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Comparison of characteristics, water quality and health risk assessment of trace elements in surface water and groundwater in China

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ABSTRACT

Water quality is closely associated with human health and socio-economic sustainable development. With the increasingly intensive anthropogenic activities, pollutants especially trace elements, enter water aquatic system and cause harm to humans. This study conducted the first systematic comparison on the pollution status of surface water and groundwater in China. Water quality and health risk assessment of 12 trace elements were evaluated according to the water quality index (WQI), hazard quotient (HQ), hazard index (HI), and carcinogenic risk (CR). The results showed that the average values of trace elements in the majority of surface water were higher than those in groundwater. The WQI values demonstrated that 86.02% of surface water and 83.11% of groundwater were suitable for drinking water. Arsenic served as the predominant pollutant and contributed significantly to cause the non-carcinogenic risk on human health in both surface water and groundwater, children were more vulnerable to the adverse effects than adults. In surface water, the priority non-carcinogenic risk sites were mainly distributed in Anning River, Taizi River, Middle reaches of Huai River and Jilin section of Songhua River. For groundwater, the high-risk sites were located in Huhhot Basin, Kuitun, Jianghan Plain, Datong Basin and Yucheng County. Arsenic posed potential carcinogenic risk to local resident in some water sites, and it presented higher in groundwater than surface water. More concerns should be paid on Songhua, Yangtze and Huai River Basins, in addition, Pearl, Southeast Coastal, Southwest and Northwest River Basins still need attention. The future work should be carried out more extensive range of the water sites and long-term monitoring in China. In particular, more attention should be dedicated to assess high As water bodies.

1. Introduction

Water is widely regarded as the most essential of all our natural resources (Vorosmarty et al., 2010), water resources and water quality are important and indispensable for human survival, ecological environment, and economic and regional sustainable development (Long and Luo, 2020; Ma et al., 2020; Xiao et al., 2019). Water security and water shortages have become widespread problems, increasingly adversely impacted a significant portion of the world's population (Vorosmarty et al., 2010; Wang et al., 2017a, 2017b). China has low per capita water availability basis approximately 2068 m^3, amounting to only 34% of the world average at 6016 m^3 per capita per year, and unevenly distributed water resources especially in northern China, which accounts for 45% of the total population but receiving 19% of total water resources (Jiang, 2015; Ma et al., 2020).

Along with the intensification of anthropogenic influences such as industrial, agricultural activities and waste discharge, and the easy accessibility to the disposal of waste waters, surface water is highly susceptible to be polluted (Sener et al., 2017; Zeng et al., 2015). In addition, the overexploitations and intensive use of groundwater have brought about the deterioration of the environment including water table decline, seawater intrusion, and land subsidence (Blanke et al., 2007). The percentage of monitored surface water sections that were unsuitable for drinking reached 30% in 2013 and 2014, the seriously polluted regions were mainly distributed in northern China, especially in Hai, Huai, Songhua and Yellow river basins. In addition to surface

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water, the quality of groundwater was also under serious polluted condition, with up to 80% of tube wells of groundwater being polluted in 2015 (Wang et al., 2017a, 2017b).

Additionally, pollutants in water bodies, especially trace elements enter into aquatic ecosystems, and subsequently into the human body through drinking water pathway (Phung et al., 2017). Some of the metals are non-degradable, persistent and toxic (Zeng et al., 2015), which would pose adverse effects to human health and ecological security along with the accumulation of these metals in the water body (Guo et al., 2014; Long and Luo, 2020; Turdi and Yang, 2016). More than 100 million people worldwide ingest excessive amounts of As through drinking water, especially in the densely populated countries of South and Southeast Asia, such as Bangladesh, India, China, Pakistan, and Vietnam. Due to high As in groundwater, hundreds of millions of people are suffering from chronic As poisoning including chronic weakness, loss of reflexes, weariness, gastritis, colitis, anorexia, weight loss, and hair loss (Bissena and Frimmel, 2003; Guo et al., 2014; Winkel et al., 2008). In China, it had been estimated that more than 200 million people were facing the utilize of unsafe water sources, a survey conducted by the Chinese Ministry of Health between 2001 and 2005 estimated that 5.6 million people faced using of excessive concentrations of As in drinking water (> 50 μg/L) (Gao et al., 2019; Rodriguez-Lado et al., 2013; Tao and Xin, 2014).

Due to the increasingly serious water pollution and the associated hazards to human health, more and more attention has been paid to the quality of drinking water. Since 1996, the Chinese government successively implemented the “Nine-Five” Plan, “Ten-Five” Plan, “Eleven-Five” Plan and “Twelve-Five” Plan for the prevention and control of water pollution in three Rivers and three Lakes, the Three Gorges Reservoir Area and its upper reaches, the Songhua River, the middle and upper reaches of the Yellow River and other river basins. In 2015, the State Council issued and implemented the Action Plan for The Prevention and Control of Water Pollution, known as the "Water 10", making a systematic effort to prevent and control water pollution, protect water ecosystems, and manage water resources; and the 19th National Congress of the CPC set the goal of building a beautiful China, which put forward higher requirements for water quality, water ecology and water security (Xu et al., 2019).

Furthermore, a large number of areas have conducted the assessment of trace elements of water quality and the potential human health risk throughout the country. In China, researchers have carried out water quality, source identification and health risk assessment of trace elements in river water, well water, shallow groundwater, and drinking water sources in most regions, such as the Chinese Loess Plateau (Xiao et al., 2019), the five major river basins including the Yangtze River Basin, Yellow River Basin, Huai River Basin, Hai River Basin and Liao River Basin (Gao et al., 2019), the North China Plain (Zhang et al., 2015), the Upper Han River (Li and Zhang, 2010), the Guanzhong Plain (Qiao et al., 2020), the middle reaches of Huaihe River (Wang et al., 2017a, 2017b), etc. These regions cover most of the densely populated areas in China, and provide a better understanding of water quality, predominant pollutant, possible sources, and the degree of potential risk for the local administration. However, most of the current research has focused on water pollution in a single area. Limited comprehensive comparison and analysis of water contamination in different regions of China has been carried out. In particular, very little information exists on the comparison of surface water and groundwater. Therefore, a systematic and comprehensive evaluation of trace elements pollution and potential risk in surface water and groundwater among diverse areas remains to be carried out.

This study was conducted to evaluate the pollution status of 12 trace elements in selected sites in surface water and groundwater from the online literature in different areas of China. The principal objectives were: (a) to analysis the pollution characteristic of trace elements in selected surface water and groundwater; (b) to determine spatial distribution of the trace elements in surface water and groundwater among different regions; (c) to evaluate the water quality in surface water and groundwater based on the Water Quality Index (WQI); (d) to assess the potential health risk for human being according to the model recommended by the US EPA; (e) to calculate the hazard quotient (HQ) and the hazard index (HI) of the trace elements through the ingestion and dermal absorption pathways, and to assess the carcinogenic risk of As. The results of this study provide a comprehensive understanding of China’s water quality for policy-makers, inform potential human risks status, shed light on the theoretical foundation for coping strategies and alleviation measures for the sustainable development and effective management of surface water and groundwater resources for the drinking water.

2. Methodology

2.1. Data collection and processing

2.1.1. Search method

Studies relating to trace elements in surface water and groundwater in China published from January 2003 to July 2020 were systematically collected from the online literature, obtaining from several well-known literature databases including Web of Science, Elsevier Science Direct, Springer, Weipu Chinese Periodical Service and China National Knowledge Infrastructure (CNKI). Three categories of defined keywords were conducted by combing “OR” within a group and “AND” among groups, the search syntax comprising: (trace element* OR multi-Element* OR metallic element* OR heavy metal*) AND (surface water OR groundwater OR river water OR well water OR level* OR content* OR quality OR water quality OR concentrations OR characteristics OR distribution OR contamination OR pollution OR health risk OR assessment OR evaluat*) AND (China OR Chinese). The eligibility criteria comprised: (*a*) the studies subject focused on trace elements in surface water and groundwater; (*b*) the study area was restricted in China; (*c*) the average contents or medium values of trace elements were specific or could be obtained by calculating; (*d*) the sampling and analytical methods and process were conducted with scientific procedures and strict quality assurance; (*e*) the papers were published in full text and not abstracts or news reports.

2.1.2. Searching and screening procedures

A total of 4761 papers were obtained according to the search strategy, three successive screening procedures were conducted to select eligible papers. First, the titles of the searching articles were reviewed to inspect whether they were relevant to the study subject, and the non-relevant titles were excluded in this step. Second, an abstract review was implemented to further guarantee the papers conformed to the eligibility criteria, and the papers not relating to the content levels of the trace elements and with only abstracts or from news reports were excluded in this step. Third, a full text review of the selected literature from the second step was carried out to thoroughly check whether they accorded with the screening criteria. The papers involving the data of mean contents or medium values of trace elements could not be extracted were excluded in this step. Finally, a total of 94 papers on surface water including river water, reservoir water and stream water, and 77 papers relating to groundwater including well water (hand pump well, or dug well), shallow groundwater, and deep groundwater were selected (Table S1), and the distribution of different river basins in China is presented in Fig. S1.

2.1.3. Data extraction and analysis methods

The data of 12 trace elements including Aluminum (Al), Arsenic (As), Barium (Ba), Cadmium (Cd), Cobalt (Co), Chromium (Cr), Copper (Cu), Iron (Fe), Manganese (Mn), Nickel (Ni), Lead (Pb), and Zinc (Zn) were extracted from the screening papers, with the information comprising study region, year, authors, the pH value, mean contents or medium values, and the number of samples, as shown in Table S1. In addition,

Table 1
Statistics of chemical parameters and parameters for WQI calculation in surface water and groundwater in China (μg/L).

Parameters	Surface water				Groundwater				Parameters for WQI		
	Min	Max	Ave	SD	Min	Max	Ave	SD	CSS[a]	Weight (w_i)	Relative weight (W_i)
pH	6.20	8.64	7.71	0.56	6.57	8.09	7.53	0.37	6.5–8.5	4	0.095
Al	2.94	9908	459.87	220.39	1.84	800	75.51	175.36	200	2	0.048
As	0.06	48.05	5.65	7.80	0.06	262.43	16.81	45.88	10	5	0.119
Ba	12	310	81.96	74.63	4.71	733	133.78	165.70	700	2	0.048
Cd	0.01	61.74	3.54	11.08	0.01	15.23	1.69	3.18	5	5	0.119
Co	0.04	42.49	2.05	7.44	0.09	60	4.81	13.93	50[b]	1	0.024
Cr	0.10	624	25.43	86.86	0.08	49.70	6.55	9.05	50	5	0.119
Cu	0.10	1100	30.14	134.79	0.06	223	16.66	42.58	1000	2	0.048
Fe	1.44	68479	2186.84	11373.76	0.18	3071	450.27	659.33	300	4	0.095
Mn	0.30	2745	139.70	498.14	0.21	1300	223.17	305.56	100	5	0.119
Ni	0.18	600	18.49	83.48	0.09	62.00	8.07	11.22	20	1	0.024
Pb	0.01	690	29.29	102.77	0.01	136.50	9.58	23.08	10	5	0.119
Zn	0.38	10600	366.23	1644.55	0.21	7400	303.61	1325.05	1000	1	0.024

[a] Standards for drinking water quality, the national standard of the P.R. China (GB5749-2006) (The Ministry of Health of the P.R. China, et al., 2007).
[b] Co standard is from World Health Organization (WHO) standard (WHO, 2011).

the value of minimum, maximum, average and standard deviation of the elements were calculated (Table 1).

The concentrations of trace elements (Al, As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn) were determined by inductively coupled plasma mass spectrometry (ICP-MS) or inductively coupled plasma atomic emission spectrometry (ICP-AES). Total As was determined using a hydride generation-atomic fluorescence spectrometer (HG-AFS) in some papers. All the calibration curves were linear with a correlation coefficient higher than 0.99. Reagent blank, parallel samples and duplication samples were conducted to ensure the accuracy of the analysis processing, errors of more than 5% were excluded.

2.2. Water quality assessment

The water quality index (WQI) was applied to assess the trace elements pollution level in the water for drinking purpose, the specific illustration and calculation methods are presented as follows (Gao et al., 2019; Xiao et al., 2019):

$$\mathrm{WQI} = \sum \left[W_i \times \left(\frac{C_i}{S_i} \right) \times 100 \right] \quad (1)$$

Where $W_i = w_i/\Sigma w_i$, which represents the relative weight, w_i is the weight attributed to the target element according to its relative apperceptive effects on human health and significance in terms of potability (Meng et al., 2016; Sener et al., 2017). As, Cd, Cr, Mn, and Pb were assigned the highest weight of 5, due to these elements can have particularly detrimental effects on water quality, such as toxicity. The parameters pH and Fe were assigned a weight of 4, for their importance to water quality; Al, Ba and Cu were assigned a weight of 2; Co, Ni and Zn were assigned a weight of 1, because of their low impact on water quality (Gao et al., 2019). C_i was the measured trace element concentrations, and S_i was the standard for Chinese drinking water. The number 100 refers to the constant. Water quality can be divided into five

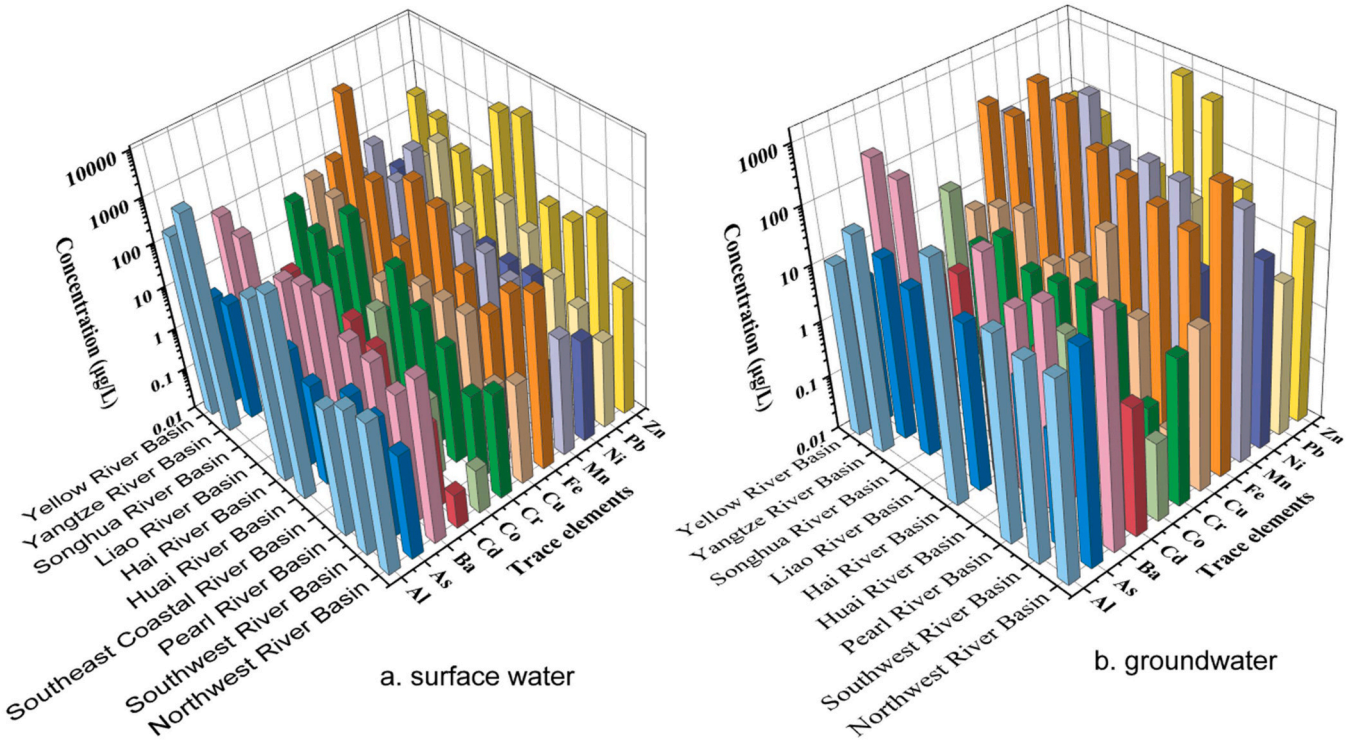

Fig. 1. Three-dimensional histogram of mean contents of trace elements in surface water (a) and groundwater (b) in different River basins.

different classifications according to the WQI values: when WQI < 50, the water quality is excellent; when $50 \leq$ WQI < 100, water quality is good; when $100 \leq$ WQI < 200, water quality is poor; when $200 \leq$ WQI < 300, water quality is very poor; when WQI ≥ 300, water is deemed undrinkable.

2.3. Human health risk assessment

Direct ingestion and dermal absorption by skin (showering/bathing and swimming) are usually considered to be the common exposure pathways for human beings (Gao et al., 2019; Xiao et al., 2019). The exposure dose and health risk assessment method are conducted based on the model recommended by the US EPA (2004).

The exposure dose for direct ingestion ($ADD_{ingestion}$) and dermal absorption (ADD_{dermal}) are evaluated by the following equation,

$$ADD_{ingestion} = \frac{C_w \times IR \times EF \times ED}{BW \times AT} \quad (2)$$

$$ADD_{dermal} = \frac{C_w \times SA \times K_p \times ET \times EF \times ED \times 10^{-3}}{BW \times AT} \quad (3)$$

where $ADD_{ingestion}$ and ADD_{dermal} represent the average daily dose via ingestion and dermal adsorption (μg/kg/day), respectively; C_w is the mean concentrations of trace elements in each water sample (μg/L); IR is the ingestion rate, 2.0 L/day for adults and 0.64 L/day for children, respectively; EF is the exposure frequency (350 days/year); ED is the exposure duration, 30 years for adults and 6 years for children, respectively; BW is the average body weight, 70 kg for adults and 15 kg for children; AT is the average time (for non-carcinogens, AT = ED × 365 days/year; for carcinogens, AT = 365 days/year × 70 years); SA is the exposed skin area, 18,000 cm^2 for adults and 6600 cm^2 for children; and ET is the exposure time, 0.58 and 1 h/day for adults and children, respectively. All of these exposure parameters were referenced from (Wang et al., 2017a, 2017b) and the US EPA (2004). K_p is the dermal permeability coefficient in water (cm/h), 0.001 for Al, As, Ba, Cd, Mn, Fe, and Cu (Wang et al., 2017a, 2017b; Zeng et al., 2015), 0.0001 for Pb (Zeng et al., 2015), 0.0002 for Ni and 0.0004 for Co (Wang et al., 2017a, 2017b), 0.002 for Cr and 0.0006 for Zn (Zeng et al., 2015).

The potential non-carcinogenic risk was assessed by the following equation:

$$HQ = \frac{ADD}{RfD} \quad (4)$$

$$RfD_{dermal} = RfD \times ABS_g \quad (5)$$

$$HI = \sum (HQ_{ing} + HQ_{derm}) \quad (6)$$

Where *HQ* is the hazard quotient through ingestion or dermal absorption of water (unitless); *RfD* is the oral toxicity reference dose for a specific metal, the values are listed in Table S2, ABS_g is the gastrointestinal absorption factor (dimensionless), the reference values are obtained from USA. Risk-based concentration table (US EPA, 2009). HI is the hazard index (unitless), which refers to the potential non-carcinogenic risk posed by all heavy metals. When HQ or HI > 1, the adverse effect on human health effects might be likely occur; however, when HQ or HI < 1, indicating that there are no adverse effects on human health (US EPA, 2004). The HI classification is presented as follows: HI < 0.1, negligible non- carcinogenic risk; $0.1 \leq$ HI < 1, low non-carcinogenic risk; $1 \leq$ HI < 4, moderate non-carcinogenic risk; $4 \leq$ HI, high non-carcinogenic risk (Alver, 2019; Rahim et al., 2019).

Potential carcinogenic risk level caused by carcinogenic chemical pollutants was calculated by the following formula,

$$CR = ADD \times SF \quad (7)$$

$$TCR = \sum CR \quad (8)$$

Where CR is carcinogenic risk, SF is the oral cancer slope factor $(mg/kg/day)^{-1}$, TCR refers to the carcinogenic risk posed by all heavy metals. When CR or TCR $< 10^{-6}$, indicating the carcinogenic risk could be negligible, CR or TCR $> 10^{-4}$, high risk of causing cancer on human beings, $10^{-6} <$ CR or TCR $< 10^{-4}$, there is an acceptable risk to human beings (US EPA, 2004). It should be pointed out that it varied among different regions, since the discrepancy of the exposure population, in some case it may be high and served as an unacceptable risk.

2.4. Statistical analysis

Basic statistical analysis such as the mean value, standard deviation, WQI, *HQ* and HI were evaluated based on excel. All the statistical procedures were conducted using Microsoft Office 2016 for Windows. The spatial distribution maps of the sampling sites, WQI and HI were drawn by using ArcGIS 10.5 (ESRI Inc, USA). Variation and spatial distribution, carcinogenic risk of trace elements were conducted by origin 2018 (OriginLab, USA).

3. Results and discussion

3.1. General characteristics of the surface water and groundwater

The general characteristics of selected water parameters in surface water and groundwater are showed in Table 1. The pH values of surface water ranged from 6.20 to 8.64, with an average value of 7.71, and for groundwater the values varied between 6.57 and 8.09, with a mean value of 7.53, which was lower in comparison with surface water. Most of the water sites presented alkaline characteristics. The pH variation of groundwater was slightly higher than surface water.

According to the median or mean values, trace elements in water bodies could be divided into three categories: low abundant ($<$ 1 μg/L), moderate abundant (1–100 μg/L), most abundant ($>$ 100 μg/L) (Gao et al., 2019; Xiao et al., 2019). From Table 1, in surface water, the mean concentrations of As, Ba, Cd, Co, Cr, Cu, Ni and Pb were in the range of 1–100 μg/L, and could be classified as moderate abundant elements. The concentrations of Al, Fe, Mn and Zn were $>$ 100 μg/L, which were classified as most abundant elements. In groundwater, moderate abundant elements, such as Al, As, Cd, Co, Cr, Cu, Ni and Pb, the concentrations of which ranged among 1–100 μg/L; most abundant elements, mainly were comprised of Ba, Fe, Mn and Zn, the concentrations of which were $>$ 100 μg/L. The average values of trace elements descended in the trend of Fe $>$ Al $>$ Zn $>$ Mn $>$ Ba $>$ Cu $>$ Pb $>$ Cr $>$ Ni $>$ As $>$ Cd $>$ Co for surface water, and Fe $>$ Zn $>$ Mn $>$ Ba $>$ Al $>$ As $>$ Cu $>$ Pb $>$ Ni $>$ Cr $>$ Co $>$ Cd for groundwater. In addition, the average values of Al, Cd, Cr, Cu, Fe, Ni, Pb, and Zn in surface water were higher than groundwater, while the mean values of As, Ba, Co and Mn in groundwater were lager than surface water. The results suggested that most of mean values of trace elements in surface water were higher than those in groundwater, it could manifest that surface water exhibited relatively more severe pollution by trace elements compared with groundwater. In addition, the SD values of Zn, Mn, Al and Fe in both surface water and groundwater were lager comparing with the other elements, indicating those elements were greatly influenced by human activities.

The spatial distribution of trace elements in surface water and groundwater are presented in Fig. S2. As was shown in Fig. S2a, the higher concentrations of trace elements in surface water were distributed in the following areas: Al in Baisha River Basin, Yangtze River, Dezhou Irrigation District, Middle reaches of Huaihe River and Yellow River Delta; As in Diaojiang River, Lanmuchang area, Upper Han River, Tianjin, Hengyang section of Xiangjiang River and Xiangjiang River; Ba in Southern Shaanxi, Yellow River Delta, Danjiangkou Reservoir,

Table 2
Comparison of trace elements concentrations in surface water and groundwater in this study with others in the world (μg/L).

Area/Surface water	Al	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	References
China	459.87	5.65	81.96	3.54	2.05	25.43	30.14	2186.84	139.70	18.49	29.29	366.23	This study
Stare Miasto reservoir, Poland	38.6	2.07	—	0.08	0.369	0.223	6.45	—	267.9	2.64	1.55	16.9	Siepak and Sojka (2017)
Mississippi River, USA	3	—	1	0.01	0.03	0.01	0.4	2.3	9.5	0.07	0.03	1.4	Bussan et al. (2017)
Douro River estuary, Portugal	322.6	—	19.24	0.11	0.71	2.12	16.26	—	—	5.93	7.25	244.7	Ribeiro et al. (2018)
Dudh Koshi River, Nepal	—	3.68	9.49	0.009	0.8	3.10	1.78	—	28.24	2.74	1.82	16.74	Paudyal et al. (2015)
Tabriz area, Iran	64.4	11.4	—	9.4	—	35.4	—	83.50	118.80	—	3.8	44.5	Barzegar et al. (2019)
Seine river, France	—	0.75	27.3	31	—	—	2.23	—	6.26	—	353.6	—	Elbaz-Poulichet et al. (2006)
Tigris River, Turkey	—	0.63	—	0.044	—	25.41	17.1	158.16	—	24.54	2.82	12.01	Varol et al. (2013)
Damodar River, India	—	—	—	9	2	16	18	—	33	52	10	89	Pal and Maiti (2018)
Buriganga River, Bangladesh	—	134	—	59	199	114	239	612	157	150	119	332	Bhuiyan et al. (2015)
Koel River, India	—	—	—	—	8.667	10.889	6.667	481.78	30.33	24.778	1.667	31.556	Sundaray (2010)
Catalan River, Spain	—	2.9	4.356	1.2	3.455	3.455	1.3	—	—	2.7	2.2	1.9	Carafa et al. (2011)
Ganga River, India	—	—	—	0.005	—	—	0.01	0.8	0.26	0.14	0.12	0.06	Aktar et al. (2010)
World average	—	0.62	23	0.08	0.148	0.7	1.48	66	34	0.801	0.079	0.6	Gaillardet et al. (2014)
Area/Groundwater	Al	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	References
China	75.51	16.81	133.79	1.69	4.81	6.55	16.66	450.27	223.17	8.07	9.58	303.61	This study
Bisha Area, Saudi Arabia	—	0.95	—	0.29	0.05	0.52	0.41	—	1.73	0.63	0.67	8.38	Alqahtani et al. (2020)
Nuevo León, Mexico	—	0.57	—	0.06	1.21	1.21	1.21	—	5.3	—	0.6	51	Mora et al. (2017)
Rangpur district, Bangladesh	27.44	8.8	44.55	—	—	—	—	7726.46	684.48	—	—	33.29	Towfiqul Islam et al. (2017)
Ischia Island, Italy	—	175.66	11.13	0.12	0.37	6.02	157.83	8418.7	462.25	4.83	4.14	137	Daniele (2003)
Tuticorin City, India	84.59	15.8	12.13	0.3	2	13.09	31.66	123.03	40.32	5.19	4.38	203.49	Kumar et al. (2012)
Nalgonda District, India	6.2	4.04	143.61	9.28	18.54	6.2	8.4	25.48	—	39.25	—	771.65	Purushotham et al. (2017)
Andhra Pradesh, India	52.43	0.19	20.21	0.03	0.08	12.54	1.49	80.52	4.37	1.23	20.21	41.52	Dar et al. (2017)
Pesarlanka Island, India	—	114.3	101.6	1.62	9.07	—	11.5	154.51	444.2	7.35	26.21	453.5	Mondal et al. (2010)
Hindon-Yamuna interfluve region, India	2194	1.7	—	0.8	2.8	67.4	900	728	—	68.9	117	1282.6	Alam and Umar (2013)
Tokyo, Japan	5.2	1.8	—	0.1	—	1.8	3.7	31	82	32.9	0.3	—	Thuyet et al. (2016)
Abakaliki area, Nigeria	—	5	—	5	31	23	520	264	8433	14	9	931	Okogbue and Ukpai (2013)

"—" refers no data.

Dezhou Irrigation District and Middle reaches of Huaihe River; Cd in Middle reaches of Huaihe River, Hunchun basin, Yancheng Coastal Area and Huayuan River; Co in Middle reaches of Huaihe River, Upper Han River, Lanmuchang area and Zoige Alpine Basin; Cr in Taizi River, Liangtan River, Middle reaches of Huaihe River, Qingjiang River, Upper Han River and Songhua River; Cu in Anning River, Zoige Alpine Basin, Southwest of Dongguan City, Jilin section of Songhua River, Hengyang section of Xiangjiang River, Middle reaches of Huaihe River and Middle Xijiang River; Fe in Baisha River Basin, Yangtze River, Tianjin, Huayuan River, North Lake of Jining, Handan city and its nearby area, Handan, Daye Region and Middle reaches of Huaihe River; Mn in Baisha River Basin, Taizi River, Huayuan River, Tianjin, Dongjiang River, Yellow River Delta and Dezhou Irrigation District; Ni in Anning River, Southwest of Dongguan City, Middle reaches of Huaihe River, Boai County and Ningbo; Pb in Anning River, Jilin section of Songhua River, Middle reaches of Huaihe River, North Lake of Jining, Yancheng Coastal Area, Huayuan River and Liangtan River; and finally, Zn in Dezhou Irrigation District, Middle reaches of Huaihe River, Yellow River Delta, Anning River, Huayuan River, Mt. Yulong region and Diaojiang River. In general, the hotspots of the higher concentrations of trace elements in surface water were mainly distributed in Middle reaches of Huaihe River, Upper Han River, Dezhou Irrigation District, Yellow River Delta, Huayuan River, Baisha River Basin and Xiangjiang River, which were mainly situated in the middle and lower reaches of Yellow River and Yangtze River basin, and upper reaches such as Yarlung Tsangpo, Lhasa River and Lancangjiang River exhibited lighter pollution, which might attributed to the intensive industrial and agricultural activities in the middle and lower reaches of the river basin.

For groundwater (Fig. S2b), the higher concentrations of trace elements were distributed in the following areas: Al in Dezhou Irrigation District, Jiangjin District, A site in North China and Foshan; As in Huhhot Basin, Kuitun, Jianghan Plain and Datong Basin, Chang-Zhu-Tan urban agglomeration, Baotou City, Tianjin, Guiyang Region and Loess Plateau; Ba in Loess Plateau, Chang-Zhu-Tan urban agglomeration, Jiangjin District, Dezhou Irrigation District, Huhhot Basin and Mayang; Cd in Changzhou, Hunchun basin, Yucheng County, Poyang Lake Plain and Shenyang North New Area; Co in A Mining area in Central Hunan, Loess Plateau, Jining City, Vegetable fields in north Henan Province, Boai County and coastal area of Shenzhen; Cr in Yucheng County, Sewage Irrigation Area in Pearl River Delta, Changzhou, Baoji and A site in North China and Guanzhong Plain; Cu in Huaibei Plain, Huainan City and Changzhou; Fe in Western Kashi Region, eastern part of Songnen Plain, Qingjiang River Basin, Shenyang North New Area and Yerqiang River Basin; Mn in Northern Suburb of Xuzhou City, A site in North China, Shenyang North New Area, coastal area of Shenzhen, eastern part of Songnen Plain and Kashi Region; Ni in Kashi Region, A mining district in north Anhui, Bosten Lake Basin and Boai County; Pb in Yucheng County, An area in southern Hunan Province, Northern Suburb of Xuzhou City, Baoji, Kuitun and Changzhou; Zn in Laiyang City, Dezhou Irrigation District, Pingyi County, Lower Reaches of Yellow River and Dingji coal mine. Overall, the hotspots of the higher concentrations of trace elements in groundwater were mainly distributed in Loess Plateau, Dezhou Irrigation District, Chang-Zhu-Tan urban agglomeration, coastal area in Shenzhen, eastern part of Songnen Plain, Kashi Region, Changzhou and Baoji, which shared certain identical area with the surface water such as Dezhou Irrigation District.

The mean values of trace elements in water body exhibited significantly discrepancy among different river basins (Fig. 1). For surface water, Yangtze River Basin presented higher Al, Fe and Ni than other river basins, As was higher in Pearl River Basin, Southwest River Basin, and Hai River Basin, with the mean values being 9.31, 6.89 and 6.16 μg/L, respectively; Ba in Yellow River Basin, Huai River Basin and Yangtze River Basin showed higher pollution, Cd, Co and Zn were higher in Huai River Basin, Cr (mean, 210.67 μg/L) and Mn (mean, 803.88 μg/L) presented higher in Liao River Basin, and Songhua River Basin and Huai River Basin presented higher Pb pollution than other river basins, the mean values were 194.72 μg/L and 67.33 μg/L, respectively.

With regards to groundwater, Al and Zn were higher in Hai River Basin, with the mean values being 185.31 μg/L and 1076.80 μg/L, respectively. As showed higher in Northwest River Basin, Yangtze River Basin, the mean values were 57.40 μg/L and 18.20 μg/L, respectively. Songhua River Basin presented higher Cd, Cr and Fe pollution than other river basins, the mean values were 6.82 μg/L, 10.88 μg/L and 1622.31 μg/L, respectively. Higher Ba was in Yellow River Basin and Yangtze River Basin, and higher Co in Yellow River Basin, Cu in Huai River Basin and Songhua River Basin, Mn in Liao River Basin and Songhua River Basin, Ni in Northwest River Basin and Huai River Basin, Pb in Huai River Basin and Yangtze River Basin were exhibited.

In general, from a river basin perspective, Yangtze River Basin, Songhua River Basin and Huai River Basin manifested more serious pollution, while comparably speaking, Hai River Basin, Northwest River Basin, Southwest River Basin, Pearl River Basin, and Liao River Basin presented relatively lighter pollution. A large number of heavy chemical enterprises and megacities with a population of more than a million concentrated along the Yangtze River basin, such as the largest comprehensive industrial base Hu-Ning-Hang Industrial Base in China, large steel industrial base including Panzhihua, Wuhan, Maanshan and Chongqing, which posed serious pollution to surface water. The Songhua River Basin was the vital water pollution control basin in China, it was a very important heavy industry production base in China, the vast majority of industrial enterprises such as metallurgy, iron and steel industry were distributed along the river basin, secondly, Songhua River Basin served as China's famous commodity grain production base, the applying of fertilizers and pesticides were large amount, and the discharge of domestic sewage and garbage, all these factors led to serious pollution in Songhua River Basin (Wang, 2017). Since 2004, the Huai River basin had become one of the worst rivers in China's rivers, it served as an important grain production base and nonferrous mining and coal mining areas, non-point source pollution caused by agricultural production, point source pollution caused by industrial production and concentrated pollution caused by urban development, all of which caused the deterioration of the ecological environment and water bodies in the Huaihe River Basin (Yu, 2017).

Furthermore, limited by the level of economic development, the domestic sewage treatment facilities in rural areas were weak, the residents were lack of the awareness of water environment protection, and the state and local governments had not yet established and improved the wastewater resource utilization system, the random discharge of domestic sewage and industrial wastewater caused serious pollution to water bodies in rural areas.

Additionally, in this study, relatively fewer sample points were selected in some river basins, therefore, the results might not well reflect the comprehensive pollution situation of trace elements over the China's river basins, while the information still has certain scientific significance for reference.

By contrast, Table 2 illustrated that the mean values of all the selected trace elements in surface water were higher than those of the world averages (Gaillardet et al., 2014), with the exception of Al which has no average value. The mean values of Ba, Fe and Zn in this study were higher than all other selected surface water worldwide, and the concentrations of other nine elements were larger than most of the selected surface water and less than minority of the comparing waters. For example, As was larger than Stare Miasto reservoir in Poland (Siepak and Sojka, 2017), Dudh Koshi River in Nepal (Paudyal et al., 2015), Seine river in France (Elbaz-Poulichet et al., 2006), Tigris River in Turkey (Varol et al., 2013) and Catalan River in Spain (Carafa et al., 2011), but less than surface water of Tabriz area in Iran (Barzegar et al., 2019) and Buriganga River in Bangladesh (Bhuiyan et al., 2015); Cd was larger than Stare Miasto reservoir in Poland, Mississippi River in USA (Bussan et al., 2017), Douro River estuary in Portugal (Ribeiro et al., 2018), Dudh Koshi River in Nepal, Tigris River in Turkey, Catalan River in Spain and Ganga River in India (Aktar et al., 2010), and was less than surface waters of Tabriz area in Iran, Seine river in France, Damodar River in India (Pal and Maiti, 2018), and Buriganga River in Bangladesh; Cr was larger than most of the other water, such as Koel River in India (Sundaray, 2010); Pb was less than Seine River and Buriganga River, while was larger than the other surface water.

With regards to the groundwater, the average concentrations of Al, As, Ba, Cd, Co, Fe, Mn, Ni Pb and Zn were higher than most of the selected groundwater, such as Al was larger than those of all the groundwater in worldwide except for Tuticorin City in India (Kumar et al., 2012) and Hindon-Yamuna interfluve region in India (Alam and

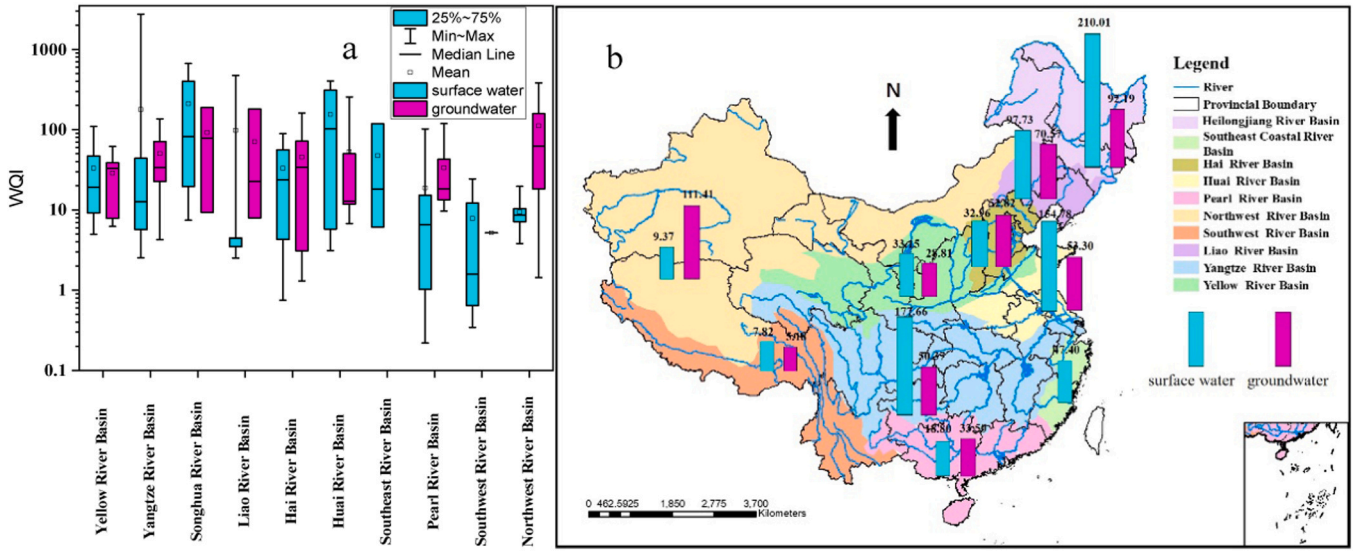

Fig. 2. Box plots of WQI (a) and mean WQI value (b) in surface water and groundwater in different river basins.

Umar, 2013), As was larger than those of all the groundwater in the world expect Ischia Island in Italy (Daniele, 2003) and Pesarlanka Island in India (Mondal et al., 2010), Mn was larger than those of Bisha Area in Saudi Arabia (Alqahtani et al., 2020), Nuevo León in Mexico (Mora et al., 2017), Tuticorin City in India (Kumar et al., 2012), Andhra Pradesh in India (Dar et al., 2017) and Tokyo in Japan (Thuyet et al., 2016), while was less than Rangpur district in Bangladesh (Towfiqul Islam et al., 2017), Ischia Island in Italy, Pesarlanka Island in India and Abakaliki area in Nigeria (Okogbue and Ukpai, 2013). The average concentrations of Cr and Cu were less than most of the selected groundwater and larger than minority of the comparing groundwater in the world, for example, Cr was larger than Bisha Area, Nuevo León, Ischia Island, Nalgonda District and Tokyo, while was less than the other water, Cu was larger than most of the groundwater in the world, except for those of Ischia Island, Tuticorin City, Hindon-Yamuna interfluve region, and Abakaliki area.

3.2. Water quality assessment

Compared with the drinking water guidelines of Chinese State Standards (GB5749-2006) (Table 1), the mean pH values of most of the selected surface water and groundwater were in the range of 6.5–8.5 formulated by CSS, with exception of two surface water sites in Chinese Loess Plateau (8.64) and Tianjin (8.53), indicating that most of the water pH values were suitable for human drinking in this study. The concentrations of Ba, Co and Cu in surface water and Co, Cr, Cu and Ni in groundwater were within the permissible limits set by CSS for drinking purposes. The proportions of trace elements in surface water exceeding the CSS were Al 20%, As 15.49%, Ba 3.70%, Cd 5.56%, Cr 6.85%, Cu 1.20%, Fe 28.57%, Mn 12.96%, Ni 9.26%, Pb 17.95%, and Zn 5.95%, respectively; with regards to trace elements in groundwater, the exceeding proportions were Al 5%, As 21.82%, Ba 5.56%, Cd 9.26%, Co 5.56%, Fe 42.86%, Mn 50.94%, Ni 8.11%, Pb 17.24% and Zn 3.39%, respectively. The major pollution elements were Al, As, Fe and Pb for surface water, and As, Fe and Mn for groundwater. The highest Al was in Baisha River Basin (9908 μg/L) in Guizhou province and As in Diaojiang River (48.05 μg/L) in surface water, for groundwater, highest Al was in Dezhou Irrigation District (800 μg/L) and As in Huhhot Basin (262.43 μg/L).

Additionally, the WQI was used to further determine the water quality in surface water and groundwater for drinking purposes, according to the Eq. (1), and the parameters for WQI were listed in Table 1, Fig. S3 presented the water quality classifications in surface water and groundwater in this study. The WQI values for surface water (Fig. S3a) varied between 0.22 and 2746.60, with the mean value of 86.02, indicating good water quality based on the WQI classifications standards; the proportions of excellent water quality, good water quality, poor water quality and undrinkable water quality were 78.49%, 7.53%, 6.45% and 7.53%, respectively, manifesting that the majority of the surface water in this study was suitable for drinking water except for the Huayuan River (367.56), Anning River (927.75), Baisha River Basin (2746.60), Jilin section of Songhua River (668.61), Taizi River (473.72), Middle reaches of Huaihe River (403.84) and Yancheng Coastal Area (310.15). For groundwater (Fig. S3b), the WQI values ranged from 1.31 to 382.61 with an average of 56.64, the mean water quality presented good level. According to the WQI classifications standards, the water quality could be classified five levels including excellent water quality, good water quality, poor water quality, very poor water quality and undrinkable water quality, the proportions of which were 67.53%, 15.58%, 11.69%, 3.90% and 1.30%, respectively; the very poor water quality sites distributed in a mining district in north Anhui (255.86), Northern Suburb of Xuzhou City (200.03) and Kuitun (289.16), and the water quality in Huhhot Basin (382.61) reached undrinkable grade.

In general, 86.02% of surface water and 83.11% of groundwater in this study were suitable for human drinking water, while the minority of undrinkable water and poor water sites should also be paid more attention by the local administration, for they were unsuitable for drinking water, and might induce adverse effect to human beings. In addition, groundwater manifested more proportion of poor water quality sites compared with surface water.

The highest WQI value was located in Baisha River Basin (2746.60), with the high concentrations of Al (9908 μg/L), Fe (68479 μg/L), and Mn (2745 μg/L), which was attributed to intensive mining areas distributing in the basin. The original coal mining in the area and the water from the old kiln was discharged randomly without treatment, and gangue was piled up at will without comprehensive treatment. In addition, large areas of agricultural activity and dense urban settlements established upstream were also likely the composition of stable source of pollution in the Baisha River Basin (Liu et al., 2020). The high WQI in the middle reaches of Huaihe River might be derived from the intensive anthropogenic activities (industrial, agricultural, and domestic) and economic developments along the regions, such as the extensive mining activities and coal-fired power plants in Huainan city, which generated approximately 10 Mt of coal combustion annually that were rich in trace elements (Wang et al., 2017a, 2017b). The mean concentration of As in groundwater of Jianghan Plain was up to 109 μg/L, with the highest As concentration reaching as high as 2330 μg/L, which should be taken seriously attention, for the high arsenic groundwater mainly emerged in most of residential wells in the region (Gan et al., 2014). High concentrations of Fe (2840 μg/L) and Mn (730 μg/L) were a typical characteristic in groundwater of Songnen Plain, the phenomenon of which might be fundamentally attributed to the interaction between groundwater and rock, and can be enhanced by nitrogen and organics infiltrated into the aquifer from earth surface (Zhai et al., 2019). The mean concentrations of Mn was up to 730 μg/L in groundwater of coastal area in Shenzhen, and was especially higher in summer season, which might be induced by the depletion of oxygen in groundwater and the reducing environment in low pH water (Chen et al., 2007). In summary, anthropogenic activities and the native environment were the main causes of the high concentrations of trace elements and high WQI of water quality, and could ultimately pose adverse effects on aquatic ecosystem and human health (Cao et al., 2019).

As was illustrated in Fig. 2, for surface water, the WQI values in Yangtze River Basin, and Huai River Basin varied from 2.54 to 2746.60, and 3.13–403.84, respectively, with the mean values of 177.66, and 154.78, respectively, all of which reached poor water quality; the WQI values in Songhua River Basin ranged 7.44–668.61, with the mean values being 210.01, reaching very poor water quality. For groundwater, the WQI values Northwest River Basin varied between 1.44 and 382.61, the mean value was 111.41, which belonged to poor water quality. Intensive anthropogenic activities such as industrial and agricultural production, residents' lives and economic development in these regions could contribute to the accumulation of trace elements in water bodies, which ultimately generate water quality deteriorates. Such as the anthropogenic activities including domestic sewage, coal mining, and coal fired power plants in Huai River Basin could result in the deterioration of water quality (Wang et al., 2015).

The mean WQI values of surface water and groundwater in Liao River Basin, Hai River Basin, Yellow River Basin, Southwest River Basin and Pearl River Basin were below 100, belonging to excellent or good water quality. The majority of the river basins manifested excellent water quality or good water quality, especially in the Northwest River Basin, Southwest River Basin and upper river stream, which presented excellent water quality. In general, comparatively, rivers in the South relatively demonstrated a better water quality than the North, it was reported that the percentages of poor quality surface water sections in Hai, Huai, Songhua and Yellow river basins in 2014 were 65%, 65%, 51% and 44%, respectively (Wang et al., 2017a, 2017b), which may be attributed to the abundance of water resources in the South, and low resilience to absorb and mitigate pollution based on the scarcity and pollution interaction in the North (Jiang, 2015).

However, it should be illustrated that although the mean WQI values

Table 3
Hazard quotient (HQ), Hazard Index (HI) and cancer risk (CR) for trace elements in surface water and groundwater.

Element	$HQ_{ingestion}$		HQ_{dermal}		HI		TCR	
	Child	Adult	Child	Adult	Child	Adult	Child	Adult
Surface water								
Al	1.88E-2	1.26E-2	9.70E-4	3.29E-4	1.98E-2	1.29E-2		
As	7.65E-1	5.12E-1	8.30E-3	2.81E-3	7.73E-1	5.15E-1	3.02E-5	1.00E-4
Ba	1.68E-2	1.12E-2	2.47E-3	8.37E-4	1.92E-2	1.21E-2		
Cd	2.90E-1	1.94E-1	5.98E-2	2.03E-2	3.50E-1	2.14E-1		
Co	2.79E-1	1.87E-1	5.76E-3	1.95E-3	2.85E-1	1.89E-1		
Cr	3.46E-1	2.31E-1	2.85E-1	9.66E-2	6.31E-1	3.28E-1		
Cu	3.08E-2	2.06E-2	1.59E-3	5.38E-4	3.24E-2	2.12E-2		
Fe	1.28E-1	8.56E-2	6.59E-3	2.23E-3	1.34E-1	8.78E-2		
Mn	2.38E-1	1.59E-1	6.14E-2	2.08E-2	3.00E-1	1.80E-1		
Ni	3.78E-2	2.53E-2	1.95E-3	6.61E-4	3.98E-2	2.60E-2		
Pb	8.56E-1	5.73E-1	2.94E-3	9.97E-4	8.59E-1	5.74E-1		
Zn	4.94E-2	3.31E-2	1.53E-3	5.18E-4	5.09E-2	3.36E-2		
Groundwater								
Al	3.09E-3	2.07E-3	1.59E-4	5.40E-5	3.25E-3	2.12E-3		
As	2.29	1.53	2.49E-2	8.43E-3	2.32	1.54	9.06E-5	3.00E-4
Ba	2.74E-2	1.83E-2	4.03E-3	1.37E-3	3.14E-2	1.97E-2		
Cd	1.38E-1	9.25E-2	2.85E-2	9.66E-3	1.67E-1	1.02E-1		
Co	6.56E-1	4.39E-1	1.35E-2	4.58E-3	6.70E-1	4.44E-1		
Cr	8.93E-2	5.98E-2	7.36E-2	2.50E-2	1.63E-1	8.47E-2		
Cu	1.70E-2	1.14E-2	8.79E-4	2.98E-4	1.79E-2	1.17E-2		
Fe	2.63E-2	1.76E-2	1.36E-3	4.60E-4	2.77E-2	1.81E-2		
Mn	3.80E-1	2.55E-1	9.81E-2	3.32E-2	4.79E-1	2.88E-1		
Ni	1.65E-2	1.11E-2	8.52E-4	2.89E-4	1.74E-2	1.13E-2		
Pb	2.80E-1	1.88E-1	9.63E-4	3.26E-4	2.81E-1	1.88E-1		
Zn	4.14E-2	2.77E-2	1.28E-3	4.34E-4	4.27E-2	2.82E-2		

of some river basins were under safe drinking levels, several sites were still need attention, for example, the WQI value in Taizi River reached 473.72, to the point that was undrinkable degree, which may be attributed to the sewage discharged by steel enterprises in Taizi River flowing through Benxi urban area in Liao River Basin, where the highest Mn content reached 6370 μg/L (Bu et al., 2017). The groundwater sites of Northern Suburb of Xuzhou City and Yucheng County in Huai River Basin, the WQI of which were 255.86 and 200.03, respectively, indicating very poor water quality. The Pb and Mn in the shallow groundwater in Northern Suburb of Xuzhou City were seriously out of standard, and the mean Mn concentration was 1300 μg/L, which indicated that the shallow groundwater had been polluted, and the Pb pollution had affected the whole study area, anthropogenic influence, and the presence of a large number of Fe-Mn nodules in the aquifer may be the causes of water pollution in this area (Zhang et al., 2009). In Yucheng County, the highest concentration of Cr and Pb in groundwater were 171.4 and 253.9 μg/L, respectively, which may be related to local factors (such as pesticide and fertilizer application) in the planting area (Wang and Ma, 2011).

Additionally, groundwater was still a large part of the people living drinking water source, roughly 70% of Chinese people relied on groundwater for drinking water, for example, in Hai River basin, it contributed 65–67% of the total water supply (Jiang, 2015; Qiu, 2011). It was necessary to carry out systematic evaluation of heavy metal elements in shallow groundwater in order to protect the health of local people and the sustainable development of social economy.

3.3. Health risk assessment

Table 3 presented the hazard quotient (HQ), hazard Index (HI) and cancer risk (CR) for trace elements in surface water and groundwater in this study. The selected trace elements had the potential to pose adverse risk to human health with the increasing of the concentrations of pollutants in water body if contaminated water was used for potable supply

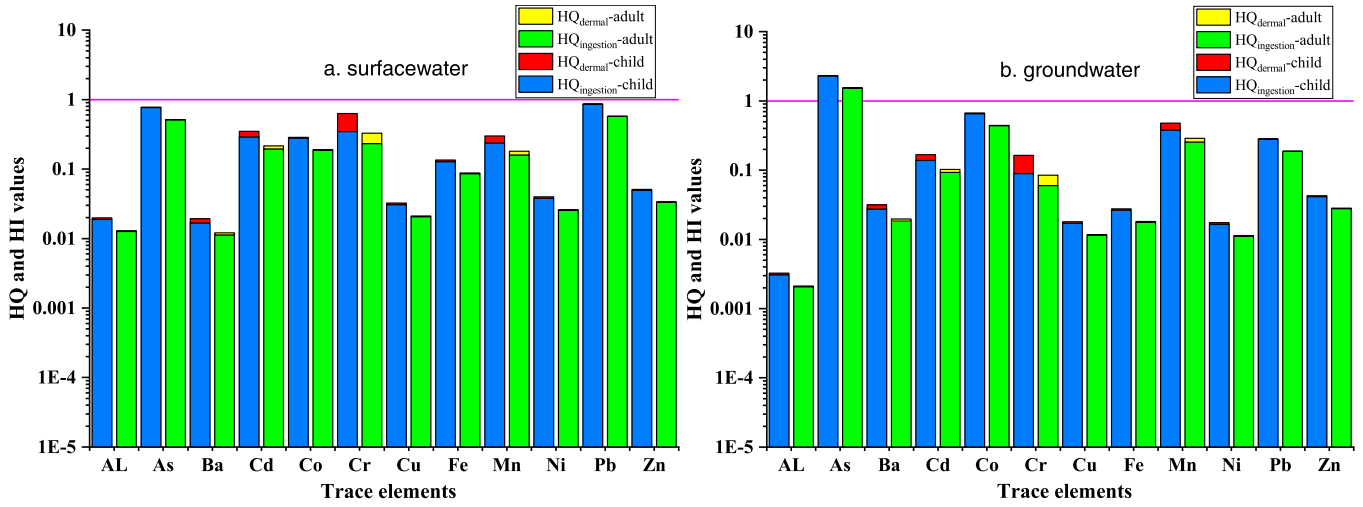

Fig. 3. The $HQ_{ingestion}$, HQ_{dermal} and HI values for child and adult in surface water (a) and groundwater (b).

(Hussain et al., 2019), while several elements could pose carcinogenic risk to human health such As, Cd, Cr and Pb, the non-carcinogenic risk of the 12 trace elements and carcinogenic risk of As through ingestion and dermal pathways for children and adults were calculated according to the health risk assessment model recommended by the US EPA.

3.3.1. Non-carcinogenic risk

As was illustrated in Table 3, for surface water, the mean values of $HQ_{ingestion}$, HQ_{dermal}, HI of the selected trace elements for children and adults were all below 1, indicating no adverse effects and potential non-carcinogenic risk on human health. For groundwater, the mean values of $HQ_{ingestion}$ of As and HI for both children and adults were above 1, indicating it might cause adverse effects and non-carcinogenic risk to human health, and the average values of $HQ_{ingestion}$, HQ_{dermal}, HI of the other elements for both children and adult were below 1, within the safe limits. As shown in Fig. 3, in both surface water and groundwater, As served as the significant pollutant to cause the non-carcinogenic risk to human health, the values of $HQ_{ingestion}$ of the trace elements were higher than those of HQ_{dermal} for both children and adults, which manifested that ingestion was the principal pathway to pose adverse effects and cause non-carcinogenic risk on human body in water system, which was consistent with the previous research (Zhang et al., 2015). Furthermore, $HQ_{ingestion}$, HQ_{dermal}, HI values for children were higher with compared with adults, implying that children were more vulnerable to the adverse effects of trace elements than adults when exposed from the same water medium, which was confirmed by the former study conducted by Gao et al. (2019) and Xiao et al. (2019). In addition, the HI values of As for adult and children in groundwater were larger than 1, indicating non-carcinogenic risk might induce on human being, and it presented higher risk in groundwater than surface water.

Excessive consumption or long-term chronic exposure of trace elements might induce toxic effects. For example, previous studies had manifested that long-term and low-dose Cd exposure was associated with nephrotoxicity, osteoporosis and neurotoxicity, in addition, Cd may play a role in prostate cancer by destroying androgen receptors (Neslund-Dudas et al., 2018; Qing et al., 2020). Chromium is highly toxic and considered to be a class A carcinogen (Coetzee et al., 2018), Cr (VI) is biologically highly toxic among the chromium forms, and it could deter the normal physiological functions of cellular components and generate mutagenic, teratogenic, and carcinogenic effects in biological systems (Dhal et al., 2013; Mishra and Bharagava, 2016). Exposure to cobalt compounds can induce toxicity in human lung cells such as cytotoxic and genotoxic to human lung epithelial cells (Xie et al., 2016). Lead exposure may cause nephropathy, affective disorders, decline in intelligence, memory and cognitive deficits, especially result in more severe neurological damage in pediatric exposure (Mason et al., 2014). Zinc is considered to associate with apoptosis, neuronal injury and neurodegenerative diseases (Plum et al., 2010).

For the two groups, the majority of the HI values in the both water bodies were below 1, indicating no non-carcinogenic risk on human health, and children suffered more serious risk than adults. In surface water, for children and adults, the high non-carcinogenic risk sites were mainly distributed in Anning River, Taizi River, Middle reaches of Huaihe River and Jilin section of Songhua River, the HI values of those sites were 25.52, 21.76, 18.95 and 16.56 for child, and 16.65, 11.89, 12.19 and 11.07 for adults, respectively. With regards to groundwater, the high non-carcinogenic risk sites were mainly distributed in Huhhot Basin, Kuitun, Jianghan Plain, Datong Basin and Yucheng County, with the HI values of 37.11, 30.06, 15.03, 7.20 and 6.36 for children, and 24.66, 20.01, 10.01, 4.77 and 4.01 for adults, respectively. In general, they constituted the predominant non-carcinogenic risk sites, which were not suitable for drinking water source and should be paid much more attention. The pollution source of the high risk sites might be attributed to the intensive human activity, industrial and agricultural activities, and it should be strengthened the control measures by the management department. For example, high concentrations of As and Ni appeared in the middle reaches of the Anning River were closely related to the exploitation of mineral resources, under the influence of the Taihe vanadium titanomagnetite (Zhu et al., 2018). The high contents of heavy metals in Taizi River were mainly caused by point source pollution from industry and urban life, it received the large amount of living and production waste water and sewage when flowing through the urban area of Benxi, especially the waste water from Benxi Iron and Steel Group (Wang et al., 2019). The pollution source characterizations of trace elements in Middle reaches of Huaihe River were associated with the spatial heterogeneity of human activities such as industry, agriculture, household and economic development in the area (Wang et al., 2017a, 2017b). The discharge of industrial, agricultural and domestic sewage seriously polluted the water quality of Jilin section of Songhua River (Liu et al., 2009). In some groundwater sites, hydrogeochemical properties in aquifer affected the release and mobilization of elements in groundwater such as Huhhot Basin, Kuitun, Jianghan Plain and Datong Basin, which were related to the pH, reducing condition, or geological conditions, etc.

Additionally, the HI values in some sites were in the range of 0.1–1, although showing low non-carcinogenic risk, they might pose adverse risk on human health in the long time, therefore, those sites should also be attached great importance to, such as Yellow River Delta, Yangtze

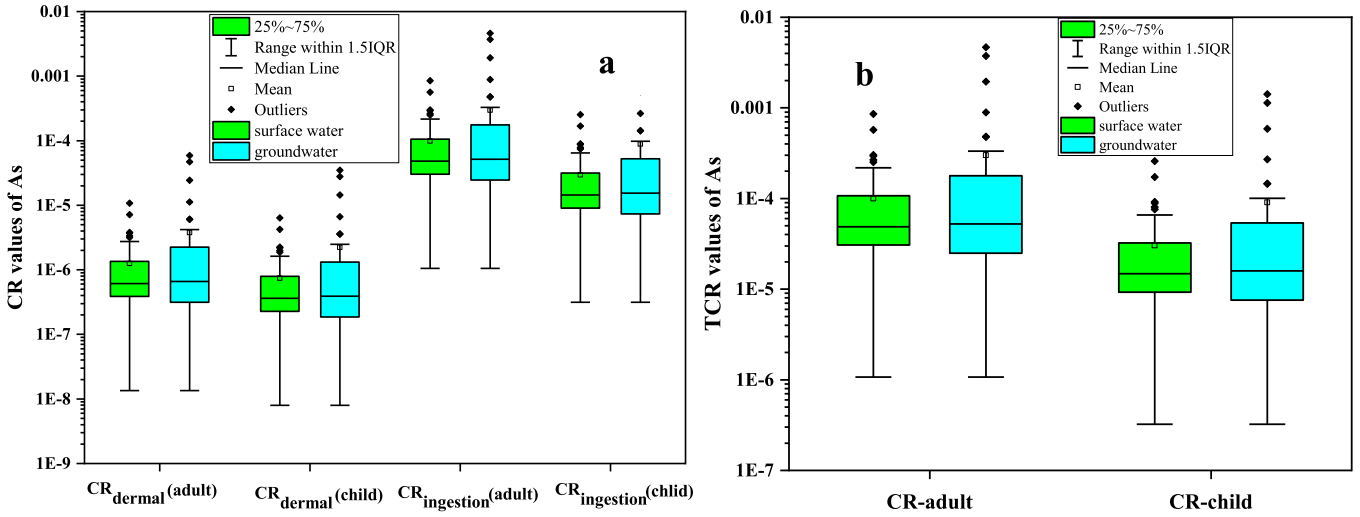

Fig. 4. CR_{dermal} and $CR_{ingestion}$ values (a) and TCR values (b) of As for child and adult in surface water and groundwater.

River Basin, Handan, Poyang Lake, and Guanzhong Plain.

Moreover, groundwater manifested more risk sites than surface water, and it was served as the main drinking water sources in many regions around China especially in the vast rural areas. However, most research was focused on surface water pollution and less on the groundwater, yet the quality of the groundwater was particularly important for the health of the population. Therefore, a wide range of groundwater research should be carried out in the future, especially in remote rural areas that lack water purification facilities and regular monitoring.

Finally, more concerns were paid on the Yellow, Yangtze, Huai and Pearl River Basins, while it is lack sufficient attention to the Northwest and Southwest River Basins, although the population and economic development of which are inferior to the central and eastern regions, they are the source of Chinese large rivers and the main area of inland rivers, the water quality in those regions still cannot be ignored. Therefore, subsequent studies and water quality monitoring in Northwest and Southwest River Basin should also be given more attention.

3.3.2. Carcinogenic risk

Among the selected elements, As served as the significant contributor to pose adverse effects on the human body, therefore, the carcinogenic risk of As were calculated in this study (Table 3). The carcinogenic risk of Pb was not assessed because it was classified as probable human carcinogen by US EPA. According to the CR classification recommended by the US EPA, carcinogenic risk values of As (Fig. 4a) for children and adults through dermal pathway in surface water and groundwater were in the range of $7.94 \times 10^{-9} - 5.89 \times 10^{-5}$, all of which were below the value 1×10^{-4}, indicating acceptable risk or negligible risk. While through the ingestion pathway, the CR values varied from $3.16 \times 10^{-7} - 1.68 \times 10^{-4}$ for children and $8.45 \times 10^{-6} - 5.64 \times 10^{-4}$ for adults in surface water, and $3.16 \times 10^{-7} - 5.73 \times 10^{-4}$ for children and $1.06 \times 10^{-6} - 4.62 \times 10^{-3}$ for adults in groundwater, some of the values exceeded the threshold 1×10^{-4}, belonging to high carcinogenic risk. In addition, the risk caused by ingestion pathway were higher than dermal ingestion.

The range of TCR values of As in surface water for children and adults were in the range of $2.24 \times 10^{-7} - 1.73 \times 10^{-4}$ and $1.07 \times 10^{-6} - 8.57 \times 10^{-4}$ (Fig. 4b), respectively, for groundwater, they were $3.24 \times 10^{-7} - 5.88 \times 10^{-4}$ and $1.07 \times 10^{-6} - 4.68 \times 10^{-3}$, respectively, some of the values were larger than 1×10^{-4}, manifesting high carcinogenic risk. The mean TCR values of As for children and adults in surface water were 3.02×10^{-5} and 1.00×10^{-4}, respectively, and they were 9.06×10^{-5} and 3.00×10^{-4} in groundwater (Table 3), respectively, indicating that high carcinogenic risk might occur on adults groups. Additionally, the mean carcinogenic risk of As in groundwater sites presented higher than those in surface water, and adults suffered higher carcinogenic risk than children groups, which was contrary with the non-carcinogenic risk, which might be attributed to larger quantity of drinking water in daily life for adults with compared to children.

In this study, the high risk sites were mainly located in Lanmuchang area, Diaojiang River in surface water, and for groundwater, they were in Baotou City, Jianghan Plain, Chang-Zhu-Tan urban agglomeration, Datong Basin, Huhhot Basin, and Kuitun, which were also the high As concentration regions, for example, the average As contents in Huhhot Basin and Kuitun were 262.43 μg/L and 210.19 μg/L, respectively, exceeding the Chinese national standard of 50 μg/L, the highest arsenic contents were 1480 μg/L and 381 μg/L in the two sites, respectively; mean As concents were 109 μg/L and 50.13 μg/L in Jianghan Plain and Datong Basin, respectively, with the highest As concentration being 2330 μg/L (Gan et al., 2014) and 452 μg/L (Huq et al., 2018), respectively. It was speculated that Fe and Mn oxides/hydroxides were potential sources of As, due to the moderate to strong associations between dissolved As and Fe and Mn in groundwater, and the reduction of As(V) to As(III) could promote the desorption of As through the strong reductant of Fe(II) (Gan et al., 2014), the bacterial reductive suspension of Mnoxy-hydroxides could contribute to the release of As (Huq et al., 2018). High concentrations of As in groundwater of Huhhot Basin were associated with strongly reducing conditions characterized by high concentrations of dissolved Fe, Mn, NH_4, DOC and As(III) and relatively low concentrations of SO_4 (Smedley et al., 2003), reductive dissolution of Fe and Mn oxides/hydroxides under reducing conditions in groundwater of Jianghan Plain was the main possible mechanism of As release (Gan et al., 2014). Furthermore, antigenic siderite and pyrite immobilize arsenic in groundwater with high arsenic concentrations, the main reason for arsenic releasing from aquifer sediments was the dissolution of reducing iron oxide (Long and Luo, 2020). In addition, hydrogeological conditions posed a significant impact on pollutants in water, for example, arsenic-rich veins and lacustrine sediments, or high arsenic anomaly zones formed by geological and hydrothermal conditions that were conducive to the storage and enrichment of arsenic elements may be the reasons for the high arsenic content in the groundwater of Kuitun, Xinjiang (Luo et al., 2011). Additionally, anthropogenic factors also influenced the mobility of As such as mining and smelting activities, the use of arsenical pesticides in agriculture and As bearing sulfide minerals in sulfuric acid manufacturing (Gao et al., 2019), etc.

It had been confirmed that long-term exposed to arsenic through contaminated drinking water could induce potentially carcinogenic health risk such as the cancers of liver, lung, bladder, kidney and skin, and together with non-cancer outcomes including cardiovascular diseases, hypertension, neuropathy (Li and Zhang, 2010; Phung et al., 2017). Especially for the susceptible children populations, it has been proved that exposure to As has been linked to neurobehavioral defects during the early years of life (Calatayud et al., 2019). A significant negative effect on neurodevelopment might occur in children with arsenic exposure between the age 5–15 years of ages, which affected Full Scale IQ specifically in verbal and performance domains (Rodriguez-Barranco et al., 2013). A study conducted in Bangladesh suggested that children exposed to high As drinking water ($>$ 50 μg/L) might result in the decreased intellectual function such as obtaining poorer full-scale and performance scores than those exposed to low As drinking water ($<$ 5.5 μg/L) (Wasserman et al., 2004).

Concentrations of As more than 10 μg/L in groundwater had been widely found in China mainland covering 19 provinces, it was estimated that up to 19.6 million people were under the unhealthy condition of consuming high-arsenic groundwater, and drinking water endemic arsenism was distributed in 8 provinces and municipalities, high arsenic exposure population with the drinking water arsenic over 50 μg/L was estimated to 523 thousand (Guo et al., 2014; Jin et al., 2003; Rodriguez-Lado et al., 2013). It had induced significant health problems in large number of human beings inhabiting in the contaminated locations. In addition, except for the sites that As concentration surpassed 10 μg/L, the low As concentration sites should also be paid attention for the accumulation and non-biodegradable of the toxic element.

Therefore, the monitoring of As concentrations in water body for the drinking purpose should be strengthened and paid more attention especially for the vulnerable children groups by the local administration department. Furthermore, other trace metals such as Cd, Cr, Pb and Ni should also be brought into the special attention, for they were non-biodegradable in aquatic ecosystem and could also pose larger risk to human being even if their concentrations meet the drinking water standards. It was considered that contaminants such as nitrate/nitrite and chromium in drinking water posed a major risk for cancers of the digestive system, for example, in the Huai River Basin, there seemed to be a significant spatial correlation between the mortality of digestive system cancer and water pollution in rural areas (Jiang, 2015).

Additionally, in some severe contamination regions, local inhabitants should be encouraged to develop healthy drinking habits through conducting educational campaigns to raise their awareness of the dangers of harmful elements in drinking water, and the necessity to transformate drinking water resources especially avoiding the ingestion of groundwater, or taking effective measures to remove toxic metals

from the water body.

4. Conclusions

This study conducted a comprehensive analysis on dissolved trace elements in surface water and groundwater in China acquiring from the online literature. The results showed that most of mean values of trace elements in surface water were higher than those in groundwater. From the perspective of watershed distribution, it synthetically presented more serious pollution in Yangtze River Basin, Songhua River Basin and Huai River Basin. The majority of the surface water and groundwater sites selected in this study were suitable for drinking water purpose, with the exception of the minority of undrinkable water and poor water sites such as Baisha River Basin, the middle reaches of Huai River, Taizi River, Jilin section of Songhua River, and Huhhot Basin, etc.

Due to the high non-carcinogenic risk sites were mainly situated in Anning River, Taizi River, Middle reaches of Huaihe River, and Jilin section of Songhua River, Huhhot Basin, Kuitun, Jianghan Plain, Datong Basin, and Yucheng County, more efforts should be concentrated in these regions, while the lower risk sites in Northwest River Basin and Southwest River Basin still need attention. Arsenic contributed significantly to cause the non-carcinogenic risk on human health, ingestion was the predominant exposure pathway to pose adverse effects on human body in water system, children were more vulnerable to the adverse effects of trace elements comparing with adults. It should be paid much more attention to the predominant risk sites such as Lanmuchang area, Diaojiang River, Baotou City, Jianghan Plain, Chang-Zhu-Tan urban agglomeration, Datong Basin, Huhhot Basin, and Kuitun. The carcinogenic risk of As presented higher in groundwater than surface water.

In addition, the calculation of the risk assessment based on the average values of the trace elements, the body weight and the daily intake vary among different regions and groups, etc., which might result in the uncertainties of the potential risk posed by these elements. Furthermore, it should be noted that in the case of metals, their toxicity is also affected by valence, mobility, bioavailability and other factors, and ultimately affect the health risk they pose to humans, therefore, it may be incomplete through only taking into account the total concentrations.

Furthermore, the risk of arsenic exposure should be comprehensively assessed from drinking water, diet and other exposure routes. The data and the sampling sites collected in this study are limited and the relatively few sample sites in water body may not well reflect the overall contamination situation of trace elements in surface water and groundwater around China, in addition, some of the published articles focused on the heavily polluted areas, using these data to analyze the elements pollution level of surface water and groundwater in China may lead to the comprehensive evaluation of the pollution level to be high. Although there are shortcomings, our study still provides certain scientific references for the systematic comparation and understanding of the pollution status of trace elements in surface water and groundwater in different basins in China.

Finally, we provide the proposal that the future research should be carried out more extensive investigation on river basin water quality, in particular, for the drinking purpose water resource of groundwater in vast rural areas, due to most dig wells were drilled privately following no drinking water standard and lack of water purification facility. It should also strengthen the long-term monitoring of the major river basin especially with the high As water body. It is essential to transform the drinking water sources for good water quality in poor water region, and assess the risk caused by the diet exposure pathway.

CRediT authorship contribution statement

Shuangmei Tong: Conceptualization, Methodology, Data curation, Writing - original draft, Writing - review & editing. **Hairong Li**: Visualization, Funding acquisition. **Muyesaier Tudi**: Supervision. **Xing Yuan**: Software, Validation. **Linsheng Yang**: Reviewing, Editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2021.112283.

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