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Comparison of Organic Matter Properties and Disinfection By-Product Formation between the Typical Groundwater and Surface Water

Chonghua Xue ¹, Ying Yu ^{2,3} and Xin Huang ^{2,*}

- ¹ Key Laboratory of Urban Stormwater System and Water Environment, Ministry of Education, Beijing University of Civil Engineering and Architecture, Beijing 102616, China; xuechonghua@bucea.edu.cn
- ² Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China; yingyu_st@rcees.ac.cn
- ³ University of Chinese Academy of Sciences, Beijing 100049, China
- * Correspondence: huangxin1212@163.com or xinhuang@rcees.ac.cn

Abstract: The disinfection by-product (DBP) formation was affected by the dissolved organic matter (DOM). Therefore, the DOM properties and DBP formation potential of the two most widely used source waters: groundwater (GW) and surface water (SW), were comparatively studied in this work. The results suggested that the GW mainly consisted of protein-like organics with smaller molecular weight (Mw) less than 3000 Da, while the SW contained the humic- and fulvic-like substances with larger Mw. The tap water DBP concentration of GW as source water was lower than that of SW as well as the cytotoxic index (CI). The total DBP formation potential of the SW under chlorine and chloramine disinfection was higher than that of GW, especially the trihalomethanes (THMs) and haloacetic acid (HAAs). The higher THM and HAA formation potential of the SW was mainly attributed to the relatively hydrophobic and aromatic humic and fulvic substances. The halonitromethanes (HANs) formation was mainly due to the less hydrophobic protein-like components with smaller Mw. In addition, the total CI of the GW was lower than the SW under both chlorine and chloramine disinfection. Therefore, for the DBPs control, using the GW as source water was more beneficial to human health.

Keywords: organic matter properties; disinfection by-products; surface water; groundwater



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1. Introduction

Disinfection by-products formed by the chemical disinfection with dissolved organic matter (DOM) is a vital issue in drinking-water production. It has been reported by many studies that the DBPs have high cytotoxicity, mutagenicity, and carcinogenicity [1,2]. On account of the non-negligible health risks and widespread occurrence of DBPs in drinking water, some countries and organizations have regulated some common DBPs including trihalomethanes (THMs) and haloacetic acid (HAAs) [3]. In addition, nitrogenous disinfection by-products, including haloacetonitriles (HANs) etc., have attracted much attention in recent years because they have higher cytotoxicity and genotoxicity than traditional carbonous disinfection by-products even though they emerge in lower concentrations than THMs and HAAs in drinking water [4,5].

In practice, though DBP formation by chlorine disinfection is inevitable [6], it still could be controlled by the alteration of the DOM properties and quantity, which is suggested to be the predominant precursor of the DBPs [7]. Therefore, the effects of many water treatment technologies on the DOM and DBP control performance have been studied [8–10]. Enhanced coagulation and activated carbon (AC) adsorption are also suggested as two efficient technologies to control DBPs by removing the DOM. Zhao et al. [11] have suggested that THM's formation potential (THMFP) and HAAFP can be decreased by 51% and 59%,

respectively, through the enhanced coagulation with polyaluminum chloride. The granular AC (GAC) contactor in a two-stage biofiltration process and a sand biofilter coupled with a biologically-active GAC contactor was reported to account for $60.63\% \pm 16.64\%$ of the total DBPFP removal [12]. Other treatment technologies, such as advanced oxidation, could also alter the DOM properties, and thus reduced the DBP formation. The UV/H₂O₂ process, which could form the non-selective hydroxyl radical (HO•), has proven that it can significantly degrade phenicol antibiotics including florfenicol, chloramphenicol, and thiamphenicol [13], and the related TCMFP formed in post-chlorination decreased with UV/H₂O₂ treatment compared to UV [14]. Chin and Berube (2005) suggested that the UV/O₃ could achieve about 50% total organic carbon mineralization rated and about 80% and 70% reductions in THMFP and HAAFP, respectively [15].

Except for the effect of the treatment process, the water source is also an important factor affecting the water matrix due to the geological variations and heterogeneity of NOM [16,17]. Wang et al. [18] compared the components and DBPFP of the algae organic matter (AOM) and NOM from a typical eutrophic lake and suggested that AOM were mainly composed of aromatic protein-like and soluble microbial by-product-like matters, while NOM were chiefly contributed from humic acid-like and soluble microbial by-product-like materials, and AOM displayed higher N-DBPFPs than NOM. Xu et al. [19] identified the surface water DOM in northeastern China by PARAFAC, and suggested that DBPFP was significantly correlated with four humic- and fulvic-like components and the source and types of DOM affect the formation of DBPs. Jutaporn et al. [20] separated the DOM from two water sources—a surface water and a secondary treated wastewater effluent—and studied the DBPFP from different DOM species. They found that the removal of the humic-like component and protein-like component correlated with THMFP and HAAFP removal by MIEX[®] resins, respectively. Here, similar studies which also focused on the relationships between source water and DBP formation are summarized and their differences concluded with our study as shown in Table 1. However, there are limited studies focused on their different DOM characteristics and DBPFP, especially for the three-dimensional excitation emission matrix (3D-EEM) character and DBP toxicity.

Table 1. The comparison of this work with other similar studies.

Source Water	Similar Results	Difference	Reference
SW and GW using Yellow River water	SW achieved much higher DBP concentrated in the finished water.	Lack of tap water data and toxicity evaluation, and did not compare the DBPs with the source water NOM characteristics.	[21]
Sea (SWR), river (RWR), and reservoir (RVR) water	The THMFP: RSR > SWR > RVR for chlorination SWR > RSR > RVR for ClO ₂ treatment.	Lack of tap water data and toxicity data, and did not compare the DBPs with the source water NOM characteristics.	[22]
Taihu Lake water	19 possible precursors of the discovered DBPs were detected, with the aromatic compounds being a major group.	Mainly focused on the unknown DBPs detection and only one surface source water.	[23]
Reservoir water	The source, types, and humification degree of DOM affect the formation of DBPs.	Only the surface source water and lacks the toxicity evaluation.	[19]

Therefore, this study was aimed to comparatively investigate the water characteristics and the DBP formation of the typical GW and SW. The water properties of the treated effluent from the water treatment plants using GW and SW as source water were characterized by the 3D-EEM and molecular weight (Mw). The DBP concentration of the real tap waters from these two treatment plants and their DBPFP by the chlorine and chloramine were

also comparatively analyzed. In addition, the cytotoxicity of the formed DBPs was also calculated to evaluate their health risk.

2. Materials and Methods

2.1. Test Water

During this study, the water samples (GW and SW) used for water properties characterization were collected from the influent water and finished water by two water treatment plants using GW and SW water before disinfection. The DBPFP test was conducted with the finished water from the water treatment plants before disinfection. The tap water GW and SW were collected from the coverage area of these two different water treatment plants for the real DBP analysis. These two water treatment plants (finished water) and their corresponding distribution systems (tap water) are located in Beijing City in northern China.

2.2. Water Characterization

Basic water quality indexes including turbidity, pH, total dissolved solids (TDS), $\text{NH}_3\text{-N}$ etc., were measured according to the National Environmental Quality Standards for Surface Water (Environmental Quality Standards for Surface Water (GB3838–2002)). The DOM in treated water samples of GW and SW was characterized by the UV254, dissolved organic carbon (DOC), the 3D-EEM fluorescence spectrum and molecular weight (Mw) distribution using a UV-6100 double beam spectrophotometer at 254 nm using 1 cm quartz cells, TOC analyzer (TOC-VCPH, Shimadzu, Japan), luminescence spectrometer (F-4500, Hitachi, Japan), excitation (Ex) and emission (Em) ranges, which were set at 200–400 nm and 220–550 nm with scan steps of 5 nm and using HPSEC consisting of a high-performance liquid chromatography system (Waters 1525, Waters, Milford, MA, USA) and a size exclusion chromatography column (TSKgel G3000SWXL, Tokyo, Japan), respectively [24].

2.3. DBP Analysis

The tap water GW and SW filtered through 0.45 μm membrane filters were directly used for the DBP measurement. The stock solution and experiment procedure were referenced from a previous study [25]. For the DBPFP experiment, the water samples were reacted with 0.1 mM chlorine or chloramine for 72 h, after that the residual chlorine was quenched by the $\text{Na}_2\text{S}_2\text{O}_3$ with a molar ratio of 1:1.2. Then, the sample was filtered with a 0.45 μm membrane filter before the DBPs measurement.

DBP analysis was conducted on a gas chromatograph with an electron capture detector (GC/ECD) (Agilent 7890, Agilent Technologies, Santa Clara, CA, USA); the column used was an HP-5 fused silica capillary column (30 m \times 0.25 mm I.D. with 0.25 μm film thickness) based on USEPA Standard Methods 551.1 and 552.3. For the HAAs analysis, the samples were pretreated with an extraction/derivatization procedure by MTBE and acid methanol (Fang et al. 2010). In this study, THMs include trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and tribromomethane (TBM); HAAs include monochloroacetic acid (MCAA), monobromoacetic acid (MBAA), dichloroacetic acid (DCAA), dibromoacetic acid (DBAA), and trichloroacetic acid (TCAA); and HANs include dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), bromochloroacetonitrile (BCAN) and dibromoacetonitrile (DBAN).

2.4. Toxicity Evaluation

To evaluate the different disinfection agent health risks in different raw water, the DBP cytotoxicity indexes (CI) of evaluated DBPs were referenced as the product of unit toxicity indexes and DBP concentrations [26]. The reciprocal of LC50 values for THMs, HAAs, and HANs was suggested as the unit cytotoxicity index (CI) values [2]. The calculated CI data presented were calculated based on the method reported in the previous study [27] on the toxic potential of both regulated and emerging DBPs.

3. Results and Discussion

3.1. Comparison of Organic Matter Characteristics

Table 2 exhibits the basic water characteristics of the GW and SW samples. It should be noted that the GW has higher TDS and lower pH. The DOC of the GW sample was lower than that of SW, which was consistent with the previous study [28] which suggested the DOM of the uncontaminated GW was around 1 mg L^{-1} . The N content of the GW was almost the same as the SW water. Specifically, the $\text{NH}_3\text{-N}$ of the GW sample was slightly greater than that of the SW, while the $\text{NO}_3\text{-N}$ of the GW sample was less. The effluent Mw and 3D-EEM distribution of the GW and SW are shown in Figures 1 and 2, respectively. It should be noted that the organic matter in the GW had a smaller Mw, ranged from 1000 to 3000 Da, while the Mw of the SW organics ranged from 2000 to 10,000 Da. In addition, there was only one peak in the GW, and for the SW, three main peaks around 2000, 4000, and 7000 Da were presented, which may suggest that the organic matter in the GW was more simplex.

Table 2. Water index of groundwater (GW) and surface water (SW).

Water Sample	GW		SW	
	Effluent	Influent	Effluent	Influent
Turbidity (NTU)	0.21	0.23	0.24	2.70
pH	7.43	7.45	8.26	8.41
TDS	274	280	156	140
$\text{NH}_3\text{-N}$ (mg/L as N)	0.12	0.12	0.09	0.16
$\text{NO}_3\text{-N}$ (mg/L as N)	9.58	10.31	11.62	15.32
DOC (mg/L)	1.97	2.01	2.23	3.48
NO_3^- (mg/L)	42.44	45.66	51.48	69.22

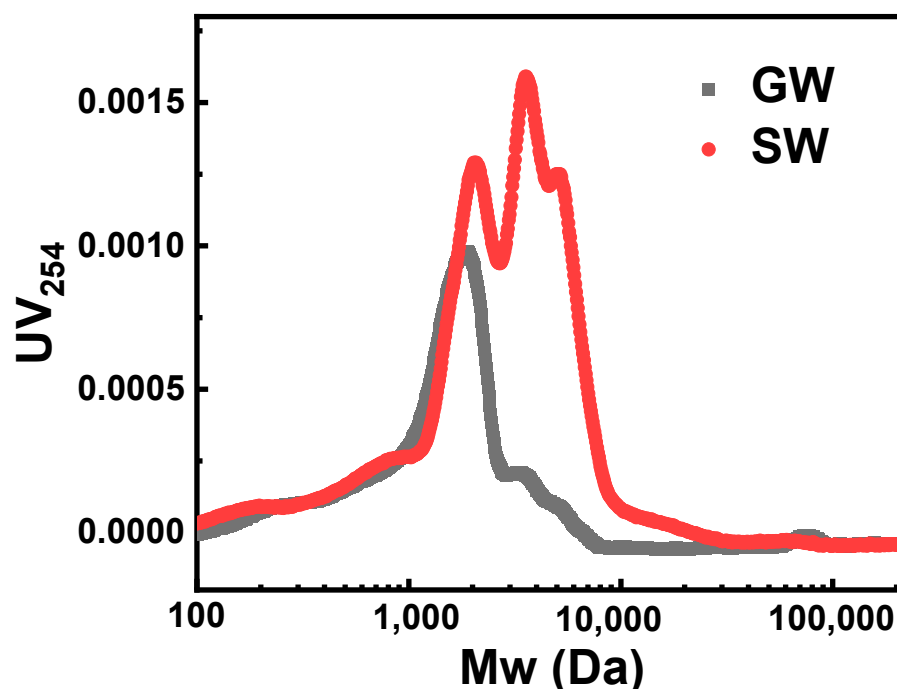


Figure 1. The Mw distribution of the groundwater and surface water.

As shown in Figure 2, the 3D-EEM intensity of the GW was much smaller than that of the SW, which was correlated with the DOC results. As suggested by the previous study [29], the 3D-EEM could be divided into five regions based on the differences of Ex-Em wavelengths of organic matters: region I (Ex220–250 nm, Em280–330 nm) was regarded as tyrosine; region II (Ex220–250 nm, Em330–380 nm) was correlated to tryptophan; region

III (Ex220–250 nm, Em380–450 nm) was related to the fulvic acid-like materials; region IV (Ex250–400 nm, Em380–500 nm) was related to the soluble microbial by-product-like substance; and region V (Ex250–400 nm, Em280–380 nm) was linked to the humic acid-like organics. It should be noted that the GW contained mainly the tyrosine and tryptophan-like organic matter and very little fulvic and microbial by product-like substance. For the SW, similarly it mainly consisted of the tyrosine and tryptophan-like organic matter. Differently, it contained some humic-like organics which did not appear in the GW sample, and the fulvic acid-like materials of the SW were slightly higher than that in the GW sample. In addition, the ratio of fluorescence intensity at Ex 370 nm/Em 450 nm and Ex 370 nm/Em 500 nm could indicate the source of the organic contaminant of GW [30]. Since the ratio was as low as 1, it could be suggested the GW sample was not interfered with by the exogenous pollution. Thus, these two samples could well represent the GW and SW.

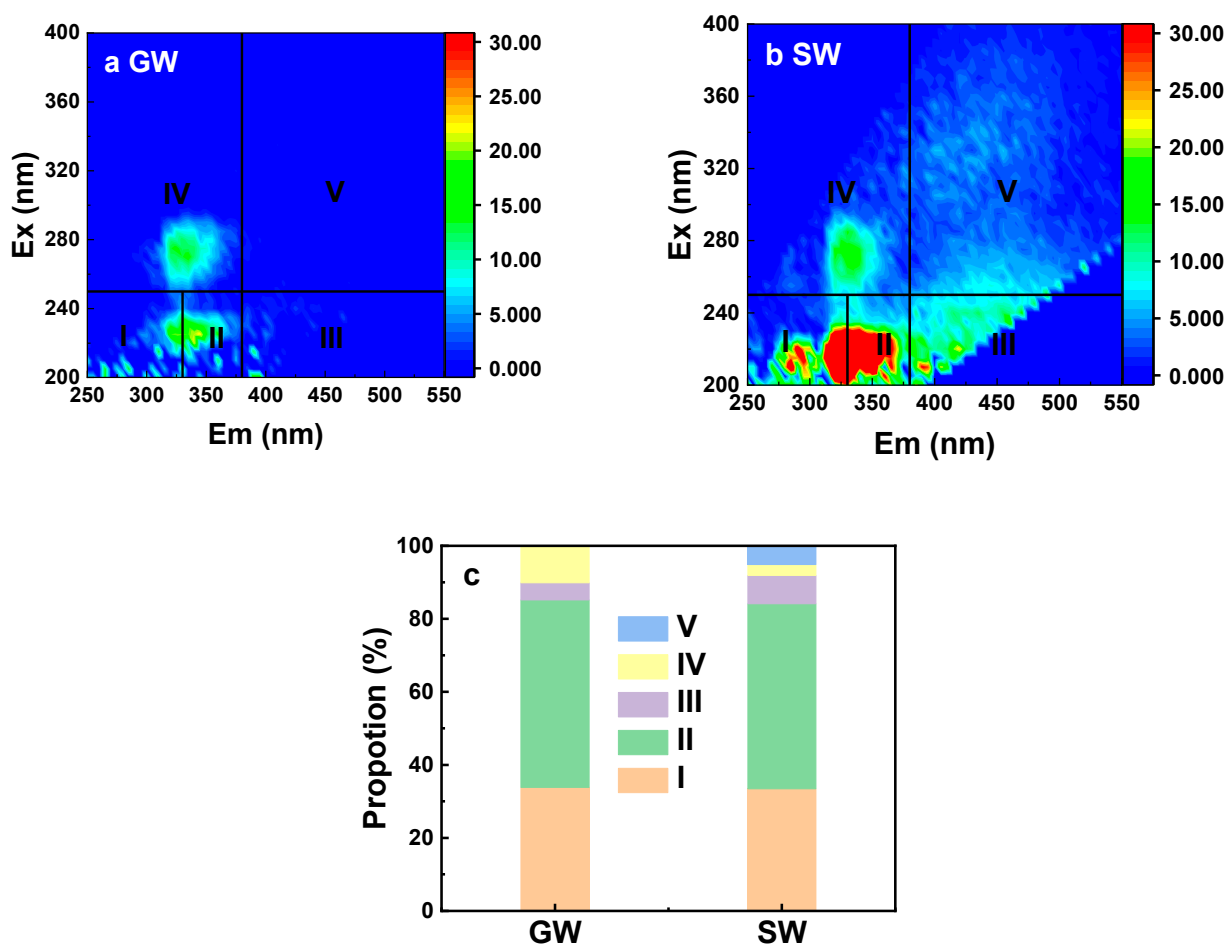


Figure 2. The 3D-EEM spectra of the GW (a) and SW (b); the fluorescence regional integration (FRI) of GW and SW (c).

3.2. Comparison of the DBPs in the Tap Water

The different tap water samples sourced from these two water treatment plants were collected and the contained DBPs were analyzed as shown in Figure 3. It should be noted that the DBP concentrations in the SW were higher than those in the GW, especially the HAAs. The HAA concentration in the SW was 15 $\mu\text{g/L}$ higher than that in the GW. For the THMs, the TCM was the most in both the GW and SW. The TCM concentration was higher in the SW, while other THM concentrations including BDCM, DBCM, and TBM were slightly higher in the GW. For the HAAs, the TCAA and DCAA were the most and much higher in the SW, while other HAAs, which contained Br, were slightly higher than that in the GW. For the HANs, the DCAN concentration was slightly higher in the SW than

that in the GW, while the DBAN and BCAN was higher in the GW than in the SW. Above all, it should be noted that all of the DBPs containing Br were higher in the GW, implying the GW contained more Br⁻.

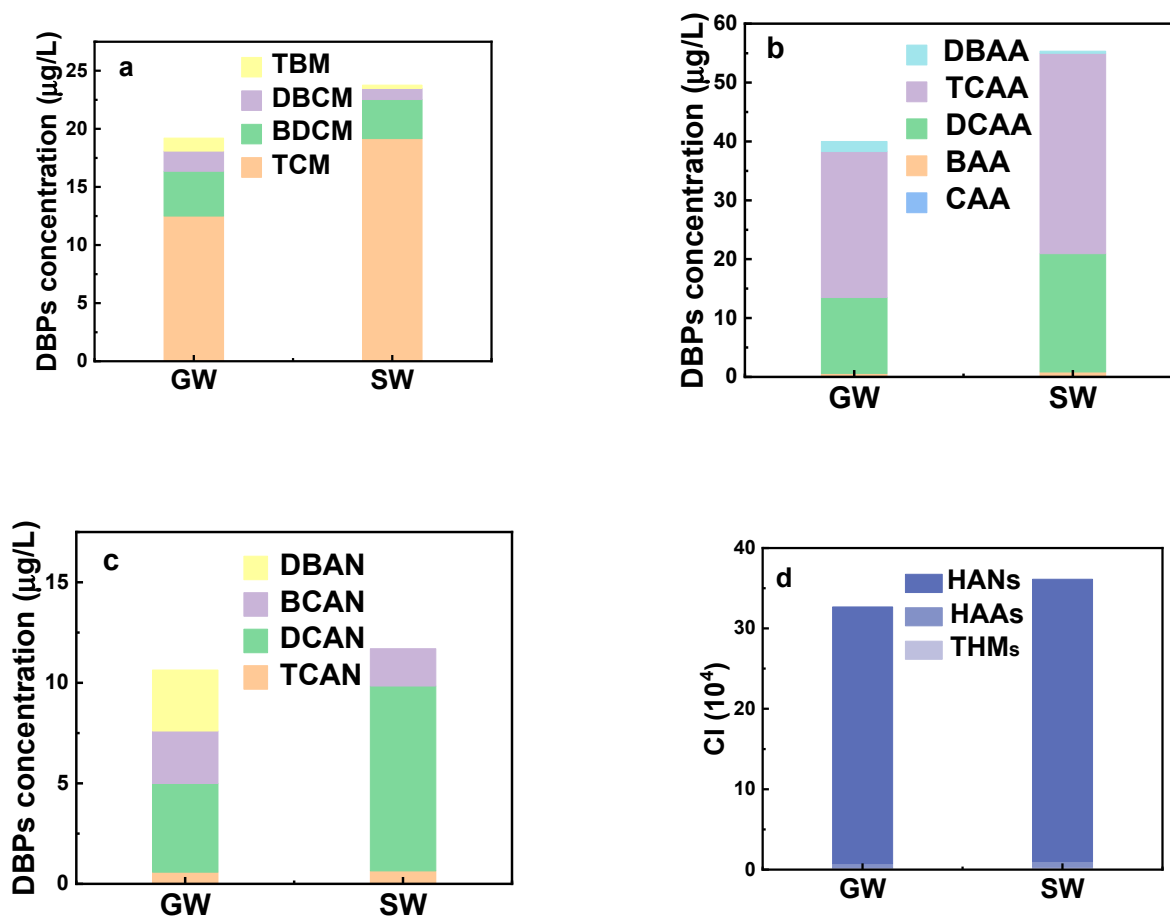


Figure 3. The DBP concentrations in the tap water from different water sources: (a) THMs; (b) HAAs; (c) HANs; (d) the cytotoxicity indexes of THMs, HAAs, and HANs.

The calculated total mean CI values of the DBPs in GW and SW are shown in Figure 3d. It should be noted that the total mean CI of GW was significantly lower than that of SW. In addition, compared with THMs and HAAs, HANs contributed the most to the CI. All of the DBP concentrations in the GW were lower than those in the SW, and thus the calculated CI value was smaller for the GW. As mentioned in Section 3.1, the tyrosine and tryptophan-like substances were higher in the SW and the humic- and fulvic-like substances were only detected in the SW. The total DOC of the SW was also greater than that of the GW, and thus the fact that more DBPs were generated was reasonable. To further compare the DBPs formed by the GW and SW, their DBPFP per DOC was analyzed in the following section.

3.3. Comparison of DBPFPs by Chlorine and Chloramine

The DBPFP by the chlorine and chloramine were shown in Figures 4 and 5. It should be noted that the DBPs formed by chlorine for both the GW and SW were higher than those by chloramine, even the HANs which contained N. In addition, the distribution of HAAs and HANs was different. For THMs, TCM was the main one for both chlorine and chloramine disinfection due to the low Br content in both the GW and the SW. For HAAs, the CAA concentration was much higher for chlorine disinfection than chloramine disinfection. For the HANs, the DCAN was the dominant one for chlorine disinfection, while the TCAN was the dominant one for the chloramine disinfection. Generally, the DBPFP of the SW was significantly higher than that of the GW, especially for the chloramine

disinfection. It should be noted that, although the N-NH_3 concentration in the GW was slightly higher than that in the SW, the HAN concentrations, especially DCAN, of the GW were lower in both chlorine and chloramine disinfection. Previous studies suggested that the reactions between ammonia and the excessive chlorine could form NO_3^- and other products, which may contribute to the DCAN formation [31]. The NO_3^- concentration in the SW was slightly higher, which may result in the higher HAN concentration. For the THMs and HANs, the DBPFP for the SW water was more than twice that of the DBPFP for the GW. For the THMs, the TCM was the dominant one in the SW, while in the GW its proportion was low. Even for the chloramine disinfection, the TCM was not present in the GW. For the HAAs, TCAA and CAA were the most two DBPs in the SW, while in the GW the proportion of DCAA was higher, especially under the chloramine disinfection. The hydrophobic and higher aromatic contents have been suggested to be the main procedure of the THMs and HAAs by previous studies. As suggested in Section 3.1, the SW contained more humic and fulvic substances, which were more hydrophobic and aromatic; therefore more THMs and HAAs were generated. For the HANs, for both chlorine and chloramine disinfection, the differences between these two water samples were relatively small. As suggested by the previous study, the N contained DBP formation was more related to hydrophilic substances with smaller Mw [32]. As suggested by Figures 2 and 3, though the total DOM in the GW was less, the DOM in the GW was mainly proteins with smaller Mw. Compared to the humic and fulvic components, the protein-like materials were less hydrophobic; thus the gap for the precursor for the HANs were smaller, and thus the gap between the HANs formed by the GW and SW were not as much as the THMs and HAAs.

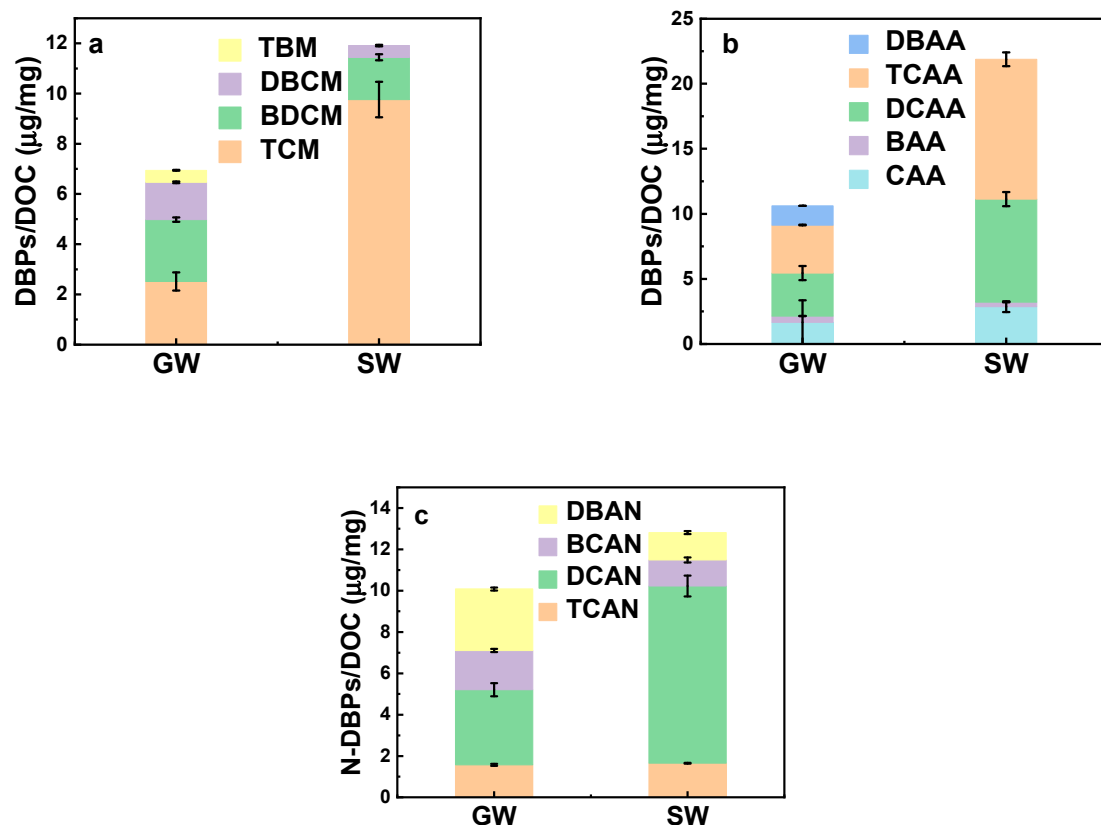


Figure 4. The DBPFP of the GW and SW by chlorine disinfection: (a) THMs; (b) HAAs; (c) HANs.

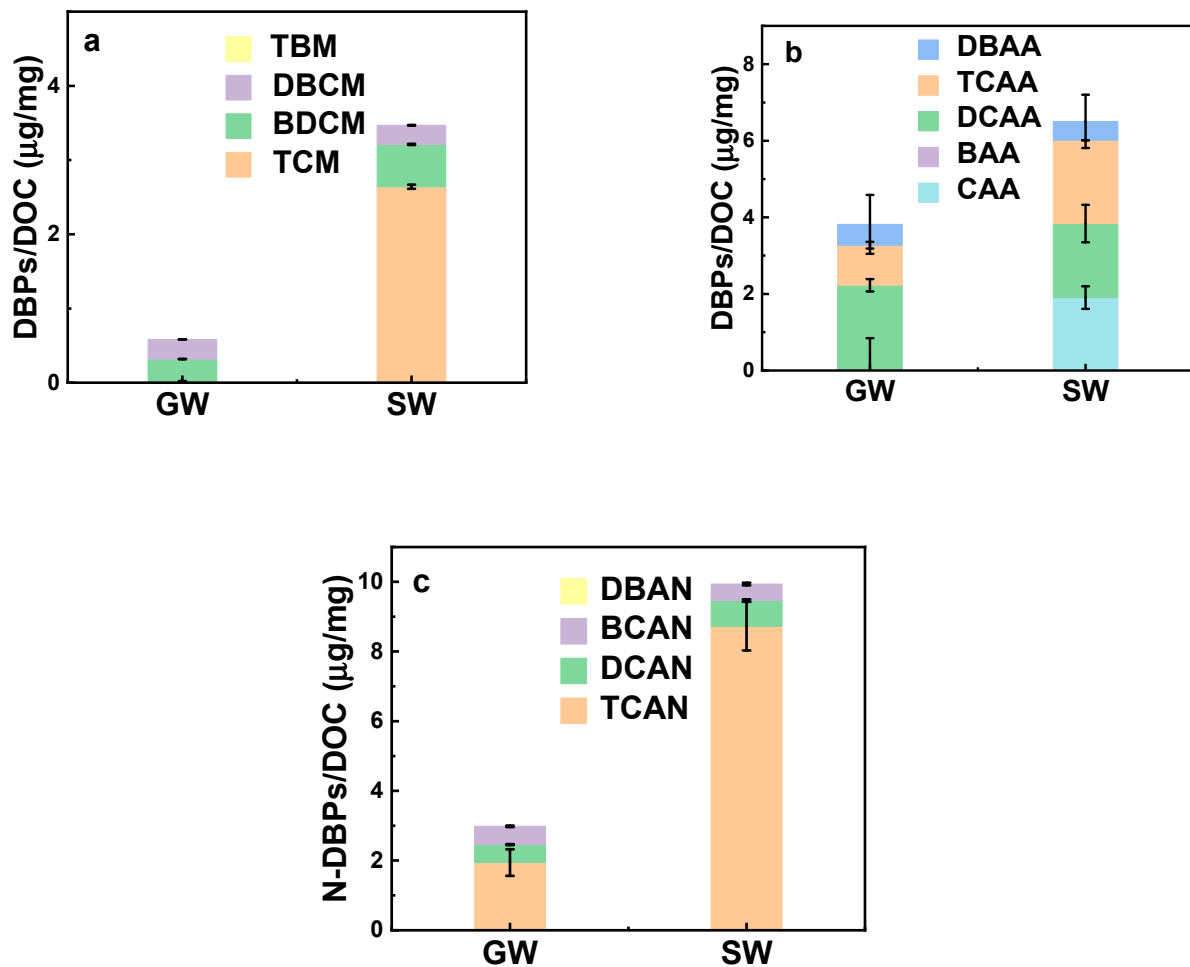


Figure 5. The DBPFP of the GW and SW by chloramine disinfection: (a) THMs; (b) HAAs; (c) HANs.

The CI of the GW and SW under chlorine and chloramine disinfection were also calculated as shown in Figure 6. Similarly, the CI of the GW and SW was dominated by the HANs. The HAAs and THMs contribute a little to the CI index. Generally, the CI indexes of the chloramine disinfection were significantly lower than those of the chlorine disinfection, especially for the GW by the chloramine disinfection. The CI index of the GW under chloramine disinfection was less than 10×10^4 , which was only one third of that under chlorine disinfection, which suggested that the NOM in the GW water was less reactive to the chloramine compared with chlorine. As discussed in Section 3.2, the DOM in the GW was mainly the tyrosine and tryptophan-like organic matter and microbial by-product-like substance. The GW did not contain any HA-like materials, which might indicate that the HA-like substance was more reactive to chlorine as well as chloramine, and therefore the DBPFP per mg DOC of the SW was much higher than that of the GW.

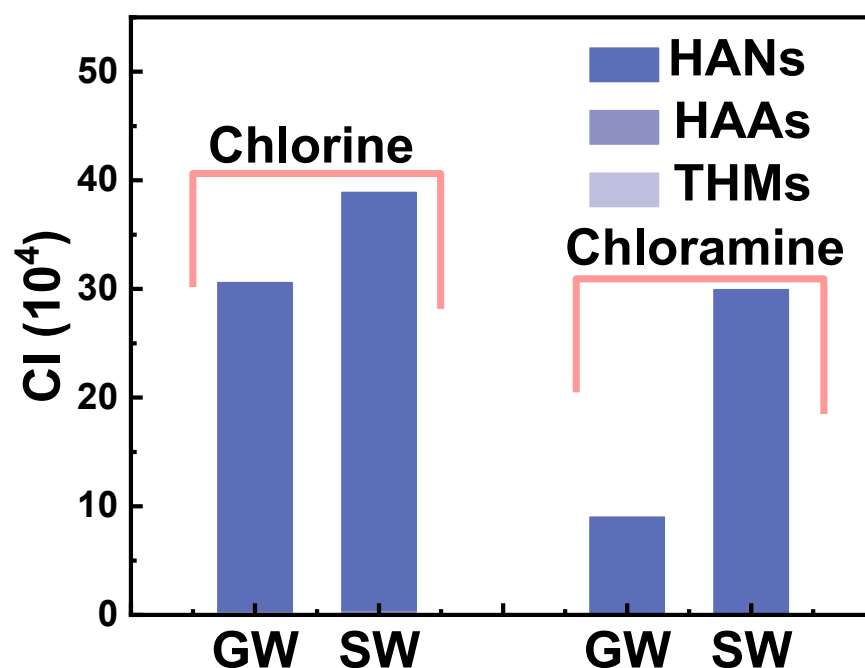


Figure 6. Cytotoxicity indexes of THMs, HAAs, and HANs of GW and SW by chlorine and chloramine disinfection.

4. Conclusions

This study compared the DOM properties, DBP formation, and DBP toxicity of GW and SW. The results showed that the GW contained low Mw, tyrosine and tryptophan-like organic matter, and microbial by-product-like substance, and little humic and fulvic substances with high Mw. Therefore, the DBP concentration and CI index were low for the GW. The DBPFP per mg DOC for the GW was also lower than that for the SW under both chlorine and chloramine disinfection. The SW achieved higher DBPFP due to its high proportion of humic and fulvic fractions, which was suggested to be the main procedure of the DBPs, and the CI value was also lower for the GW. Therefore, for the DBP control, using GW as source water led to low health risk.

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