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Spatial assessment of tap water safety in China and associated health impacts

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1 **Abstract:**

2 The quality of municipal water supplies is of fundamental importance to public health and national
3 security. Here, we assess the tap water quality in 31 provinces across China and examine the effects
4 of natural and anthropogenic conditions on water quality and associated health risks. Precipitation
5 is a crucial factor influencing the organic matter content and ionic conductivity in tap water,
6 especially for arid and semi-arid regions. Although the concentration of disinfection byproducts
7 (DBPs) is closely related to the organic matter content, the occurrence of highly toxic DBPs was
8 more significantly affected by anthropogenic factors such as economic development and pollution
9 emission. We confirmed nanofiltration as an effective point-of-use treatment to reduce the adverse
10 effects of DBPs in public water supplies. Since DBPs in tap water is a long-term global problem,
11 our results highlight the potential health hazards of drinking water brought about by social
12 development and conclude that countries and regions with rapid development might face high DBP
13 toxicity.

Introduction

The provision of safe and reliable supply of drinking water is of fundamental importance to society ^{1,2}. According to the World Health Organization (WHO), the sixth goal for sustainable development in accordance with Agenda 2030 is water quality. Conventional water treatment by coagulation-sedimentation-filtration-disinfection, the most widely used drinking water treatment process in China, can remove 10%-60% of dissolved organic matter (DOM) ^{3,4}. However, disinfection byproducts (DBPs) are formed from the reaction between chlorine-based disinfectants and DOM; these byproducts are associated with adverse health outcomes (e.g. bladder cancer), as reported in epidemiological and toxicological studies ⁵⁻⁹. The adverse health outcomes associated with DBPs represent a long-term public health problem with regard to drinking water ¹⁰.

While several studies have evaluated the occurrence of DBPs in tap water, these studies were conducted mainly in local regions and cities ¹¹⁻¹³. Only a limited number of nationwide DBP surveys have been reported — in the UK ¹⁴, the USA ¹⁵ and China ¹⁶ — approximately a decade ago. In China, the regular monitoring of drinking water quality is carried out by local authorities (Water Supply Bureaus), but this typically includes only routine parameters such as turbidity and pH. As a consequence, statistical data related to national tap water quality, especially exposure to DBPs via tap water and the impact of DBPs on public health, remain unclear and elusive.

China has a large land area of more than 9.6 million square kilometers, with tremendous variations in climate, topography, population density and level of economic development. These differences might lead to diverse composition of DOM in soil and water in different regions. For instance, climate has been found to account for the majority of soil DOM heterogeneity ¹⁷, and to determine the regional variation of the forest carbon stock ¹⁸. Considering that about one-quarter of the net carbon fixation on land enters the dissolved organic carbon (DOC) pool of natural waters ¹⁹, the impact of climate stress on water quality is inevitable. In addition to natural factors, human activities have also resulted in environmental challenges; this connection is referred to as the coupling between economic prosperity and ecological impact ²⁰. A recent study has indicated that

lakes in China's populated regions are facing an imbalance of nitrogen and phosphorus due to the discharge of domestic sewage²¹. While these studies have shown the effects of environmental stressors on soil or natural waters, it is unclear whether the variations in natural conditions and human activities have significant effects on the quality of tap water.

In this study, we conducted a comprehensive analysis of the quality of tap water samples from 31 provinces in China (excluding Hong Kong, Macao and Taiwan) (**Figure S1**). Our study provides answers to the following questions: (i) What is the spatial distribution of tap water quality across China? (ii) What are the correlations between natural and anthropogenic factors and tap water quality distribution? (iii) Do spatial differences in tap water quality lead to health risk disparity? (iv) How might we deal with the health risks of poor-quality tap water? This is the first study to discuss water safety issue at a large spatial scale. The results of this study generate a unique database of national tap water quality, particularly DBPs, and provide a *prima facie* insight on the relationship between multiple factors and drinking water safety.

Results

Precipitation has a dual effect on total organic carbon

Total organic carbon (TOC) was used to evaluate the organic content of tap water; the distribution pattern is shown in **Figure 1a**. The TOC of tap water in different regions was greatly affected by the quality of local water sources. According to the national surface water quality report (<http://www.cnemc.cn/jcbg/qgdbsszyb/>), surface water pollution was the most serious in Songhua River and Hai River (**Figure 1b**), with 36.4% and 38.8% monitoring points, respectively, not suitable for domestic drinking water source (water quality > III, the Environmental Quality Standards for Surface Water are shown in **SI, Table S1**). As a consequence, high TOC values were observed in the Songhua River Basins. However, the levels of TOC in the Hai River Basin were relatively lower than those in surrounding areas, because most cities in the Hai River Basin used water from the Changjiang River as water source rather than local source waters. This pattern of

use is known as the South-to-North Water Diversion Project (**Figure S2**). It was reported that in 2020 the middle route of the South-to-North Water Diversion project (eastern, middle and western routes) supplied 8.3 billion cubic meters of water for Beijing, Tianjin as well as more than 20 large and medium-sized cities in Henan and Hebei provinces. Therefore, the water diversion engineering works have not only significantly relieved the water resource pressure of the Hai River Basin ^{22,23}, but also improved the drinking water quality in these receiving areas.

In addition to the TOC differences caused by different water sources, the accumulation of organic matter along the same river also led to an increase of TOC from upstream to downstream. For instance, the TOC was observed to increase gradually along the main stream of the Changjiang (Yangtze) River (**Figure S3**), the longest and largest river in China, which suggests accumulation of DOM when pollution exceeds the self-purification capacity of the water body. Moreover, the intense human activity (**Figure S4**) in eastern regions resulted in more nutrient pollutants (e.g. phosphorus and nitrogen) from domestic wastewaters to water bodies, and thus the microbial activity in these water bodies was significant ²⁴. A previous study found an increase of low-molecular-weight organics from upstream to downstream of the Changjiang River; the main component of organics changed from humic substances with terrestrial origins to microbial-derived protein-like biopolymers ²⁵. The latter were more difficult to remove in drinking water treatment ²⁶. Therefore, the variation of DOM composition is also responsible for the higher TOC value in eastern regions.

Precipitation is another crucial natural factor that accounts for the variation of TOC. Precipitation has a dual effect on TOC: (1) atmospheric particulate matter and soil organic matter can be scavenged by precipitation and contribute DOM to runoff waters, and (2) conversely, heavy precipitation can dilute the DOM in the runoff waters. From the correlation between precipitation and the TOC of tap water (**Figure 1c**), it can be seen that TOC increases with precipitation for the northern samples, but changes little with precipitation in the southern samples. The boundary between northern and southern China follows the Qinling-Huai River line, which coincides with

the 800 mm precipitation line that divides humid and sub-humid regions (**Figure S5**). For northern cities, TOC has a significant positive correlation with precipitation due to DOM scavenging from soil and atmosphere. In contrast, the dilution effect offsets the DOM scavenging in southern cities, and the TOC variation is much smaller than that within northern cities.

Figure 1. TOC in tap water. (a) Spatial distribution of TOC (averaged by province) in China. (b) Surface water quality and the average TOC in nine basins. Detailed information about the water quality level (I-V and >V) appears in SI, **Table S1**. (c) Variation of TOC with annual precipitation (in 2019), where the grey line is the linear fitting of results. The color indicates the annual precipitation (blue: >800 mm, red: < 800 mm), and the shape indicates the location (triangles: south; circles: north).

Soil erosion and seawater intrusion lead to high ionic conductivity of tap water

The variation of electric conductivity in tap water across China is shown in **Figure 2a**. Among the eight regions, middle Yellow River (MYR) and north coast (NC) have relatively higher conductivity (**Figure 2b**). Considering that conventional drinking water treatment has little effect on the removal of background ions, the variation in conductivity is mainly ascribed to different water sources. Tap water using Yellow River (YR) or underground water (UW) as water sources has significantly higher conductivity than others (OT) (**Figure S6**), and cities with such levels of

water conductivity were mainly located in the north-west (NW), middle Yellow River (MYR) and north coast (NC) regions. However, Henan province (tagged in **Figure 2a**), with water from the South-to-North Water Diversion project as part of its drinking water source (**Figure S2**), was found to have a lower conductivity than other provinces located at the middle Yellow River region, which once again proved that the south-to-North Water Diversion project has improved the tap water quality of cities along its route.

The Yellow River basin is known to experience intense soil erosion due to the combined impact of natural processes and human activity¹. The annual runoff of the Yellow River (Tongguan Hydrological Station) in 2019 was 41.56 billion cubic meters, with 168 million tons of annual sediment load (equivalent to an average concentration of ~4 g/L). According to the monitoring data of the gauging stations from 2009 to 2019, the sediment load increased markedly in the middle reach of the river as it crosses the Loess Plateau (**Figure 2c**)²⁷, and a similar large rise in conductivity (for samples using Yellow River as a water source) was also evident in this region. Loess Plateau, located in the arid and semi-arid region of China, is an important inorganic carbon (calcium carbonate) reservoir, and its soil inorganic carbon (SIC) stock was calculated to be 2.1 times greater than its soil organic carbon (SOC) stock²⁸. Therefore, it is likely that inorganic carbon was introduced to the Yellow River through soil erosion and led to the increase of calcium in the river. As shown in **Figure S7**, calcium ions were the main metal ions in tap water, accounting for 54.5% of the total metal ion concentration. For coastal regions, seawater intrusion and soil salinization were responsible for high conductivity. According to the Bulletin of China Marine Disaster (<http://www.mnr.gov.cn/sj/sjfw/hy/gbagg/zghyzhgb/>), seawater intrusion is serious in the coastal plain area of the Bohai Sea, and the extent of intrusion increased from 2015 to 2018. In contrast, the extent of intrusion in most coastal areas of the Yellow Sea, the East China Sea and the South China Sea was relatively small. As a result, Liaoning, Hebei, Shandong and Jiangsu provinces were found to have higher conductivity than other coastal provinces. Conductivity was found to decrease with precipitation in both northern and southern China (**Figure 2d**). Considering

that the TOC increased with annual precipitation in northern China, we deduced a negative relationship between TOC and conductivity in this region. Previous studies have suggested that when the landscape varied from desert to forest along a precipitation gradient, the root biomass and SOC content increased considerably, while the SIC content significantly decreased²⁹. Our study found that this conclusion not only applied to soils, but also to tap water in arid and semi-arid regions, indicating the connection between the soil and water environments.

Figure 2. Conductivity in tap water. (a) Spatial distribution of tap water conductivity (averaged by province) in China. (b) Boxplot of conductivity in eight economic regions. The shape of points indicates the water sources, and the color indicates eight economic regions: north-east (NE), north coast (NC), east coast (EC), south coast (SC), middle Yellow River (MYR), middle Changjiang River (MCR), north-west (NW), south-west (SW), where boundaries are shown in the SI, **Figure S1**. (c) The conductivity of tap water using the Yellow River as the water source (blue), and the sediment load of the Yellow River at the main gauging station (red). The x-axis is the longitude of sampled cities and the gauging stations. The sediment load data were ten-year averages (2009-2019) from the China River Sediment Bulletin (2019). (d) Variation of conductivity with annual precipitation (in 2019), where the grey line is the linear fitting of results. The color indicates the annual precipitation (blue: >800 mm, red: < 800 mm), and the shape indicates the location (triangle: South; circle: North). Note: 810 $\mu\text{S/cm}$ is equivalent to 518 mg/L total dissolved solids (TDS).

DBP concentrations vary widely across China

At present, trihalomethanes (THMs) and haloacetic acids (HAAs) are the two most detected and

regulated groups of DBPs, although hundreds of other DBPs have been identified and some are of greater potential toxicity³⁰⁻³⁴. In this study, four THMs, seven HAAs and four nitrogen-containing DBPs (N-DBPs) were detected in tap water samples from 103 cities; the spatial distribution of these three kinds of DBPs are shown in **Figures S8-S10**. The average totals of THMs, HAAs and N-DBPs were 54.82 µg/L, 16.07 µg/L and 3.85 µg/L, respectively (**Figure 3a**). We found that 88.3% of the samples were below the THMs maximum level for drinking water in China (100 µg/L), and 100% of the samples met the requirement for HAAs. The north-east (NE) region exhibited the highest concentration of total DBPs, followed by middle Changjiang River (MCR) and coastal regions. In contrast, the western regions (north-west (NW), south-west (SW) and middle of Yellow River (MYR)) had relatively lower DBP concentrations, which is partially due to the water sources in those regions being mainly underground water with fewer DBP precursors³⁵. Among the four largest cities, Beijing, Guangzhou and Shenzhen were found to have high DBPs (**Figure 3c**). Shanghai, located at the estuary of the Changjiang River, however, had a very low DBP concentration. According to the "13th Five-Year Plan of Shanghai Water Supply System", Shanghai is conducting a program of upgrading its water treatment and supply facilities. By the end of 2020, 60% of the city's water plants had adopted the combination of ozone and biological activated carbon (biofiltration) for enhanced water treatment. Ozone-biofiltration has been shown to significantly decrease DBP precursors in previous studies³⁶⁻³⁸. Other treatment processes such as advanced oxidation (e.g. O₃, O₃-H₂O₂, UV-H₂O₂), nanofiltration and their combination were also reported to lower the DBP formation potential prior to chlorination³⁹⁻⁴¹. These results highlight the fact that advanced or extended treatment is necessary for the control of DBPs in tap water. This economically viable conclusion should be promoted in other first-tier or second-tier cities facing the risk of high DBP formation.

Figure 3. Occurrence of DBPs in tap water samples. (a) Boxplots of three categories of DBPs (total THMs, HAAs and N-DBPs) in tap water samples of 103 cities. (b) Average concentrations of THMs, HAAs and N-DBPs in eight economic regions (see caption of Figure 2 for the names of the economic regions). The color indicates different DBPs: THMs (red), HAAs (blue) and N-DBPs (grey). (c) Spatial distribution of DBP concentration (averaged by province) in China as a whole and in its four largest cities.

Areas with high bladder cancer incidence have higher concentrations of DBPs

High exposure to DBPs was reported to be related to adverse health outcomes^{42,43}. A landmark study three decades ago concluded that exposure to DBPs in drinking water is associated with 10–40% excess risk of cancers of the bladder, colon and rectum⁴⁴. Since then, numerous epidemiological studies were conducted to reveal the association between chlorinated DBP exposure and specific cancer outcomes; the studies have aroused considerable concern⁴⁵. Most of these studies used a case-control method focused on individuals, concluding that the case group had higher DBP exposure than the control group.

In this work, we discussed this issue from the spatial perspective. We took a closer look at 46 cities with both tumor registration information (from China's cancer registry annual report (2018))

and DBPs data (measured in this study), and found that cities with a higher incidence rate for bladder cancer were characterized by higher DBP concentration in tap water (Figure 4a-4c). Specifically, 20 cities with an age-standardized rate (ASR) higher than the national average had a median DBP concentration of 103.43 µg/L, which was significantly higher than the remaining 26 cities with a median DBP concentration of 71.45 µg/L ($p < 0.01$, Wilcoxon test). Linear regression further showed a log-linear relationship between the concentration of DBPs and the bladder cancer incidence (Figure 4d-4f). When considering the gender difference, males had a higher incidence of bladder cancer than females, but both rates were strongly associated with the concentration of DBPs. The relationships between DBPs and rectal and colon cancers were also considered (Figure S11); however, the statistical results are only on the verge of significance. Unlike consistent association between DBP exposure and bladder cancer risk, results for colon and rectal cancers were controversial⁴⁶.

It should be emphasized that, considering the lack of a large sample size, the above discussion only highlights the potential link between DBPs in tap water to bladder cancer. Other confounding variables such as tobacco consumption, dietary and genetic factors are also responsible for cancer disparities⁴⁵. More detailed studies are needed to understand which populations or regions in China are more vulnerable to the health impacts of DBPs.

Figure 4. DBP association with bladder cancer. (a-c) Comparison of DBP concentrations for cities with bladder cancer rate higher (high group) or lower (low group) than the national average: (a) all gender, (b) female, (c) male. Results from Wilcoxon tests showed significant differences ($p < 0.05$) between high group and low group. Details of cancer rates in 46 cities were provided in SI, **Table S2**. (d-f) Scatter plot showing the correlation between bladder cancer indexes and DBP concentration, where the red line is the logarithmic linear fitting of results: (d) all gender, (e) female, (f) male. The cancer indexes are: ASR-age standardized rate, F.ASR-female age standardized rate, M.ASR-male age standardized rate.

The cytotoxicity level of DBPs shows spatial aggregation

In addition to the cancer risk, DBPs were also reported to have cytotoxicity. Based on the lethal concentration 50% (LC_{50}) value from a previous study⁴⁷, the total toxicity of DBPs in tap water samples was calculated (see Methods). The north coast (NC) and east coast (EC) regions were found to have the greatest DBP toxicity risk (**Figure 5a**). Global spatial autocorrelation (Moran's I) suggested that the total toxicity was spatially aggregated (SI, **Figure S12**). A Local Moran statistic was constructed to further identify local clusters and local spatial outliers of DBP total toxicity (**Figure 5b**). Shandong, Jiangsu, Zhejiang and Anhui provinces were the cores of a high-high cluster, while Xinjiang, Tibet, Qinghai, Sichuan and Yunnan provinces were the cores of low-low cluster. Nonetheless, Shanghai (high-low point) and Chongqing (low-high point) were two spatial outliers, suggesting the total toxicity was significantly lower (in Shanghai) or higher

(in Chongqing) than their adjacent provinces. As discussed earlier, Shanghai had substantially low DBP concentration due to its wide adoption of advanced treatment processes. Chongqing, one of the four municipalities directly under the central government, is the largest industrial and commercial city in southwest China. The higher level of DBP toxicity observed here indicated that anthropogenic factors also affected the water quality, which will be discussed in the subsequent section.

Anthropogenic factors are strongly linked to DBP toxicity

The above discussion involved TOC, conductivity, DBPs and DBP toxicity. The heat map shown in **Figure S13** depicts the Pearson coefficients between each two water quality indexes, with the indexes being ordered via hierarchical clustering. The results show that toxicity was highly correlated with Br-containing DBPs (Br-DBPs), while the correlation between TOC and non-brominated DBPs (Cl-DBPs) was greater (**Figure S14**). Therefore, although most organic matter formed Cl-DBPs during chlorination, the Br-DBPs with lower concentration contributed more to the total toxicity of DBPs. When considering the effect of natural and anthropogenic factors on water quality, we found a negative relationship between conductivity and natural factors such as annual precipitation and forest coverage, while anthropogenic factors such as gross domestic product (GDP) and waste emission showed a significant positive relationship with DBP toxicity (**Table S4**). Therefore, partial least squares path modeling (PLS-PM) analysis was further performed to examine the complex relationships among two natural factors, ten anthropogenic factors, three DOM indexes, conductivity, six Cl-DBPs, nine Br-DBPs and the total toxicity of DBPs (**Figure 5c**, **Figure S15** and **Table S4**). The DBP toxicity was more significantly affected by Br-DBPs than Cl-DBPs. A previous study also showed that both cytotoxicity and genotoxicity were highly correlated with total organic bromine, but were weakly or inversely correlated with total organic chlorine⁴⁸. In contrast, DOM had a stronger and more significant effect on Cl-DBPs than Br-DBPs. The effects of conductivity on the concentration of Cl- and Br-DBPs were opposite. A possible reason is that areas with high conductivity, such as coastal regions, had a high

concentration of bromide in their water environment due to seawater intrusion or bromine industrial production, thus promoting the formation of Br-DBPs during chlorine-based disinfection³⁵. Natural factors (precipitation and forest coverage rate) had weak correlation with DBP concentration and toxicity. In contrast, anthropogenic factors were significantly correlated with Br-DBPs, and therefore had a strong indirect effect on DBP toxicity (Figure 5d). This result implies that human activities, such as industrial manufacturing, might change the properties of DOM in water and lead to the increase of DBP toxicity in drinking water. As a consequence, countries or regions with rapid economic development and high pollution emissions might face a higher risk of DBP toxicity exposure. Therefore, potential health hazards in drinking water caused by social development should be emphasized in future studies.

Figure 5. Calculated DBP cytotoxicity. (a) Average toxicity of THMs, HAAs and N-DBPs in eight economic regions. The color indicates different DBPs: THMs (red), HAAs (blue) and N-DBPs (grey). (b) Local Moran cluster map of DBP total toxicity. Strictly speaking, the locations shown as significant on the significance

and cluster maps are not the actual clusters, but rather the cores of a cluster. In contrast, in the case of spatial outliers, the locations are the actual locations of interest; (c) PLS-PM describing the relationships between water quality and environmental factors. The composite variable DOM, natural factors, social factors, Cl-DBPs and Br-DBPs are explained in **Table S4**. Larger path coefficients are shown as wider arrows, and blue and yellow colors indicate positive and negative effects, respectively. Path coefficients and coefficients of determination (R^2) were calculated after 999 bootstraps, and significance levels are indicated by *** $p < 0.001$, ** $p < 0.01$ and * $p < 0.05$. The degree of fit was 0.70; (d) Standardized direct and indirect mean effects on total toxicity derived from the partial least-squares path models.

Geographically close regions have similar water quality

Principal component analysis (PCA) was conducted to demonstrate the relationship between samples from different regions (**Figure 6a**). The aforementioned indexes (TOC, conductivity, 15 DBP compounds and DBP toxicity) as well as indexes calculated from UV-visible spectra and excitation-emission matrix (EEM) spectrofluorometry (**Table S3**) were included. We observed that TOC, total fluorescence (TF), A_{245} and most chlorinated DBPs are distributed along the first principal axis, while most brominated DBPs, DBP toxicity and conductivity clustered at the second principal axis, which was inconsistent with the results in **Figure 5a**. The samples were geographically divided into eight regions as before. North-west (NW, dark grey) and south-west (SW, light grey) samples were primarily clustered at the third quadrant, suggesting they had relatively lower concentrations of DBPs and DOM. North-east (NE, dark red) samples, located at the positive side of the first principal axis, had a significantly higher concentration of chlorinated DBPs and DOM than other regions. Samples from coastal regions (especially for the east coast (EC)) had a higher value at the second principal component, implying they had more brominated DBPs and exhibited greater DBP toxicity. For coastal regions, the landward expansion of seawater or underground salt water (directly related to seawater) along an aquifer might elevate the bromide concentration in the drinking water source, and result in more Br-DBP formation and greater total toxicity^{35,48}. Moreover, Shandong province and Jiangsu province dominate the production of brominated chemical products such as brominated flame retardant⁴⁹. A previous study observed relatively high concentrations of brominated flame retardant in the rivers near Laizhou Bay⁵⁰,

which is one of three bays in the Bohai Sea and the largest bay in Shandong Province. Therefore, the brominated chemical products in surface water may not only cause adverse effects to human health by direct exposure⁵¹, but also by increasing the possibility of higher Br-DBPs formation as well as the associated DBPs toxicity.

Permutational multivariate analysis of variance (PERMANOVA) was used to further evaluate the significance of differences among the eight regions (**Figure 6b**). North-east (NE) and south-west (SW) samples showed significant differences from samples from other regions. However, there were some linkages between the remaining six regions: (1) northern samples (NW, MYR, NC connected by Yellow River); (2) eastern samples (NC, MCR and EC connected by Changjiang River and the South-to-North Water Diversion Project); (3) southern samples (EC, SC and MCR connected by waterbodies in the middle and lower reaches of Changjiang River) exhibited insignificant differences. These results imply that geographically close areas tend to exhibit similar water quality. The correlation between water quality and geographical location is demonstrated in **Figure 6c**. Here, we used two indexes (namely, ‘*north*’ and ‘*east*’ calculated by the method described in SI, **Text S3**) to describe the relative location of the sample point in China, with higher value indicating farther north or east. We saw that the ‘*east*’ index had a more significant correlation with all water indexes except for conductivity, suggesting that the east-west divide in water quality is more pronounced than the north-south divide. The conductivity of tap water was greatly affected by precipitation, and therefore significantly correlated with ‘*north*’ as discussed above. The results of multiple linear regression suggested that geographical location can explain 18.9% to 47.0% of the variation in different water indexes. TOC had the highest explained variance, while DBP concentration is affected by the water treatment and disinfection parameters (disinfectant type and dosage), and thus had a lower explained variance.

Figure 6. Overview of water quality and environmental factors. (a) PCA analysis of water quality index. The color of points indicates different regions. The indexes include total organic carbon (TOC), UV absorbance at 254 nm (A_{254}) and 300 nm (A_{300}), specific UV absorbance (SUVA), ratio of absorbance at 250 nm to 365 nm (E2:E3), total fluorescence (TF), five specific TF (R1-R5), humification index (HIX), biological index (BIX), fluorescence index (FI), and EEM peaks (A, B, C, M, T). The explanations of these water quality indexes are summarized in **Table S3**. (b) Difference in water quality indexes in eight regions by permutational multivariate analysis of variance (PERMANOVA), where the color denotes the significance level. (c) Contribution of geographical location (north and east direction) to water qualities based on correlation and multiple regression models. The heat map shows the Pearson correlation coefficients for geographical location and water quality. The asterisks denote the significance levels: *** $p < 0.001$, ** $p < 0.01$ and * $p < 0.05$. The bar plot shows the total contribution of geographical location to the interpretation of water qualities (obtained by multiple linear regression).

Nanofiltration effectively improves water quality

With the deterioration of the quality of water resources and the heightened demand for high-quality drinking water, interest in using domestic water purifiers to improve tap water quality and reduce adverse health effects has been growing rapidly in recent years ⁵². Compared to

conventional water treatment, membrane filtration can produce water of significantly higher quality with no chemical addition. Here, we used two nanofiltration membranes with different rejection rates (namely, NF270 and NF90) to test the improvement in water quality by nanofiltration. The results (**Figure 7a**) show that NF270 and NF90 on average decreased conductivity by 12.8% and 57.7%, respectively. In conventional drinking water treatment, hydrophobic substances with large molecular weight are readily removed⁵³, leaving small and hydrophilic substances in the treated drinking water; such substances are difficult to retain by hydrophilic NF membranes³⁸. Therefore, the removal rate of TOC was only 20.5% and 55.2% for NF270 and NF90, respectively. For DBPs, although their molecular weights (MW) are very low (< 300 Da), their hydrophobic nature resulted in a high removal rate by nanofiltration, with 66.2% and 78.2% for NF270 and NF90, respectively.

The removal rates of 15 DBP compounds are shown in **Figure 7b**. Generally, molecules with more halogen atoms had a higher retention rate, and Br-DBPs were easier to remove than Cl-DBPs for DBPs with identical halogenation degrees due to their larger MW. The negative charge of HAAs favored their removal via electrostatic repulsion by the negatively charged NF membranes, but due to their hydrophilicity they were not removed by adsorption and partitioning onto the NF membranes as was the case with the hydrophobic THM molecules^{53,54}. Some studies also used reverse osmosis (RO) or even multistage RO to remove DBPs^{55,56}, but one inevitable disadvantage of RO lies in the unfavorable balance between minerals and pollutants. Here, we found that NF is sufficiently capable of removing a great portion of DBPs, thus significantly decreasing the total toxicity of DBPs (**Figure 7c**) while leaving some minerals in place. In comparing the two NF membranes, the results suggest that using NF270 as a domestic treatment could be more effective due to its comparable DBP removal rate, but nearly double water permeability compared to NF90 (SI, **Table S5**).

Figure 7. Water quality improvement by nanofiltration. (a) Removal rate of TOC, conductivity, DBPs and total toxicity by two NF membranes: NF270 (“loose” NF) and NF90 (“tight” NF). (b) Removal rate of different types of DBPs (THMs, trihalomethanes; HAAs, haloacetic acid; N-DBPs, nitrogen-containing DBPs) by the two NF membranes. Detailed information about these DBPs is given in [Table S6](#). (c) Total toxicity of DBPs in tap water before (left) and after NF270 (middle) or after NF90 (right). The calculation of total toxicity was shown in Methods.

Discussion

This is one of the first studies to assess tap water quality at a large spatial scale and to discuss the driving factors as well as the adverse health outcomes. The findings revealed a significant variation of tap water quality across China in terms of organic content, background ions and disinfection byproducts (DBPs). The results further demonstrate the influence of natural and

anthropogenic factors on tap water quality, while also confirming the association between DBPs and specific cancers from the perspective of spatial distribution. In addition, the feasibility of nanofiltration (NF) to improve water quality was tested, suggesting that NF can be used as a point-of-use treatment to reduce the health risks of DBP exposure without removing necessary minerals as RO does.

Based on the assessment of tap water from 103 cities, the spatial distribution pattern of tap water quality was identified. The total organic carbon (TOC) and conductivity in tap water are closely related to water source, with precipitation playing an essential role in their spatial diversity, especially for northern China where its annual precipitation of less than 800 mm. Therefore, arid and semi-arid regions face not only water shortages but also poor drinking water quality. The South-to-North Water Diversion project conducted by the Chinese government provided a solution. Our results suggest that the water diversion engineering works have not only significantly relieved the water resource pressure on the Hai River Basin, but also improved the drinking water quality in this region. The concentration of DBPs was significantly correlated with the organic content (**Figure S14**). Therefore, the north-east (NE) region and the middle Changjiang River (MCR) harbored a high concentration of DBPs, while the west regions (north-west (NW) and south-west (SW)) faced less DBP exposure. However, the toxicity of DBPs was not only determined by the DBP concentration, but was highly affected by their composition. Key areas with significantly higher or lower DBP toxicity were identified. High toxic areas clustered at east coastal regions, while low toxic areas were distributed at west regions. Moreover, we found that the total toxicity of DBPs was strongly affected by anthropogenic factors such as economic strength and pollution emission. Therefore, it is necessary to improve the coverage of advanced drinking water treatment in economically developed cities, to make up for the problems of drinking water safety caused by economic development.

While several alternative disinfectants have been proposed to reduce DBPs, chlorine-based disinfection (i.e., chlorine, chloramine and chlorine dioxide) continues to be the mainstream

disinfection method in drinking water treatment in China. Hence, chlorinated DBPs in drinking water and their associated health risks will remain a long-term problem. Scientists have associated DBP exposure to a variety of adverse health outcomes, including specific cancers (bladder, colon and rectal), low birth weight, and miscarriages. Among these, bladder cancer has the most consistent association with DBPs in previous studies. Our study confirmed that areas with high DBPs had a significantly higher incidence of bladder cancer from a spatial distribution perspective, but the results for colon and rectal cancer were not as significant as bladder cancer. It is worth noting that although more than 600 emerging DBPs have been reported in the literature, less than 100 have undergone quantitative occurrence or health effect studies⁵⁷. Most of the epidemiological studies used THMs and/or haloacetic acids (HAAs) to estimate DBPs exposure, but epidemiological data related to emerging DBP exposure is particularly scarce. Therefore, it is still unclear whether emerging DBPs have other health effects on humans, and whether the use of other chlorine-containing disinfectants will enhance the formation of unregulated DBPs with higher health risks.

The results in this study represent a first attempt to consider drinking water safety issues and the pattern of spatial variation in tap water quality in terms of DBP occurrence on a national scale. However, there are still some uncertainties that deserve further investigation. First, although we tried to collect as many water samples as possible to provide accurate information on national tap water quality, the spatial and temporal variations of DBPs in drinking water make it difficult to estimate actual human exposure. Second, due to the lack of records, it is challenging to conduct time series analysis and explore the evolution of tap water quality over time. High-resolution data (both spatial and temporal) can provide stronger evidence to identify the key factors affecting tap water quality, and important data for researchers to reveal the relationship between tap water and human health. Therefore, it is recommended that researchers and practitioners should cooperate to perform more intensive and detailed surveys of DBPs and other contaminants at a national level, and a database of drinking water quality should be established to provide data support for related

studies.

Materials and methods

Chemicals and materials

Methyl tert-butyl ether (MTBE) and methanol were, respectively, liquid chromatography grade and high-performance liquid chromatography (HPLC) grade; they were purchased from J.T. Baker, USA. Anhydrous sodium sulfate (Na_2SO_4), sulfuric acid (H_2SO_4) and sodium bicarbonate (NaHCO_3) were obtained from Sinopharm Chemical Reagent Co., Ltd, China.

Sample collection and nanofiltration

Household tap water samples were collected from 103 cities, located within 31 administrative provinces and eight regions (see SI, **Figure S1**) during March to April, 2021. At sampling points, we collected as many samples as possible to ensure the reliability of the results. However, for some remote areas, such as northwestern China, only a small number of principal cities (i.e. the capital of each province) were sampled, as rural areas with low population densities typically have no centralized water supply system. For each sampling point, two replicate water samples were collected, and the tap faucets were not equipped with a filter or other water purification unit. Prior to sampling, the faucet was opened for at least five minutes, the sample bottles were rinsed three times and then filled to just overflowing. All collected samples were transferred to the laboratory in ice packs as soon as possible, and stored at 4 °C before use.

Nanofiltration tests were conducted using flat sheet poly (piperazine-amide) (PA) membranes (NF270 and NF90, RisingSun Membrane Technology (Beijing) Co., Ltd., China) in a dead-end filtration apparatus (Amicon 8400, Millipore, USA). The properties of these two membranes are given in the Supporting Information (**Table S5**). Before use, the membranes were immersed in deionized (DI) water for at least 72 hours to remove organic residuals during production. The operational applied pressure was set at 4 bar using high-purity nitrogen (99.99%, Haike Corp.,

China). The filtrate was collected after filtering 100 mL tap water, in order to avoid the influence of initial adsorption.

Instrumental analysis

A UV-visible spectrophotometer (UV-2600, Shimadzu, Japan) and a three-dimensional excitation-emission matrix (EEM) spectrofluorometer (F-4600FL, Hitachi, Japan) were used to characterize the chromophore and fluorescent organic substances in water samples, respectively. Total organic carbon (TOC) and conductivity were measured using a TOC analyzer (TOC-Vwp, Shimadzu, Japan) and a conductivity meter (FiveEasy Plus, Mettler Toledo, Switzerland). The concentration of metal ions was measured via Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES, Shimadzu, Japan) after microwave digestion.

DBP extraction methods followed the standard methods^{58,59}, and details of these methods are described in the SI (**Text S1**). Each sample was prepared in duplicate. The concentration of DBPs was measured by gas chromatography (Clarus 590, PerkinElmer, U.S.) with an electron capture detector (ECD). Conditions for the analyses were as follows: (1) THMs and N-DBPs, injector temperature 200 °C, column temperature 35 °C (holding four minutes) to 260 °C (10 °C/min), detector temperature 290 °C; (2) HAAs, injector temperature 200 °C, column temperature 35 °C (holding four minutes) to 65 °C (2 °C/min), detector temperature 290 °C.

Data analysis

Data concerning environmental stress factors were collected from different types of databases, including the China Statistical Yearbook, China Statistical Yearbook on Environment and others (SI, **Table S4**). The age standard rates (ASR) for bladder, colon and rectal cancers (SI, **Table S2**) were obtained from the China cancer registry annual report (2018). The toxicity of DBPs were calculated by summing the individual toxic potency-weighted DBP concentrations (Equation 1). The LC₅₀ values were obtained from published literature⁴⁷ and are provided in the Supplementary Information (**Table S6**).

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$$\text{CHO cytotoxicity index} = \sum_1^n \frac{[\text{DBP}]_i}{\text{LC}_{50_i}} \quad (1)$$

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Spatial mapping was performed with ArcMap 10.6 for the 31 provinces, excluding Hong Kong, Macao, and Taiwan due to sample unavailability. Data analyses were performed with the R software (4.0.5). EEM data were corrected and the optical parameters (SI, **Table S3**) were calculated from UV absorbance and corrected EEM data via the R package (named staRdom). Additionally, EEM data were divided into five regions and the volumetric integration under the EEM within each region was calculated according to previous study ⁶⁰. Principal component analysis (PCA) and PERMANOVA were performed by using the R package (named vegan) Function lm from the R package statistics was used to conduct the linear regression. A partial Mantel test was performed to examine the correlation between location and tap water qualities using the R package (named psych). Partial least squares path modeling (PLS-PM) analysis was performed via R package (named plspm) to explore the direct and indirect relationships among environmental variables and water qualities.

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Geoda (1.18.0) was used to perform spatial autocorrelation analysis for the 31 sampled provinces ⁶¹. Moran's I statistic is the most commonly used indicator of global spatial autocorrelation, which is a cross-product statistic between a variable and its spatial lag, with the variable expressed in deviations from its mean. For an observation at location i, this is expressed as $z_i = x_i - \bar{x}$, where $\bar{x}$ is the mean of variable x.

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Moran's I statistic is then calculated as follows:

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$$I = \frac{\sum_i \sum_j w_{ij} z_i \cdot z_j / S_0}{\sum_i z_i^2 / n}$$

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with w_{ij} as the elements of the spatial weight matrix, $S_0 = \sum_i \sum_j w_{ij}$ as the sum of all the weights, and n as the number of observations.

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Global spatial autocorrelation is designed to reject the null hypothesis of spatial randomness in favor of an alternative of clustering. The Local Moran statistic was further performed to identify

local clusters and local spatial outliers. It should be noted that the cluster maps are not the actual clusters, but the cores of a cluster. In contrast, in the case of spatial outliers, they are the actual locations of interest. More information can be found on the statistical software website (<https://geodacenter.github.io/documentation.html>).

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