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Dissolved organic matter fractions and disinfection by-product formation potential from major raw waters in the water-receiving areas of south-to-north water diversion project, China

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ABSTRACT

The raw waters from the Yellow River and the Danjiangkou Reservoir are the two most important sources of water in the water-receiving areas of the south-to-north water diversion project (SNWDP). This paper compared the constitution of dissolved organic matter (DOM) in the two waters based on XAD resins isolation, ultrafiltration fractionation, and the disinfection by-product formation potential of each fraction. The dissolved organic carbon (DOC), UV_{254}, trihalomethanes, and nitrogenous disinfection by-product (N-DBP) formation potential of the raw water from the Yellow River were higher than those from the Danjiangkou Reservoir. The molecular weight (MW) < 1 kDa fraction contained the most DOM and dominated the major THM formation potential (THMFP) and N-DBP formation potential (N-DBPFP) in both waters. The hydrophilic fraction possessed the highest proportion in the Yellow River water, while the transphilic fraction was predominant in the Danjiangkou Reservoir water. In both waters, the hydrophobic fraction owned the highest THMFP and the hydrophilic fraction contained the highest N-DBPFP. Meanwhile, the molecular weight (MW) < 1 kDa and hydrophilic fractions contributed more bromated THMFP than the other fractions. Very toxic bromoform, bromochloroacetonitrile, and dibromoacetonitrile were only detected in DOM fractions from the Yellow River water. Therefore, the hydrophobic/hydrophilic organic compounds with low MW in both raw waters and bromide in Yellow River water should receive attention during the upgrading and reforming of drinking water treatment processes for adapting waters with different qualities in the water-receiving areas of SNWDP.

Keywords: South-to-north water diversion project; Dissolved organic matter; Molecular weight; Hydrophobic–hydrophilic; Disinfection by-product; Formation potential

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1. Introduction

Natural dissolved organic matter (DOM) is an extremely complex mixture of organic compounds, which is a major contributor to dissolved organic carbon (DOC) in water. DOM with complex structures and functional groups tend to be the precursors of carcinogenic disinfection by-products (DBPs) in chlorinated or chloraminated waters [1,2]. It is reported that the removal of DBP precursors before disinfection is a more effective and economical way than altering disinfection conditions or introducing other advanced technology to remove DBPs after their formation [3,4]. To minimize the formation of hazardous DBPs, it is necessary to isolate and fractionate the DOM based on different physicochemical properties (such as molecular weight (MW) and hydrophobic–hydrophilic) for characterization and reactivity studies [5]. Knowledge of DBP formation is important for identifying potential DBP precursors and for selecting better treatment strategies.

Ultrafiltration (UF) is one of the prevalent technologies used to isolate DOM into different MW fractions. Although MW distribution of DOM varies from source to source and can be affected by season and climate change [6,7], many studies have shown that low MW precursors played an important role in DBP formation [8–11]. Depending on adsorption affinity for synthetic resins (XAD-4 and XAD-8), DOM can be fractionated into hydrophobic, transphilic, and hydrophilic fractions [12–14]. Many studies compared the hydrophobic–hydrophilic features of DOM in the formation of DBPs [15]. In general, hydrophobic DOM was found to be a more important precursor for the formation of trihalomethanes (THMs) and haloacetic acids than the corresponding hydrophilic DOM [16,17]. Dissolved organic nitrogen (DON), as a small portion of DOM, is primarily composed of amino acids, amino sugars, amides, peptides, and heterocyclic-N compounds (e.g. pyrimidine, imidazole, purine, and porphyrins) [18]. Its low MW and strong hydrophilic feature make DON the crucial precursor of N-DBPs [18].

In order to balance the nation's water supply, China's South-to-North Water Diversion Project (SNWDP) aims to relieve the drought-ridden north of China by diverting water from the Yangtze River. Once it is completed, about 45 billion cubic meters of water will be transferred yearly to the areas with a population of 300 million. As one of the three parts of SNWDP, the middle route will come into service in 2014. As Fig. 1 shows, the middle route diverts water starting from the Danjiangkou Reservoir, flowing through Hubei, Henan, and Hebei Provinces to Beijing and Tianjin Cities in China. This great project can solve the problem of drinking water shortage for 110 million people. Currently, the Yellow River is the primary source of water for most residents along the middle route, especially in Henan Province. Even after the completion of the middle route of SNWDP, many drinking water treatment plants (DWTPs) will still take the Yellow River water as their source or spare source water. However, different characteristics exist between the Yellow River and Danjingkou Reservoir waters. Therefore, it is a new challenge for DWTPs to adopt different water treatment processes to accommodate those two sources of water in the water-receiving areas of SNWDP. The primary objectives of this research were to investigate the MW distribution and the hydrophobic–hydrophilic feature of DOM in these two water sources to reveal the relationship between disinfection by-product formation potential (DBPFP) and each DOM fraction. This study can provide technical support for the upgrade and reform of DWTPs in the water-receiving areas of SNWDP.

2. Materials and methods

2.1. Sample collection and preparation

In this study, water sample from the Yellow River was collected from a DWTP located in Zhengzhou City, China (34.76°N, 113.57°E, sample site 1 in Fig. 1) in January, 2013. Raw water from the Danjiangkou Reservoir was collected at a monitoring station in Danjiangkou City, China (32.67°N, 111.49°E, sample site 2 in Fig. 1). After collection, the raw water samples were immediately transported to the laboratory on ice. Upon arrival, the samples were filtered

Table 1
Quality parameters of raw water samples from the Yellow River and Danjiangkou Reservoir

Parameters	Yellow River raw water	Danjiangkou Reservoir raw water
pH	8.24	8.07
UV_{254} (cm^{-1})	0.058	0.048
NH_4^+-N (mg/L)	0.11	<0.02
$NO_3^-NO_3{}^-$-N (mg/L)	1.26	0.35
NO_2^--N (mg/L)	<0.001	<0.001
DOC (mg/L)	2.23	1.96
Cl^- (mg/L)	73.45	2.35
Br^- (mg/L)	0.060	0.038
SO_4^{2-} (mg/L)	25.78	3.73
PO_4^{3-} (mg/L)	<0.02	<0.02

Note: UV_{254}: Ultraviolet absorbance at 254 nm; DOC: dissolved organic carbon.

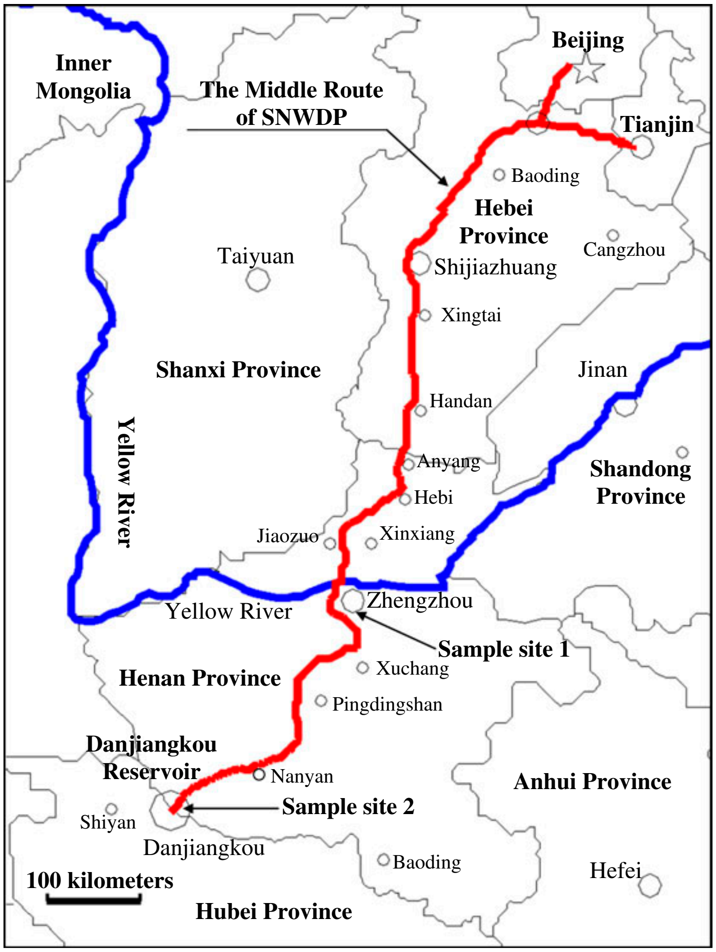

Fig. 1. Engineering presentation drawing of the middle route of China's SNWDP.

through 0.45-μm membrane filters (Millipore, USA) to remove suspended solids and then stored at 4℃ in the dark until usage. The quality parameters of these two raw water samples are shown in Table 1.

2.2. Chemicals

THMs mix standard solutions including chloroform (CF, CAS No. 67-66-3), bromodichloromethane (BDCM, CAS No. 75-27-4), dibromochloromethane (DBCM, CAS No. 124-48-1), and bromoform (BF, CAS No. 75-25-2) were purchased from Sigma–Aldrich (USA). Nitrogenous disinfection by-product (N-DBP) standard solutions including chloropicrin (TCNM, CAS No. 76-06-2), dichloroacetonitrile (DCAN, CAS No. 3018-12-0), trichloroacetonitrile (TCAN, CAS No. 545-06-2), bromochloroacetonitrile (BCAN, CAS No. 83463-62-1), and dibromoacetonitrile (DBAN, CAS No. 3252-43-5) were purchased from Sigma–Aldrich (USA). Sodium hypochlorite (NaOCl) solution (available chlorine 4.00–4.99%) was also purchased from Sigma–Aldrich (USA). Analytical grade reagents including NaOH, $Na_2S_2O_3$, KH_2PO4, anhydrous Na_2SO_4, and H_2SO_4 were purchased from Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China), without further purification. A seven-anion stock standard (F^-, Cl^-, Br^-, NO_2^-, NO_3^-, SO_4^{2-}, and PO_4^{3-}) was purchased from Shanghai ANPEL Scientific Instrument Co. Ltd (China). All solutions were prepared with ultrapure water produced from a Milli-Q water purification system (Millipore, USA). HPLC grade methyl tert-butyl ether (MTBE) was obtained from J.T. Baker (USA).

2.3. DOM isolation using UF membranes

DOM in raw water was isolated into five groups after filtration using a series of cellulose-derivative UF membranes (Millipore, USA) with MW cutoffs of 30, 10, 5, and 1 kDa, respectively, in dead-end stirred cells (Millipore, USA). As described in our previous study [19], Milli-Q water was used to pass through the membranes to remove any possible leached organics until UV_{254} in the percolate was less than 0.001 cm^{-1} before starting the DOM separation. Highly purified nitrogen (99.999%) was used to pressurize the filtration process (~0.15 MPa), and the percolates were collected and stored at 4℃ until analysis.

2.4. DOM fractionation using XAD resins

Water samples were acidified to pH 2 using concentrated sulfuric acid and then passed through XAD-8 resin followed by XAD-4 resin [12]. Effluent from the XAD-4 resin was collectedand referred to the hydrophilic fraction. The fraction referred to the hydrophobic DOM was retained by XAD-8 resin and can be eluted out with 0.1 M NaOH. The XAD-4 resin retained the transphilic fraction, which was also eluted with 0.1 M NaOH. The pH of the three fractions was adjusted to 7 using sulfuric acid or sodium hydroxide immediately after the elution, and the volumes of all fractions were adjusted to the initial sample volume. Before extraction, XAD-4 and XAD-8 resins were activated by methanol for 24 h and then swashed by Milli-Q water until UV_{254} of the percolates was less than 0.001 cm^{-1}.

2.5. Analytical methods

DOC, UV_{254}, trihalomethane formation potential (THMFP), and nitrogenous disinfection by-product formation potential (N-DBPFP) were analyzed for all fractions of DOM in the raw waters from the Yellow River and Danjiangkou Reservoir. The DOC and total dissolve nitrogen concentrations were determined by a Shimadzu TOC-VCSH analyzer with a TNM-1 TN unit (Shimadzu, Japan). The detection limit of DOC was 0.1 mg C/L. UV_{254} was measured by a spectrophotometer (SQ-4802 UV–vis spectrophotometer, UNICO, Shanghai) using a 1-cm quartz cell. Anions were analyzed using an ion chromatography (Dionex ICS-2000, USA) equipped with a conductivity detector, a Dionex AS11-HC analytical column (250 × 4.0 mm id), and a Dionex AG11-HC guard column (50 × 4.0 mm id). The THMFP and N-DBPFP experiments were conducted following the procedure described in our previous study [20]. The residual chlorine of the samples was quenched by NH_4Cl [21]. THMs and N-DBPs were quantified by liquid/liquid extraction with MTBE followed by gas chromatography (GC-2010, Shimadzu, Japan) with electron capture detection, and a fused silica capillary column (HP-5, 30 m × 0.25 mm id, 1 μm film thickness) according to the USEPA Method 551.1 [21]. The detection limit of CF, BDCM, DBCM, BF, TCNM, DCAN, TCAN, BCAN, and DBAN was 0.09, 0.03, 0.03, 0.05, 0.03, 0.05, 0.02, 0.03, and 0.03 μg/L, respectively.

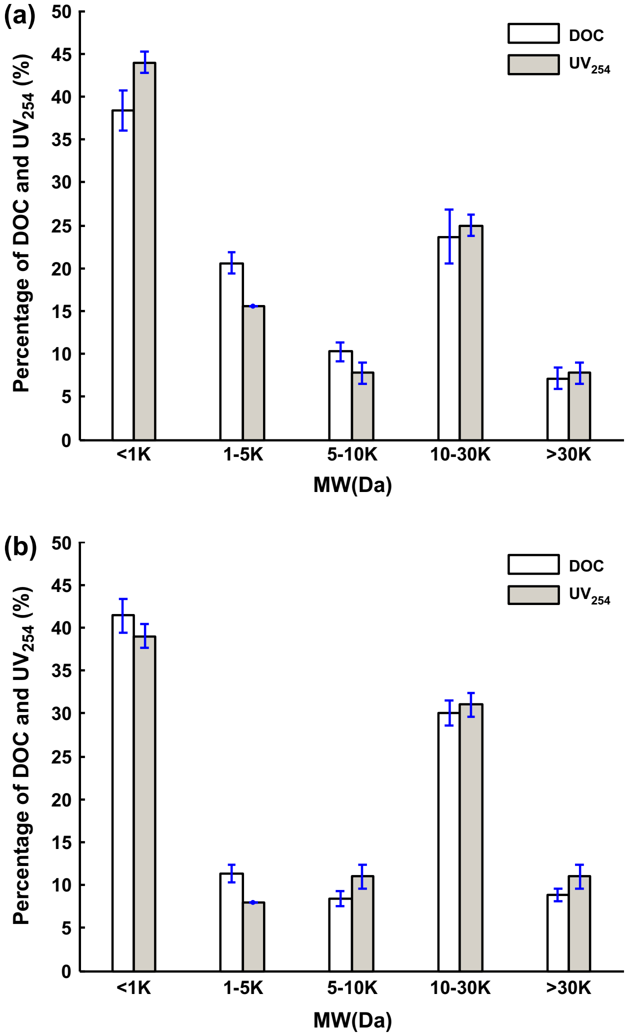

Fig. 2. MW distribution of DOM in the raw waters from the Yellow River (a) and Danjiangkou Reservoir (b).

3. Results and discussion

3.1. Characteristics of raw waters and DOM fractions

In this study, DOC and UV_{254} were selected as the main water quality parameters representing the content of organic matter. The average DOC concentration and UV_{254} of the Yellow River water were 2.23 mg C/L and 0.058 cm^{-1}, respectively. The corresponding values of raw water from the Danjiangkou Reservoir were 1.96 mg C/L and 0.048 cm^{-1}.

Fig. 2(a) and (b) show the DOC and UV_{254} of each MW fraction of DOM in the raw waters from the Yellow River and Danjiangkou Reservoir, respectively. Taking DOC as the index reflecting the contents of DOM, the MW < 1 kDa fraction in the Yellow River and Danjiangkou Reservoir occupied 38.4 and 41.4% of the total DOM, respectively, which were the dominant fractions in those two waters. In the raw water from the Yellow River, the MW fractions of both 1–5 and 10–30 kDa contributed more than 20% of the total DOM. In comparison, the MW fraction of 1–5 kDa in the raw water from the Danjiangkou Reservoir contributed to a much smaller percentage of DOM compared with MW fraction of 10–30 kDa, which constituted 30.1% of the DOM. The percentages of MW > 30 kDa fraction in both raw waters were less than 10%. Because the MW < 5 kDa fraction accounted for almost 60% of the total DOC concentration, it was concluded that most DOMs in those two raw waters were composed of small molecules, while the MW fraction of 10–30 kDa could not be neglected. Similar trends were also observed for UV_{254} of each MW fraction in those two waters. Many other studies also reported that the low-MW DOM was predominant for source water samples [12,14,17]. It is not easy to remove low-MW DOM during the processes of coagulation and sedimentation [11]. Therefore, it is very important to enhance the removal of low-MW DOM during the upgrading and reforming of drinking water treatment processes.

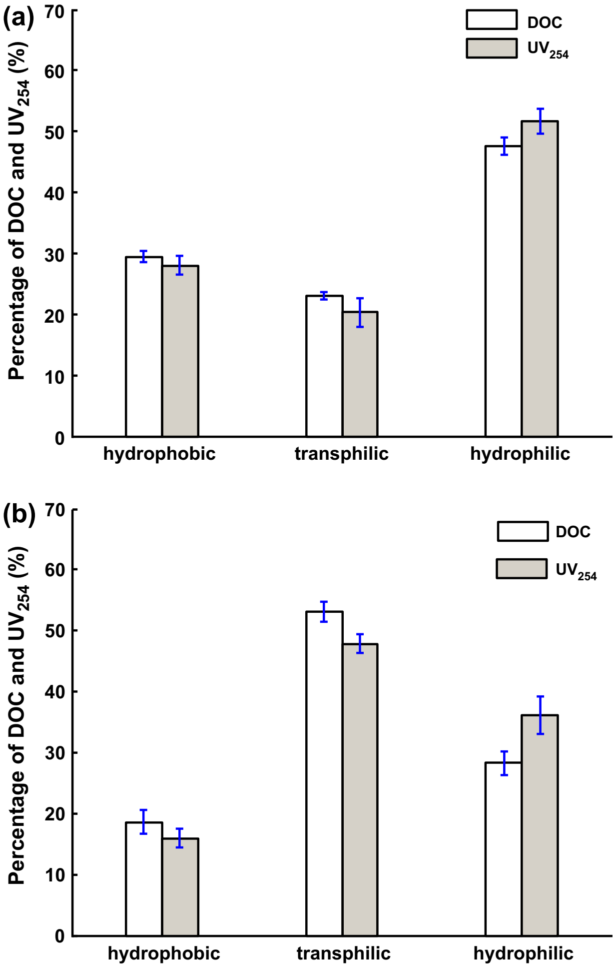

Fig. 3. Hydrophobic–hydrophilic feature of dissolved organic matter (DOM) in the raw waters from the Yellow River (a) and Danjiangkou Reservoir (b).

DOMs in raw waters from the Yellow River and Danjiangkou Reservoir were fractionated into three fractions using XAD-4 and XAD-8 resins. Fig. 3(a) and (b) show the distribution of the hydrophobic, transphilic, and hydrophilic fractions of DOM in those two waters. Taking DOC, for example, the hydrophilic fraction accounts for 47.5% of DOM, which was the dominant fraction in the Yellow River water. The results are consistent with the previous studies [11,22]. The other two basic fractions including transphilic and hydrophilic fractions approximated 25% of DOM in the Yellow River water. In the raw water from the Danjiangkou Reservoir, transphilic DOM was the major fraction containing 53.1% of the total DOC, and hydrophobic and hydrophilic fractions constituted 18.6 and 28.3% of total DOC, respectively. The sum of transphilic and hydrophobic fractions in the raw water from the Danjiangkou Reservoir was up to 70%, which indicated that humic-like contents accounted for the major part of DOM in Danjiangkou raw water. Because of more and more involvement of human activities in the Yellow River Basin, it was enriched with the hydrophilic DOM, thus leading to much higher hydrophilic fraction. UV_{254} of each hydrophobic–hydrophilic fraction had the similar trend like DOC in those two waters. The hydrophobic DOM is made up of mostly humic acid and fulvic acid, and the hydrophilic DOM contained more carbohydrate and amino acid [14,23,24]. Therefore, the Yellow River water contained more carbohydrate and amino acid, and the raw water from Danjiangkou Reservoir had relatively higher proportion of humic acid and fulvic acid.

3.2. *DBPFP of each DOM fraction*

In order to evaluate the DOM in both raw waters on a more fundamental level, the relationship between individual DOM fraction and DBP formation potential was investigated during the chlorination of both waters.

3.2.1. THMFP of each DOM fraction

Four THM species were formed (chloroform (CF), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform (BF)) in the Yellow River water during chlorination experiments. The concentration of BF in the Danjiangkou Reservoir water was below the detection limit. After chlorination, the THMFPs in raw waters from the Yellow River and the Danjiangkou Reservoir were 382.11 and 279.10 μg/L, respectively. Compared with CF, the formation of BDCM and DBCM were much lower in these two waters. However, the concentrations of BDCM and DBCM in the Yellow River water were 64.12 and 17.47 μg/L, respectively, which were much higher than those in the Danjiangkou Reservoir water (12.96 and 0.98 μg/L). The more formation of bromated THMs in the Yellow River was owing to the higher bromide concentration in the raw water from the Yellow River, which was confirmed in Table 1.

Fig. 4(a) shows the THMFP of each MW fraction of DOM in raw waters from the Yellow River and Danjiangkou Reservoir. The MW < 1 kDa fraction contained the maximum THMFP in both raw waters, which was consistent with the results of other studies [25–27]. The THMFP of MW < 1 kDa fraction in the Yellow River water and Danjiangkou water was 203.41 and 144.47 μg/L, respectively. The MW < 1 kDa fraction also had higher BDCM and DBCM formation in both raw waters. It was concluded that DOM fractions with lower MW were more reactive with bromide. In the raw water from the Yellow River, the MW fractions of 1–5, 5–10, 10–30 k, and > 30 kDa contained roughly similar THMFP (42.33, 44.49, 49.87, and 42.01 μg/L). However, the THMFP in the raw water from the Danjiangkou Reservoir decreased in the order of 10–30 k > 30 k > 5–10 k > 1–5 kDa. Generally, high-MW fractions of DOM are mainly aliphatic matters in nature, while low-MW fractions contain more aromatic and carboxyl groups, which are more reactive to the formation of THMs during chlorination [25].

Fig. 4(b) shows the THMFP of the hydrophobic, transphilic, and hydrophilic DOM fractions in the Yellow River and Danjiangkou Reservoir waters. As the figure shows, the hydrophobic DOM fraction got the highest THMFP during chlorination of these two waters (177.83 and 154.20 μg/L for the Yellow River and Danjiangkou Reservoir waters, respectively). The results indicated that the hydrophobic fraction was highly reactive with chlorine to form THMs. On the other hand, BDCM, DBCM, and BF were only detected in the hydrophilic DOM fraction. Some researchers [28] demonstrated that the hydrophilic aliphatic compounds easily reacted with bromide to form bromated THMs. Due to the higher bromide concentration in the Yellow River water, the hydrophilic DOM fraction produced more BDCM and DBCM compared with the Danjiangkou Reservoir water. The THMFP in the DOM fractions decreased in the order of hydrophobic > hydrophilic > transphilic for both waters. It was concluded that the hydrophobic DOM contained the most THM precursors in the raw waters from the Yellow River and Danjiangkou Reservoir. Other researchers [10,11,22] also had the same conclusion that hydrophobic DOM was the main THM precursor due to the richer possession of aromatic compounds [17,22]. Because hydrophobic DOM can be preferably removed by coagulation process, the risk of THMFP

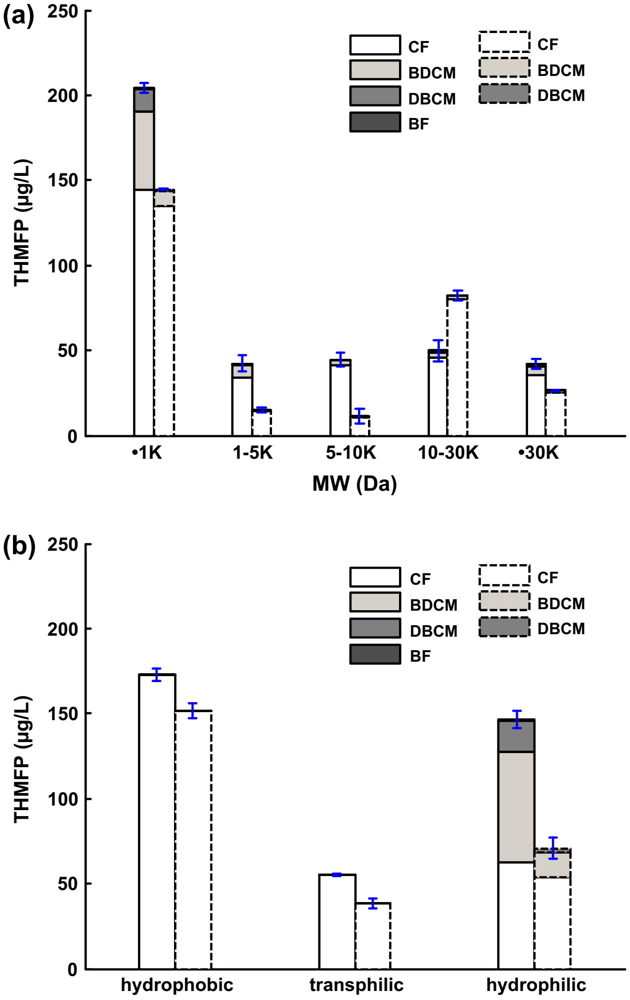

Fig. 4. THMFP of the MW fractions (a) and hydrophobic–hydrophilic fractions (b) of DOM in the raw waters from the Yellow River and Danjiangkou Reservoir. Bars with solid line: DOM fractions in the raw water from the Yellow River; bars with dashed line: DOM fractions in the raw water from the Danjiangkou Reservoir.

from the hydrophobic DOM could be effectively controlled [23]. However, the hydrophilic DOM possessing almost one third of THMFP in both waters showed limited removal during conventional water treatment processes, which could cause negative influence on the safety of drinking water [20].

3.2.2. N-DBPFP of each DOM fraction

Five N-DBPs (chloropicrin (TCNM), dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), bromochloroacetonitrile (BCAN), and dibromoacetonitrile (DBAN)) were detected in the raw water from the Yellow River, whereas only TCNM, DCAN, and TCAN were detected in the raw water from the Danjingkou Reservoir after chlorination experiments. In addition, the N-DBPFP in the Yellow River water was higher than that in the Danjiangkou Reservoir water, which was in accordance with the results of THMFP discussed in the previous section. The N-DBPFP in these two waters was 13.49 and 6.79 μg/L, respectively. Compared with other N-DBPFP, TCNM was the highest in the raw waters from the Yellow River and Danjiangkou Reservoir (8.13 and 6.25 μg/L respectively). The formation of DCAN and TCAN in the Yellow River water was higher than those in the Danjiangkou Reservoir water. Two important components of HANs, BCAN, and DBAN were only detected in the raw water from the Yellow River, with concentrations of 1.86 and 1.27 μg/L, respectively.

Fig. 5(a) shows the N-DBPFP of each MW fraction of DOM in the raw waters from the Yellow River and Danjiangkou Reservoir. The MW < 1 kDa fraction owned the highest N-DBPFP in both waters during chlorination (>75% of the total N-DBPFP), with concentrations of 10.43 and 6.02 μg/L for the Yellow River and Danjiangkou Reservoir waters, respectively. In these two raw waters, the concentrations of TCNM and TCAN were much higher in the MW <1 kDa fraction than those in the other MW fractions. Additionally, the MW < 1 kDa fraction in raw water from the Yellow River formed much more TCNM, DCAN, and TCAN than those in the raw water from the Danjiangkou Reservoir. In the Yellow River water, the MW fractions of <1 kDa and >30 kDa were the main contributors to the formation of BCAN and DBAN. The concentrations of BCAN and DBAN in the MW < 1 kDa fraction in the Yellow River water were 1.01 and 0.75 μg/L, respectively, which were nearly twice as much as those in the MW > 30 kDa fraction. As reported, DON with low MW such as amino acids, peptides, purines, and pyrimidines is the major precursor of HANs [29], which could explain why the MW < 1 kDa fraction produced the most N-DBPFP in both waters.

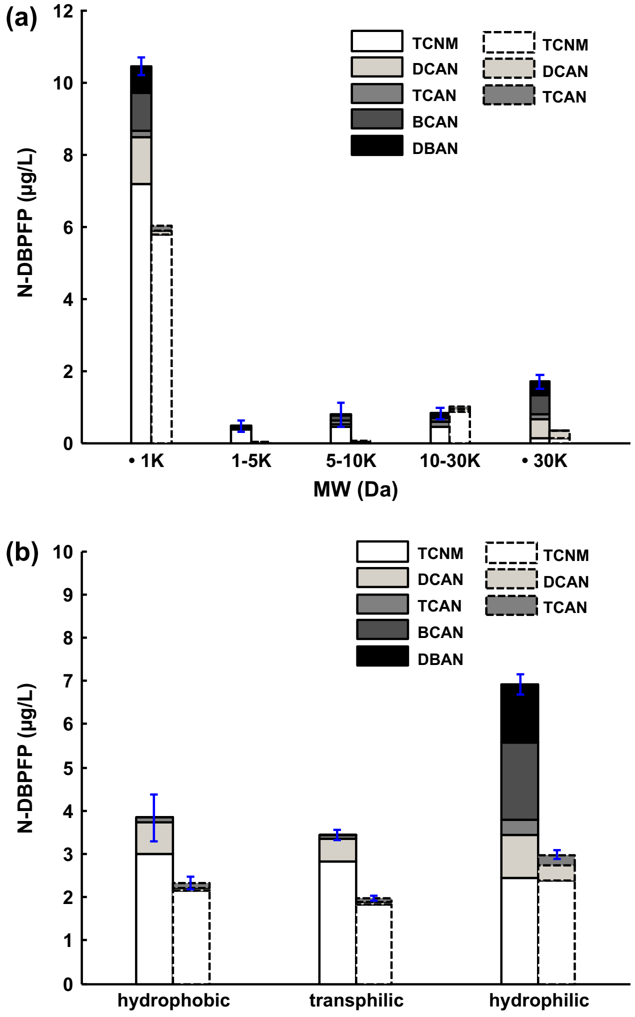

Fig. 5. N-DBPFP of the MW fractions (a) and hydrophobic–hydrophilic fractions (b) of DOM in the raw waters from the Yellow River and Danjiangkou Reservoir. Bars with solid line: fractions of DOM in the raw water from the Yellow River; bars with dashed line: fractions of DOM in the raw water from Danjiangkou Reservoir.

Fig. 5(b) shows the N-DBPFP of the hydrophobic, transphilic, and hydrophilic DOM fractions in the raw waters from the Yellow River and Danjiangkou Reservoir. Hydrophilic DOM fraction had the highest N-DBPFP in both waters, with concentrations of 6.92 and 2.98 μg/L for the Yellow River and Danjiangkou Reservoir waters, respectively. BCAN and DBAN were only detected in the hydrophilic DOM fraction in the Yellow River water with concentrations of 1.79 and 1.36 μg/L, respectively. It was found that the total N-DBPFP and their components decreased in the order of hydrophilic > hydrophobic > transphilic in both waters. The hydrophilic and hydrophobic DOM fractions were the two main contributors to the formation of N-DBPs, which accounted for more than 75% in those two waters. It was concluded that hydrophilic and hydrophobic organic matter with low MW (<1 kDa) were significant N-DBP precursors in the Yellow River and Danjiangkou Reservoir waters.

4. Conclusions

(1) The MW < 1 kDa fraction contributed to the most DOM in both raw waters. The hydrophilic and transphilic fractions dominated the raw water from the Yellow River and Danjiangkou Reservoir, respectively.

(2) THMs were derived mostly from the MW < 1 kDa and hydrophobic fractions, while the hydrophilic and hydrophobic DOM fractions were the major N-DBP precursors during chlorination of both raw waters.

(3) Hydrophilic DOM fraction with low MW (< 1 kDa) produced more toxic bromated DBPs during chlorination in the presence of bromide in both raw waters. Compared with the raw water from the Danjiangkou Reservoir, the Yellow River water showed higher concentration of bromide and bromated DBPFP after chlorination, which should receive special attention during the process of water source switching.

(4) Water treatment processes need to be improved in order to enhance the removal of hydrophobic and hydrophilic DOMs to minimize the risk of DBPFP in both waters. Because conventional drinking water treatment processes could not remove the hydrophilic and low-MW DOM effectively, pretreatment or advanced water treatment processes such as pre-ozonation, enhanced coagulation, ozone/granular activated carbon, or nanofiltration should be evaluated to promote the removal of DOM during the upgrading and reforming of original processes.

(5) The knowledge gained from this research will guide the upgrade and reform of DWTPs in the water-receiving areas of SNWDP for using the two raw waters with very different physicochemical properties.

Acknowledgments

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