exceed its MCL during the long time delivery of water in the pipelines, as cautioned by its FP in Fig. 2b.

2.3 Formation of N-DBPs

Figure 3 shows the concentrations of N-DBPs in the finished water and their FPs in the filtered water of the

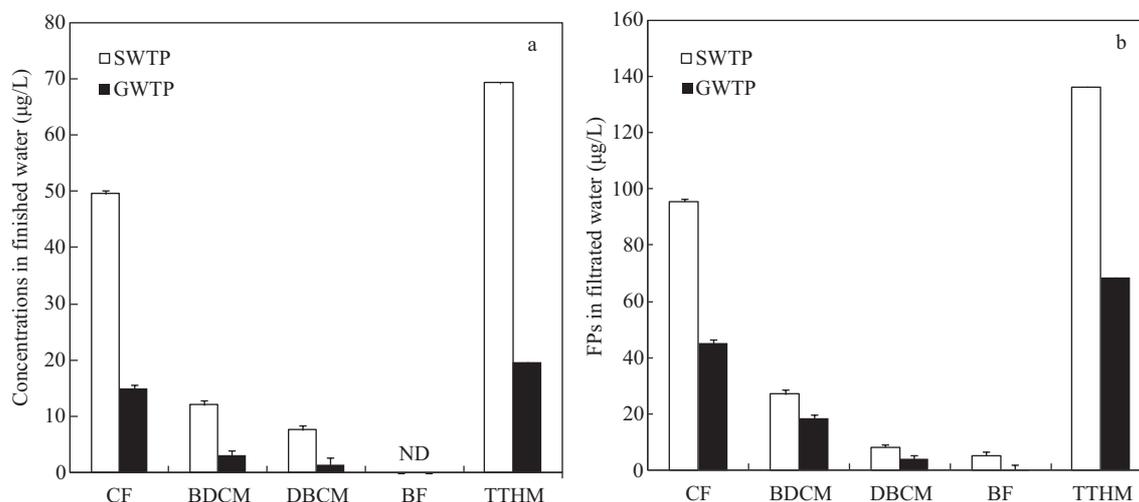


Fig. 1 Four THMs (CF, BDCM, DBCM, BF) and total THM concentrations in the finished water (a) and THM formation potentials (FPs) in the filtered water (b) of the SWTP and GWTP. Error bars represent the standard deviation of triplicate measurements. ND: not detected.

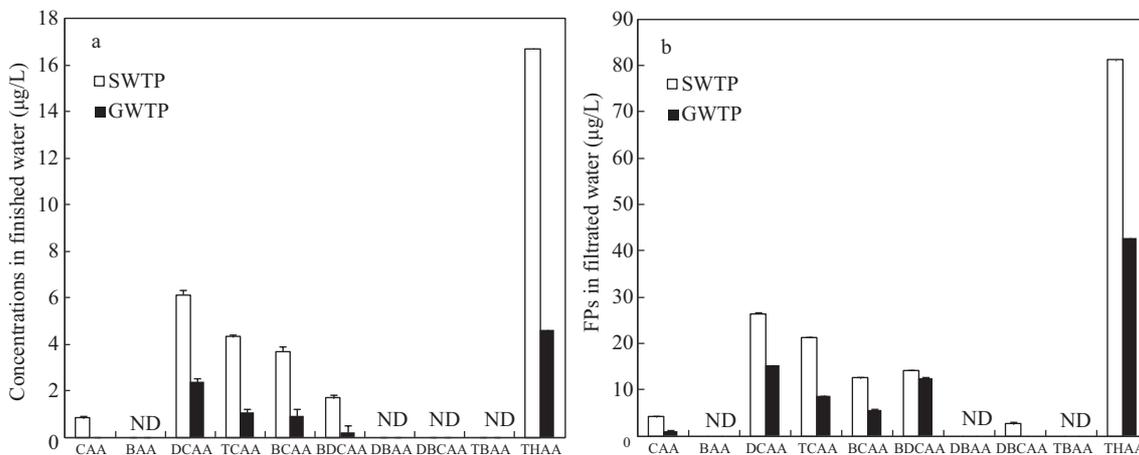


Fig. 2 Nine HAAs and total HAA (THAA) concentrations in the finished water (a) and HAA FPs in the filtered water (b) of the SWTP and GWTP. Error bars represent the standard deviation of triplicate measurements.

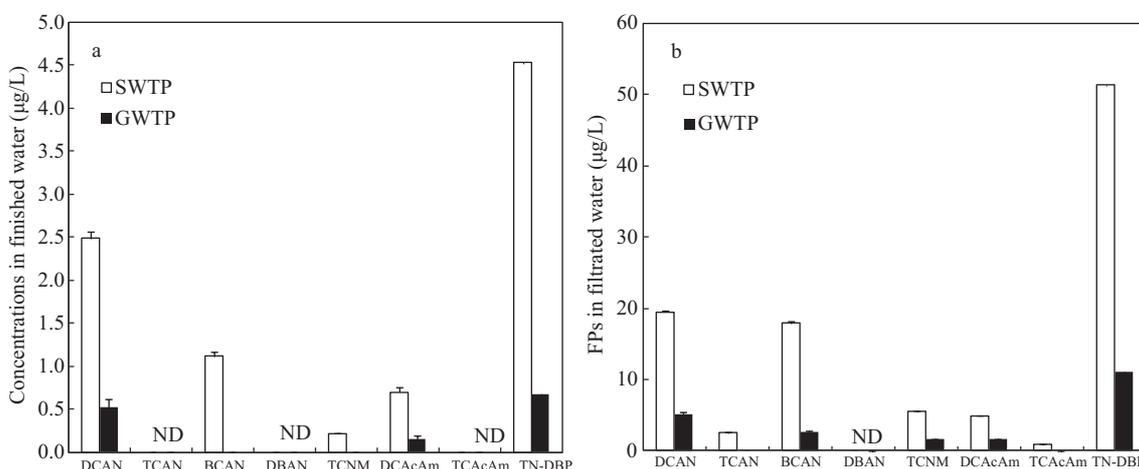


Fig. 3 N-DBP concentrations in the finished water (a) and N-DBP FPs in the filtered water (b) of the SWTP and GWTP. Error bars represent the standard deviation of triplicate measurements.

SWTP and GWTP. DCAN, BCAN, TCNM and DCACAm were all detected in the finished water of the SWTP at a concentration level of 0.3–2.5 µg/L, whereas only DCAN and DCACAm were detected in the GWTP at a notably decreased concentration level of 0.2–0.5 µg/L. Additionally, the N-DBP FPs in the finished water of the SWTP were also higher than those of the GWTP, which was in accordance with the results of C-DBPs such as THMs (Fig. 1) and HAAs (Fig. 2). The previous observations imply that a portion of the DON plays an essential role in the formation of DCACAm and DCAN, where that portion is probably watershed-specific, and the presence of DON may act as an indicator (but not predictor per se) of N-DBP FP (Chu et al., 2010). The concentration of DON, which is believed to be the main precursor of N-DBPs (Lee et al., 2007), was much higher in the source water of the SWTP (i.e., 1.00 µg/L) than that of the GWTP (i.e., 0.19 µg/L). This mainly accounted for the significant difference in the N-DBP concentrations of the two WTPs. A low SUVA value means that the organic matter in water is mostly comprised of non-humics, and has low hydrophobicity and low molecular weights, usually resulting in poor removal of organics (Edzwald and Tobiason, 1999; Roe et al., 2008). The lower SUVA values in the finished water of the SWTP may, to some extent, also explain the higher

concentrations and FPs of N-DBPs in the SWTP.

2.4 Formation of volatile chlorinated compounds

The concentrations of eight chlorinated volatile compounds in the finished water of the two WTPs are shown in Fig. 4. DCM, 1,2-dichloroethane,

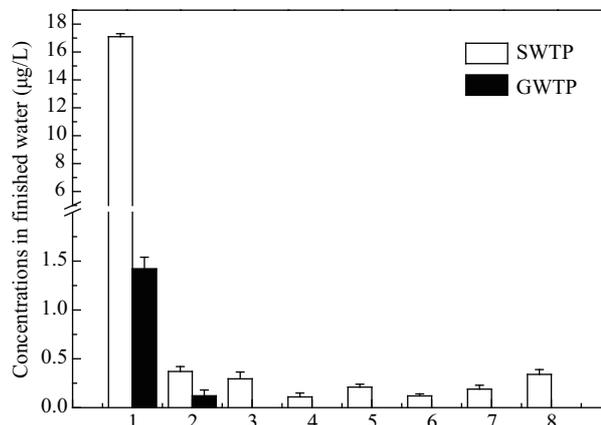


Fig. 4 Chlorinated volatile DBP concentrations in the finished water of the SWTP and GWTP. Error bars represent the standard deviation of triplicate measurements. 1: DOM; 2:1,2-dichloroethane; 3: tetrachloroethylene; 4: chlorobenzene; 5: 1,2-dichlorobenzene; 6: 1,4-dichlorobenzene; 7: 1,2,3-trichlorobenzene; 8: 1,2,4-trichlorobenzene.

tetrachloroethylene, chlorobenzene, 1,2-dichlorobenzene, 1,4-dichlorobenzene, 1,2,3-trichlorobenzene and 1,2,4-trichlorobenzene, were all detected in the finished water of the SWTP, whereas only DCM and 1,2-dichloroethane were detected in the finished water of the GWTP. These volatile compounds were undetectable in the filtered water of both the SWTP and GWTP before disinfection, which preliminarily indicated that DCM, 1,2-dichloroethane, tetrachloroethylene, chlorobenzene, 1,2-dichlorobenzene, 1,4-dichlorobenzene, 1,2,3-trichlorobenzene and 1,2,4-trichlorobenzene were probably formed during chlorination disinfection. Therefore, to better control the concentration of these eight volatile compounds to meet their corresponding MCLs, WTPs should pay more attention to disinfection processes. In the study, with the exception of DCM, the concentrations of the other seven chlorinated volatile compounds were much lower than their corresponding MCLs, i.e., 20, 30, 40, 300, 1000, 300 and 20 $\mu\text{g/L}$ for 1,2-dichloroethane, tetrachloroethylene, chlorobenzene, 1,2-dichlorobenzene, 1,4-dichlorobenzene, and the sum of 1,2,3-trichlorobenzene and 1,2,4-trichlorobenzene, respectively. It is noted that the concentration of DCM (17.1 $\mu\text{g/L}$) in the finished water of the SWTP was close to its MCL (20 $\mu\text{g/L}$). Thus, it is necessary to strengthen the long-term monitoring of DCM in the finished water, and also optimize the treatment processes to reduce its formation.

2.5 Formation of brominated DBPs

The formation of brominated DBPs has attracted more attention because they are generally more toxic than their chlorinated analogues (WHO, 2000). The bromine incorporation factor (BIF) is used as an index to describe the proportion of the DBPs that can be partially or totally brominated. The BIFs for THMs, tri-HAAs (THAAs), di-HAAs (DHAAs), and di-HANs (DHANs) are defined by the following equations:

$$\text{BIF(THMs)} = \frac{[\text{BDCM}] + 2[\text{DBCM}] + 3[\text{BF}]}{[\text{CF}] + [\text{BDCM}] + [\text{DBCM}] + [\text{BF}]} \quad (1)$$

$$\text{BIF(THAAs)} = \frac{[\text{BDCAA}] + 2[\text{DBCAA}] + 3[\text{TBAA}]}{[\text{TCAA}] + [\text{BDCAA}] + [\text{DBCAA}] + [\text{TBAA}]} \quad (2)$$

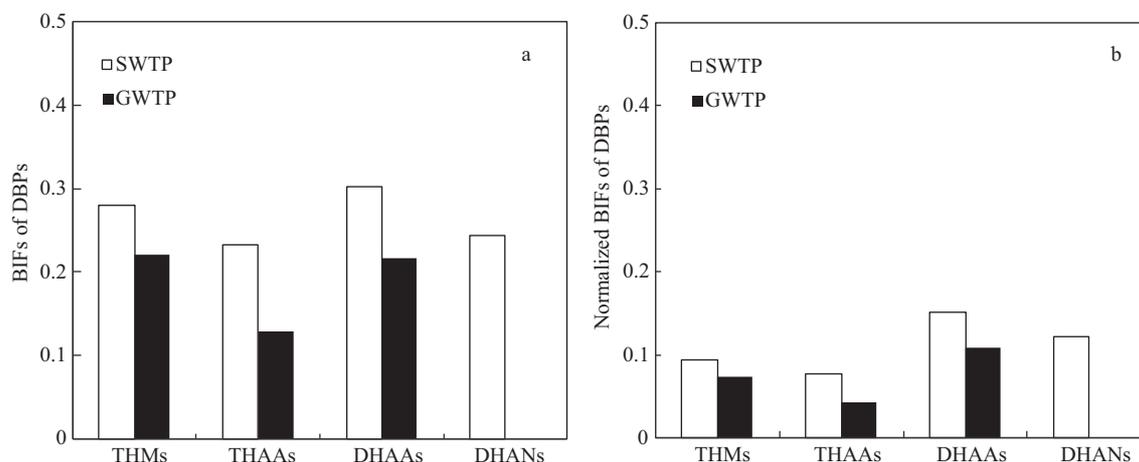


Fig. 5 Raw (a) and normalized (b) bromine incorporation factor (BIFs) of THMs, THAAs, DHAAs and DHANs.

$$\text{BIF(DHAAs)} = \frac{[\text{BCAA}] + 2[\text{DBAA}]}{[\text{DCAA}] + [\text{BCAA}] + [\text{DBAA}]} \quad (3)$$

$$\text{BIF(DHANs)} = \frac{[\text{BCAN}] + 2[\text{DBAN}]}{[\text{DCAN}] + [\text{BCAN}] + [\text{DBAN}]} \quad (4)$$

As shown in the above Eqs. (1)–(4), the BIF values describe the molar contribution of the brominated THMs, THAAs, DHAAs and DHANs. Depending on the degree of bromine substitution, BIF can range from 0 (no brominated species) to 3 (pure BF and pure TBAA) for THMs and THAAs, and 2 (pure DBAA and DBAN) for DHAAs and DHANs. Figure 5a shows that the calculated BIF values of the selected DBPs were mainly below 0.3, indicating that CF, DCAA, TCAA and DCAN were the dominant species in THMs, DHAAs, THAAs and DHANs, respectively. If one divides BIF by the number of halogens (i.e., 3 in THMs and THAAs; 2 in DHAAs and DHANs), the normalized BIF will range from 0 to 1. The normalized BIFs for THMs, THAAs, DHAAs and DHANs are shown in Fig. 5b. For the SWTP, more brominated DHAAs and DHANs were formed than brominated THMs and THAAs. It was also reported that more bromine was incorporated into DHANs than into THMs in the post-chlorinated and post-chloraminated waters in some previous studies (Chu et al., 2011b; Obolensky and Singer, 2005). For the GWTP, no brominated DHANs were detected, and the normalized BIF of the DHAAs was also higher than those of the THMs and THAAs. Therefore, bromine appeared to be more readily incorporated into dihalogenated, rather than trihalogenated, DBPs in the studied WTPs.

3 Conclusions

The raw water quality plays a crucial role in the formation of DBPs during chlorination after the conventional water treatment processes (i.e., coagulation, sedimentation and filtration). During the sampling period, the concentrations of 4 THMs (CF, BDCM, DBCM, and BF), 9 HAAs (CAA, DCAA, TCAA, BAA, BCAA, BDCAA, DBAA, DBCAA, and TBAA) and some other regulated volatile chlorinated compounds (1,2-dichloroethane, tetrachloroethylene, chlorobenzene, 1,2-dichlorobenzene, 1,4-dichlorobenzene, 1,2,3-trichlorobenzene and 1,2,4-

trichlorobenzene) were all significantly lower than their MCLs. However, some N-DBPs were detected, a few of which had higher concentrations in the studied SWTP than what was reported in some previous studies. The concentration of DCM was found to approach its MCL, which necessitates the long-term monitoring of this compound. In addition, this study seriously cautions that the concentrations of CF, TTHM and THAA in SWTP might exceed their MCLs during the long time delivery of water in the pipelines, as indicated by their FP data. Further study on the risk assessment and long-term monitoring of C-DBPs, N-DBPs and volatile chlorinated compounds should be carried out in China, especially in source water with high precursor loadings.

Acknowledgments

This work was supported by the National Major Science and Technology Project on Water Pollution Control and Management of China (No. 2009ZX07424-003), the National Natural Science Foundation of China (No. 51108327) and the State Key Laboratory of Pollution Control and Resource Reuse Foundation (No. PCRRY11015).

References

- Bellar T A, Lichtenberg J J, Kroner R C, 1974. The occurrence of organohalides in chlorinated drinking waters. *Journal of the American Water Works Association*, 66(12): 703–706.
- Chu W H, Gao N Y, Deng Y, Krasner S W, 2010. Precursors of dichloroacetamide, an emerging nitrogenous DBP formed during chlorination or chloramination. *Environmental Science and Technology*, 44(10): 3908–3912.
- Chu W H, Gao N Y, Templeton M R, Yin D Q, 2011b. Formation of nitrogenous disinfection by-products from pre-chloramination. *Chemosphere*, 85(7): 1187–1191.
- Chu W H, Gao N Y, Templeton M R, Yin D Q, 2011a. Comparison of inclined plate sedimentation and dissolved air flotation for the minimisation of subsequent nitrogenous disinfection by-product formation. *Chemosphere*, 83(5): 647–651.
- Chu W H, Gao N Y, Krasner S W, Templeton M R, Yin D Q, 2012. Formation of halogenated C-, N-DBPs from chlor(am)ination and UV irradiation of tyrosine in drinking water. *Environmental Pollution*, 161(15): 8–14.
- Edzward J K, Tobiasson J E, 1999. Enhanced coagulation: US requirements and a broader view. *Water Science and Technology*, 40(9): 63–70.
- Goslan E H, Krasner S W, Bower M, Rocks S A, Holmes P, Levy L S et al., 2009. A comparison of disinfection by-products found in chlorinated and chloraminated drinking waters in Scotland. *Water Research*, 43(18): 4698–4706.
- Krasner S W, McGuire M J, Jacangelo J G, Patania N L, Reagan K M, Aieta E M, 1989. Occurrence of disinfection by-products in US drinking water. *Journal of the American Water Works Association*, 81(8): 41–53.
- Krasner S W, Weinberg H S, Richardson S D, Pastor S J, Chinn R, Scilimenti M J et al., 2006. Occurrence of a new generation of disinfection byproducts. *Environmental Science and Technology*, 40(23): 7175–7185.
- Krasner S W, Scilimenti M J, Mitch W A, Westerhoff P, Dotson A A, 2007a. Wastewater and Algal Derived N-DBPs. AWWA, Denver, CO, USA. AWWA Annual Conference.
- Krasner S W, Scilimenti M J, Mitch W, Westerhoff P, Dotson A, 2007b. Using formation potential tests to elucidate the reactivity of DBP precursors with chlorine versus with chloramines. In: 2007 AWWA Water Quality Technology Conference, Denver, CO.
- Lee W, Westerhoff P, Croué J P, 2007. Dissolved organic nitrogen as a precursor for chloroform, dichloroacetonitrile, n-nitrosodimethylamine, and trichloronitromethane. *Environmental Science and Technology*, 41(15): 5485–5490.
- McGuire M J, McLain J L, Obolensky A, 2002. Information collection rule data analysis. AwwaRF and AWWA, Denver, CO, USA.
- Mitch W A, Krasner S W, Westerhoff P, Dotson A, 2009. Occurrence and formation of nitrogenous disinfection by-products. Water Research Foundation, Denver, CO, USA.
- Muellner M G, Wagner E D, McCalla K, Richardson S D, Woo Y T, Plewa M J, 2007. Haloacetonitriles vs. regulated haloacetic acids: Are nitrogen-containing DBPs more toxic? *Environmental Science and Technology*, 41(2): 645–651.
- Obolensky A, Singer P C, 2005. Halogen substitution patterns among disinfection byproducts in the information collection rule database. *Environmental Science and Technology*, 39(8): 2719–2730.
- Plewa M J, Wagner E D, Jazwierska P, Richardson S D, Chen P H, McKague A B, 2004. Halonitromethane drinking water disinfection byproducts: Chemical characterization and mammalian cell cytotoxicity and genotoxicity. *Environmental Science and Technology*, 38(1): 62–68.
- Plewa M J, Muellner M G, Richardson S D, Fasano F, Buettner K M, Woo Y T et al., 2008. Occurrence, synthesis, and mammalian cell cytotoxicity and genotoxicity of haloacetamides: An emerging class of nitrogenous drinking water disinfection byproducts. *Environmental Science and Technology*, 42(3): 955–961.
- Roe J, Baker A, Bridgeman J, 2008. Relating organic matter character to trihalomethanes formation potential: A data mining approach. *Water Science Technology*, 8(6): 717–723.
- Simpson K L, Hayes K P, 1998. Drinking water disinfection byproducts: An Australian perspective. *Water Research*, 32(5): 1522–1528.
- Williams D T, LeBel G L, Benoit F M, 1995. A national survey of chlorinated disinfection by-products in canadian drinking water Canada, environmental health Directorate, Health Canada, 1995.
- WHO (World Health Organization), 2000. Disinfectants and disinfectant by-products, environmental health criteria 216, international programme on chemical safety.
- Zhang J Z, Yu J W, An W, Liu J, Wang Y J, Chen Y J et al., 2011. Characterization of disinfection byproduct formation potential in 13 source waters in China. *Journal of Environmental Sciences*, 23(2): 183–188.

JOURNAL OF ENVIRONMENTAL SCIENCES

Editors-in-chief

Hongxiao Tang

Associate Editors-in-chief

Nigel Bell Jiuhui Qu Shu Tao Po-Keung Wong Yahui Zhuang

Editorial board

R. M. Atlas University of Louisville USA	Alan Baker The University of Melbourne Australia	Nigel Bell Imperial College London United Kingdom	Tongbin Chen Chinese Academy of Sciences China
Maohong Fan University of Wyoming Wyoming, USA	Jingyun Fang Peking University China	Lam Kin-Che The Chinese University of Hong Kong, China	Pinjing He Tongji University China
Chihpin Huang "National" Chiao Tung University Taiwan, China	Jan Japenga Alterra Green World Research The Netherlands	David Jenkins University of California Berkeley USA	Guibin Jiang Chinese Academy of Sciences China
K. W. Kim Gwangju Institute of Science and Technology, Korea	Clark C. K. Liu University of Hawaii USA	Anton Moser Technical University Graz Austria	Alex L. Murray University of York Canada
Yi Qian Tsinghua University China	Jiuhui Qu Chinese Academy of Sciences China	Sheikh Raisuddin Hamdard University India	Ian Singleton University of Newcastle upon Tyne United Kingdom
Hongxiao Tang Chinese Academy of Sciences China	Shu Tao Peking University China	Yasutake Teraoka Kyushu University Japan	Chunxia Wang Chinese Academy of Sciences China
Rusong Wang Chinese Academy of Sciences China	Xuejun Wang Peking University China	Brian A. Whitton University of Durham United Kingdom	Po-Keung Wong The Chinese University of Hong Kong, China
Min Yang Chinese Academy of Sciences China	Zhifeng Yang Beijing Normal University China	Hanqing Yu University of Science and Technology of China	Zhongtang Yu Ohio State University USA
Yongping Zeng Chinese Academy of Sciences China	Qixing Zhou Chinese Academy of Sciences China	Lizhong Zhu Zhejiang University China	Yahui Zhuang Chinese Academy of Sciences China

Editorial office

Qingcai Feng (Executive Editor) Zixuan Wang (Editor) Suqin Liu (Editor) Zhengang Mao (Editor)
Christine J Watts (English Editor)

Journal of Environmental Sciences (Established in 1989)

Vol. 24 No. 7 2012

Supervised by	Chinese Academy of Sciences	Published by	Science Press, Beijing, China
Sponsored by	Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences	Distributed by	Elsevier Limited, The Netherlands
Edited by	Editorial Office of Journal of Environmental Sciences (JES) P. O. Box 2871, Beijing 100085, China Tel: 86-10-62920553; http://www.jesc.ac.cn E-mail: jesc@263.net , jesc@rcees.ac.cn	Domestic	Science Press, 16 Donghuangchenggen North Street, Beijing 100717, China Local Post Offices through China
Editor-in-chief	Hongxiao Tang	Foreign	Elsevier Limited http://www.elsevier.com/locate/jes
CN 11-2629/X	Domestic postcode: 2-580	Printed by	Beijing Beilin Printing House, 100083, China
		Domestic price per issue	RMB ¥ 110.00

ISSN 1001-0742



9 771001 074123