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Water quality in the Tibetan Plateau: Major ions and trace elements in rivers of the "Water Tower of Asia"

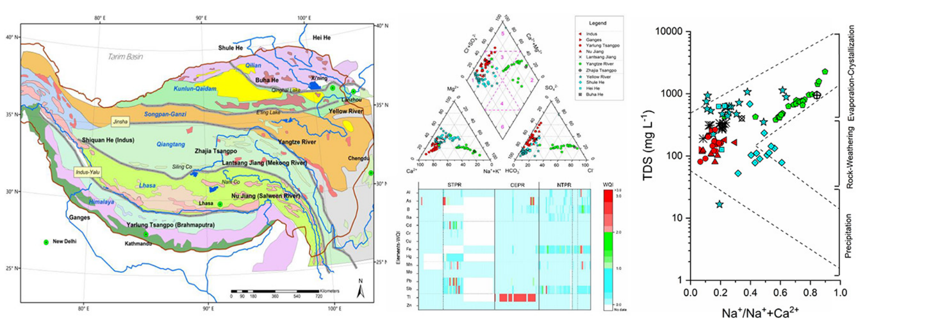
Bin Qu [a,b,c,d,*], Yulan Zhang [e], Shichang Kang [e,f], Mika Sillanpää [g]

[a] Yale-NUIST Center on Atmospheric Environment, International Joint Laboratory on Climate and Environment Change (ILCEC), Nanjing University of Information Science & Technology, Nanjing 210044, China
[b] Jiangsu Key Laboratory of Agricultural Meteorology, Nanjing University of Information Science & Technology, Nanjing 210044, China
[c] Key Laboratory of Meteorological Disaster, Ministry of Education (KLME)/Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters (CIC-FEMD), Nanjing University of Information Science & Technology, Nanjing 210044, China
[d] Department of Geography, National University of Singapore, Singapore 119260, Singapore
[e] State Key Laboratory of Cryospheric Sciences, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences, Lanzhou, Gansu 730000, China
[f] CAS Center for Excellence in Tibetan Plateau Earth Sciences, Chinese Academy of Sciences, Beijing 100085, China
[g] Laboratory of Green Chemistry, Lappeenranta University of Technology, Sammonkatu 12, Mikkeli 50130, Finland

HIGHLIGHTS

- Natural processes dominate the water chemistry in rivers of the Tibetan Plateau.
- Human activities have certain impacts on the water quality in the Tibetan rivers.
- Waters in parts of the rivers of the Tibetan Plateau are not safe for drinking.

GRAPHICAL ABSTRACT



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ABSTRACT

As the "Water Tower of Asia", rivers originating from the Tibetan Plateau provide water resources for more than one billion residents in both its local and surrounding areas. With respect to the essential role that this region plays in terms of water resources in Asia, we provide an overview of the mechanisms governing the water quality, including the major ions and trace elements release, in eleven rivers of the Tibetan Plateau. Overall, the rivers running on the Tibetan Plateau reflect an alkaline aquatic environment, with an average pH of 8.5; and the total dissolved solids (TDS, ~339 mg L^{-1}) are much higher than the global average value. Over 80% of the water ionic budget in the rivers of the plateau is comprised of Ca^{2+}, Mg^{2+}, HCO_3^- and SO_4^{2-}. The main mechanisms that control the river water chemistry on the Tibetan Plateau are natural processes and present a visible spatial heterogeneity. For instance, in rivers of the southern Tibetan Plateau, the water quality is mainly controlled by the rock-weathering, while rivers of the central and northern Tibetan Plateau are also largely affected by evaporation-crystallization processes. In general, most of the rivers on the Tibetan Plateau are uncontaminated and still in a pristine condition. However, it should be noted that due to the natural process such as rock-weathering and groundwater leaching, and anthropogenic activities such as urbanization and mining operations, the concentrations of several toxic elements (e.g., As, Cd, Pb, Mn, Hg and Tl) in some of the basins are higher than

* Corresponding author at: Yale-NUIST Center on Atmospheric Environment, International Joint Laboratory on Climate and Environment Change (ILCEC), Nanjing University of Information Science & Technology, Nanjing 210044, China.
E-mail address: bin.qu@nuist.edu.cn (B. Qu).

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the China national standard (GB) and the World Health Organization (WHO) guidelines for drinking water. With increasing anthropogenic activities on the plateau and changes in the river basins, it is necessary to conduct the long-term monitoring of the river water chemistry of this climate-sensitive and eco-fragile region.

1. Introduction

The Tibetan Plateau is the largest elevated plateau in Central and East Asia (Yao et al., 2012). It extends approximately 2500 km from east to west and 1000 km from north to south, covering an area of almost 2% of the Earth's land surface (Zhang et al., 2002). With an average elevation exceeding 4000 m above sea level (a.s.l.), the Tibetan Plateau has a peculiarly cold climate for its latitude (26°N–40°N) and has developed the largest glacial ice volume in the middle-latitude region of the Earth (Xu et al., 2008; Yao et al., 2012). Because of its high elevation and storage of a huge amount of solid water, the Tibetan Plateau serves as the "Water Tower of Asia", which feeds several large rivers (e.g., the Yarlung Tsangpo, the Yellow River, the Yangtze River) in Asia (Huang et al., 2008; Jiang and Huang, 2004). There are >1.4 billion people living in China, Pakistan, Nepal, Bangladesh, Vietnam, Thailand and India who depend on the waters originating from this region (Immerzeel et al., 2010); hence, the water chemistry in the Tibetan Plateau is an important issue for the residents in both its local and surrounding areas.

Chemical substances transported by rivers have been studied using geochemical budgets for more than one hundred years (Forel, 1892). The most abundant materials dissolved in rivers are ions that originate from terrestrial and atmospheric systems, which are mainly calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), bicarbonate (HCO_3^-), sulfate (SO_4^{2-}), chloride (Cl^-) and nitrate (NO_3^-) (Meybeck and Ragu, 1995). The ionic chemistry of rivers can provide essential information about both the processes affecting the continental surface (e.g., rock weathering, atmospheric precipitation, evaporation-crystallization) and the amount of natural material carried by river water bodies (Gibbs, 1970; Meybeck, 1982). In addition to studying dissolved elements such as lead (Pb), cadmium (Cd) and mercury (Hg), scientists can also investigate the influences of anthropogenic activities on the water chemistry in rivers (Huang et al., 2009; Meybeck and Helmer, 1989; Novotny and Olem, 1994).

Due the essential role that the Tibetan Plateau plays in terms of water resources in Asia, intensive efforts have been dedicated to studying the chemical compositions of its water in recent decades. It has been revealed that the contents of dissolved salts in the major rivers of the Tibetan Plateau were relatively high compared to those of waters from other parts of the world (Huang et al., 2009; Jiang et al., 2015; Pant et al., 2018; Qu et al., 2017; Zhang et al., 2015). Due to the anthropogenic impacts (i.e., mining activities) occurring on the southern plateau, some pollution has been identified in the Yarlung Tsangpo basin (Huang et al., 2010; Huang et al., 2009; Qu et al., 2017). Additionally, groundwater and sediment leaching have also affected the water quality in the rivers of the central and southern plateau (Huang et al., 2011; Jiang et al., 2009; Li et al., 2009). Most previous studies have focused on the large rivers in the eastern and southern plateau, which exhibit large runoff and flow transnationally. However, few studies have focused on the inland rivers in the arid northern and western plateau, despite the crucial role that they play in the livelihood of local residents. Considering that the rivers on the Tibetan Plateau are the headwaters of rivers that provide water for more than one billion people in High Asia and its surrounding areas, we have conducted a series of field trips over the past five years and compiled a dataset of the water chemical compositions in eleven main rivers, including those running on the remote plateau. This study will provide an overview of the mechanisms of water quality, including the major ions and trace element release, in the rivers of the Tibetan Plateau. Moreover, the potential risks for decreased water quality are addressed to provide a caution of the water quality of this essential water source region of Asia.

2. Materials and methods

2.1. Geology of the major river catchments

In this study, the water major ions and trace element transport in eleven rivers on the Tibetan Plateau were investigated (Fig. 1, Table 1). Most of these rivers originate from mountain glaciers and serve as the "Water Tower of Asia" in the uplifted region of the Tibetan Plateau. On a geological time scale, the Tibetan Plateau has formed six major east-west blocks (Yin and Harrison, 2000). These blocks include the Himalaya Block and Lhasa Block, which are located in the southern Tibetan Plateau; the Qiangtang Block, which is developed in the central region and extends from west to east in the plateau; and the Songpan-Ganzi Block, Kunlun-Qaidam Block and Qilian Block, which are located in the northern Tibetan Plateau (Fig. 1). The large rivers (e.g., the Indus, Yarlung Tsangpo and Yangtze rivers) on the Tibetan Plateau are usually developed along the sutures between the tectonic blocks forming the plateau. For example, the Indus and Yarlung Tsangpo rivers run along the Indus-Yalu suture between the Himalaya Block and the Lhasa Block, while the Yangtze River was developed along the Jinsha suture between the Lhasa Block and the Qiangtang Block. Different blocks exhibit different terranes reflecting the tectonic formation of the plateau, and the rivers of the Tibetan Plateau run over different bedrock (Fig. 1). Generally, the water chemistry of rivers in remote areas is largely dominated by the bedrock within the catchments. Due to the geological differences between the catchments, we group the eleven rivers into three river systems: 1) the South Tibetan Plateau River system (STPR), which includes the Indus, Ganges, Yarlung Tsangpo, Nujiang and Lantsang Jiang rivers, which run on the Himalaya Block and the Lhasa Block; 2) the Central and East Plateau River system (CEPR), which includes the Yangtze and Zhajia Tsangpo rivers, which run on the Qiangtang Block; and 3) the North Tibetan Plateau River system (NTPR), which includes the Yellow, Shule He, Hei He and Buha He rivers, which run on the blocks of the northern plateau. The selected rivers drain an area that is larger than half of the plateau (Fig. 1).

2.2. Data preparation

Because water chemistry can change rapidly due to natural and anthropogenic factors, the data collected in this study were obtained during the *Go-West Campaign* (Economy, 2002) in the year 2000 and afterwards (Table 1). A data set from over one hundred sampling sites (112 for ions and 134 for elements) was compiled from the eleven rivers on the Tibetan Plateau, and detailed data are listed in Tables S1 and S2.

The general parameters of water quality (e.g., water temperature, pH, EC, TDS, ORP and turbidity) in this study were measured with a Wagtech CP1000 portable water testing kit (Potalab®+, Palintest Ltd., England) during the sampling work in the field. The accuracy of the probes/sensors for water temperature is ±0.5 °C; that for pH is ±0.01; that for TDS is ±1% Full Scale +1 digit. Due to the major role that they play in the area's water resources, the rivers of the Tibetan Plateau were investigated for their water chemistry. Water samples were collected against the river current at a depth of approximately 10 cm; they were then filtered by polypropylene membrane (0.2 μm) and stored in polypropylene bottles (15 mL). The samples for elemental

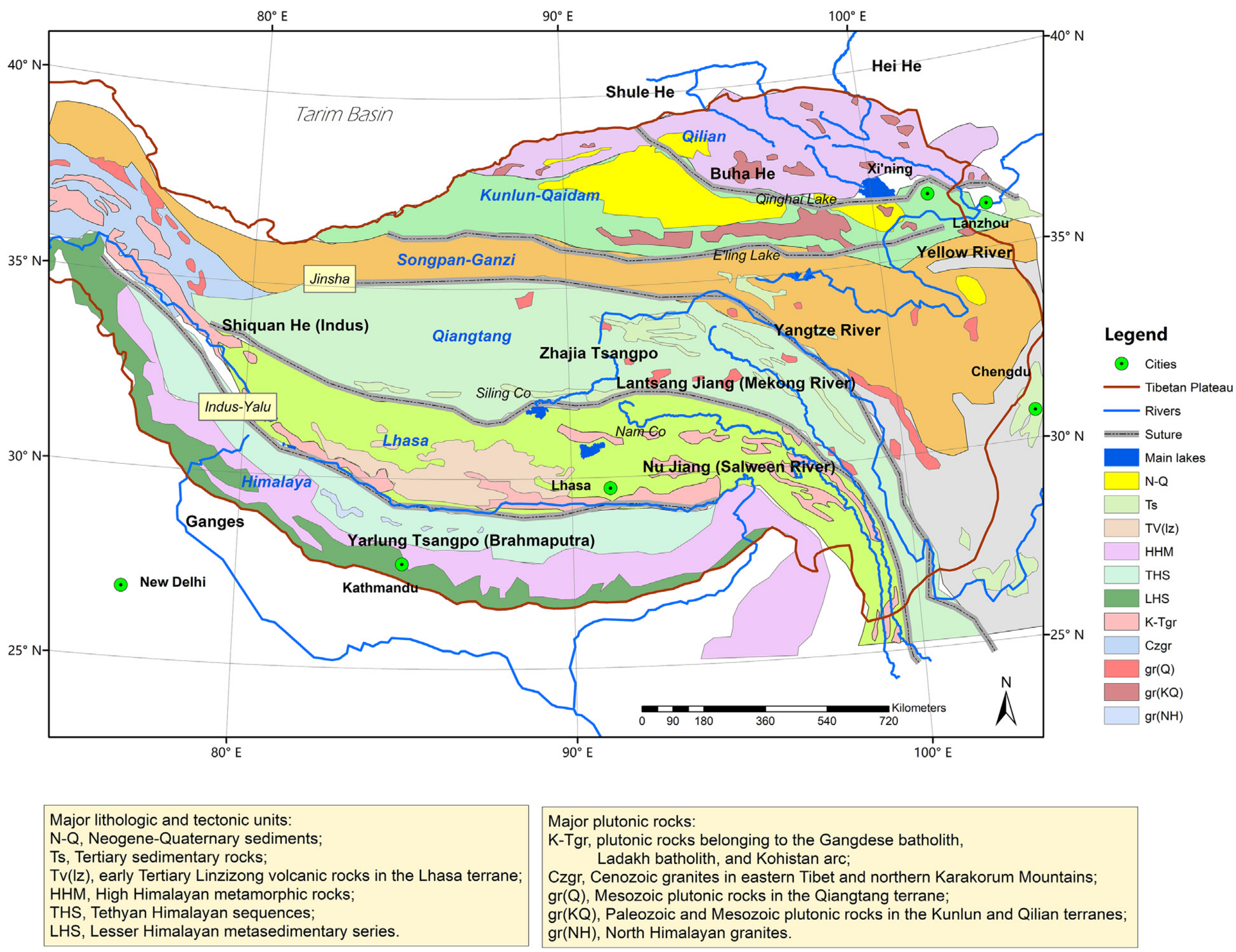

Fig. 1. Map of the studied rivers and geology of the river catchments on the Tibetan Plateau. The geological map was extracted from Yin and Harrison (2000) and generated by ArcGIS 10.2 (ESRI®).

measurements were acidified with HNO_3^- (ultrapure, pH < 2) and stored at 4 °C until laboratory analysis. The concentrations of cations (Na^+, K^+, Ca^{2+}, Mg^{2+} and NH_4^+,) and anions (Cl^-, NO_3^-, SO_4^{2-} and F^-) in water samples were detected by Dionex Ion Chromatography System (ICS) 2000 and 2500 analyses in the State Key Laboratory of Cryospheric Sciences, Chinese Academy of Sciences. HCO_3^- contents were determined using the contents of other ions with the charge balance method, which has been proven to be reliable within 10% in rivers draining on the Tibetan Plateau (Galy and France-Lanord, 1999; Wu et al., 2008). Element samples were measured within 30 days of collection using an Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES). The Hg contents in the water samples were analyzed by cold vapor atomic fluorescence spectroscopy (CVAFS) based on the US EPA Method 1631 (version E) (US EPA, 2002). We employed an Analytik-Jena Hg analyzer (Analytik-Jena Corporation Inc., Jena, Germany) to measure the concentrations of Hg in the waters, and these measurements were conducted in a metal-free class 100 ultra-clean laboratory in the Institute of Tibetan Plateau Research, Chinese Academy of Sciences (Huang et al., 2012).

2.3. Assessment of water quality: comprehensive pollution index

A comprehensive pollution index, namely, the water quality index (WQI), was used to evaluate the combined effects of several toxic

Table 1
Length, drainage area and annual runoff of the selected rivers on the Tibetan Plateau[a].

	River	Length (km)	Drainage area (km^2)	Annual runoff (m^3)	Data source
STPR	Shiquan River (Indus)	430	27,000	7.00E+08	This study
	Ganges[b]	250	19,600	8.93E+09	This study
	Yarlung Tsangpo (Brahmaputra)	2057	240,480	1.65E+11	(Qu et al., 2017)
	Nu Jiang (Salween)	1393	103,330	3.59E+10	(Huang et al., 2009)
	Lantsang Jiang (Mekong)	1308	84,220	2.93E+10	(Huang et al., 2009)
CEPR	Yangtze River	1206	158,400	1.79E+10	(Qu et al., 2015)
	Zhajia Tsangpo	463	12,793	8.86E+08	This study
NTPR	Yellow River	1694	152,500	2.07E+10	This study
	Shule He	736	27,730	1.37E+09	This study
	Hei He	488	35,600	1.65E+09	This study
	Buha He	386	14,384	8.18E+08	This study

[a] Data from MWR (2015) except as noted;
[b] Data at Rishikesh station from Khan et al. (2018).

elements considered detrimental to aquatic ecosystems. The WQI is estimated as follows:

$$WQI = \frac{1}{n}\sum_{i=1}^{n} A_i = \frac{1}{n}\sum_{i=1}^{n} C_i/Q_i$$

where C_i is the concentration of toxic elements examined in the water sample and Q_i is the limit value of the water quality standard of the elements. In addition, the China national standard (GB) (MOH&SAC, 2006) and the World Health Organization (WHO, 2011) for surface water were used as limit values, and the lower limit of water quality standard of the toxic elements from the GB and the WHO were chosen if the standards are different from the two guidelines. The comprehensive pollution index consists of four classes. Class 1 (practically unpolluted): WQI≤1; Class 2 (slightly polluted): 1<WQI≤2; Class 3 (moderately polluted): 2<WQI≤3; and Class 4 (heavily polluted): WQI>3 (Ma et al., 2014).

3. Major ions in rivers of the Tibetan Plateau

The rocks and soils on the Tibetan Plateau, which is the youngest plateau in the world, are experiencing intensive erosion within their catchments. It was proposed that due to the uplift of the continent and steep slopes, the sediment derived from rivers in the Himalayan region and its environs accounted for almost half of the world's total load to deltas (Meade, 1996). Due to the lack of precipitation and strong evaporation in most parts of the plateau, the rivers running on the Tibetan Plateau present much higher ion concentrations than the rivers in other parts of the world (Fig. 2). The total dissolved solids (TDS) in the rivers of the Tibetan Plateau ranged from 137 mg L^{-1} to over 900 mg L^{-1}, with an average of ~380 mg L^{-1}; this value is almost four times higher than the world mean level (Huang et al., 2009; Wetzel, 1975). With different hydrological (e.g., unevenly distributed precipitation and runoff) and geological conditions, the concentrations of major ions in river waters, such as Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, SO_4^{2-} and HCO_3^-, presented a clear spatial distribution on the Tibetan Plateau (Huang et al., 2009; Ramanathan, 2007).

3.1. Spatial variations in the major ions of rivers on the Tibetan Plateau

The rivers of the STPR (Indus, Ganges, Yarlung Tsangpo Nu Jiang and Lantsang Jiang) are distributed on the southern Tibetan Plateau along the Himalaya and the Lhasa Block (Fig. 1). Most of the rivers in this region presented relatively homogeneous ion compositions (Fig. 2). The TDS in the rivers of the STPR ranged from 66 mg L^{-1} to 343 mg L^{-1} and mainly comprised Ca^{2+}, Mg^{2+}, HCO_3^- and SO_4^{2-} (Fig. 2). The concentrations of most major ions (e.g., SO_4^{2-}) in the STPR are higher than the world's average levels, while the Cl^- contents are fairly low (Fig. 2). To date, most studies have claimed that the Cl^- transported in the rivers of the world is contributed by cyclic salts and is expected to decrease with increasing distance from the sea, except where there are sedimentary halite deposits (Stallard and Edmond, 1981). The water chemistry in the rivers of the Himalaya region is inevitably affected by precipitation, with the major contribution of rainfall occurring during the monsoon season (Liu, 1999). However, as the roof of the world, the Himalaya blocks most of the water vapor flux from the Indian Ocean to the Tibetan Plateau and reduces the atmospheric transported Cl^- in the southern Tibetan Plateau (Shi and Yang, 1985). In addition to the barely halite deposits along the Himalaya (Yin and Harrison, 2000), the concentrations of Cl^- in rivers of the STPR are lower than those in most other rivers in the world.

The rivers of the CEPR (the Yangtze River and the Zhajia Tsangpo) run within the central and eastern Tibetan Plateau (Fig. 1). The TDS in the rivers of the CEPR ranged from 382 to 2239 mg L^{-1}, with an average value of 736 mg L^{-1}. The contents of ions in the rivers of this region were much higher than those in the other rivers on the Tibetan Plateau and the world average level (Fig. 2). For instance, the average concentrations of SO_4^{2-} in the rivers of the Zhajia Tsangpo and upstream of the Yangtze River were ~122 mg L^{-1}, i.e., almost 15 times higher than the world median level. Elevated concentrations of Na^+ and Cl^- were also found to be the dominant ions comprising the high TDS in the rivers of the CEPR, especially in the source upstream of the Yangtze River (Tuotuo He). The average concentrations of Na^+ and Cl^- in the CEPR were 150 mg L^{-1} and 223 mg L^{-1}, respectively, which are >20 times higher than those observed in the other rivers on the Tibetan Plateau.

The rivers of the NTPR (the Yellow River, the Shule He, the Hei He and the Buha He) are mainly distributed on the northern Tibetan Plateau. Similar to the other rivers of the CEPR and STPR, their major ion concentrations are also higher than those in other rivers around the world (Fig. 2). Due to the relatively arid climate within their catchments, the TDS in the rivers of the NTPR are generally higher than those in the rivers of the STPR. Ca^{2+}, Mg^{2+}, HCO_3^- and SO_4^{2-} are the dominant components in the rivers of the NTPR. Notably, in the upstream region of the Yellow River, the HCO_3^- concentrations were as high as 385 mg L^{-1}, which is the highest value observed among the rivers of the Tibetan Plateau and >7 times higher than the global median value.

3.2. Characterization of major ion compositions in rivers of the Tibetan Plateau

The milli-equivalent percentages (meq%) of major ions are plotted on a Piper (1944) diagram and further projected onto the central diamond field to evaluate the hydrogeochemical facies and types of these

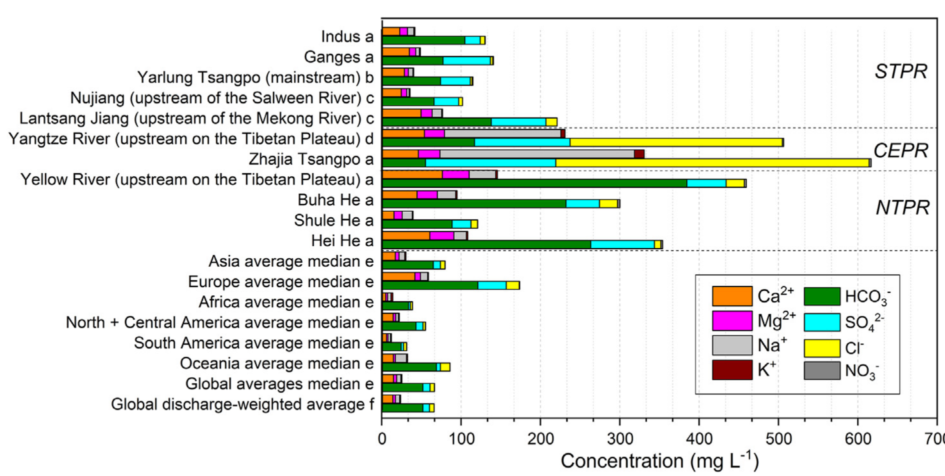

Fig. 2. Average concentrations of major ions in the rivers on the Tibetan Plateau and around the world. Data source: a. this study; b. Qu et al. (2017); c. Huang et al. (2009); d. Qu et al. (2015); e. Meybeck and Ragu (2012); f. Meybeck and Helmer (1989).

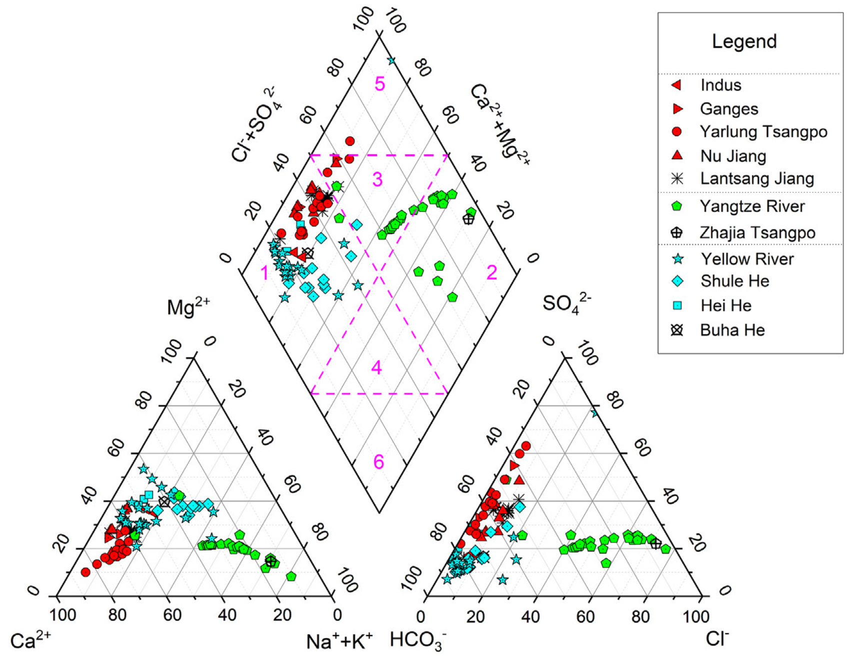

Fig. 3. Piper diagram of the relative equivalent proportions of major ions for the river waters of the Tibetan Plateau.

water samples (Fig. 3 and Table S1). On the cation plot, most of the samples from the STPR (the Indus, the Ganges, the Yarung Tsangpo, the Nujiang and the Lantsang Jiang, in red symbols, Fig. 3) lie in the lower left corner, indicating the dominance of calcium in the river water. The samples from the rivers of the NTPR (the Yellow River, the Shule He, the Buha He and the Hei He, in green symbols, Fig. 3) showed relatively higher concentrations of Mg^{2+}. The anion diagram shows that most of the water samples from the STPR and NTPR fall in the lower left corner, near the HCO_3^- apex (Fig. 3), again signifying their carbonate-dominated lithology. Nevertheless, the majority of the water samples from the rivers of the Zhajia Tsangpo and the source region of the Yangtze River in the CEPR are more enriched with Na^+ and Cl^- compared to the downstream samples, demonstrating the possible influence of evaporites spreading though the catchments.

The results plotted in the central diamond field of the Piper diagram show the overall characteristics of the river water chemistry in these samples: the dominance of alkaline earth elements (Ca^{2+} and Mg^{2+}) over alkaline elements (Na^+ and K^+) and that of weak acids (HCO_3^-) over strong acids (Cl^- and SO_4^{2-}). Generally, six sub-fields can be identified in the diamond Piper diagram: 1) Ca-HCO_3, 2) Na-Cl, 3) mixed Ca-Mg-Cl, 4) mixed Ca-Na-HCO_3, 5) Ca-Cl, and 6) Na-HCO_3 (Khadka and Ramanathan, 2013). The hydrogeochemical results showed that most rivers of the Tibetan Plateau can be generally classified into three types (1, 2, and 3, Fig. 3), thus indicating their carbonate-dominant lithology. For example, ~70% of the water samples belong to the Ca-HCO_3 type, ~14% belong to the Na-Cl type (mostly from the Zhajia Tsangpo and the source region of the Yangtze River of the CEPR), and ~15% belong to the Ca-Mg-Cl type (Table S1). The elevated concentrations of Cl^- upstream of the Yangtze River are likely attributed to the widespread evaporites and groundwater present in the central-east Tibetan Plateau (Cao, 2013; Qu et al., 2015).

In general, significant differences between the hydrogeochemical facies of rivers were observed over the entire Tibetan Plateau, which clearly indicates that most of the major ions are of natural origin. For instance, the cations and anions in the rivers of the STPR and NTPR plot in the Ca^{2+}, Mg^{2+} and HCO_3^-, SO_4^{2-} corners, respectively, suggesting a Ca-Mg-HCO_3-SO_4 river water type in these regions. Most of the samples in the rivers of the Zhajia Tsangpo and the source area of the Yangtze River plot as Na-Cl types. Moreover, flowing downstream, the concentrations of Na^+ and Cl^- in the Yangtze River decrease, and its river water changes to the Ca-Mg-Cl type downstream of the plateau (Fig. 3 and Table S1).

3.3. Major sources and controlling factors of the ions in the rivers of the Tibetan Plateau

The mechanisms controlling the water chemistry in rivers include natural processes (e.g., rock weathering, atmospheric precipitation, evaporation-crystallization, groundwater leaching) and anthropogenic activities (e.g., agricultural and industrial activities, urbanization) (Gibbs, 1970; Meybeck, 2003). Changes in climate and land use in the headwater region of the Tibetan Plateau may introduce variations in the chemical composition of the downstream river water (Chen et al., 2002). Therefore, when identifying the major sources and controlling factors of water chemistry, it is necessary to differentiate human inputs from natural inputs to the river basins (Meybeck, 2003; Tong et al., 2016). For instance, in the Yarlung Tsangpo, Yellow, Yangtze and Heihe rivers, which are characterized by relatively intense human activities, such as agriculture, industry, and urbanization (Tong et al., 2016), their concentrations of NO_3^- were higher than those in the other rivers on the Tibetan Plateau (Fig. 2 and Table S1). These results are also supported by the absence of correlations between NO_3^- and other hydrogeochemical variables (Huang et al., 2009; Pant et al., 2018).

The natural mechanisms that control the hydrogeochemistry of surface water can be inferred from the Gibbs diagram, which was used to identify three endmembers (Fig. 4). (1) Endmember 1 has low TDS (<10 mg L^{-1}) concentrations and high weight ratios of $Na^+/(Na^+ + Ca^{2+})$ and $Cl^-/(Cl^- + HCO_3^-)$ (0.5–1) and is located in the lower right corner of the schematic diagram; (2) Endmember 2 has medium TDS (70–300 mg L^{-1}) concentrations and low weight ratios of $Na^+/(Na^+ + Ca^{2+})$ and $Cl^-/(Cl^- + HCO_3^-)$ (<0.5) and is located to the left of center and demonstrates the predominance of rock weathering; (3) Endmember 3 has high TDS (>300 mg L^{-1}) concentrations and high weight ratios of $Na^+/(Na^+ + Ca^{2+})$ and $Cl^-/(Cl^- + HCO_3^-)$ (0.5–1) and is located in the upper right corner, indicating the dominance of evaporite dissolution (Gibbs, 1970; Pant et al., 2018).

Plotting the samples collected from the rivers of the Tibetan Plateau on the Gibbs diagram (Fig. 4) illustrates that rock weathering and

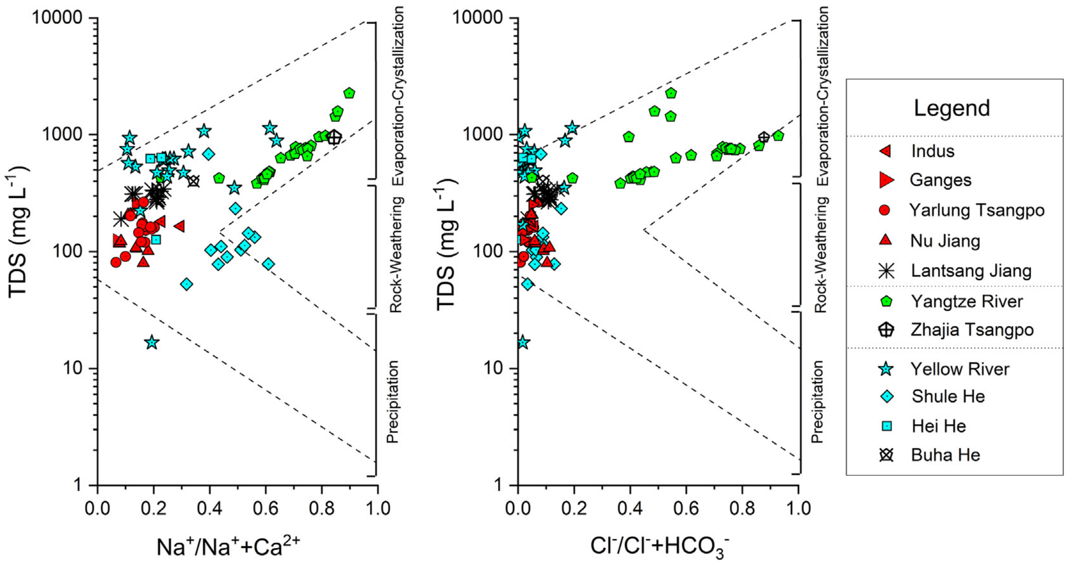

Fig. 4. Gibbs diagram (weight ratios) of major ion compositions in the rivers of the Tibetan Plateau. Detailed data are listed in Table S1.

evaporation-crystallization are the dominant sources of ions in the rivers of the plateau. For instance, most of the rivers in the STPR and NTPR were characterized by relatively moderate TDS and low ratios of $Na^+/(Na^++Ca^{2+})$ and $Cl^-/(Cl^- + HCO_3^-)$, thus indicating the dominance of rock weathering (Fig. 4). However, the higher concentrations of TDS and wide ranges of $Na^+/(Na^+ + Ca^{2+})$ and $Cl^-/(Cl^- + HCO_3^-)$ ratios in the rivers of the CEPR (except the Lantsang Jiang), compared to those of the other rivers on the Tibetan Plateau, provided evidence that evaporation-crystallization processes were a major controlling factor of the hydrogeochemistry in this region (Fig. 4). In other words, the ion sources in the river water of the Tibetan Plateau are mainly derived from bedrock (e.g., Ca-, Na- and K-mineral weathering and dissolution) rather than precipitation. To identify the origins of the major ions produced by the chemical weathering of bedrock, the ionic ratios of the water samples obtained from the rivers of the Tibetan Plateau are shown in Table 2.

The water samples from the rivers of the STPR plot towards the carbonate endmember on the Mixing diagram (Fig. 5), suggesting that carbonate weathering plays a crucial role in the major ion chemistry of the southern Tibetan Plateau. The high mean molar ratios of $(Ca^{2+} + Mg^{2+})/(Na^+ + K^+)$ (~9.11) and $HCO_3^-/(Na^+ + K^+)$ (~6.02) also confirm that the basin is dominated by the weathering of calcite and dolomite minerals (Table 2). Most of the waters from the rivers of the CEPR are located near the evaporite and silicate endmembers, indicating that the sampling sites likely located near environments characterized by evaporite dissolution and high degrees of silicate weathering (Fig. 4). Most of the waters from the rivers of the NTPR are located near the carbonate and silicate endmembers. Generally, carbonates have higher solubilities (12–40 times) than silicates and are more susceptible to weathering under natural conditions (Meybeck, 1987). In the present study, the relatively high grand mean ratio of $Ca^{2+}+Mg^{2+}/Tz^+$ (~0.80) and low ratio of Na^++K^+/Tz^+ (~0.20) in the rivers of the Tibetan Plateau suggest that the weathering of silicates and evaporates is less intense than that of carbonates in the rivers of the NTPR (Table 2). This conclusion is also supported by the absence of significant correlations between Si and Na^+, K^+, Ca^{2+} and Mg^{2+} in the rivers of the southern Tibetan Plateau (Huang et al., 2011; Huang et al., 2009).

Furthermore, a proton source is required for the chemical weathering of carbonate rocks. Thus, the relative importance of two proton-producing reactions, i.e., carbonation and sulfide oxidation, can be explained on the basis of $HCO_3^-/(HCO_3^- + SO_4^{2-})$ ratios (C-ratio). If the C-ratio is <0.50, the coupled chemical reactions of both carbonate dissolution and sulfide oxidation are indicated, whereas if the ratio is close to 1, exclusively carbonation reactions occur, and protons are derived from the dissociation of CO_2 from atmospheric inputs (Pant et al., 2018). In the present study, the grand mean C-ratio of the rivers of the Tibetan Plateau was 0.65 (>0.50), indicating the importance of carbonate and CO_2 dissolution in their proton-producing mechanisms. Additionally, the high ratio of Ca^{2+}/SO_4^{2-} (~2.2) observed in the rivers of the Tibetan Plateau further confirms that H_2SO_4 does not replace

Table 2
Ionic equivalent ratios of various hydrogeochemical attributes (used for this study) from the rivers of the Tibetan Plateau.

	Rivers	$Ca^{2+}+Mg^{2+}/Tz^+$	Na^++K^+/Tz^+	$HCO_3^-/HCO_3^- + SO_4^{2-}$	Ca^{2+}/SO_4^{2-}	Na^+/Cl^-	K^+/Cl^-	$Ca^{2+}+Mg^{2+}/Na^+ + K^+$	$HCO_3^-/Na^+ + K^+$
STPR	Indus	0.84	0.16	0.81	2.75	2.05	0.22	5.20	4.64
	Ganges	0.92	0.08	0.55	1.65	2.14	0.57	11.63	6.93
	Yarlung Tsangpo	0.88	0.12	0.61	2.07	4.03	0.39	7.77	5.20
	Nu Jiang	0.91	0.09	0.63	2.03	1.04	0.15	11.39	7.33
	Lantsang Jiang	0.87	0.13	0.62	1.85	1.24	0.11	7.48	4.87
	Grand mean-STPR	0.88	0.12	0.62	2.00	2.24	0.24	8.58	5.64
CEPR	Yangtze River	0.48	0.52	0.49	1.20	1.09	0.03	1.13	0.61
	Zhajia Tsangpo	0.29	0.71	0.21	0.67	0.96	0.03	1.03	2.03
	Grand mean-CEPR	0.48	0.52	0.48	1.18	1.08	0.03	1.11	0.59
NTPR	Yellow River	0.83	0.17	0.86	3.98	3.41	0.20	6.94	6.45
	Buha He	0.80	0.20	0.81	2.49	1.57	0.07	1.07	2.07
	Hei He	0.88	0.12	0.74	1.96	5.95	0.37	7.65	6.18
	Shule He	0.72	0.28	0.79	2.17	2.72	0.10	2.75	2.69
	Grand mean-NTPR	0.80	0.20	0.83	3.21	3.35	0.18	5.60	5.17
	Grand mean-Tibetan Plateau	0.74	0.26	0.65	2.18	2.30	0.16	5.50	4.08

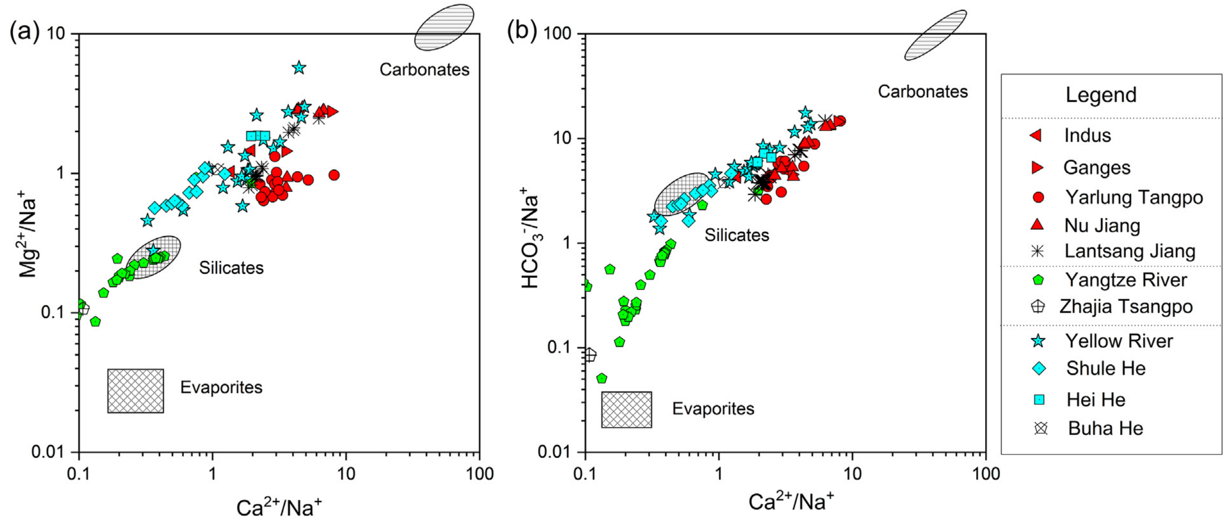

Fig. 5. Mixing diagram of the Na-normalized molar ratios of (a) Ca^{2+} versus Mg^{2+} and (b) Ca^{2+} versus HCO_3^- in the rivers of the Tibetan Plateau. The data for the three endmembers, i.e., carbonates, silicates and evaporites, are obtained from Gaillardet et al. (1999).

H_2CO_3 as a major source of protons for rock weathering, as has also been discussed elsewhere (Singh et al., 2014). The spatiotemporal variability in the major ions chemistry of the river basin is illustrated by the mixing diagrams of the Na-normalized molar ratios (μeq) of Ca^{2+} versus HCO_3^- and Ca^{2+} versus Mg^{2+}. The main feature of the water derived from the semi-arid/arid environments of the river basin is the depletion of HCO_3^- compared to the concentrations of Ca^{2+} and Mg^{2+} (Gaillardet et al., 1999).

In addition to rock weathering, which controls the water ionic chemistry in the rivers of the Tibetan Plateau, groundwater also plays a crucial role in the ion budget, as numerous geothermal springs and mineral-rich alpine lakes are distributed over the central and southern Tibetan Plateau (Liu, 1999; Zhao Ping, 2000). For instance, in the sites near Yangbajing of the Yarlung Tsangpo, the average Cl^- concentration (>3 mg L^{-1}, Table S1) was higher than the average content of the entire Yarlung Tsangpo basin (~2 mg L^{-1}, Table S1). This is mainly due to the existence of chloride-rich geothermal springs in the Yangbajing area, where the concentrations of Cl^- reach as high as 562.1 mg L^{-1} (Zhao Ping, 2000). In addition, the mean molar ratios of Na^+/Cl^- (~2.3) and K^+/Cl^- (~0.16) in the rivers of the Tibetan Plateau are much higher than their corresponding ratios in marine water ($Na^+/Cl^- = 0.86$ and $K^+/Cl^- = 0.02$) (Table 2). This fact reveals that the contribution of atmospheric inputs is very low in the rivers within the plateau (Singh et al., 2014). Given that the Na^+/Cl^- ratio is larger than 1 (Fig. 2 and Table S1) and that silicate weathering is less intense, the Na^+ in these samples is believed to be released mainly from local Na-bearing salts (e.g., marine evaporates that are spread throughout the plateau, Fig. 1) rather than from sea salts (Meybeck, 1987; Yin and Harrison, 2000).

In summary, most of the ionic correlations indicate that the major ionic chemistry in the rivers of the Tibetan Plateau is mainly regulated by carbonate weathering and evaporite dissolution, followed by some contributions from silicate weathering and ground water leaching. And spatial heterogeneity is visible in the rivers running on the different tectonic blocks/bedrocks of the Tibetan Plateau. With respect to the ionic compositions and their ratios, the high concentrations of Ca^{2+}, Mg^{2+} and HCO_3^- in the rivers of the STPR and NTPR are most likely derived from the weathering of carbonates and silicates. The Na^+ and K^+ contents in the rivers of the Tibetan Plateau can be identified as originating from the dissolution of evaporites and the weathering of silicate minerals. Furthermore, the abundant Na^+, Cl^- and SO_4^{2-} contents in the rivers of the CEPR are derived from the dissolution of evaporates (e.g., halite, sulfide oxidation and soft sulphate minerals such as gypsum) (Galy and France-Lanord, 1999). Whereas in some parts of the rivers (e.g., Yangbajing to the Yarlung Tsangpo) on the plateau, the ionic compositions (i.e., Cl^-) are significantly affected by the ground water.

4. Dissolved trace elements and water quality assessment in rivers of the Tibetan Plateau

4.1. Dissolved trace elements in rivers of the Tibetan Plateau

Trace elements are characterized by concentrations of lower than 1 mg L^{-1} in natural waters such as rivers and lakes (Gaillardet et al., 2003). Despite their low concentrations in natural waters, trace elements play essential roles in human health (WHO, 2011). Here, we will discuss the characteristics and possible sources of trace elements in the rivers of the Tibetan Plateau, as well as their potential risks to the residents of this region. The concentrations of 20 elements measured in the rivers of the Tibetan Plateau are listed in Table 3.

Generally, the trace element contents in the river waters on the Tibetan Plateau varied with differences in lithological weathering, water alkalinity, groundwater supply, and the influence of anthropogenic activities (Huang et al., 2009; Qu et al., 2015; Qu et al., 2017; Zhang et al., 2015). As a result of the mountain climate due to the high altitude, soils are weakly developed on the Tibetan Plateau, and the contents of dissolved elements in most rivers (except the Yarlung Tsangpo) in the STPR are lower than those in most other rivers around the world (Qu et al., 2017). In particular, in the source area of the Ganges, the concentrations of most elements were so low that they were not detected. It is widely accepted that the levels of trace elements in global surface water are strongly governed by alkalinity due to its influence on solubility (Dupré et al., 1996; Gaillardet et al., 2003). The river waters on the Tibetan Plateau have relatively high alkalinity, with pH values of 7.9–9.3 (Table S1). Elevated pH values usually reflect the low solubility of elements in river water, especially for the metal elements (e.g., Fe), which are more likely to be deposited in sediment than dissolved in waters in an alkaline aquatic environment. Despite the low concentrations of most of the studied elements in the river waters of the southern Tibetan Plateau (STPR, Table 2), trace elements such as boron (B), barium (Ba), titanium (Ti), rubidium (Rb), strontium (Sr) and uranium (U) presented elevated concentrations in the rivers of the NTPR (Tables 3, S2). Considering the relatively low degree of weathering of the bedrock within the catchment, this phenomenon is mainly due to the high amount of evaporation occurring in this arid region. High concentrations of arsenic (As) were observed in the rivers of the Yarlung

Table 3
Concentrations of selected elements in the rivers on the Tibetan Plateau.

		Al μg L^{-1}	As μg L^{-1}	B μg L^{-1}	Ba μg L^{-1}	Cd μg L^{-1}	Cr μg L^{-1}	Cu μg L^{-1}	Fe μg L^{-1}	Hg μg L^{-1}	Li μg L^{-1}
STPR	Indus	<8.8	13.7	10.4	5.6	N.D.	2	0.4	N.D.	–	134.7
	Ganges	<0.2	N.D.	<0.04	N.D.	N.D.	N.D.	N.D.	0.4	–	N.D.
	Yarlung Tsangpo	20.6	10.5	–	12	1	2.7	1.4	–	1.46–4.99	33
	Nujiang (Salween)	20.7	–	–	–	–	–	–	19.7	–	10.1
	Lantsang Jiang (Mekong)	14.75	–	–	–	–	–	–	2.55	–	17.55
CEPR	Yangtze River	–	N.D.	66.3	46.9	–	–	–	–	1.7	–
	Zhajia Tsangpo	38.8	5.7	–	43.8	N.D.	2.4	0.8	–	–	454.4
NTPR	Yellow River	10.2	1.2	68.5	90.9	<0.007	1.8	1	154.1	–	8.2
	Buha He	18.2	0.9	135.1	110.4	N.D.	2	1.4	197.5	–	11.2
	Hei He	10.5	0.8	78.4	78.1	<0.01	1.2	0.9	186.6	–	8.4
	Shule He	31.4	1.4	234.5	52.7	<0.004	2	0.8	198.1	–	20.5
World-avg.		32	0.62	10.2	23	0.08	0.7	1.48	66	–	1.84

		Mn μg L^{-1}	Mo μg L^{-1}	Pb μg L^{-1}	Rb μg L^{-1}	Sb μg L^{-1}	Sr μg L^{-1}	Ti μg L^{-1}	Tl μg L^{-1}	U μg L^{-1}	Zn μg L^{-1}
STPR	Indus	0.4	N.D.	N.D.	1.9	N.D.	89.8	0.3	<0.005	1.6	0.2
	Ganges	N.D.	N.D.	N.D.	0.1	N.D.	0.1	<0.03	<0.002	N.D.	N.D.
	Yarlung Tsangpo	12.8	1.2	5.6	3.4	3.4	149.5	7.8	<0.007	2.2	9.8
	Nu Jiang (Salween)	3.8	1.3	–	–	–	–	0.75	–	–	5.2
	Lantsang Jiang (Mekong)	1.7	1.2	–	–	–	–	0.55	–	–	4.3
CEPR	Yangtze River	–	–	N.D.	605	–	755	1.9	5.3	–	–
	Zhajia Tsangpo	0.3	–	0	23.4	–	1295	1.1	–	1.7	0.4
NTPR	Yellow River	3.3	0.6	0.1	0.6	1.4	276.9	0.9	0	1.8	4.4
	Buha He	3.8	0.6	<0.05	0.6	1	364.9	1.1	N.D.	1.6	3.7
	Hei He	2.6	1.4	<0.02	1.4	0.2	366.1	0.7	<0.004	3.2	1.7
	Shule He	6.3	2.3	0.1	0.9	1.9	433.6	1.7	<0.01	3.6	1.5
World-avg.		34	0.4	0.08	1.63	0.07	60	0.49	–	0.37	0.6

* – means no data available; N.D. means not detected, indicating that concentrations are below the detection limit of the instrument; detailed data are listed in Table S2; world average data are from (Gaillardet et al., 2003).

Tsangpo (10.5 μg L^{-1}) and the source area of the Indus (13.7 μg L^{-1}). It was found that most high concentrations of As appeared in the rivers of the central and southern Tibetan Plateau, where numerous arsenic-rich springs were distributed (Huang et al., 2011; Li et al., 2014; Zhao Ping, 2000). Therefore, groundwater is also an important source of elements in the rivers of the Tibetan Plateau. Furthermore, with the existence of lithium-rich saline lakes in the central plateau (Zhao, 2003), it was found that the concentrations of lithium (Li) was fairly high in the Zhajia Tsangpo (454.4 μg L^{-1}). Additionally, due to the existence of Rb mineral-rich brine and saline lakes (Sun et al., 1994), the average Rb concentration upstream of the Yangtze River was found to be as high as 654.6 μg L^{-1}, which is two orders of magnitude higher than the median value (1.6 μg/L) of world stream water (Gaillardet et al., 2003).

It should be noted that the concentrations of heavy metals such as Cd, Cr, Pb and Zn in the river of the Yarlung Tsangpo are much higher (3–10 times) than those of the other rivers on the Tibetan Plateau and around the world (Table 3). The elevated concentrations of heavy metals observed in the regional riverine element budgets of the rivers of the Tibetan Plateau usually suggest an anthropogenic contribution. With abundant mineral deposits (e.g., copper ore and lead zinc ore) distributed in the Yarlung Tsangpo basin (Qu et al., 2007; She et al., 2005), mineral activities have polluted its rivers (Huang et al., 2010). Furthermore, municipal waste from many parts of the plateau has been dumped into the rivers without adequate management for many decades, and solid waste is usually piled in landfills near towns or city settlements (Jiang et al., 2009). The influence of the discharge of municipal wastewater has been detected based on the elevated concentrations of dissolved nitrogen in the rivers of the Tibetan Plateau (Huang et al., 2011; Qu et al., 2017). Therefore, municipal waste discharge is also a threat to the river waters on the Tibetan Plateau (Huang et al., 2011; Huang et al., 2009; Qu et al., 2017). For instance, the nitrogen concentrations in the rivers that flow through cities are higher than those of most other rivers running on the plateau (Table S1), such as the Yellow River near Lanzhou (5.5 mg L^{-1}), the Yarlung Tsangpo near Shigatse (2.8 mg L^{-1}) and Hei He near Zhangye (3.8 mg L^{-1}). In addition to the direct discharge of pollutants from anthropogenic activities, such as mining operations and municipal wastewater, trace elements can also be transported by the atmosphere in the form of wind-blown soil particles, fossil fuels (e.g., coal and oil) and biomass burning (Gaillardet et al., 2003). The atmospheric input of trace elements can be significant on both local and global scales (Guo et al., 2015; Pirrone et al., 2010; Tripathee et al., 2014; Zhang et al., 2012). However, because aerosols can travel over long distances in the atmosphere, it is difficult to determine the exact quantity of atmospheric source elements in the river waters of the plateau based on present-day sampling. Further studies are needed to address the impact of anthropogenic aerosols on the release of elements from the rivers of the Tibetan Plateau.

4.2. Water quality assessment of rivers of the Tibetan Plateau

Given that these rivers represent major water resources for regional inhabitants, an assessment of their water quality was conducted. The standards used for the assessment in this work included the drinking water guidelines from GB (MOH&SAC, 2006) and the WHO (WHO, 2011) (Table S3).

The rivers on the Tibetan Plateau, especially the main streams, were found to be highly turbid (turbidity >500 NTU) during the high-flow period (Huang et al., 2009). The waters in most rivers on the Tibetan Plateau were characterized by high alkalinity. According to the drinking water guidelines established by the WHO and GB, the concentrations of most ions and elements in the rivers of the southern Tibetan Plateau, that is, the STPR (e.g., Indus, Ganges, Yarlung Tsangpo and Nu Jiang), were within the maximum desirable limits and safe for drinking. In the rivers of the CEPR and NTPR, the concentrations of ions, particularly

Cl^-, were much higher than those of the guidelines of both the WHO and GB. For instance, the high concentrations of Cl^- in other rivers of the STPR and NTPR, such as Buhe He and the Yellow River, were 3–5 times higher than those of the WHO and GB guidelines for drinking water. Furthermore, due to the numerous saline lakes and springs spread throughout the central Tibetan Plateau, the average Cl^- concentrations in the rivers of Zhajia Tsangpo and upstream of the Yangtze River were as high as 394 mg L^{-1} and 267.5 mg L^{-1}, respectively, which were >40 times higher than the permissible drinking water threshold. In natural water, high concentrations of Cl^- usually mean high levels of Na^+. We found that the Na^+ contents in the Zhajia Tsangpo and source area of the Yangtze River exceeded 300 mg L^{-1}, which were >70 times higher than those of the WHO and GB guidelines.

In addition to the essential role that ions play in the water quality assessment of the rivers of the Tibetan Plateau, elements, especially heavy metal and toxic elements such as As and Hg, also play an important role in human health. Similar to major ions, trace elements in the surface water of rivers also originate from natural processes (e.g., rock weathering and precipitation) and anthropogenic activities (e.g., mining). Most of the WQI values of the heavy metals and toxic elements (Fig. 6) in the rivers of the Tibetan Plateau were practically unpolluted (WQI < 1), suggesting the relatively pristine water quality of the entire plateau. However, the spatial variation observed in the WQI values requires attention to be paid to some potential metal pollution risks. For instance, As, Cd, Mn and Pb, and pollution was found in the rivers of the STPR, Fe and Al pollution was mainly found in the rivers of the NTPR, and thallium (Tl) pollution was found in the CEPR (Fig. 6).

Arsenic (As) is colorless, tasteless and odorless but has been proven to be toxic at very low concentrations (10 μg L^{-1}) (WHO, 2011). The concentrations of As in the rivers of the central and southern plateau, particularly in the Indus, Yarlung Tsangpo and source area of the Yangtze River, were found to be three times higher than the permissible guideline for drinking water (Fig. 6 and Table S2). Arsenic contamination in the rivers of the Tibetan Plateau is mainly due to the contributions of arsenic-rich soils and geothermal springs distributed in this region (Huang et al., 2011; Li et al., 2014; Sheng et al., 2012). Both Hg and Ti are dangerous toxic metallic elements for human health, despite their trace abundances in the Earth's crust (Haxel et al., 2002). Although the concentrations of Hg in the waters of the Yarlung Tsangpo (1.46–4.99 ng L^{-1}) (Zheng et al., 2010) and Yangtze River (2.59 ng L^{-1}) were under the drinking water guideline values of the WHO and GB (Table S2), they were found to be higher than the global average value (0.07 ng L^{-1}) (Huheey et al., 1983; Porterfield, 1984). Tl was also found in the rivers of the Tibetan Plateau, especially upstream of the Yangtze River. The average concentration was as high as 4.2 μg L^{-1}, which is much higher than the guideline for safe drinking water established by the GB (WQI is >3). The sources of Hg and Ti pollution in the rivers of the Tibetan Plateau are still unclear. However, Hg and Ti are usually deposited with copper, lead, zinc, and other heavy-metal-sulfide ores (Chen et al., 2009; Feng and Qiu, 2008; Zhang et al., 2006). The Tibetan Plateau contains the largest copper ores in China; these ores are developed along sutures, where large rivers have also developed (Chen et al., 2009; Yin and Harrison, 2000). Therefore, we can infer that the ores (i.e., copper) along the catchments should contribute to the Hg and Tl pollution in the rivers of the Tibetan Plateau.

In addition to the water quality contamination caused by natural processes, such as groundwater leaching and rock weathering, anthropogenic factors, such as mining and urbanization, have also been proven to affect the regional chemical budgets in the rivers of the Tibetan Plateau in recent decades. Metal pollution (e.g., Cu, Pb, Zn, Mn, Fe, Sb and Al) has been identified in rivers in the socio-economically developed regions of the Tibetan Plateau (Huang et al., 2010; Qu et al., 2017). The concentrations of toxic elements such as Cd, Mn and Pb in Lhasa and Shigatse, the two largest cities in the Tibetan Autonomous Region, were found to be 1–3 times higher than the safety threshold values for drinking water (Table S2). A similar phenomenon also occurred in the rivers of the NTPR, where Fe and Al pollution was found (Fig. 6) due to the mining activities in this region (Chen et al., 2009).

Moreover, it should be noted that the elemental concentrations studied in this work were obtained from filtered water samples. Although previous work in rivers along the Himalaya demonstrated that element concentrations were relatively similar between filtered and unfiltered water samples, the results still showed underestimated values in the filtered samples (Zhang et al., 2015). Due to the alkaline pH values

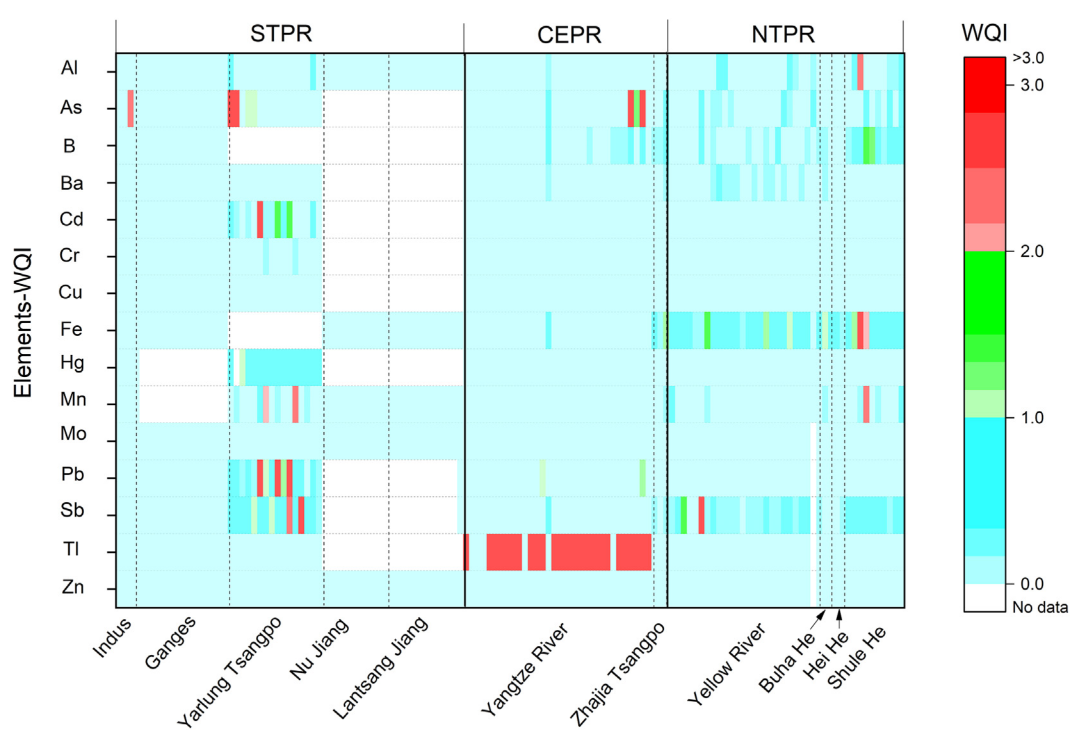

Fig. 6. The pollution index of selected trace elements in the rivers of the Tibetan Plateau.

of these watercourses, the heavy metal ions derived from weathering and acid mining leachates are instantly converted to colloidal or particulate matter when they mix with river water (Förstner and Wittmann, 2012). For instance, the total Hg (including dissolved Hg and particulate Hg) concentrations in the Yarlung Tsangpo were almost two times higher than their dissolved Hg contents (Zheng et al., 2010). Furthermore, considering the existence of numerous mining activities in this region, the total heavy metal contents in the rivers are likely much higher. This is of prime importance, as the colloidal and particulate fraction is transported downstream, where the water is then used for the irrigation of agricultural streams. Once these heavy metals enter agricultural fields, they can be taken up by crops, thus further threatening human health (Förstner and Wittmann, 2012; Zhang et al., 2010).

5. Conclusions and perspectives

This study provides an overview of the mechanisms of the water quality, including the major ions and trace element release, in eleven major rivers of the Tibetan Plateau. In the relatively undisturbed environments on the Tibetan Plateau, the water chemistry of these rivers was found to be largely influenced by the compositions of the bedrock within the catchments. The water ionic chemical compositions of the rivers of the Tibetan Plateau are significantly affected by natural processes, such as rock weathering and evaporation-crystallization. Elevated levels of TDS in the rivers of the eastern and southern plateau are mainly composed of Ca^{2+} and HCO_3^- derived from the weathering of carbonates. In the rivers of the central Tibetan Plateau (e.g., the Zhajia Tsangpo and source of the Yangtze River), high concentrations of Na^+ and Cl^- were found as a result of the distribution of numerous saline lakes and hot springs in this area.

The waters in the rivers on the plateau presented pH values varying from 8.3 to 9.3 (Table S1). This alkaline aquatic environment led to low concentrations of elements in rivers on the Tibetan Plateau. The elements in the natural river waters of the plateau are generally derived from bedrock weathering, and they appeared to vary greatly with different lithologies in each river basin. However, although they represent a major water resource for local residents, the waters in some of the rivers on the plateau were found to be unsuitable for drinking directly due to their high concentrations of toxic/harmful elements caused by natural processes (e.g., saline lakes, arsenic-rich springs in the source area of the Yellow River, Yangtze River and Yarlung Tsangpo) as well as anthropogenic activities (e.g., mining operations, municipal wastewater drainage in the central Yarlung Tsangpo). In addition, due to the alkaline environment of these watercourses, the heavy metal ions derived from weathering and acid mining leachates are instantly converted to colloidal or particulate matter when they mix with river water, therefore, the heavy metal pollution in rivers of the Tibetan Plateau might be underestimated in this study and further work is warranted in this field. Moreover, human activities in both local and surrounding areas have led to the generalized perturbation of elemental abundances in the atmosphere, soils, and waters of the Tibetan Plateau. Hence, with respect to the essential role that rivers on the Tibetan Plateau play in terms of water resources in Asia, long-term and intensive observations of the qualitative/quantitative nature of pollutants are necessary in the rivers of this eco-fragile region.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.08.316.

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