

Spatial characterization, risk assessment, and statistical source identification of the dissolved trace elements in the Ganjiang River—feeding tributary of the Poyang Lake, China

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Received: 9 December 2015 / Accepted: 25 October 2016 / Published online: 14 November 2016
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Abstract Surface water samples were collected from 20 sampling sites throughout the Ganjiang River during pre-monsoon, monsoon, and post-monsoon seasons, and the concentrations of dissolved trace elements were determined by inductively coupled plasma-mass spectrometry (ICP-MS) for the spatial and seasonal variations, risk assessment, source identification, and categorization for risk area. The result demonstrated that concentrations of the elements exhibited significant seasonality. The high total element concentrations were detected at sites close to the intensive mining and urban activities. The concentrations of the elements were under the permissible limits as prescribed by related standards with a few exceptions. The most of heavy metal pollution index (HPI) values were lower than the critical index limit, indicating the basically clean water used as habitat for aquatic life. As was identified as the priority pollutant of non-carcinogenic and carcinogenic concerns, and the inhabitants ingesting the surface water at particular site might be subjected to the integrated health risks for exposure to the mixed trace elements. Multivariate statistical analyses confirmed that Zn, As, Cd, and Tl were derived from mining and urban activities; V,

Cd, and Pb exhibited mixed origin; and Co, Ni, and Cu mainly resulted from natural processes. Three categorized risk areas corresponded to high, moderate, and low risks, respectively. As a whole, the upstream of the Ganjiang River was identified as the high-risk area relatively.

Keywords Ganjiang River · Trace element · Surface water · Risk assessment · HPI · Source identification · Risk categorization

Introduction

With escalating human populations and economic development, water quality is deteriorated by various types of contaminations. Unfortunately, more than 0.3 billion people rely heavily on detrimental water sources, which are contaminated with untreated industrial pollutants, sewage, and agricultural chemicals (World Bank 2007; Zhang et al. 2009). These water sources may cause serious health effects. About 11% of digestive cancer cases are in a relationship with chemical contaminants in drinking water worldwide (WHO-UNDP 2001). Trace elements, which are contributed to surface water from natural input and anthropogenic activities, are ubiquitous substances and one of the most common pollutants (Ali et al. 2013; Gao et al. 2015). These elements, especially some of the heavy metals, are non-degradable, persistence, bioaccumulative, and toxic (Varol 2011; Jiang et al. 2012). Once entering in the water environment, heavy metals deteriorate water quality and cause adverse effects on human health via drinking water directly (Asante et al. 2007; Kavcar et al. 2009; Muhammad et al. 2010) and then accumulate in upper tropic level species via food chain and pose a great threat to human health indirectly (Qiu 2015; Tong et al. 2015). It is noteworthy that trace elements in water environment are prone

Responsible Editor: Philippe Garrigues

Electronic supplementary material The online version of this article (doi:10.1007/s11356-016-7988-z) contains supplementary material, which is available to authorized users.

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to migration and can easily spread to other agents and regions and damage biodiversity of aqueous ecosystem eventually (Zorer et al. 2008; Suresh et al. 2012; Ma et al. 2013). Therefore, to understand, the levels of trace elements in the water environment were quite important for water management and human health.

Trace elements in water environment are derived from a variety of natural processes as well as anthropogenic influences, which enhance the level of element concentrations in water. Natural processes include dry and wet deposition, reductive dissolution, and soil erosion and rock weathering (Krishna et al. 2009; Iqbal et al. 2013; Zeng et al. 2015). Different from the natural processes, anthropogenic influences related to human activities behave varied sources, such as municipal sewage and industrial wastewater discharge, mining and mineral processing, agricultural fertilizer and pesticide leaching, and automobile exhaust emissions (Nouri et al. 2008; Varol and Şen 2012; Cheng et al. 2014). In a particular river basin, apportionment and identification of anthropogenic and/or natural sources of trace elements play an important role on managing water resources (Li and Zhang 2010). Simultaneously, seasonal variations in these natural processes and anthropogenic influences have strong effects on river water quality (Cidu and Biddau 2007; Giri and Singh 2014a). Therefore, it is imperative to characterize the seasonal variability of surface water quality for evaluating temporal changes of water pollution from anthropogenic or natural contributions.

The Ganjiang River, one of the main tributaries of the Yangtze River, is undertaken as one of the major sources of drinking water for the residents of water basin and also serves for aquaculture, irrigation, and fishing. The Ganjiang River basin is quite well known for its abundant mineral resources. Unfortunately, industrial and mining activities have a strong possibility on heavy metal pollution in the Ganjiang River. Therefore, its water quality is of great concern. The limited capabilities of water treatment on top of deficient constraint mechanism on these activities result in a steady deterioration in the water quality level. To our best knowledge, previous studies have characterized water chemistry (Li et al. 2007) and heavy metal enrichment in sediments of the river (Ji et al. 2014), whilst there is no sufficient information available regarding trace elements in the water from the Ganjiang River, and their variations in different hydrological periods remain unclear. Knowledge of spatial and seasonal distribution of dissolved trace elements in the Ganjiang River makes great contribution to understand the effect of mining, urban development, and other land use activities on water quality. In view of the previous facts, we report the extensive study on dissolved trace element levels of the Ganjiang River accomplished through regular monitoring in three seasons at 20 sites to understand the spatial and seasonal variations of selected elements and evaluate the water suitability for drinking purpose and aquatic life habitat. Ultimately, these results would

provide the baseline data for developing water conservation and management strategies in the Ganjiang River basin.

Material and methods

Study area

The Ganjiang River, the main tributary of the Yangtze River, is in central China's Jiangxi Province. It rises in Shicheng County and traverses a total distance of about 823 km, and its mainstream runs through three major cities including Ganzhou, Ji'an, and Nanchang before discharging into the Yangtze River through the Poyang Lake, which is the largest freshwater lake in China.

The Ganjiang River basin holds a total area of 82,809 km² and sustains a population of 18 million at least and is characterized as the subtropical monsoon climate. The Ganjiang River is also the largest and most important tributary of the Poyang Lake and accounts for about 51.5% of the watershed area of the Poyang Lake (Fig. 1). Geological strata from the oldest to youngest are Middle Proterozoic, Upper Proterozoic, Sinian, Cambrian, Ordovician, Carboniferous, Devonian, Permian, Triassic, Jurassic, Cretaceous, Tertiary, and Quaternary. The main lithologic units include sand slate, phyllites, siltstone, slate, limestone, sandstone, limestone, shale, mudstone, and sandstone. The Ganjiang River plays an important role and serves as the major water source for the local residents for various usages; i.e., 80% of farmland was irrigated from the Ganjiang River, and the flooding irrigation is the main irrigation practice (Wu et al. 2014). However, it is confronted with many non-ferrous mines (upstream), rare earth mines (upstream), and metal-associated industries (Wu 2012). Therefore, it is necessary to evaluate the surface water quality and the human health risk via digesting water and environment risk for aquatic life in the Ganjiang River.

Sample collection

Three sampling campaigns were conducted in 20 sites reasonably representing the riverine system's water quality throughout the Ganjiang River in February, June, and November 2013 (pre-monsoon, monsoon, and post-monsoon correspondingly). The sampling sites were arranged at the mainstream, some important tributaries, and the confluences of the mainstream and tributaries (Fig. 1). The site numbers from 1 to 20 hereafter are referred to as S1 to S20, whose detailed information is listed in Table S1. In each site, three replicates (250 mL) were taken about 100 m apart and collected at a depth of approximately 10 cm using previously acid-washed high-density polyethylene (HDPE) containers, which were subsequently well mixed. These mixed samples were immediately filtered

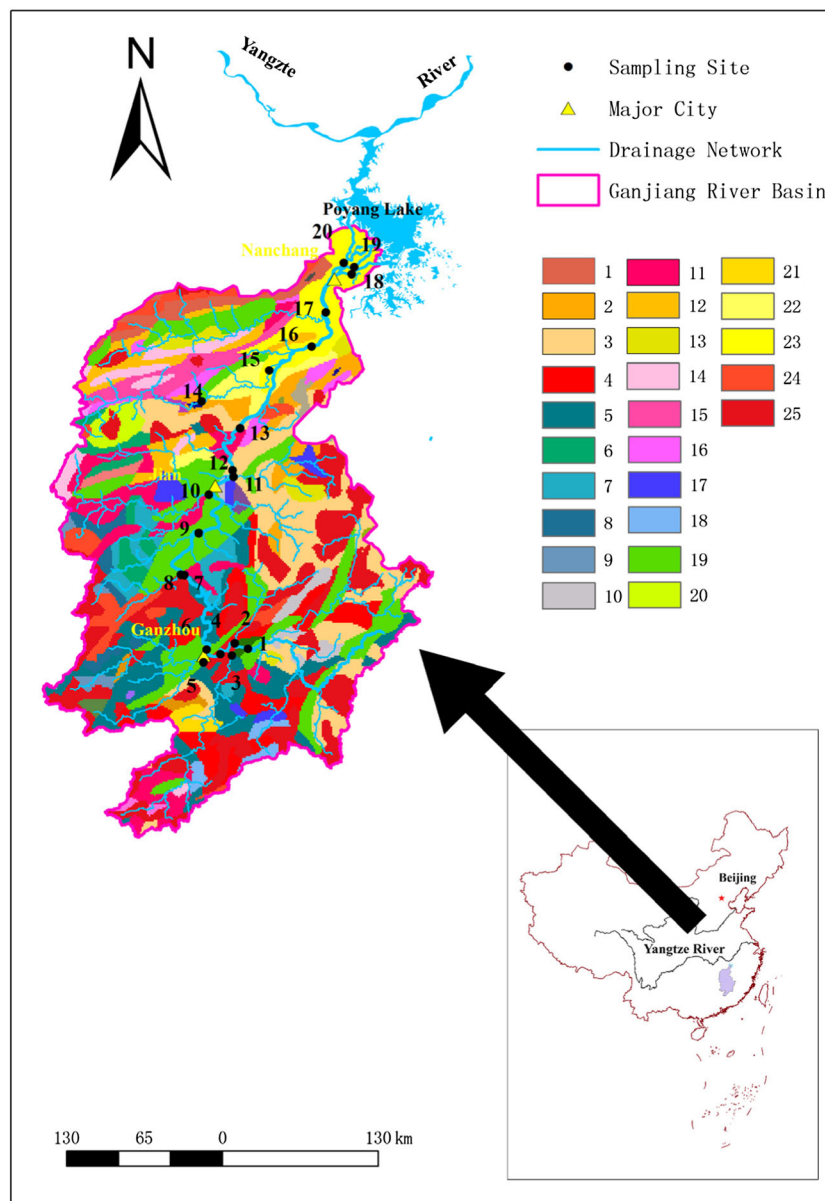


Fig. 1 Geology sketch of the Ganjiang River basin with sampling sites. *1* Tuffaceous sand slate, phyllites, and metasandstone (Middle Proterozoic); *2* tuffaceous phyllite, carbon-bearing phyllite, and siltstone (Upper Proterozoic); *3* sedimentary phyllite, sedimentary tuff, sand slate (Sinian); *4* sedimentary tuff, tuffaceous sand slate, and silicalite slate (Sinian); *5* sandstone, slate, and carbon-bearing slate (Cambrian); *6* sand slate, tuffaceous sandstone, and carbonaceous silicalite slate (Cambrian); *7* interbedded sand slate, mixed carbon-bearing slate (Cambrian); *8* sand slate, carbon-bearing slate, and silicalite-bearing slate (Ordovician); *9* sand slate, and carbonaceous silicalite slate (Ordovician); *10* glutenite, sandshale, argillaceous limestone, and carbonaceous shale (Carboniferous); *11* sandshale, siltstone, and mudstone (Devonian); *12*

limestone, carbon-bearing limestone, silicalite, carbonaceous mudstone, and carbonaceous shale (Permian); *13* limestone, silicalite limestone, sandshale, and mudstone (Permian); *14* grayish-green mudstone, siltstone, limestone, and dolomite limestone (Triassic); *15* brown-red mudstone, siltstone, and calcareous shale (Triassic); *16* glutenite, sandshale, mudstone, and carbonaceous shale (Triassic); *17* feldspar-quartz sandstone, and sandshale (Jurassic); *18* sandshale, glutenite, tufa, rhyolite, andesite, and basalt (Jurassic); *19* brick-red sandstone, glutenite, and siltstone (Cretaceous); *20* varicolored sandshale, tufa, and lava tuff (Cretaceous); *21* glutenite and glutenite (Tertiary); *22* brick-red glutenite and siltstone (Tertiary); *23* loose alluvium (Quaternary); *24* granodiorite; *25* granite

through a prewashed 0.45- μm nitrocellulose filter, and the initial filtration was thrown away to clean the container. The remaining portion was stored and acidified to pH <2 with suprapure nitric acid in situ later. After that, the samples were kept in an ice box and then transported to the laboratory and stored in a fridge before analysis. The locations referred as

longitude and latitude of each site were positioned by a GPS, and general physical and chemical properties such as temperature, pH, dissolved oxygen (DO), and electrical conductivity (EC) were determined in situ, which are listed in Table S2. The environmental conditions were also documented during the fieldwork.

Determination of trace elements

To explore the effect of anthropogenic activities on the risk assessment of the dissolved trace elements in the surface water of Ganjiang River, the selected trace elements adhered to the two following criteria: (1) they are the common pollution elements in the aquatic ecosystem (Li et al. 2008; Li and Zhang 2010; Li et al. 2013; Zeng et al. 2015) and (2) they are possibly the indicators of pollution source in the Ganjiang River drainage basin (Liu et al. 2014; Zhang et al. 2014). Taking these two criteria into consideration, V, Cr, Co, Ni, Cu, Zn, As, Cd, Tl, and Pb were determined by inductively coupled plasma-mass spectrometry (ICP-MS, Thermo Fisher X series), in which these elements were monitored by standard sampling (multi-element standard, lot number B9035086). The limit of detection (LOD) was 0.018 µg/L for V, 0.019 µg/L for Cr, 0.007 µg/L for Co, 0.435 µg/L for Ni, 0.143 µg/L for Cu, 0.042 µg/L for Zn, 0.004 µg/L for Cd, and 0.003 µg/L for Pb. Recovery rates of trace elements spiked in water fell into the range from 89 to 110%. In order to assess the absence of any contamination occurring from reagents and materials, whole procedure blank tests were performed on double-deionized water (Milli-Q System, Millipore) to keep matrix interference (blank) <2%. Triplicates of sample analysis yielded a relative standard deviation (RSD) of <5%. Otherwise, the determinations were done again until all data reach the standard.

Heavy metal pollution index

The heavy metal pollution index (HPI) was generally adopted to represent the total quality of water with respect to trace element (Mohan et al. 1996; Giri and Singh 2014b). It is based on weighted arithmetic quality mean method and developed from two basic steps. The first step was to identify weightage to selected parameter by establishing the rating scale, which is an arbitrary value (range from 0 to 1) calculated by making values inversely proportional to the recommended standard for the corresponding parameter (Mohan et al. 1996). The second step was to select the pollution parameter on which the index is to be based. The HPI was calculated as the following expressions:

$$HPI = \frac{\sum_{i=1}^n W_i Q_i}{\sum_{i=1}^n W_i} \tag{1}$$

where Q_i is the sub-index of the i th parameter, W_i is the unit weightage of the i th parameter, and n is the number of parameters.

The sub-index (Q_i) of the parameter was expressed in the following equation:

$$Q_i = \sum_{i=1}^n \frac{\{M_i(-)I_i\}}{(S_i-I_i)} \times 100 \tag{2}$$

where M_i is the monitored value of heavy metal of i th parameter, I_i is the ideal/baseline value of the i th parameter, and S_i is the standard value of the i th parameter. The sign (-) only demonstrates numerical difference, ignoring the algebraic sense.

The HPI was always used for evaluating water quality served as drinking (Mohan et al. 1996; Prasad and Bose 2001; Edet and Offiong 2002; Giri and Singh 2014b). In the present study, the HPI was calculated for evaluating the water quality used as habitat for aquatic life. Therefore, the weightage (W_i) was taken as the inverse of S_i namely criterion maximum concentration (CMC; for protection of aquatic life) and I_i -adopted criterion continuous concentration (CCC) as ideal value of the selected element. The specific parameters of HPI in this study are presented in Table S3. The calculated HPI relates to Cr, Ni, Cu, As, Cd, and Pb due to lack of the corresponding recommended standard for other elements. The critical pollution level of the HPI value is similarly set at 100 given by Prasad and Bose (2001).

Health risk assessment via water consumption

Trace elements in environment entering into the human body mainly came down to three pathways, including direct ingestion, inhalation through the mouth and nose, and dermal absorption through skin exposure (Wu et al. 2009; Li and Zhang. 2010). With respect to trace elements in water, two exposure pathways namely ingestion and dermal absorption play the most important roles (Giri and Singh 2014a; Zeng et al. 2015). According to the risk guidelines of USEPA (2004), the dose received through the individual pathway was calculated by the following expressions:

$$E_{ing} = \frac{C_w \times IR \times EF \times ED}{BW \times AT} \tag{3}$$

$$E_{derm} = \frac{C_w \times SA \times K_p \times ET \times EF \times ED \times CF}{BW \times AT} \tag{4}$$

The values, descriptions, and sources of parameters in the previous formulas are summarized in Table S4.

Risk characterization was quantified by non-carcinogenic and carcinogenic risks (Li and Zhang 2010). The potential non-carcinogenic risks, reflected by the hazard quotient (HQ), were calculated as the quotient between the environmental exposure (ingestion, dermal) and the reference dose. If the HQ is greater than one, there might be concern for non-carcinogenic effects (Giri and Singh 2014a). The hazard index (HI) reflected the total potential non-carcinogenic risks posed by more than one pathway, which was the sum of the HQs from all applicable pathways. Values of HI above the unity were considered a

potential for an adverse effect on human health or the necessity for further study (USEPA 2004). Similarly, the excess cancer incidence (ELCR) is estimated as the incremental probability of an individual developing any type of cancer over a lifetime due to carcinogenic element exposure under specific scenarios, calculated by multiplying the daily exposure and the cancer slope factor (SF) (Alves et al. 2014). An acceptable level of carcinogenic risks by the USEPA ranged 10^{-6} to 10^{-4} (Rodriguez-Proteau and Grant 2005).

Statistical analyses

Data were statistically analyzed by using the statistical software package PASW 17.0 (SPSS Inc., Chicago, USA). Analysis of variance (ANOVA) was applied to test the significant temporal differences. Principal component analysis (PCA) was used to identify overlying factors that analyze the interaction and the relationship between the different variables (Li and Zhang 2010). In factor analysis (FA), an orthogonal varimax rotation was used to rotate the axes for maximizing the variation among the variables and factors with eigenvalues greater than 1 were extracted (Lu 2010). To examine the suitability of the raw data for PCA/FA, Kaiser-Meyer-Olkin (KMO) and Bartlett's sphericity tests were performed (Varol et al. 2013). Reduction of variables is accepted when the KMO values are >0.5 and $p < 0.05$ (Merkle et al. 1998). Pearson's coefficient was used to test the interrelationship among the selected variables with statistical significance set a priori at $p < 0.05$. Cluster analysis (CA) was applied to cluster the similar studied elements and sampling sites (spatial variability). In the present study, hierarchical agglomerative CA was performed on normalized data set to test the variables' (case) similarity or nearness

using the squared Euclidean distance with between-group linkage method.

Results and discussion

Distribution of trace elements in surface water

The concentrations of individual and total trace elements for 20 sites and three seasons are presented in and Table 1 and Fig. 2. Taking it as a whole, the trace elements exhibited significant fluctuations in the surface water samples varied with different sites and seasons. The total concentrations of trace elements in surface water from the Ganjiang River ranged from 14.49 (S4) to 156.15 $\mu\text{g/L}$ (S5) and followed in order of abundance by pre-monsoon (46.21 $\mu\text{g/L}$), post-monsoon (37.12 $\mu\text{g/L}$), and monsoon (19.28 $\mu\text{g/L}$). The high concentration level of Zn in the surface water made the greatest contribution to total concentration in either site or season, followed by that of Cu (3.86 $\mu\text{g/L}$), Ni (2.99 $\mu\text{g/L}$), and As (2.80 $\mu\text{g/L}$). These three elements accounted for 85.98% of the total concentrations. However, the lowest average level was found for Tl (0.03 $\mu\text{g/L}$). With respect to individual, Zn, Cu, Ni, As, and Pb were the most abundant elements in the surface water, whereas Co, Cd, and Tl were the least abundant at any seasons. The levels of all individuals in the water at pre- and post-monsoon were significantly higher than those at monsoon and behaved significant seasonality (Table 1 and Fig. 2). The diluting effect due to great precipitation in monsoon season was the most plausible explanation for the lowest total concentrations and all individuals owning their lowest concentrations (Li and Zhang 2010; Gao et al. 2015). The concentrations of the dissolved trace elements significantly

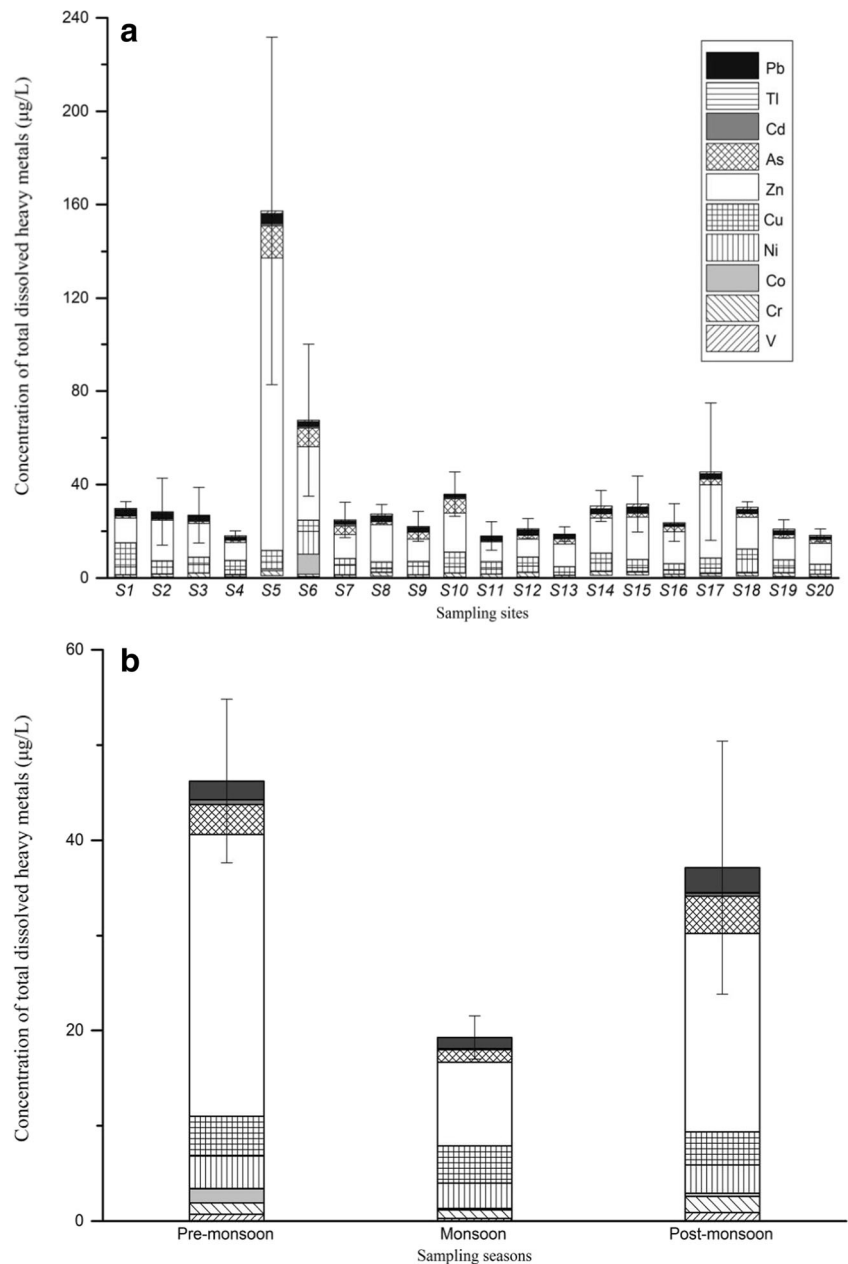
Table 1 Statistical characteristics of the trace element concentrations in surface water of the Ganjiang River

	Pre-monsoon				Monsoon				Post-monsoon			
	Max ($\mu\text{g/L}$)	Min ($\mu\text{g/L}$)	Mean ($\mu\text{g/L}$)	SD	Max ($\mu\text{g/L}$)	Min ($\mu\text{g/L}$)	Mean ($\mu\text{g/L}$)	SD	Max ($\mu\text{g/L}$)	Min ($\mu\text{g/L}$)	Mean ($\mu\text{g/L}$)	SD
V	1.87	0.14	0.73 a	0.51	0.75	0.05	0.28 b	0.15	2.09	0.11	0.90 a	0.50
Cr	3.41	0.30	1.19 a	0.76	2.57	0.15	0.90 a	0.82	2.58	0.87	1.70 b	0.42
Co	22.11	0.10	0.42 a	4.86	1.75	0.044	0.085 a	0.37	1.45	0.10	0.34 a	0.33
Ni	15.82	0.77	3.42 a	2.60	9.33	0.73	2.62 a	2.02	9.09	0.49	2.17 a	2.60
Cu	9.54	1.63	4.18 a	2.18	21.77	1.02	2.70 a	4.93	6.42	1.80	3.48 a	1.49
Zn	106.90	4.94	29.61 a	26.87	21.69	3.16	8.81 b	5.55	253.20	4.13	9.36 b	54.83
As	18.71	0.34	3.16 a	1.53	4.75	0.15	1.32 b	1.13	18.36	0.38	3.92 a	4.16
Cd	1.73	0.08	0.486 a	0.464	0.298	0.015	0.111 b	0.068	2.25	0.028	0.136 b	0.548
Tl ^a	77.70	0.42	28.14 a	19.32	29.65	2.92	15.13 b	8.11	134.80	7.60	45.97 a	31.51
Pb	7.26	1.01	1.92 ab	1.41	1.80	0.68	1.15 a	0.31	4.64	0.91	2.61 b	1.08

^a Nanogram per liter

The data with different letters have significant differences in rows

Fig. 2 Total concentrations (mean ± SE) of ten trace elements in the different sampling site (a) and time (b) of the Ganjiang River, China



declined even after suffering from severe flood (Li et al. 2014b). Some individuals such as Zn and Cd presented significantly higher level at per-monsoon season compared to the post-monsoon season with significant higher level of Cr (Table 1). As shown in Fig. 2, the gradually higher concentrations found at S5 (156.15 µg/L) compared with S4 (14.49 µg/L), particularly for Zn, As, and Cd, might likely be attributed to wastewater discharging from tungsten ore dressing plants located at the upper reaches of the Zhang River (Zhang et al. 2014). As the confluence of large volumes of non-contaminated river water, such as the Ping River, Tao River, and Gong River, a decreasing trend from S5 to S6 was observed. Some sites with high level of total trace elements, such as S6 (66.94 µg/L), S10 (35.51 µg/L), and S17 (44.61 µg/L), distributed around areas

with high urbanization (Ganzhou, Ji’an, and Nanchang), which produced a large amount of sewage and industrial wastewater. Therefore, natural process and anthropogenic activities were responsible for governing trace elements in the Ganjiang River.

The mean concentrations of all the selected elements were below the permissible limit drinking water guidelines established by WHO (2008), USEPA (2009), EC (1998), and Chinese standards (CMH 2007) (Table 2). Regarding spatial concentrations of trace elements with guidelines, V, Cr, Co, Ni, Cu, Tl, and Pb at all sites never exceeded WHO, USEPA, EC, and Chinese standards. As concentrations were higher than EC, WHO, and USEPA drinking water limits in 5% of the total 60 samples. Zn concentrations were higher than EC drinking water limits in 3.3% of the total samples.

Table 2 Comparative levels of trace elements in the overlying water in the Ganjiang River with other rivers in the world and guidelines

Site	V	Cr	Co	Ni	Cu	Zn	As	Cd	Tl ^b	Pb	References	
Poyang Lake basin	Mean	0.63	1.26	0.66	2.99	3.86	19.76	2.80	0.306	29.75	1.89	Present study
	SD	0.49	0.75	2.84	2.65	3.18	35.84	3.88	0.438	24.97	1.19	
Raohe River, China		2.68				87.57	32.35				3.11	Hu et al. (2012)
Fuhe River, China				4.76	6.00	2.83	2.80	0.085			1.80	Li et al. (2014a)
Xiushui River, China				0.67	3.30	2.78	3.16	0.100			1.77	Li et al. (2014a)
Xinjiang River, China				1.90	5.28	6.14	13.80	1.29			1.53	Li et al. (2014a)
Poyang Lake		5.02			6.11	22.72					6.55	Hu et al. (2012)
Yangtze River, China		3.93	0.41	1.50	2.57	7.06	5.96	0.145			1.12	Yang et al. (2014)
Upper Han River, China	69.71	8.11	2.23	1.71	13.37		14.16	2.295			9.20	Li and Zhang (2010)
Xiangjiang River, China		6.61			20.33	84.57	12.24	1.34			2.29	Zeng et al. (2015)
Background values ^a	0.23	0.26	0.24	0.18	0.63	0.68	0.86	0.015			0.76	Zhang and Zhou (1992)
Gomti River, India		63.33		44.17	21.50	66.50		23.50			17.67	Gupta et al. (2014)
Tarkwa stream River, Ghana	0.46	0.52	1.30		2.65	138.00	18	0.04	<0.01	0.85		Asante et al. (2007)
Tigris River, Turkey		25.41		24.54	17.10	12.01	0.63	0.044			2.82	Varol et al. (2013)
Sacramento River, USA	2.43	0.24	0.77	1.30	43.77	84.52	0.99	0.573	26.27	0.24		Taylor et al. (2012)
Pardo River, Brazil	1.50	<0.51		5.70	2.53	12.45	<1.17	<0.05	130.00	2.54		Alves et al. (2014)
World average	0.71	0.70	0.148	0.801	1.48	0.60	0.62	0.08			0.079	Gaillardet et al. (2003)
Water quality criteria for drinking												
WHO		50		70	2000		10	3		10		WHO (2008)
Chinese standards (GB5749-2006)		50		20	1000	1000	10	5	100	10		CMH (2007)
MCLG 0		100			1300	2000	0	5	50	0		USEPA (2009)
MCL 10		100			1300	2000	10	5	200	15		
EC		50		20	2000	100	10	5		10		EC (1998)

^a Background values of trace metals in the source area of the Yangtze River

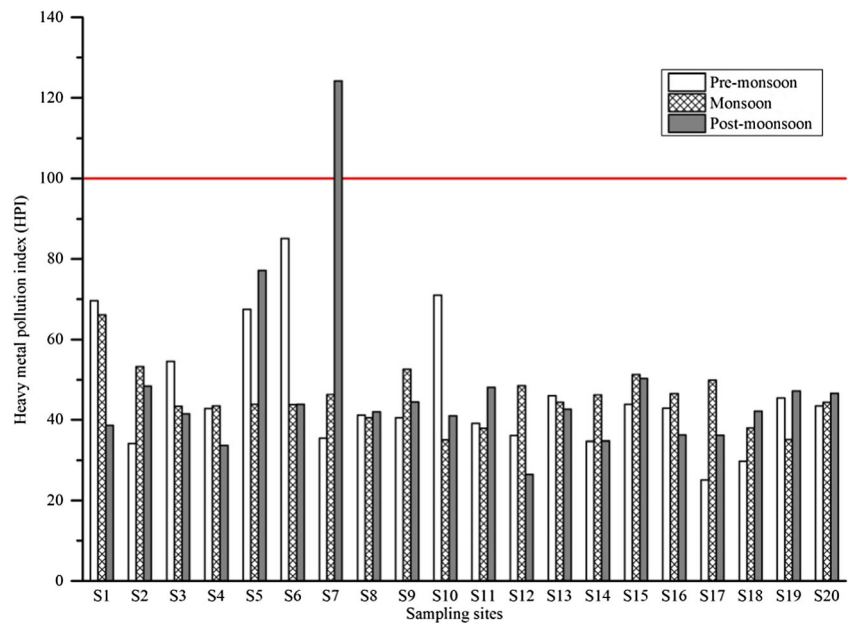
^b Nanogram per liter

As the primary tributary of the Poyang Lake basin, it was revealed that the trace elements in surface water from the Ganjiang River were in the moderate concentration level relatively comparing other rivers in the basin (Table 2). Also, as the main tributary of the Yangtze River basin, the concentrations of the trace elements were observed in a relatively low level when the trunk stream (Yangtze River) and tributary (Han River and Xiangjiang River) were considered. However, our results showed relatively higher concentrations than background values in the headwater from the Yangtze River, where it was affected by dominant physical weathering and little anthropogenic disturbances (Zhang and Zhou 1992). When compared with the trace elements from other streams or rivers abroad, it could be observed that the concentrations of the trace elements did not behave obvious characteristics and are comparable with other studies; however, the mean concentrations of all trace elements except V in the surface water from Ganjiang River were much higher than their corresponding world average concentrations for the natural river systems (Table 2). This was especially true for the cases of Zn and Pb, which were 32.93 times and 23.92 times of the world average concentrations, respectively.

Risk assessment on water quality used as habitat for aquatic life

The HPI, calculated by use of the element concentrations and surface water quality guidelines of USEPA (CMC and CCC values), was applied to evaluate the risk level of water quality used as habitat for aquatic life with respect to trace element in surface water from the Ganjiang River. In computing the HPI for the present day, Cr, Ni, Cu, As, Cd, and Pb were used (Table S1). The computed HPI values are presented in Fig. 3. The critical pollution index value is 100, below which, the overall pollution level should be considered acceptable for an aquatic ecosystem (Prasad and Bose 2001). The HPI value ranged from 26.44 (S12, pre-monsoon) to 124.25 (S7, post-monsoon). All sites except S7 were lower than 100 referred to as critical pollution level. The means of HPI at pre-monsoon, monsoon, and post-monsoon were 46.39, 45.29, and 47.26, respectively, and presented minimum difference. This indicated that the surface water from the Ganjiang River was basically clean, which can be used as habitat for aquatic life.

Fig. 3 The HPI for three seasons and 20 sites along the Ganjiang River



Health risk for exposure to the trace elements in surface water

The risk level of the trace elements in surface water from the Ganjiang River was evaluated by health risk assessment, which is estimated as the occurrence probability of adverse health effects in relation to chronic (non-carcinogenic) and carcinogenic over a specified time period (Lim et al. 2008). Table 3 and Figs. 4 and 5 present the HQ, hazard index (HI), and carcinogenic risk of trace elements via oral and dermal pathways for child and adult.

The level of the mean HQs increased in the order of Zn < Cu < V < Ni < Cr < Cd < Pb < Co < Tl < As, regardless of whether adult or child. This indicated that As and Tl were the

major contributors to the exposures of the inhabitants, while Zn and Cu were the least participants. The degree of risk was obviously lower for adult (Table 3), indicating that the adults were less vulnerable to suffer the risks of the non-carcinogenic effects than the children, which was consistent with the results in the other previous studies (Li and Zhang 2010; Zeng et al. 2015). In contrast to oral intake, dermal pathway could basically be considered as negligible, with values ranging from 1.17×10^{-8} (Zn for adult at site 4) to 9.38×10^{-5} mg/kg day (Co for child at site 6). $HQ_{ingestion}$ (HQ by ingestion) of all the studied elements was below unity for both adult and child, suggesting that these elements posed little hazard. However, $HQ_{ingestion}$ of As was nearly 1, implying that As may cause adverse health effects and potential non-carcinogenic concern.

Table 3 Reference dose and hazard quotient for each element of the Ganjiang River, China

Element	C_w ($\mu\text{g/L}$)	RfD_{dermal} ($\mu\text{g/kg/day}$)	$RfD_{ingestion}$ ($\mu\text{g/kg/day}$)	$HQ_{ingestion}$		HQ_{dermal}		$HI = \sum HQs$		Cancer risk	
				Adult	Child	Adult	Child	Adult	Child	Adult	Child
V	0.633	0.13	5	4.11E-03	8.27E-03	7.50E-07	8.12E-07	4.11E-03	8.27E-03		
Cr	1.264	0.075	3	1.37E-02	2.76E-02	5.19E-06	5.62E-06	1.37E-02	2.76E-02		
Co	0.664	0.06	0.3	7.19E-02	1.45E-01	6.82E-06	7.39E-06	7.19E-02	1.45E-01		
Ni	2.991	0.8	20	4.85E-03	9.78E-03	1.15E-07	1.25E-07	4.85E-03	9.78E-03		
Cu	3.856	8	40	3.13E-03	6.30E-03	7.42E-08	8.04E-08	3.13E-03	6.30E-03		
Zn	19.762	60	300	2.14E-03	4.31E-03	3.04E-08	3.30E-08	2.14E-03	4.31E-03		
As non-cancer	2.804	0.285	0.3	3.03E-01	6.11E-01	1.52E-06	1.64E-06	3.03E-01	6.11E-01		
As cancer	2.804	3660	1500	2.60E-05	1.05E-05	5.06E-11	1.10E-11			2.60E-05	1.05E-05
Cd	0.306	0.025	0.5	1.99E-02	4.01E-02	1.89E-06	2.05E-06	1.99E-02	4.01E-02		
Tl	0.0297	0.010	0.010	9.65E-02	1.94E-01	4.58E-07	4.96E-07	9.65E-02	1.94E-01		
Pb	1.893	0.42	1.4	4.39E-02	8.84E-02	2.78E-06	3.01E-06	4.39E-02	8.84E-02		

Rfd reference dose of metals from risk-based concentration table (USEPA 2004) expected for Pb from WHO (2006)

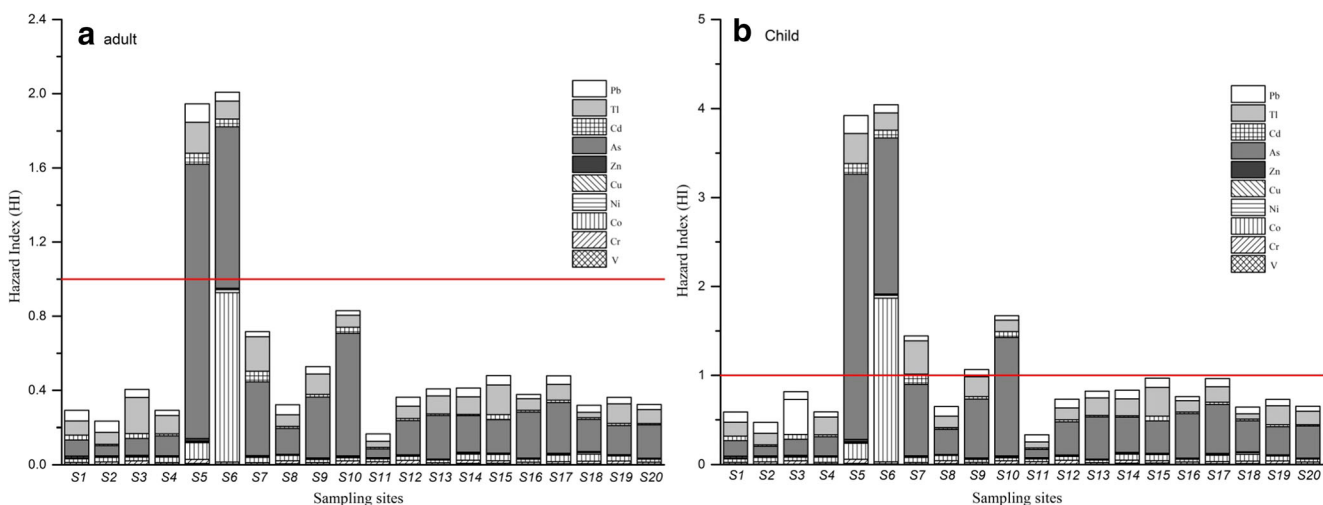


Fig. 4 Hazard index of the mixed trace elements in surface water from the Ganjiang River

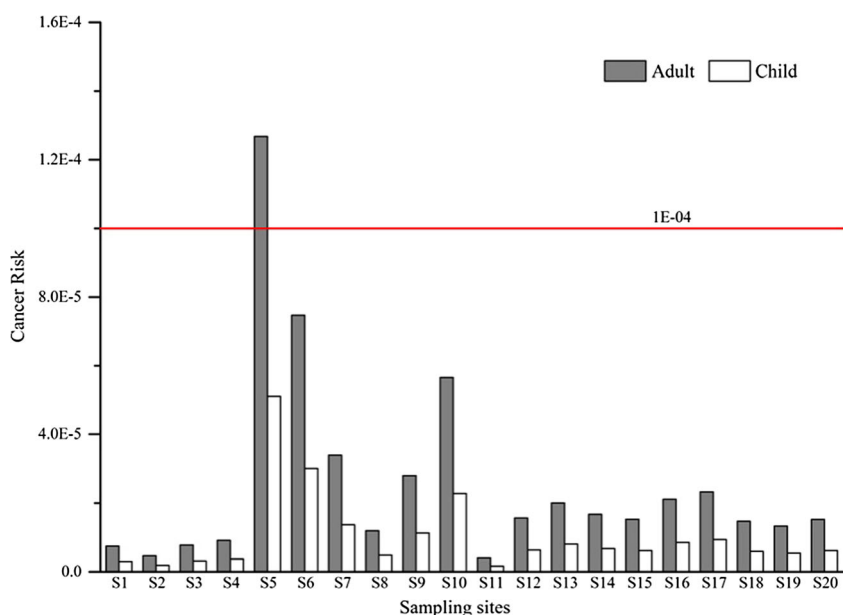
Although the concentration of Tl in surface water was little, the mean HQ corresponded to oral exposure to Tl, followed As with levels of 0.097 and 0.19 for adults and children, respectively. It was discovered that acute exposure to Tl might affect the epidermal, gastrointestinal, cardiovascular and renal systems (Alves et al. 2014). Therefore, Tl also needs to be paid attention.

The calculated HI values determined by the mixed trace element concentrations from the sampling sites were presented in the Fig. 4. The order of HI at different sites, were same to adults and children, sequenced by S6 > S5 > S10 > S7 > S9 > S15 > S17 > S14 > S13 > S3 > S16 > S12 > S19 > S20 > S8 > S18 > S4 > S1 > S2 > S11. Two sites (S5, S6) and five sites (S5, S6, S7, S9, S10) in the study areas were at risks of non-carcinogenic effects from the mixed trace elements for child and adult, respectively. The HI values for child were basically

twice as much as those for adult. Additionally, As exclusively with the HI for child and adult was the dominant contributor to non-carcinogenic concern, followed by Tl and Pb.

Carcinogenic risks were just evaluated for As due to the shortage of oral and dermal SFs for other elements (Alves et al. 2014). The carcinogenic risks for exposure to As ranged from 4.02×10^{-6} to 1.27×10^{-4} for adult and ranged from 1.62×10^{-6} to 5.11×10^{-5} for child considering both exposure pathway, indicating the adults were more vulnerable to suffer the risks of the carcinogenic effects than the children. The carcinogenic risk of As exposure in the Ganjiang River was accordance to the acceptable lifetime risks for carcinogens in drinking water (10^{-6} – 10^{-4}) expect the adults at S5 (Fig. 5). It was inferred that the probability of cancers closely related to As intake was low by ingesting surface water from the Ganjiang River over a long life time.

Fig. 5 Cancer risk of As in surface water from the Ganjiang River



As was the most important pollutant in the Ganjiang River based on the calculated HQs and carcinogenic risk assessment, which was similar to the previous studies (Li and Zhang 2010; Alves et al. 2014; Zeng et al. 2015). To date, the traditional purification process including coagulation–sedimentation, sand filtration and disinfection were employed to purification in China and its arsenic removal efficiency was taken as zero (Liu et al. 2009). Previous studies reported high arsenic intake led to potentially carcinogenic effects such as the cancers of bladder, liver, kidney, lung, and skin, and other adverse health effects including hypertension, skin lesions, neuropathy, diabetes, and cardiovascular and cerebrovascular diseases (Wu et al. 2009). In addition to As, Tl was another element of concern. Although current Tl was of very little amounts in surface water from the Ganjiang River, relatively high values of HQ related to Tl exposure were also observed. Tl is widely distributed in nature (Xiao et al. 2012), being water an important medium for dispersion. Thallium has been found to be an emerging pollutant for human health (Zhou et al. 2008; Liu et al. 2010). Potentially diseases associated with epidermal, gastrointestinal, cardiovascular, and renal systems can be diversely affected by acute exposure to Tl (Alves et al. 2014). Therefore, it is required to pay more attention on arsenic and thallium for local residents at particular sites, especially for the sensitive children, than the other selected elements.

Multivariate statistical analysis of the trace elements

Principal component analysis and cluster analysis

PCA is well developed as a statistical technique to identify the different groups of elements in water that correlate and thus can be considered as having a similar behavior and a common origin (Krishna and Mohan 2014; Giri and Singh 2014a; Bu et al. 2015). The PCA loadings evolved by varimax normalized rotation with Kaiser normalization on the data set of the selected elements of surface water in Ganjiang River are presented in Table 4. The value of KMO was 0.742, and the significance of Bartlett’s sphericity test was less than 0.001, indicating that PCA would be effective for the studies (Varol 2011).

Three principal components (PCs) were extracted (eigenvalues >1) explaining about 67.30% of the total variance in the element concentration data set (Table 4). The first component (PC1), which explained 27.41% of the total variance, had strong positive loadings (>0.70) on As and Cd and moderate positive loadings (>0.50) on Zn and Tl. The second component (PC2), which accounted for 20.82% of the total variance, had strong positive loadings on V, Cr, and Pb. The third component (PC3), which elucidated 19.07% of the total variance, had strong positive loadings on Co and Ni and moderate on Cu.

Table 4 Total variance explained and component matrixes for dissolved trace elements in surface water from the Ganjiang River

Component	Initial eigenvalues			Rotation sums of squared loadings		
	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %
1	3.950	39.502	39.502	2.741	27.412	27.412
2	1.689	16.895	56.397	2.082	20.820	48.232
3	1.090	10.905	67.302	1.907	19.069	67.302
4	0.953	9.528	76.829			
5	0.702	7.024	83.854			
6	0.585	5.848	89.701			
7	0.382	3.820	93.521			
8	0.253	2.533	96.055			
9	0.219	2.185	98.240			
10	0.176	1.760	100.000			
Variables	Rotated component			Communalities		
	1	2	3			
V	0.318	0.720	0.039	0.620		
Cr	0.075	0.744	0.038	0.561		
Co	0.352	−0.150	0.783	0.759		
Ni	0.301	0.011	0.794	0.721		
Cu	−0.205	0.328	0.692	0.628		
Zn	0.679	0.301	0.087	0.559		
As	0.808	0.268	0.268	0.796		
Cd	0.850	0.036	0.303	0.816		
Tl	0.672	0.385	−0.086	0.607		
Pb	0.301	0.754	0.058	0.662		

CA provided a dendrogram (Fig. 6) in which all studied elements from the Ganjiang River were categorized into three statistically significant clusters, which were greatly similar with those from PCA. This result basically showed the validity of the result from the PCA.

Correlation matrix

The correlations among trace elements can provide viable information on sources and pathways. The correlation matrix, which was calculated by the Pearson's correlation for the trace elements in surface water from the Ganjiang River, is presented in Table 5. The correlation matrix showed that there were a number of significant positive relationships among the selected elements. The element pairs V-Zn, V-Cd, Ni-Cu, and Cd-Pb had a significantly positive correlation at $P < 0.05$ significance level, whereas the element pairs V-Cr, V-As, V-Tl, V-Pb, Cr-Tl, Cr-Pb, Co-Ni, Co-As, Co-Cd, Ni-As, Ni-Cd, Zn-As, Zn-Cd, Zn-Tl, Zn-Pb, As-Cd, As-Tl, As-Pb, Cd-Tl, and Tl-Pb had a significantly positive correlation at $P < 0.01$ significance level (Table 5). Based on the previous studies (Lu et al. 2010; Saeedi et al. 2012), it may be deduced that the elements have common, mutual dependence and identical behavior or are affected by the same factor(s) during the transport if the correlation coefficient between the elements is positive. The correlation matrix observed basically supported the result from the PCA for trace elements in surface water.

Source identification of the trace elements

Three sources of the selected trace elements in surface water from the Ganjiang River could be identified by spatial distribution, PCA, CA, and correlation coefficient analysis. Combining the previous studies and current situation in the

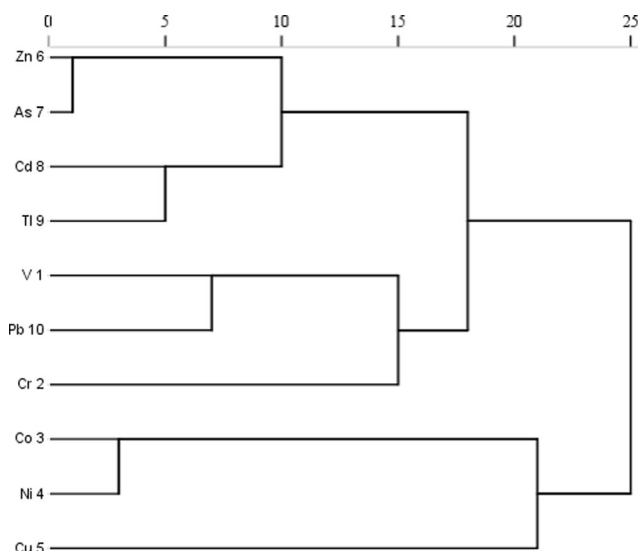


Fig. 6 Dendrogram of the studied elements from the Ganjiang River

Ganjiang basin, we deduced that Zn, As, Cd, and Tl were identified as pollutants mainly originated from the mineral exploitation and urban activities; V, Cr, and Pb mainly derived from mixed sources of anthropogenic and lithogenic origins; and Co, Ni, and Cu from lithogenic sources.

As evident from Tables 4 and 5 and Fig. 6, one group of elements, including Zn, As, Cd, and Tl, which indicated strong relationships in PCA, CA, and Pearson's correlation analysis, had a similar spatial distribution with the highly polluted sites mainly located in the upstream of the Ganjiang River. Being one of the mineralized areas, the regions in the upstream of the Ganjiang River basin were under huge pressure of resource exploitation. Wastewater discharging from tungsten ore dressing plants contains high Cd concentration (Zhang et al. 2014). It distributed some tungsten mines at the upstream of site S5, and Cd concentration in soils was high up to 52 mg/kg, which was almost 500 times the background value (0.1 mg/kg) (Liu et al. 2008). This situation inevitably affected the water quality in the downstream. The levels of Zn and As were higher at sites near the mining and urban activities. Moreover, these elements had significant spatial variations. It was inferred that Zn and As mainly originated from mining and industrial wastewater (Liu et al. 2008; Li et al. 2013). Although Tl concentration in surface water from the Ganjiang River was lower than other element concentrations, it had significantly positive relationship with Cd (0.615, $p < 0.01$) as proven by Pearson's correlation analysis. Moreover, Tl entered in the environment largely from metal mining and smelting (Tatsi et al. 2015). Therefore, we deduced that these four elements were the pollutants predominantly originated from mining and urban activities.

The second group of elements was associated with V, Cr, and Pb, which had significantly positive correlations in PCA, CA, and Pearson's correlation analysis (Tables 4 and 5 and Fig. 6). However, V and Pb behaved significantly positive correlations with elements in PC1, while Cr only had a significant relationship with Tl (Table 5). Additionally, previous studies reported that V was greatly influenced by anthropogenic activities such mining and agricultural processes (Li et al. 2008), and Pb was used as tracers of vehicles, agrochemicals, and industrial wastes (Krishna et al. 2009). Large areas of farmland were distributed along the middle and downstream of the Ganjiang River (Fig. S1), which were invested amount of agrochemicals. It was estimated that the average amount of fertilizer used in the arable land is 478.67 kg/hm² and is 2.41 times higher than the average amount of fertilizer used around the world, which is possibly the source of V and Pb in the Ganjiang River (Xie et al. 2016). Further, lead dioxide was the industrial raw material applied to lead batteries. Many battery enterprises (such as the Lvneng Electronics Technology Co., Ltd, the Huineng Electronics Technology Co., Ltd, the Shenzhou Electronics Technology Co., Ltd, and Weima power technology Co., Ltd) were attracted to

Table 5 Pearson correlation matrix of dissolved trace elements in the Ganjiang River, China

	V	Cr	Co	Ni	Cu	Zn	As	Cd	Tl	Pb
V	1.000									
Cr	<i>0.416**</i>	1.000								
Co	0.130	−0.035	1.000							
Ni	0.175	0.196	<i>0.666**</i>	1.000						
Cu	0.076	0.084	0.242	<i>0.306*</i>	1.000					
Zn	<i>0.329*</i>	0.213	0.211	0.150	0.172	1.000				
As	<i>0.398**</i>	0.240	<i>0.441**</i>	<i>0.367**</i>	0.153	<i>0.720**</i>	1.000			
Cd	<i>0.289*</i>	0.142	<i>0.449**</i>	<i>0.484**</i>	0.125	<i>0.509**</i>	<i>0.705**</i>	1.000		
Tl	<i>0.454**</i>	<i>0.372**</i>	0.044	0.243	0.002	<i>0.358**</i>	<i>0.473**</i>	<i>0.615**</i>	1.000	
Pb	<i>0.575**</i>	<i>0.382**</i>	0.114	0.067	0.198	<i>0.438**</i>	<i>0.510**</i>	<i>0.261*</i>	<i>0.366**</i>	1.000

Italic values represent correlation with significance

***Correlation significant at the 0.01 level (two tailed); correlation significant at the 0.05 level (two tailed)

locate in the Ganjiang River basin, especially in the Yichun city located in the midstream (Table S5). The environment around the enterprises contained high Pb level, which could be considered as the main anthropogenic sources of Pb discharges in the surface water (Liu et al. 2014). Therefore, this group can be attributable to mixed sources of anthropogenic and geogenic origins.

Co, Ni, and Cu in the third group, which were clearly separated from other elements in PCA and CA (Table 4 and Fig. 6), seemed to be associated to the earth’s crust and the geological formation of the area. These elements had low concentrations with small variability when the abnormal local site (site S6) was excluded. Additionally, Cu and Ni shared relatively lower pollution level among trace elements. Furthermore, these elements are therefore assumed to derive from natural contribution, also confirmed by their strong geochemical correlation (Table 5).

The categorization for risk area

Cluster analysis (CA) was generally applied to group the similar sampling sites based on the spatial variability of element concentrations in previous studies (Li and Zhang 2010; Varol et al. 2013). In our study, CA was used to categorize different risk areas on the basis of variability of risk for aquatic life (HPI) and human health (HI and cancer risk). CA rendered a dendrogram (Fig. 7), in which all 20 sampling sites along the Ganjiang River were categorized into three statistically significant clusters at $(Dlink/Dmax) \times 25 < 5$. Cluster 1 consisting of two sites associated with S5 and S6 corresponded to sites with the highest risk level for human health and relatively high-risk level for aquatic life, which were consequently taken as the high-risk area. Cluster 2 only consisting of one site (S10) corresponded to sites with moderate risk level both for aquatic life and for human health, which was grouped as moderate-risk area. The rest of sites that clustered as one

group correspond to relatively low-risk area. Moreover, sites in the same category were also regarded as having the same pollution source (Li and Zhang 2010). According to the previous results, we ascribe the three groups to mixed mining and urban activities (S5 and S6), predominantly urban activities (S10), and primarily natural contributions, respectively.

Conclusion

Among the ten selected dissolved trace elements, Zn was the most abundant element in the surface water from the Ganjiang River, whereas Tl was the least abundant. The highest total element concentrations were observed at sites where there are distributed intensive mining and urban activities. With a few exceptions, the current concentrations of trace elements in surface water from the Ganjiang River were basically in accordance to the quality standards set by China (GB5749-

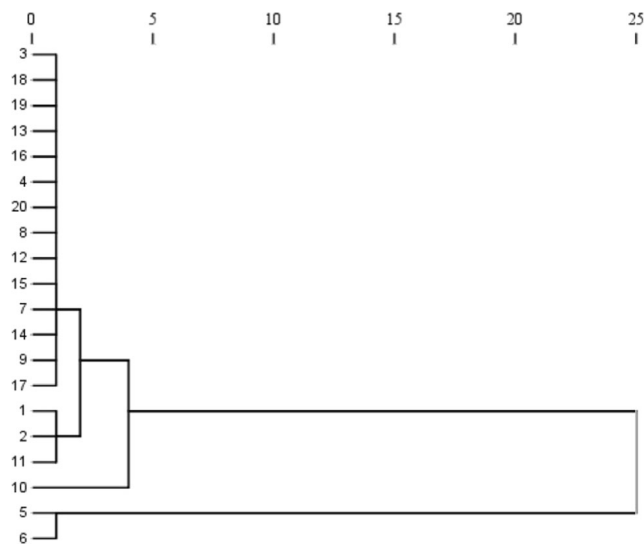


Fig. 7 Dendrogram of the sampling sites from the Ganjiang River

2006), WHO, EC, and USEPA. The water from the Ganjiang River was basically clean, which can be used as habitat for aquatic life. According to the health risk assessment, it was indicated that As and Tl are the major contributors to the exposure of the inhabitants. However, only As was identified as the priority pollutant of concern among the selected trace elements only at particular sites, causing both non-carcinogenic and carcinogenic effects. The adults were less sensitive to the exposure risks compared to children, and the oral ingestion was the primary exposure pathway. Multivariate statistical analyses revealed that different group elements were characteristics of the disparate source associated with mineral exploration, urban activities, and geogenic origins. Hierarchical agglomerative CA grouped all the sampling sites into three clusters based on the data set of exposure risk to human health and aquatic life. Most sites along the Ganjiang River were categorized as the low-risk area, while the three sites were identified as high- and moderate-risk area, respectively.

Acknowledgments This work was supported by the Opening Fund of Key Laboratory of Poyang Lake Wetland and Watershed Research (Jiangxi Normal University, Ministry of Education) under Grant PK2013002, the Collaborative Innovation Center for Major Ecological Security Issues of Jiangxi Province and Monitoring Implementation under Grant No. JXS-EW-00, and the National Natural Science Foundation of China under Grants 41401587 and 41201033.

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