

# Spatial distributions, fractionation characteristics, and ecological risk assessment of trace elements in sediments of Chaohu Lake, a large eutrophic freshwater lake in eastern China

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**Abstract** The concentrations, spatial distribution, fractionation characteristics, and potential ecological risks of trace elements (Cu, Pb, Zn, Cr, Ni, and Co) in the surface sediment samples collected from 32 sites in Chaohu Lake were investigated. The improved BCR sequential extraction procedure was applied to analyze the chemical forms of trace elements in sediments. The enrichment factor (EF), sediment quality guidelines (SQGs), potential ecological risk index (PERI), and risk assessment code (RAC) were employed to evaluate the pollution levels and the potential ecological risks. The results found that the concentrations of Cu, Pb, Zn, Cr, Ni, and Co in the surface sediments were 78.59, 36.91, 161.84, 98.87, 38.92, and 10.09 mg kg<sup>-1</sup>, respectively. The lower concentrations of Cu, Pb, Zn, Cr, and Ni were almost found in the middle part of the lake, while Co increased from the western toward the eastern parts of the lake. Cr, Ni, Co, and Zn predominantly existed in the residual fractions, with the

average values of 76.35, 59.22, 45.60, and 44.30%, respectively. Cu and Pb were mainly combined with Fe/Mn oxides in reducible fraction, with the average values of 66.4 and 69.1%, respectively. The pollution levels were different among the selected elements. Cu had the highest potential ecological risk, while Cr had the lowest potential ecological risk.

**Keywords** Chaohu lake · Trace elements · Sediments · Elements fraction · Ecological risk assessment

## Introduction

With the increasingly high requirement for water quality and the emerging water pollution, the trace elements contamination in water becomes a growing global concern in recent decades (Akinici et al. 2013; Chowdhury et al. 2016; Malferrari et al. 2009; Markert et al. 1997; Martin 2000; Nazeer et al. 2014; Pertsemli and Voutsas 2007; Santos Bermejo et al. 2003; Singh et al. 2005). The levels of trace elements concentrations provide important information for the evaluation of the quality of eco-environment including surface water, ground water, sea water, and soil. Due to their environmental persistence, biotoxicity, and accumulation in organismal tissues through the food chain (Sadiq et al. 2007; Sakan et al. 2009), trace elements contamination has been a great threat to ecosystems and human health. The trace elements in aquatic ecosystems can be sourced from both natural processes and anthropogenic activities. The anthropogenic activities include industrial effluents, domestic sewage, overland runoff, agricultural activities, and atmospheric deposition, while the natural sources are mainly through geological weathering and erosion (Diaz-de Alba et al. 2011; Mucha et al. 2003; Karageorgis et al. 2002). Trace elements from

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various sources can be accumulated in sediments of the aquatic environment and may be released back to the water column under proper conditions. Trace elements in sediments exhibit different chemical forms, combining with carbonates, sulfides, organic matter, iron and manganese oxides, and residues by various physicochemical strengths, thus demonstrating different mobility, bioavailability, biotoxicity, and ecological risk. Therefore, the detailed investigation on the element occurrence in sediments is of great significance for trace element pollution in aquatic systems.

Chaohu Lake (N 31° 43' 28"—31° 25' 28", E 117° 16' 54"—117° 51' 46") is the fifth largest freshwater lake in China with an area of 770 km<sup>2</sup> and a mean depth of 2.7 m (Wang et al. 1998, 2002). The lake basin covers an area of 13,486 km<sup>2</sup> of Anhui province and involves 13 cities and counties with a population of over 12 million. The main inflowing rivers of Chaohu Lake include Hangbu River, Paihe River, Tangxi River, Shiwuli River, Nanfei River, Zhagao River, Zhaohe River, and Baishitian River. A total of above 90% water input is derived from the eight tributaries. The water of Chaohu Lake is discharged solely through the Yuxi River and entered into the Yangtze River finally. Chaohu Lake plays an increasingly important role in drinking water supply, commercial fishing farming, industrial and agricultural irrigation water supply, navigation, flood prevention, and tourism activities. However, with rapid industrial development, urban expansion, and the increasing intensity of agricultural activities in the watershed, large amounts of contaminations were disposed to the lake and resulted in serious water and sediment pollution and degeneration of ecological service function of the lake. All of the problems have negatively affected the using functions of the lake, posing great pressure on the development of society and economy in the basin. Due to the important roles and the urgent requirement of water pollution control, Chaohu Lake was listed in “water pollution prevention and control on the three rivers and three lakes” by the state environmental protection administration of China in 2000. Previous studies were mainly related to nutrients (nitrogen and phosphorus) and eutrophication of Chaohu Lake (Wang et al. 2002; Wei et al. 2008; Yang et al. 2013; Zhang et al. 1999; Zhu et al. 1993), while trace elements in Chaohu Lake have begun to receive attention until this decade. Published studies on trace elements in sediments of Chaohu Lake mostly aimed at total concentrations of elements and the associated risk evaluation (Cao et al. 2004; He et al. 2016; Liu et al. 2012; Wang et al. 2016; Wen et al. 2012; Yin et al. 2011; Zan et al. 2011; Zhang et al. 2007), while the studies that analyzed chemical forms of elements are limited (Li et al. 2013; Liu and Shen 2014; Zhang et al. 2015). Zhang et al. reported that no obvious temporal variations of total element contents were

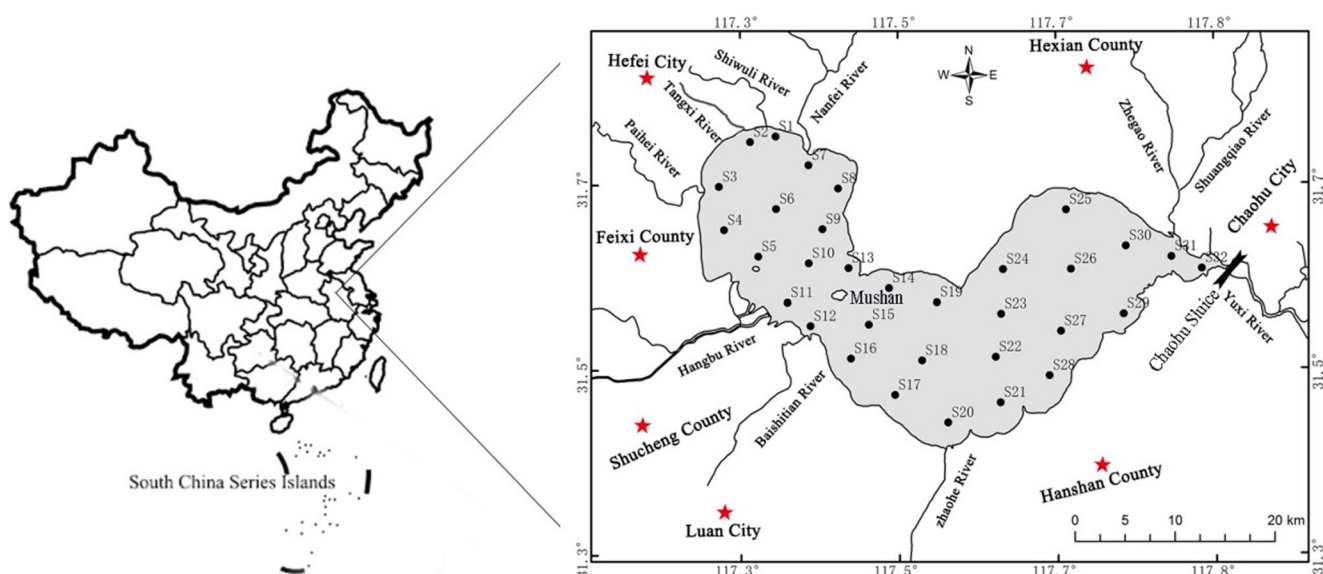
observed in a 12-consecutive-month study (Zhang et al. 2007); however, as the chemical forms can transform to each other along with the variation of environmental factors, such as pH, temperature, dissolved oxygen (DO), oxidation–reduction potential, sediment disturbances, and microbes (Chen and Lin 2001; Zhang et al. 2010), we deduce that the chemical partitioning of trace elements may vary along with seasonal alteration. Published data on trace elements in Chaohu Lake sediments covered samples collected in spring, autumn, and winter, but detailed studies of chemical forms as well as total contents of trace elements in sediments of the entire Chaohu Lake in summer are considered to be little. What is more, previous studies on potential risk assessment of trace elements in Chaohu Lake sediments were based on either total contents or chemical forms, but comprehensive assessment using multi-methods based on both total contents and chemical forms of trace elements is scarce.

Therefore, the main objectives of this study were (1) to study the spatial distributions of trace elements (Cr, Cu, Ni, Pb, Zn, and Co) in surface sediments of Chaohu Lake, (2) to analyze the chemical fractions of trace elements in surface sediments of Chaohu Lake in summer, and (3) to assess the potential ecological risks by the approaches of enrichment factor (EF), sediment quality guidelines (SQGs), and potential ecological risk index (PERI) based on total concentrations, and risk assessment code (RAC) based on chemical forms of trace elements.

## Materials and methods

### Study area

Chaohu Lake is located in the middle and lower reaches of Yangtze River, which is the longest one in China. The source rocks of the river system in Chaohu Lake basin include metamorphic rocks, volcanoclastic rock, intrusive rock, sedimentary clastic rock, and carbonate rocks, while the rocks surrounding Chaohu Lake consisted of red quartz sandstone, gray sandstone, clay or loam containing iron–manganese concretion, and gray sandy loam (Fang 1999). The lakebed of Chaohu consists of hard loess materials, and the sediments are almost entirely composed of silt (Zhang et al. 2007). The studied basin is subjected to a northern subtropical humid monsoon climate, with the annual average temperatures and the annual average precipitation of 16 °C and 1132 mm, respectively. The water column has no obvious stratification (Zan et al. 2011) due to its shallowness and the frequent winds and waves. The water of the lake flows from west to east and then enters into the Yangtze River through the Yuxi River before Chaohu Sluice was constructed in 1963 (Fig. 1). Since then, the water flow is mainly driven by the wind when



**Fig. 1** Location of sampling sites of the surface sediments from Chaohu Lake

the sluice is closed. It recovers the previous flow direction driven by the gravity with the opening of the sluice in wet season. The long-time closing of the sluice in 1 year leads to a sharp reduction of water exchange from  $1.36 \times 10^9$  to  $1.72 \times 10^8$  m<sup>3</sup> between Chaohu Lake and the Yangtze River (Wang and Tang 2010). The lake is divided into the west part and the east part by Mushan Island (Fig. 1). The water quality and the eutrophication degree are varied due to different pollution sources. The west part and the east part are at medium and light eutrophication level, respectively (Anhui environmental state bulletin 2014 and 2015). The northwest corner close to Hefei City (the capital of Anhui Province) has deteriorated into high eutrophication level, where algal density could even reach  $10^8$ – $10^9$  cells per liter water in summer.

### Sediment sampling

A total of 32 surface sediment samples were collected in Chaohu Lake (Fig. 1) in July 2014. The sampling sites were selected based on the following principles: on one hand, the sites were distributed uniformly to provide good area coverage; on the other hand, each estuary of the eight main rivers flowing into Chaohu Lake introduced above was sampled at one sampling site. Surface sediment samples (from 0 to 5 cm depth) were collected using a gravity corer (Rigo Co., Japan) with a pre-cleaned Plexiglas cylindrical tube (diameter 110 mm and length 500 mm) by the research vessel of Bureau of Chaohu Lake Basin of Anhui Province. All samples were kept in dark-colored polyethylene bags immediately after collection. Upon return to the laboratory, samples were air-dried in a laminar flow hood, sieved through nylon sieves (particle diameter < 75 μm), and stored in polyethylene bottles at 4 °C.

### Analytical methods and quality control

#### *Analysis of physicochemical properties*

The oxidation reduction potential (ORP) of the lake water (about 1 m below the water surface) of each sampling site was detected directly by using a multi-parameter controller (YSI-EXO2; ViaSat, America) when sampling on the lake. The grain size, electric conductivity (EC), and pH value of sediment samples were analyzed in the laboratory. The grain-size distribution of the fresh sediment samples was analyzed using a laser particle analyzer (Mastersizer 2000; Malvern, Britain), and then the content of clay (< 2 μm), silt (2–20 μm), and sand (> 20 μm) were calculated accordingly. EC and pH values were determined by detecting the supernatant using a conductivity meter (DDSJ-308A, Shanghai, China) and pH meter (pHSJ-3F, Shanghai, China), respectively. The supernatant was gained by extracting the dry sediment sample using deionized water ( $m_{\text{sample}}/V_{\text{water}} = 1:2.5$  for pH,  $m_{\text{sample}}/V_{\text{water}} = 1:5$  for EC) according to the methods issued in 2007 and in 2016 (Ministry of Agriculture of the People's Republic of China 2007; Ministry of Environmental Protection of the People's Republic of China 2016), respectively. The quality of pH and EC values was controlled by analyzing three replicates for each sediment sample.

#### *Analysis of trace elements*

Trace elements were partitioned into four fractions (acid-soluble fraction, reducible fraction, oxidizable fraction, residual fraction) by an improved BCR sequential extraction procedure (Nemati et al. 2011). Acid-soluble fraction includes water-leachable, ion-exchangeable, and carbonate-bound forms; reducible fraction means Fe/Mn oxide-bound fraction;

oxidizable fraction includes forms of both sulfides and organic matters; and residual fraction is bound to the silicate. The concentrations of trace elements in sediment digests and different sequential extracts were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES, Optima 2100DV; PerkinElmer). The improved BCR sequential extraction procedure (Nemati et al. 2011) are summarized as follows:

1. 1 g of each dry sediment sample was first extracted using 40 mL CH<sub>3</sub>COOH (0.11 M) by 16 h of continuous shaking at room temperature, and the extract was gravimetrically separated from the solid phase by centrifugation at 6000 rpm for 15 min.
2. The solid residue obtained from step 1 was mixed with 40 mL of NH<sub>2</sub>OH·HCl (0.1 M). After 16 h of continuous shaking at room temperature, the extract was centrifuged as described in step 1 and then decanted into a beaker.
3. The solid residue from step 2 was mixed with 10 mL of H<sub>2</sub>O<sub>2</sub> (8.8 M) and occasionally shaken at room temperature for 1 h followed by water bath warming for another 1 h at 85 ± 2 °C. This step was repeated until the volume of the mixture was reduced to 2–3 mL. Subsequently, 50 mL of NH<sub>4</sub>Ac (1 M) was added into the residue. The samples were shaken and centrifuged as described in step 1 and decanted into a clean beaker.
4. The residue from step 3 was digested with a mixture of aqua regia and HF (15 mL HCl + 5 mL HNO<sub>3</sub> + 2 mL HF), and then was dissolved entirely. Determination of total element concentrations in sediments was performed the same as the residue solid, using 1 g of each sample.

Quality control method was as follows: The instruments were checked daily with a Certified Reference Material (CRM) (GSD-9, GBW07309; Institute of Geophysical and Geochemical Exploration, China). The results indicated that the recoveries of total element content for the selected elements were in the range 93.4–105.6%. The recovery of the sequential extraction procedure (SEP) was calculated by the ratio of the summation of four fractions values of CRM to the total digestion content of CRM, and the SEP recoveries ranged from 96.01 to 103.40% for ICP-OES analysis.

**Assessment methods**

*EF pollution assessment method*

Single-element enrichment characterization in surface sediments of Chaohu Lake was evaluated by enrichment factor (EF). The EF is usually used to determine anthropogenic contributions to trace elements contents (Grant and Middleton 1990). Al has often been selected to reduce the influence of the sediment texture on trace element pollution due to its main

conservative lithogenic sources (Cao et al. 2014; Lin et al. 2016; Wang et al. 2015). The EF was calculated according to the following equation:

$$EF = \frac{(C_i/C_{Al})_{sediment}}{(C_i/C_{Al})_{background}}$$

where (C<sub>i</sub>/C<sub>Al</sub>)<sub>sediment</sub> is the element to Al ratio in sediment samples and (C<sub>i</sub>/C<sub>Al</sub>)<sub>background</sub> is the ratio in reference background. The reference concentrations of Cu, Pb, Zn, Cr, Ni, Co, and Al are 20.4, 26.6, 62, 66, 29.8, 16.3, and 67,200 mg kg<sup>-1</sup>, which are the background values of elements in soils of Anhui Province. The grading standards of EF were EF < 2, none-to-slight enrichment; 2 ≤ EF < 5, moderate enrichment; 5 ≤ EF < 20, significant enrichment; 20 ≤ EF < 40, very high enrichment; and EF ≥ 40, extremely high enrichment (Guo et al. 2015; Wang et al. 2015).

*SQG eco-risk assessment method*

The sediment quality guidelines (SQGs) were used for an assessment of the adverse biological effects of trace elements in studied surface sediments. Among numerical SQGs for freshwater ecosystems developed previously, consensus-based SQGs developed by MacDonald et al. are selected to be the reference value due to their confirmed high reliability in this study. For each contaminant of concern, the SQGs includes a threshold effect concentration (TEC) and a probable effect concentration (PEC) (Table 3) (MacDonald et al. 2000). Below the TEC, the toxicity effects of elements on organisms are negligible, and above the PEC, adverse biological effects are expected to occur frequently.

*PERI eco-risk assessment method*

The SQG method mainly evaluates the incidence of sediment toxicity by comparing the measured concentrations with the correlated TECs and PECs of trace elements, but it has a limitation in consideration of the biotoxicity and combined effects of trace elements. Hakanson proposed a PERI (potential ecological risk index) method to evaluate the ecological risk, which took the toxicology and overall levels of trace elements into consideration (Hakanson 1980). Here, the PERI was used to further assess the ecological risk of trace elements in surface sediments. The PERI is defined as follows:

$$PERI = \sum E_r^i = \sum T_r^i (C_s^i / C_n^i)$$

where PERI is the sum of potential ecological risks for all trace elements, E<sub>r</sub><sup>i</sup> is the PERI of an individual element, T<sub>r</sub><sup>i</sup> is the toxic-response factor of the interested element, C<sub>s</sub><sup>i</sup> is the measured element concentration, and C<sub>n</sub><sup>i</sup> is the reference

value of the element. The toxic-response factors for Cu, Pb, Zn, Cr, Ni, and Co are 5, 5, 1, 2, 5, and 5, respectively. The separate reference concentrations are as defined in “EF pollution assessment method” section.

The PERI value range of each risk grade is determined based on eight parameters (As, Cd, Cr, Cu, Hg, Pb, Zn, and PCBs) (Hakanson 1980). However, there are only six pollutants in this study, and the original PERI grading standard is not suitable to assess the eco-risk level in this study. The modified PERI grading standards in accordance with the number of pollutants assisted by toxicity coefficients of the pollutants were applied in this study, as Zhang et al. (2013) suggested. The classifications for  $E_r^i$  and modified PERI are shown in Table 1.

#### RAC eco-risk assessment method

The SQGs and PERI methods were based on total contents of trace elements in sediments, which cannot fully reflect the hazard to the environment. However, the behaviors including mobility, biological availability, and potential biotoxicity of trace elements in sediments are strongly influenced by their chemical forms. Therefore, the risk assessment code (RAC), which is determined by the acid-soluble fractions of trace elements, was used to assess the potential risk related to individual elements. The grading standards of RAC are shown in Table 1.

## Results and discussion

### Sediment characteristics

In order to compare the spatial difference of the determined parameters, Chaohu Lake was classified into three parts according to the lake morphology, the water area, and the hydrodynamic characteristics (Chen and Liu 2015) in this research, which were the western lake (WL; Fig. 1, S1–S13), the middle lake (ML; Fig. 1, S14–S23, S28), and the eastern lake (EL; Fig. 1, S24–S27 and S29–S32) from the west to the east.

General physical and chemical characteristics of the sediment samples as well as the corresponding ORP values of the lake water are presented in Table 2. The surface sediments of

Chaohu Lake were mainly composed of silt (67.2%), followed by sand (18.6%) and clay (14.2%). In addition, sand particles were found to be dominated by those with the equivalent diameters of 20–63  $\mu\text{m}$ . The average values of pH and EC in surface sediments of Chaohu Lake were 7.08 and 176.37  $\mu\text{S cm}^{-1}$ , respectively. The average ORP value of the water of the entire lake was 161.05 mV. The silt contents as well as the values of EC in the sediments were generally higher in WL and EL than in ML. The average pH and ORP values of three lake parts had no obvious difference.

### The concentrations and spatial distributions of trace elements

Table 3 lists the descriptive statistics of trace elements in surface sediments of Chaohu Lake. The concentrations of Cu, Pb, Zn, Cr, Ni, and Co in surface sediments collected at 32 sites ranged from 28.02 to 396.12, 17.42 to 60.28, 73.81 to 415.05, 68.28 to 129.98, 21.95 to 66.26, and 3.83 to 19.59  $\text{mg kg}^{-1}$ , respectively. The abundance of the determined elements follows an order of  $\text{Zn} > \text{Cr} > \text{Cu} > \text{Ni} > \text{Pb} > \text{Co}$ . The large coefficient of variance (Cv) indicates that the total concentrations of trace elements varied significantly among different sampling sites. This was obviously found for Cu and Zn, with the Cvs of 90.5 and 52.4%, respectively.

The spatial distributions of the total concentrations of Cu, Zn, Pb, Cr, Ni, and Co in surface sediments of Chaohu Lake are shown in Fig. 2 using contour maps. The results showed that the spatial distributions of Cu, Pb, and Zn were generally similar (Fig. 2a–c). A decrease of their concentrations is seen in the order of  $\text{WL} > \text{EL} > \text{ML}$ . The two highest concentrations for Zn, Pb, and Cu were found at S2 and S3, S1 and S8, and S2 and S3, respectively (Fig. 1). The abovementioned sites were located in the heavily polluted estuary area of the northwestern corner of Chaohu Lake. All domestic and industrial sewage from Hefei City, which was the capital of Anhui Province and carried about nearly half the population of the basin, were discharged into Chaohu Lake through the rivers distributed in the northwestern part of Chaohu Lake, leading to heavy contamination in this area. Cr and Ni were well distributed relatively in surface sediments compared with other elements in the whole Chaohu Lake. The spatial

**Table 1** The grading standards of PERI and RAC

PERI classes		RAC classes			
$E_r^i$	Risk	PERI	Risk	RAC	Risk
$E_r^i < 40$	Low risk	PERI < 25	Low risk	RAC < 1	No risk
$40 \leq E_r^i < 80$	Moderate risk	$25 \leq \text{PERI} < 50$	Moderate risk	$1 \leq \text{RAC} < 10$	Low risk
$80 \leq E_r^i < 160$	Considerable risk	$50 \leq \text{PERI} < 100$	Considerable risk	$10 \leq \text{RAC} < 30$	Moderate risk
$160 \leq E_r^i < 320$	High risk	PERI $\geq 100$	Very high risk	$30 \leq \text{RAC} < 50$	High risk
$E_r^i \geq 320$	Very high risk			RAC $\geq 50$	Very high risk

**Table 2** Grain size, pH, and electric conductivity (EC) of sediment samples, and oxidation–reduction potential (ORP) of the lake water of each sampling site

	Clay (< 2 μm) (%)	Silt (2–20 μm) (%)	Sand (20–63 μm/≥ 63 μm) (%)	pH	EC (μS/cm)	ORP (mV)
Minimum	9.42	53.48	5.41/0.00	6.74	76.90	119.00
Maximum	19.64	78.44	31.78/4.82	7.71	377.00	295.10
Lake average	14.20	67.20	17.62/0.98	7.08	176.37	161.05
Std <sup>a</sup>	3.10	8.00	8.98/1.49	0.23	76.40	32.60
WL average	13.65	71.70	14.58/0.07	7.03	218.05	161.28
ML average	14.03	59.62	24.62/1.73	7.12	119.85	158.86
EL average	14.78	67.22	16.65/1.35	7.09	186.35	163.69

<sup>a</sup> Std standard deviation

distributions of them (Fig. 2d, e) were generally similar, and their concentrations decreased in the order of EL ≈ WL > ML. Meanwhile, high concentrations of Cr and Ni were basically appeared in same sampling sites. The spatial distribution of Co exhibited a different pattern from the other five elements, with the trend of Co concentration increasing from WL toward EL (Fig. 2f). The zones with higher concentrations of Co were almost distributed in EL.

**Chemical fraction of trace elements**

Generally, trace elements that existed in non-residual forms were likely to mobilize from sediments to water column and impact organisms if environmental conditions change. Therefore, it was very important to gain the definite fraction

**Table 3** Concentrations of trace elements in surface sediments of Chaohu Lake (mg kg<sup>-1</sup>)

	Cu	Pb	Zn	Cr	Ni	Co
Minimum	28.02	17.42	73.81	68.68	21.95	3.83
Maximum	396.12	60.28	415.05	129.98	66.26	19.59
Lake average	78.59	36.91	161.84	98.87	38.92	10.09
Std <sup>a</sup>	71.16	10.95	84.74	15.94	8.70	3.87
Cv <sup>b</sup> (%)	90.5	29.7	52.4	16.1	22.4	38.4
WL average	127.38	42.96	230.58	103.83	41.10	8.98
ML average	42.24	32.37	108.39	88.39	33.70	10.31
EL average	49.30	33.31	123.63	105.22	42.57	11.61
TEC <sup>c</sup>	31.6	35.8	121	43.4	23	N/A <sup>e</sup>
PEC <sup>d</sup>	149	128	459	111	48.6	N/A
< TEC (%)	3.1	53.1	46.9	0	3.1	N/A
≥ TEC < PEC (%)	84.4	46.9	53.1	68.8	90.6	N/A
≥ PEC (%)	12.5	0	0	31.3	6.3	N/A

<sup>a</sup> Std standard deviation

<sup>b</sup> Cv coefficient of variance

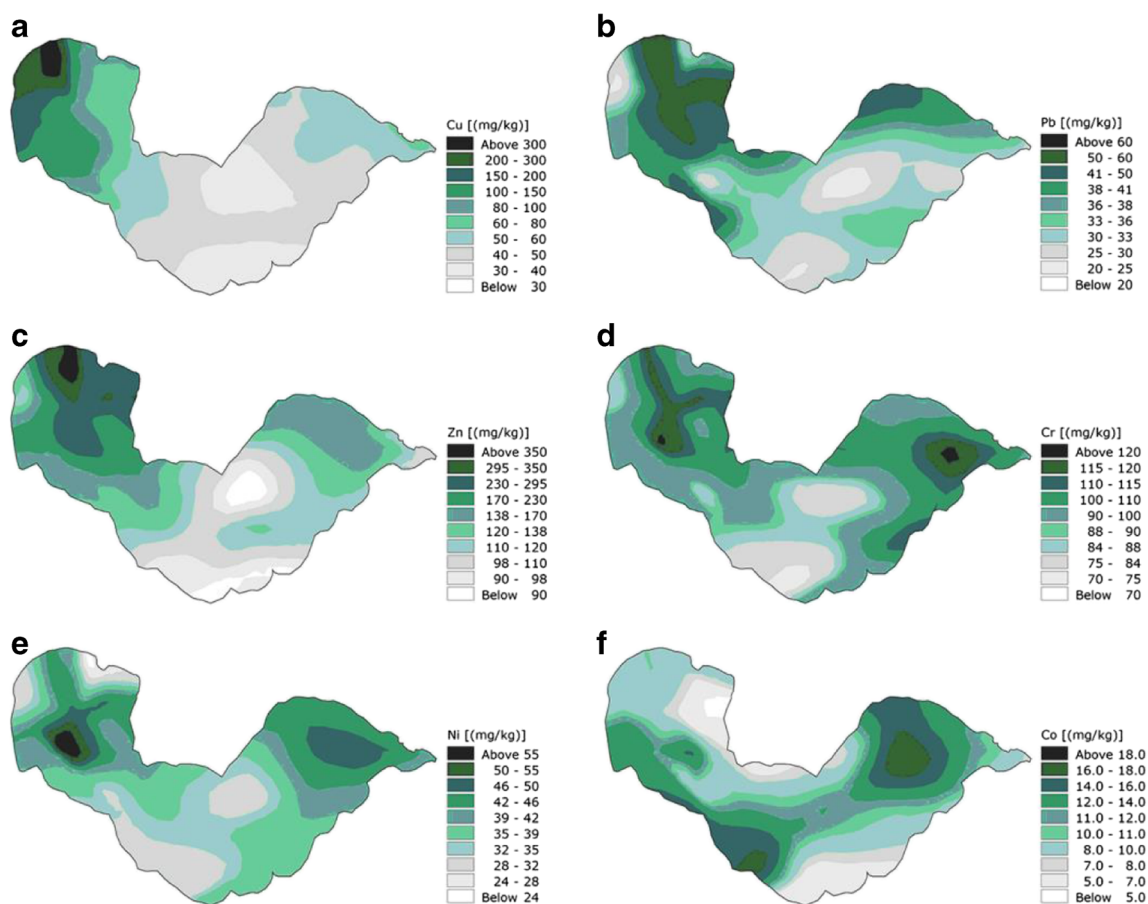
<sup>c</sup> TEC threshold effect concentration

<sup>d</sup> PEC probable effect concentration

<sup>e</sup> N/A not available

information of elements in sediments. The value ranges as well as the average value of each fraction of studied elements of 32 sediment samples are shown in Fig. 3 and Table 4, respectively.

Cu and Pb were primarily present in F2 fraction of sediments, with values of 66.4 and 69.1%, respectively. For Cu, the other three extractable fractions generally followed the order of F4 > F1 > F3 pattern, while for Pb, the order was F4 > F3 > F1. The results were similar to the study of Liu and Shen (2014). Liu and Shen reported that 34% of Cu and 59% of Pb in F2 fraction were observed as the primary chemical form in the surface sediments of the west Chaohu Lake sampled in April 2011 (on behalf of the spring) (Liu and Shen 2014). However, our results were rather different from the research of Li et al. in which Cu predominantly existed in F4 fraction (41.3%) with only 16.1% of F2 fraction, while 35.9% of Pb was present in F2 fraction as the main form in the surface sediments of the whole Chaohu Lake sampled in October 2009 (on behalf of the autumn) (Li et al. 2013). In addition, Zhang et al. reported that Cu was primarily present in F4 fraction with the value of about 50%, while Pb primarily occurred in F2 fraction with the value of about 55%, in the sediments of Chaohu Lake sampled in December 2013 (on behalf of the winter) (Zhang et al. 2015). According to the aforementioned, F2 fractions of Cu and Pb associated with Fe/Mn oxides were the predominant chemical form in summer sediments of Chaohu Lake and the values of F2 fraction were higher compared to those in other three seasons. Hydroxides of Fe/Mn can easily adsorb trace elements efficiently because of their large adsorption surface areas, especially for Pb (Ianni et al. 2010). Meanwhile, a high pH value and an oxidizing environment were promoted to form Fe/Mn oxides. According to the routine monitoring data, the average pH value of Chaohu Lake water in summer was usually higher than that in other seasons (for example, the pH values were 7.88, 8.50, 8.15, and 8.03 in spring, summer, autumn, and winter in 2014, respectively) due to frequent algae bloom outbreaks, which consumed much CO<sub>2</sub> in the aquatic system during the process of photosynthesis and led to the increasing of pH value. Moreover, the average ORP value of Chaohu

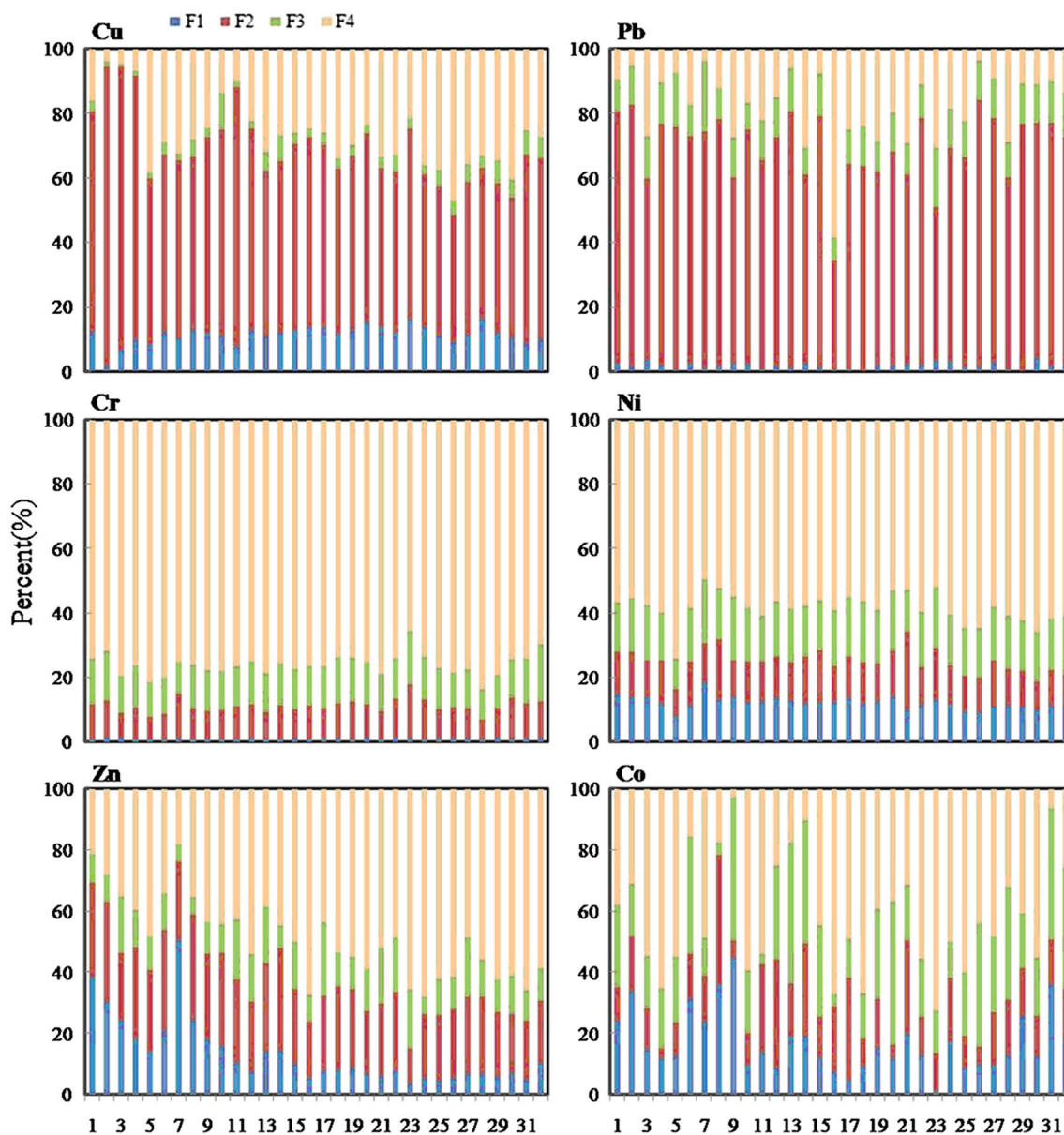


**Fig. 2** Spatial variations of total concentrations of the elements in surface sediments of Chaohu Lake

Lake water was 161 mV in summer, which provided a relatively oxidizing condition in the aquatic system. The physico-chemical conditions in surface sediments were susceptible to the water column due to the inconspicuous stratification in Chaohu Lake water. The average pH value of selected sediment samples was 7.08, indicating a weak alkaline environment which was relatively favorable for the formation of Fe/Mn oxides in surface sediments of Chaohu Lake. These reasons discussed above may explain why more proportions of F2 fraction of Cu and Pb were determined in this study. Zn was mostly associated with F4 fraction (44.26%), followed by F2, F1, and F3 fractions on average in the sediments. The main chemical form and the spatial distribution characteristic of the fractions of Zn were similar to previous studies on Chaohu Lake surface sediments (Li et al. 2013; Yu et al. 2013; Zhang et al. 2015). Interestingly, Cu, Pb, and Zn primarily existed in F2 fraction of the three bio-available forms without regard to F4 fraction. Furthermore, the percentages in each fraction of the three elements all varied greatly among sampling sites, especially for Zn. Zn was present in F1 at the highest rate (50.8%) at S7 near the Nanfei River mouth, while F4 fraction of this site was the lowest (18.3%). In contrast, Zn was present in F4 to a greatest extent (68.2%) at S24, with the relatively lower F1 fraction (5.5%) at this site. Spearman

correlation analysis was applied to demonstrate the relationships among the elements. The correlation coefficients of the total concentrations of the six elements are shown in Table 5. The results showed that Zn correlated with Pb ( $r = 0.756$ ) and Cu ( $r = 0.690$ ) significantly, while Pb had a good positive relationship with Cu ( $r = 0.475$ ). Liu et al. also reported that there existed a significant positive correlation about total concentrations among Pb, Cu, and Zn in surface sediments from Chaohu Lake and its main tributary rivers (Liu et al. 2016). Therefore, it may be suggested that Cu, Pb, and Zn might derive from similar sources, which were mainly anthropogenic sources due to their high portions of non-residual fractions.

Cr and Ni predominantly existed in F4 fraction, with the average value of 76.35 and 59.22%, respectively. The other three extractable fractions of these two elements both followed an order of F3 > F2 > F1. The results were in accordance with the studies of Li et al. (2013) and Zhang et al. (2015) (on behalf of the spring and winter, respectively). The F1 fraction of Ni was much higher than that of Cr over the whole lake, with values of Ni (8.1–18.3%) and Cr (0.9–1.3%). Moreover, the distributions of the fractions of the two elements were relatively uniform among sampling sites, which were consistent with the small Cvs of total Cr and Ni concentrations (Table 3). For total contents of Cr and Ni, high concentrations



**Fig. 3** Distribution characteristics of trace elements forms for sediments sampled at 32 sites. F1, acid-soluble fraction; F2, reducible fraction; F3, oxidizable fraction; F4, residual fraction

of Cr and Ni basically appeared in the same sampling sites. In addition, the positive correlation coefficient between Cr and Ni ( $r = 0.809$ ) suggested that Cr and Ni might derive from similar sources.

Co mainly existed in F4 fraction (45.6%). There was also an appropriate concentration of Co associated with F3 fraction. F2 fraction and F1 fraction of Co were approximately equal on the whole in Chaohu Lake. Like Zn, Cu, and Pb, the spatial distribution of Co fractions was also extremely uneven, showing F1 (1.2–44.7%), F2 (3.1–42.3%), F3 (3.6–47.1%), and F4 (2.9–72.3%). The result obtained in “The concentrations and spatial distributions of trace elements” section showed that total Co presented an entirely different distribution pattern in comparison with other five elements.

Meanwhile, the relationship of Co with other five elements demonstrated by correlation coefficients (Table 5) was very poor. Hence, the main sources of Co might be different from other five elements.

**Risk assessment**

*EF assessment*

In order to evaluate the anthropologic pollution level of the selected elements of Chaohu Lake, the EF values are calculated and listed in Table 6. The average enrichment degree of these elements in the whole lake follows an order of  $Cu > Zn > Cr > Pb > Ni > Co$ . The average EF value of Cu



**Table 4** Fraction of trace elements in surface sediments of Chaohu Lake (%)

		F1	F2	F3	F4
Cu	Range	1.67–16.50	39.31–93.05	0.67–11.10	4.01–47.09
	Mean	9.33	66.41	3.40	20.85
Pb	Range	0.01–4.26	34.15–82.89	6.92–21.59	3.61–58.53
	Mean	1.69	69.10	11.76	17.45
Cr	Range	0.73–1.51	6.18–16.52	9.33–17.83	65.60–83.76
	Mean	1.04	9.96	12.65	76.35
Ni	Range	8.08–18.34	8.27–23.88	9.58–19.97	49.54–74.07
	Mean	11.89	12.70	16.19	59.22
Zn	Range	3.60–50.77	11.51–34.32	5.51–24.00	18.28–68.18
	Mean	17.24	26.71	11.78	44.26
Co	Range	1.19–44.70	3.06–42.32	3.57–47.07	2.87–72.31
	Mean	15.05	15.84	23.47	45.63

was 6.87, reaching a significant enrichment due to the high EF values in WL. As for Zn, Cr, Pb, and Ni, the average EF values were 4.44, 2.57, 2.31, and 2.15, respectively, showing a moderate enrichment. The above five elements were considered to be polluted anthropologically. The average EF value of Co approximated 1, indicating a generally natural content level in the lake. The EF values of Cu, Pb, Zn, Cr, and Ni tended to decrease from WL to EL, while those of Co showed no obvious spatial variation. The highest value of Pb, Zn, Cr, Ni, and Co were found at S7 sample site, while that of Cu was in S3 site. Both S7 and S3 sites were located in heavily polluted estuary areas of the northwestern corner (Fig. 1) of the lake, where a high eutrophication problem existed these years.

*SQG assessment*

When compared to the TEC–PEC SQGs, the average concentrations of Cu, Pb, Zn, Cr, and Ni were between their corresponding TEC and PEC values (Table 3). The Cr concentrations in 31.3%, the Cu concentrations in 12.5%, and the Ni concentrations in 6.3% of 32 surface sediments samples in

**Table 5** Matrix of Spearman’s correlation coefficient of total concentrations of the six studied elements in surface sediments of Chaohu Lake

	Cu	Pb	Zn	Cr	Ni	Co
Cu	1					
Pb	0.475**	1				
Zn	0.690**	0.756**	1			
Cr	0.381*	0.670**	0.689**	1		
Ni	0.389*	0.636**	0.635**	0.809**	1	
Co	0.025	0.077	0.145	0.121	0.224	1

\*Correlation is significant at  $p < 0.05$  (two-tailed), \*\*correlation is significant at  $p < 0.01$  (two-tailed)

**Table 6** The EF values of the selected elements in surface sediments of Chaohu Lake

EF	Cu	Pb	Zn	Cr	Ni	Co
Minimum	2.68	1.39	2.35	1.79	1.64	0.3
Maximum	36.72	5.56	18.58	7.22	3.72	2.54
Lake average	6.87	2.31	4.44	2.57	2.15	1.07
Std <sup>a</sup>	7.38	0.82	3.12	1.01	0.46	0.55
WL average	11.63	2.74	6.55	2.86	2.33	1.01
ML average	3.82	2.23	3.20	2.49	2.09	1.22
EL average	3.33	1.73	2.71	2.21	1.96	0.95

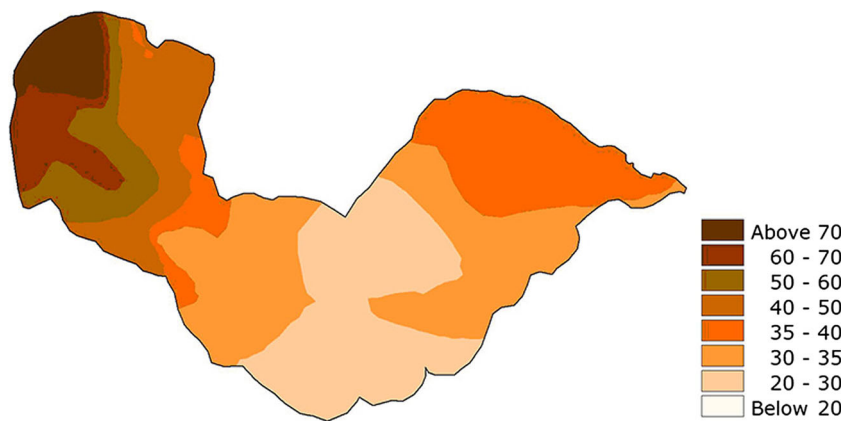
<sup>a</sup> Std standard deviation

Chaohu Lake were higher than PEC, indicating their potentially adverse biological effects. The concentrations of 84.4% of Cu, 46.9% of Pb, 53.1% of Zn, 68.8% of Cr, and 90.6% of Ni in 32 samples lie in the range between TEC and PEC. The Cu and Ni concentrations in 3.1%, the Pb concentrations in

**Table 7** Results of potential ecological risk assessment for trace elements in surface sediments of Chaohu Lake based on PERI method

Sampling site	$E_r$						PERI
	Cu	Pb	Zn	Cr	Ni	Co	
S1	25.52	11.33	6.69	3.29	6.90	2.95	56.69
S2	97.09	10.36	5.64	3.60	7.70	3.13	127.51
S3	51.41	4.28	1.67	2.56	4.86	2.55	67.33
S4	38.93	7.17	2.99	2.99	7.24	4.05	63.37
S5	37.59	8.54	4.05	3.72	11.12	3.81	68.84
S6	20.60	10.76	4.57	3.62	8.14	2.31	50.00
S7	20.04	5.50	3.67	2.86	3.68	2.52	38.27
S8	14.99	11.32	5.25	3.58	7.95	1.17	44.26
S9	16.76	6.88	2.32	2.60	6.18	1.41	36.14
S10	25.44	10.03	4.11	3.44	7.89	4.77	55.69
S11	29.31	7.12	2.40	2.76	6.24	2.19	50.02
S12	14.10	4.45	1.83	2.42	4.83	2.38	30.00
S13	14.10	7.24	3.16	3.46	6.91	2.57	37.44
S14	11.82	7.66	1.95	2.87	6.39	1.91	32.59
S15	13.83	5.96	2.43	2.97	6.31	3.62	35.12
S16	12.31	8.27	1.93	2.73	5.17	4.51	34.93
S17	10.02	5.77	1.52	2.35	5.01	5.34	30.01
S18	10.67	6.05	1.71	2.70	5.72	3.95	30.80
S19	9.59	6.54	1.71	2.45	5.53	2.80	28.61
S20	8.91	4.31	1.51	2.08	4.68	1.29	22.78
S21	8.89	5.48	1.37	2.74	6.47	1.91	26.87
S22	10.79	6.61	2.11	2.81	6.33	3.26	31.91
S23	6.87	3.27	1.19	2.19	4.36	3.78	21.66
S24	9.52	6.60	1.54	3.41	5.86	2.05	28.97
S25	11.80	8.01	2.45	2.88	7.67	4.82	37.62
S26	13.62	5.69	1.96	3.30	7.74	6.01	38.33
S27	9.78	6.08	1.91	2.75	6.67	3.04	30.23
S28	10.15	7.00	1.81	3.57	6.22	2.41	31.17
S29	10.00	5.11	1.96	3.40	6.79	3.74	31.00
S30	12.30	6.99	2.81	3.94	8.69	4.02	38.75
S31	15.86	6.56	1.65	3.19	6.96	2.71	36.93
S32	13.78	5.04	1.67	2.64	6.76	2.11	32.00
Average	19.26	6.94	2.61	3.00	6.53	3.10	41.43
Minimum	6.87	3.27	1.19	2.08	3.68	1.17	21.66
Maximum	97.09	11.33	6.69	3.94	11.12	6.01	127.51

**Fig. 4** Spatial distribution of PERI of trace elements in Chaohu Lake sediments



53.1%, and the Zn concentrations in 46.9% of the samples were below TEC, for which toxic effects on organisms were rarely expected to occur. Despite the measured Zn and Pb that were generally lower than PEC, 100% of Zn and 87.5% of Pb for the samples exceeded their background values.

**Table 8** The RAC values of trace elements in surface sediments of Chaohu Lake (%)

Sampling site	Cu	Pb	Zn	Cr	Ni	Co
S1	12.39	2.37	38.62	0.89	14.88	24.02
S2	1.67	1.38	30.42	1.31	13.89	33.80
S3	6.47	3.72	24.26	1.41	13.90	14.92
S4	9.82	1.91	18.10	0.99	11.87	11.83
S5	8.28	0.38	14.20	0.80	8.08	12.40
S6	12.19	2.41	21.03	0.76	11.38	31.16
S7	10.15	0.89	50.77	1.11	18.34	23.88
S8	12.93	1.66	24.41	0.87	13.10	35.87
S9	11.89	2.51	18.21	0.99	14.00	44.70
S10	11.05	2.07	15.82	0.99	12.05	9.62
S11	7.76	0.61	10.56	0.95	12.33	13.87
S12	12.75	1.58	7.47	1.10	13.76	8.80
S13	10.78	0.86	14.07	0.96	12.81	19.08
S14	12.05	2.34	14.27	0.98	11.92	19.14
S15	12.74	1.26	9.85	1.17	12.29	12.25
S16	13.82	0.40	5.61	0.84	12.38	7.62
S17	13.81	0.63	7.74	1.15	13.68	4.66
S18	11.61	0.01	8.16	1.01	11.77	9.29
S19	12.98	1.88	8.85	0.97	12.31	15.25
S20	15.11	1.75	6.76	1.30	14.26	11.74
S21	13.74	2.13	6.34	0.85	10.19	20.08
S22	12.07	1.97	7.72	1.33	11.50	12.75
S23	16.05	3.26	3.60	1.18	12.97	1.19
S24	13.80	2.93	5.47	0.91	11.68	17.09
S25	10.97	1.25	4.77	1.07	9.53	8.65
S26	9.10	1.37	5.66	1.05	9.07	10.04
S27	11.31	2.74	6.93	1.15	11.07	9.74
S28	16.50	0.39	6.93	0.73	11.25	12.89
S29	11.98	0.01	5.97	1.51	10.90	25.71
S30	10.36	4.26	7.11	0.94	9.68	12.51
S31	8.81	1.66	4.83	1.08	11.38	35.89
S32	10.01	2.20	10.27	1.29	11.41	17.83
Lake average	9.33	1.69	17.24	1.04	11.89	15.05
WL average	7.92	1.78	26.72	1.03	13.11	17.25
ML average	12.87	1.36	6.65	1.02	11.52	11.40
EL average	11.12	1.90	6.41	1.07	10.57	15.49

In general, the incidence of adverse effects of the above five elements in surface sediments of Chaohu Lake followed an order of Cr > Cu > Ni > Zn > Pb.

*PERI assessment*

Potential ecological risks of trace elements in Chaohu Lake surface sediments were calculated. Table 7 presents both the PERI of individual elements and the comprehensive potential ecological index of the six elements.

$E_r^i$  of each sample site for Pb, Ni, Co, Cr, and Zn were far less than 40, suggesting a low potential ecological risk. Cu in most of the sampling sites posed a low risk except for S2 and S3, showing a considerable risk and a moderate risk, respectively. The sampling sites with the highest degree of potential risks of trace elements were S2 for Cu, S1 for Pb and Zn, S30 for Cr, S5 for Ni, and S26 for Co, among which most sites were located in WL. The order of the mean potential ecological risk followed a Cu > Pb > Ni > Co > Cr > Zn pattern.

The calculated comprehensive PERI (Table 7) showed that two sediment samples of the overall 32 samples had a low risk. The selected elements in 23 sample sites were considered to pose a moderate risk, while those in six samples suggested a considerable risk. There was one sediment sample (S2) posing a high risk due to the very high content of Cu in this site. A contour map was used to show the PERIs for all sampling sites (Fig. 4). The contour map intuitively displayed a significant spatial variation of the PERIs. The levels of the risks generally decreased from WL to ML and then increased from ML to EL gradually. The area with the highest level of the risk was located in the northwestern corner of the lake. The whole spatial distribution of the PERIs matched the overall concentration tendencies for Cu, Pb, and Zn.

*RAC assessment*

The RAC values of trace elements in sediments of Chaohu Lake are listed in Table 8.

In view of the whole lake, Zn, Co, and Ni posed a moderate potential risk, while the risks of Cu, Pb, and Cr were low. The potential ecological risk broadly followed an order of  $Zn > Co > Ni > Cu > Pb > Cr$ . A similar order was also found in other studies that evaluated the potential ecological risks of trace elements in Chaohu Lake based on RAC (Liu and Shen 2014; Kong et al. 2015; Zhang et al. 2015). As for the spatial difference of each element RAC values, Cu presented a low ecological risk in WL while it posed a moderate risk in both ML and EL. The potential ecological risk of Zn in WL was moderate, which was higher than the low risk of ML and WL. The relatively high risk of Zn in WL was in accordance with its spatial distribution of overall accumulations in Chaohu Lake sediments (Fig. 2c). Zhang et al. reported that the highest concentration of Zn in the sediments of the 11 rivers flowing into Chaohu was observed in Nanfei River ( $722 \text{ mg kg}^{-1}$ , 12 times greater than the lowest value in the studied area), which flowed through Hefei City and eventually fed into the north-western lake (Zhang et al. 2015). The large amount input of Zn from Nanfei River may be the main reason leading to the high accumulation and high potential ecological risk in WL. Pb and Cr presented a low risk throughout the lake. The potential risks posed by Ni and Co were moderate in the three parts of the lake.

In summary, the selected elements were moderate to significant enrichment and were contaminated anthropologically except for Co in Chaohu Lake sediments. The eco-risk assessment results of the trace elements in Chaohu Lake sediments through three different methods were not in agreement with each other (SQGs— $Cr > Cu > Ni > Zn > Pb$ , PERI— $Cu > Pb > Ni > Co > Cr > Zn$ , RAC— $Zn > Co > Ni > Cu > Pb > Cr$ ). Therefore, we should evaluate the eco-risk grades of the elements comprehensively considering the advantages and limitations of each eco-risk assessment method combining with EF evaluation method. Cr had relatively higher potential risk according to SQG method. Nevertheless, it ranked the fifth and the last on the basis of PERI and RAC methods, respectively, due to the high background level in local soil and the low F1 fraction, respectively. What is more, the average EF value of Cr was only a little more than 2. Hence, Cr was seen as the element with the lowest eco-risk of the six elements studied in this work, posing a very low risk. Cu reached a significant enrichment degree in WL according to EF method and posed a higher risk than other elements based on SQGs and PERI methods, while it ranked a little behind by RAC values. However, in consideration of the high F2 fraction (66.41%, Table 4), some of which might transfer into F1 fraction in the event of a change in environmental conditions (pH, oxidation reduction potential, etc.), we considered Cu had the highest eco-risk of the six elements in this study with a low to moderate risk. Ranking between Cu and Cr, Zn and Ni possessed low to moderate risks, while Pb and Co presented low risks comprehensively.

## Conclusion

(1) This study analyzed the concentrations, spatial distributions, chemical fractions, and ecological risks of trace elements including Cu, Pb, Zn, Cr, Ni, and Co in surface sediments from Chaohu Lake. Their average concentrations were 78.59, 36.91, 161.84, 98.87, 38.92, and  $10.09 \text{ mg kg}^{-1}$ , respectively. (2) Except for Co, the element concentrations in ML were relatively lower than those in EL and WL, with the concentrations of Cu, Pb, and Zn decreasing in the order of  $WL > EL > ML$ , and those of Cr and Ni decreasing in the order of the  $EL \approx WL > ML$ . (3) The trace elements, other than Cu and Pb, were mainly bound in the residual fraction, with the average values of 76.40% for Cr, 59.20 for Ni, 45.60% for Co, and 44.30% for Zn. However, Cu and Pb mainly existed in the reducible fraction, with the average values of 66.4 and 69.1%, respectively, which were higher than those determined in previous studies sampled in other seasons. Among these trace elements, Cr was the least mobile element because of the lowest F1 percentage and the highest F4 percentage. (4) Taking fully into account the risk evaluating results by four different procedures, Cu, Zn, and Ni posed low to moderate risk to the ecosystem environment, while Pb, Co, and Cr presented low risk. Therefore, it is necessary to pay more attention to Cu, Zn, and Ni in the future environment management and pollution control of Chaohu Lake, especially of the western lake.

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