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Spatial–temporal distribution characteristics and source analysis of sediments in Qinghai Lake, China

Hai-lin Niu[a,b,c], Guang-wei Zhu[a,b,*], Meng-yuan Zhu[a], Bo-qiang Qin[a], San-chuan Ni[c]

[a]*State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China, emails: gwzhu@niglas.ac.cn (G.-w. Zhu), hlniu@qq.com (H.-l. Niu), myzhu@niglas.ac.cn (M.-y. Zhu), bqain@niglas.ac.cn (B.-q. Qin)*
[b]*University of Chinese Academy of Sciences, Beijing 100049, China*
[c]*College of Ecological and environmental Engineering, Qinghai University, Xining 810016, China, email: nisch@sina.com (S.-c. Ni)*

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ABSTRACT

Sediment is not only an important component of lake ecosystem, but also an important element and interface of geochemical cycle of lake eutrophication. Carrying abundant information of natural environment and mode as well as intensity of human activities on environment, lake sediments are vital to the water environment of lakes. In this paper, nutritive salt and heavy metal elements from different depths of sediments collected from eight representative sites in Qinghai Lake were analyzed, in order to demonstrate the spatial distribution law and accumulation pattern of nitrogen form, phosphorus form, organics, and typical heavy metal elements. Results showed significantly uneven spatial distribution of total nitrogen and organics in the surface sediments in Qinghai Lake, while total phosphorus in the surface sediments presented a relative equilibrium spatial distribution. Vertical distribution of nutritive salt in sediment cores differs from each other. Heavy metals in sediments have relative stable natural source and most heavy metal elements enjoy higher homology, which can be accumulated in sediments through coprecipitation or absorption. The fluctuating vertical distribution of heavy metals may be mainly caused by human activities. This research results can be served as scientific basis for water pollution and heavy metal pollution prevention in sediments in Qinghai Lake.

Keywords: Sediments; Spatial–temporal distribution; Source; Qinghai Lake

1. Introduction

As an important component of lake ecosystem, lake sediment is not only the core indicator of nutrient substance cycle in lakes, but also the major reservoir of pollutants. Accumulation and discharge of lake sediment pollution are important causes of lake pollution [1]. Nutritive salt and heavy metals get into lake water through various approaches, which will finally flocculate or subside in surface sediments [2,3]. With characteristics of strong migration resistance, heavy toxicity, and biological accumulation, they are difficulty to be discharged after getting into lakes, especially undrained lakes [4–7]. Heavy metals in surface sediments are easy to be disturbed and can be discharged into aqueous phase under certain conditions to cause secondary pollution, thus having long-term impacts on the lake ecosystem [8–10]. Considering the significance of sediments in endogenous loading mechanism of lakes, characteristics and occurrence modes of nutrients and heavy metals in sediments are the key of eutrophication studies. Sediments' "converging and sourcing" effect of nutrients in overlaying water plays an important role in not only the biological, physical, and chemical cycle of substances in the whole lake system, but also in the control of nutrient content in the lake ecosystem.

* Corresponding author.

Many researches on sediment pollution of lakes in south China (e.g., Chaohu Lake, Taihu Lake, Dian Lake, and East Lake) have been reported, which provide theoretical basis for studying and management of other lakes. The rapid industrial and agricultural development aggravates river and lake pollutions, thus requiring urgent pollution control [11,12]. Some research results on river pollution have been achieved in recent years in considering of territory and features. For example, researches on pollution situation and source of rivers running through cities (towns) provide environmental pollution information, and explorations on the pollution level of rivers running through mining areas provide basis for water environment treatment in mining areas [13–15]. Furthermore, the sediment pollution in the Pearl River, Suzhou River, Xi River, Huai River, and Yellow River as well as their tributaries was studied. Foreign associated researches were reported earlier and developed rapidly, mainly covering: (1) evaluation analysis on sediment pollution in oceans, laying a foundation to the management of polluted oceans; (2) research on characteristics of sediment pollution in rivers, finding that sediments are the source sink term of heavy metals in water environment; and (3) discussions on the effect of deposition age of sediments on their characteristics, evaluation of lake sediment pollution, and mechanism analysis of lake sediment pollution from the dynamics perspective.

Heavy metal pollution in lake sediments also attracts attentions from researches gradually. Main research contents include correlation between heavy metals and organics, heavy metal source and evaluation, as well as distribution characteristics, pollution status, and availability of heavy metals in sediments in different water bodies [16–18]. Foreign scholars studied sources of sediments and heavy metals through multivariate analysis, which discussed the relationship between heavy metal content in sediments and heavy metal load in water, sediment–water interface environmental conditions as well as properties of sediments. They explored heavy metal ion absorption mechanism of natural water as well as the absorption mechanism and model of organic acids in sediments and soil. Researches on the pollutant content in lake sediments, contaminant transport in sediment–water interface and associated influencing factors, pollutant absorption and desorption mechanism of sediments, as well as sediment quality criteria have been reported in foreign countries, achieving outstanding fruits.

Qinghai Lake, the largest undrained salt lake in inland plateau in China, is in semi-salinization development stage, which is manifested by water level fall, lake shrinking, and salified water [19–21]. It has attracted wide attentions from the public. As a typical plateau lake, Qinghai Lake has unique water environmental capacity variation, geochemical cycle of water and sediment biogenic elements, heavy metal pollution in sediments, and nutrient transformation under low temperature and frozen conditions [22,23]. In this paper, the distribution characteristics of nutritive salt and heavy metal elements on surface and vertical sediments collected from eight representative sites in Qinghai Lake were analyzed, which disclosed the current spatial distribution law and accumulation pattern of nitrogen form, phosphorus form, organics, and typical heavy metal elements in the nutritive salt. Additionally, sources of nutritive salt and heavy metals were discriminated preliminarily through factor analysis, aiming to provide scientific basis for water pollution and heavy metal pollution prevention in sediments in Qinghai.

2. Materials and methods

2.1. Study area and sampling site

Qinghai Lake lies in the northeast of Qinghai-Tibet Plateau, covering a drainage area of 29,661 km^2 and a lake area of 4,400 km^2. It has a water volume of 7.39×10^{10} m^3, average depth of 21.7 m, annual water supply of 3.63×10^9 m^3, and annual water consumption of 4.16×10^9 m^3. It has about 50 inflow runoffs which are concentrated in the north of Qinghai Lake, showing an obvious asymmetrical distribution. The 262 m long Buha River is the largest river in the Qinghai Lake basin, covering a drainage area of 14,337 km^2 and an annual runoff of 7.85×10^8 m^3 (60% runoff of the whole Qinghai Lake). Its flood season lasts from June to September. Qinghai Lake has six major inflowing rivers, namely Buha River, Shaliu River, Quanji River, Haergai River, Ganzi River, and Heima River. Among them, Haergai River and Ganzi River have become seasonal streams. According to the filed investigation results on monitoring sites arranged by Qinghai

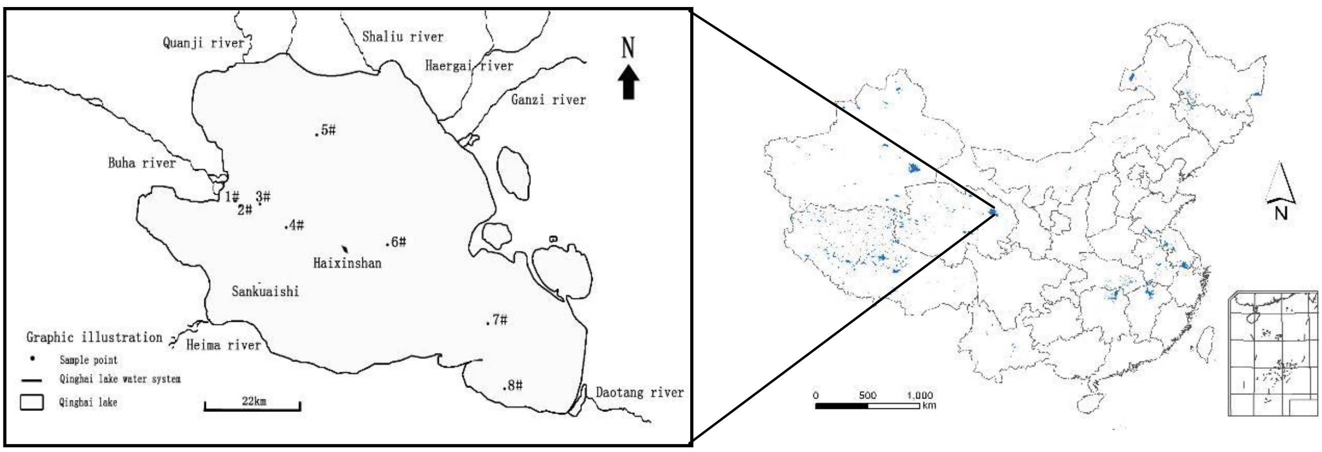


Fig. 1. The distribution chart of study area and sampling sites.

Water Environment Monitoring Center, no significant difference of water quality and outflowing rivers have been found within the Qinghai Lake. As a result, this study selected eight sampling sites in the inflowing mouth of Buha River (Fig. 1) and collected three parallel samples from each sampling site.

2.2. Sample collection and test

Three parallel sediment samples were collected from each sampling site on the surface layer (0–20 cm depth) by using an undisturbed gravity sediment collector on the middle 10 d of June 2013. Each parallel sample was tested three times. All samples were grinded by using an agate mortar after naturally dried and then screened by 2 and 0.15 mm nylon screens. Next, they were labeled for following tests. Total nitrogen (TN) in sediments was measured by potassium dichromate-sulfuric acid digestion through a semi-automatic Kjedahl apparatus. Total phosphorus (TP) and different phosphorus forms in sediments were measured by using the SMT (Standard Measurements and Testing) continuous extraction program developed under the framework of European Commission on Standardized Tests. The phosphorus content in the extraction solution was measured through phosphomolybdenum blue colorimetry. The total organic carbon (TOC) was measured by the external potassium chromate heating. After the samples were digested with hydrochloric acid, nitric acid, hydrofluoric acid, and perchloric acid successively, the heavy metal content in samples was measured by using the IRIS Intrepid II XSP inductively coupled plasma-atomic emission spectrometry (ICP-AES) (Thermo Elemental, USA). The operating parameters of IRIS Intrepid II XSP ICP-AES are frequency of high-frequency ejector: 27.12 MHz; power: 1.2 kW; observed altitude: 16 mm; carrier gas flow: 0.86 L/min; and lowest limit of detection: 0.01 mg/kg

2.3. Data processing and analysis method

The spatial–temporal distribution of sediments in Qinghai Lake was contour calculated and drawn through two-dimensional linear interpolation based on the measured data from observation points. Data analysis was accomplished by using the SPSS statistical software. The correlation analysis and factor analysis of contents of eight heavy metals in surface sediments in Qinghai Lake were conducted. The difference between these eight heavy metals and associated soil background value in Qinghai Province was compared through the single sample *t*-test within 95% confidence interval. The data were interpolated with ordinary kriging method.

3. Results and discussions

3.1. Spatial distribution characteristics of surface sediment enrichment in Qinghai Lake

Nitrogen and phosphorus contents reflect the biological productivity in lakes to certain extent. Contents and spatial distributions of TN, TP, and TOC in sediments in Qinghai Lake were shown in Figs. 2–4. TN content differs from 0.69 to 3.05 mg/L (1.85 mg/L in average). The highest content is 4.4 times of the minimum content, which represents a significant spatial distribution difference, similar with that of organics (Fig. 3). The lowest TN contents (1.27 and 1.45 g/kg) are observed in Site 1 and Site 2. The TN contents in the open water area and regions near the outflowing mouth are 1–1.5 times of the lowest content. This highest TN content is in the sampling Site 5, about two times of the lowest content.

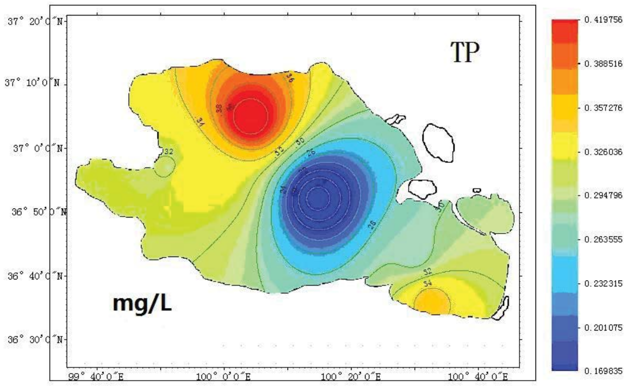


Fig. 3. The distribution of TP in surface sediment of Qinghai Lake.

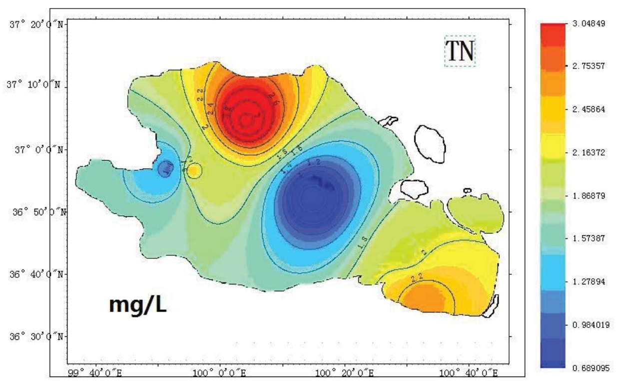


Fig. 2. The distribution of TN in surface sediment of Qinghai Lake.

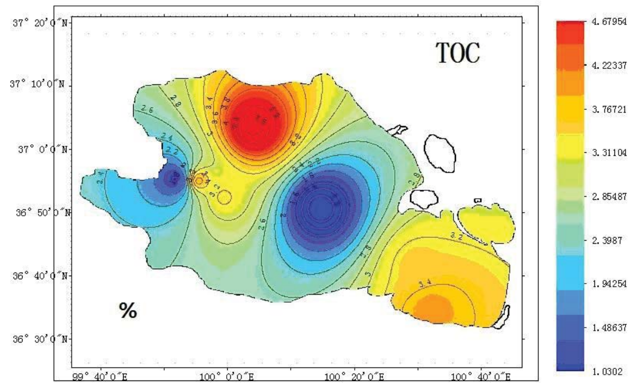


Fig. 4. The distribution of TOC in surface sediment of Qinghai Lake.

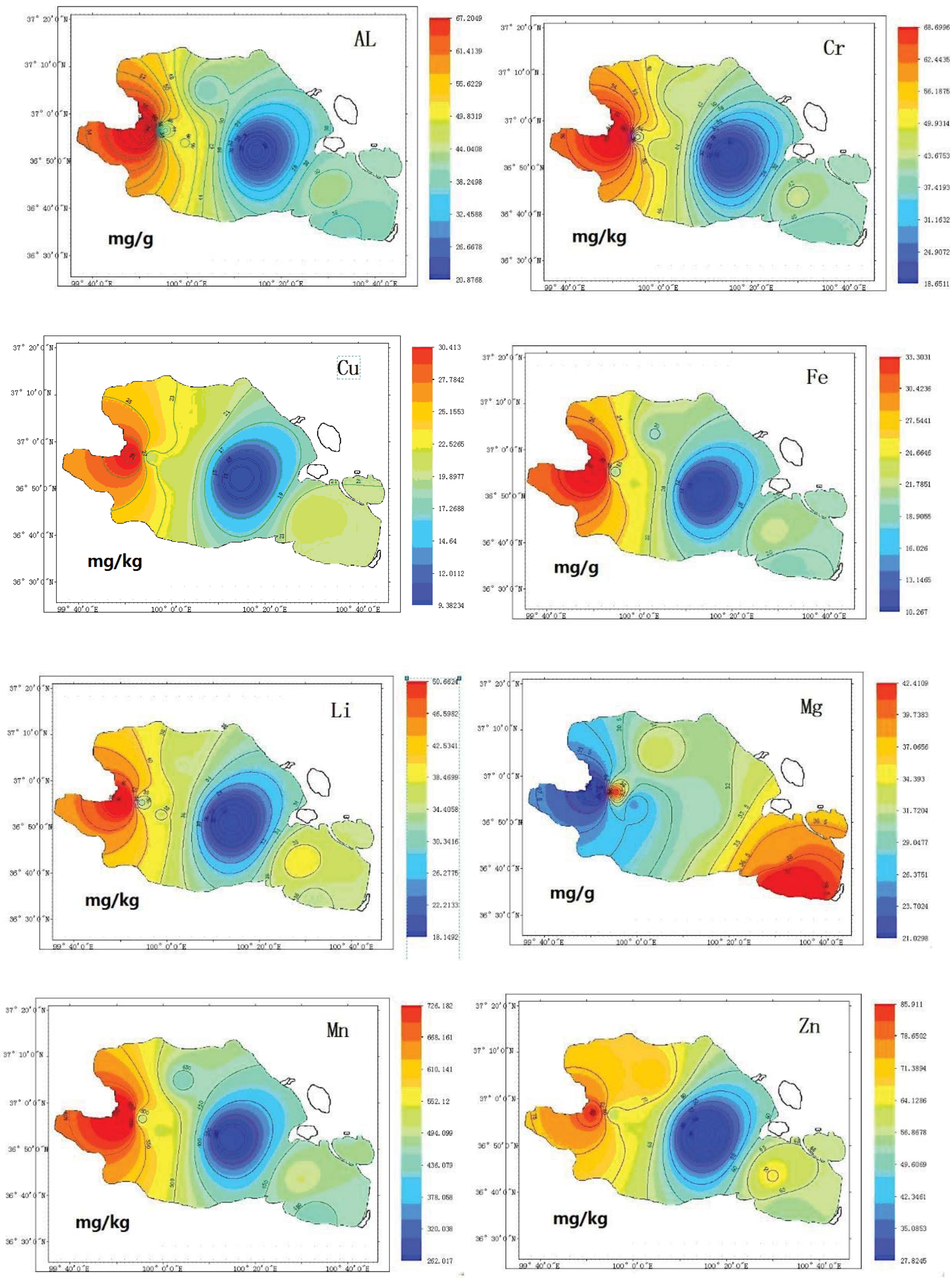


Fig. 5. The content and spatial distribution of heavy metals in surface sediment of Qinghai Lake.

Contents and spatial distributions of TP in surface sediments in Qinghai Lake were shown in Fig. 3. TP content varies from 0.17 to 0.42 mg/L (0.32 mg/L in average). Different from organics and TN, TP shows an equilibrium spatial distribution. The highest TP content is only 2.5 times of the lowest content. However, it presents distinct distribution characteristics. Regions near the Buha River mouth have higher TP content compared with the east regions. Besides, the north arm of lake and the open water area also maintain higher TP content. Obviously, the accumulation rate of TP in these regions is closely related with their geological locations.

The geochemical behaviors of nitrogen and phosphorus in lake sediments, such as migration and conversion, are influenced by various factors, especially organics. Organics in sediments mainly come from exogenous and endogenous sources. Exogenous source mainly refers to organic particles and dissolved organics carried by the external water cycle, whereas endogenous source mainly refers to organics generated from subsided dead bodies of aquatic plants, zooplanktons, and micro-organisms in the lake. Contents and spatial distribution of TOC in surface sediments in Qinghai Lake are presented in Fig. 4. North regions have higher TOC content (4.65%), while northwest and southeast regions have decreasing TOC content (1.78%) along the water flow direction. This is mainly because wastewater carrying abundant organics and nutrients enters into the Qinghai Lake through rivers on its north bank, but is difficult to reach to the northwest and southeast regions due to the weak hydrodynamic conditions and slow water velocity. The average TOC content values 2.85 mg/L with a small variable coefficient of 0.39, indicating the stable and single source of organic carbon.

A contour calculation of heavy metals at observation points in Qinghai Lake was carried out through the two-dimensional linear interpolation. Contents and spatial distributions of heavy metals in surface sediments in Qinghai Lake are exhibited in Fig. 5. Due to lakebed topography of higher at northwest and lower at southeast in Qinghai Lake, heavy metals (except for Mg) are mainly carried to and accumulated in Site 1, Site 2, and Site 3 near the inflowing mouth of Buha River, and the open water area and east regions have 1.7 times higher heavy metal contents compared with west regions. At the present stage, Cu content in surface sediments in Qinghai Lake varies from 9 to 30 mg/kg, averaging at 22.21 mg/kg. The mean contents of zinc (Zn), copper (Cu), chromium (Cr), iron (Fe), and manganese (Mn) are significantly higher than corresponding soil background value in Qinghai Province, indicating their different accumulations in lake sediments. Li content on eight sampling sites is significantly lower than soil background value and the mean Li content is only 60% of the soil background value. Heavy metal contents differ at different sampling sites to certain extent. Cu contributes the most uneven distribution, showing a variable coefficient of 41%. On the contrary, Zn shows the most equilibrium distribution, showing a variable coefficient of 15%.

3.2. Longitudinal change characteristics of sediments in Qinghai Lake

It can be seen from Fig. 6 that TP content in sediment column at Site 2 changes slightly from 0.31 to 0.34 mg/L in different depths. Site 2 is close to the inflowing mouth where suffers great impacts from inflowing river basin and strong scouring effect of water flow, thus resulting in the unstable variation of TP content. TN presents a small accumulation peak (1.46 mg/L) at 6 cm of the sediment column. A sharp increase of TN content is observed at 8–6 cm profile, which is related with excessive ammonia nitrogen brought by industrial sewage discharge at the upstream. However, TN content decreases quickly at 6–4 cm profile due to the self-cleaning

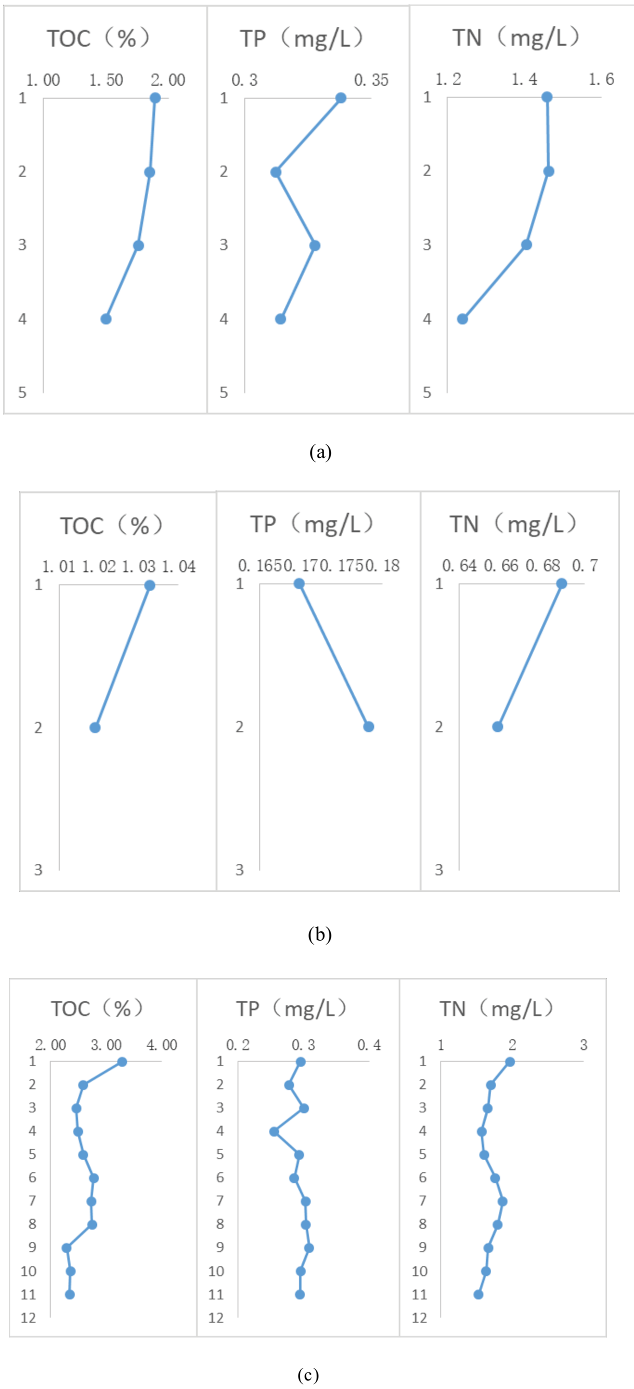
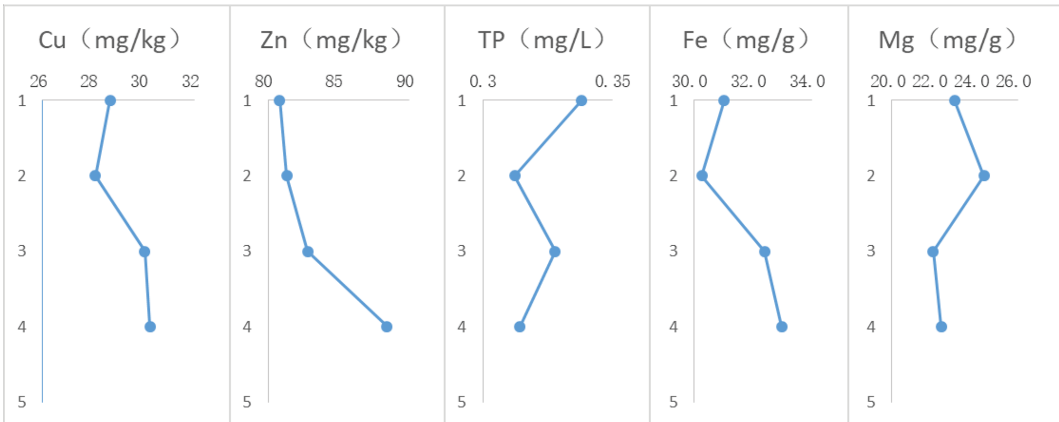


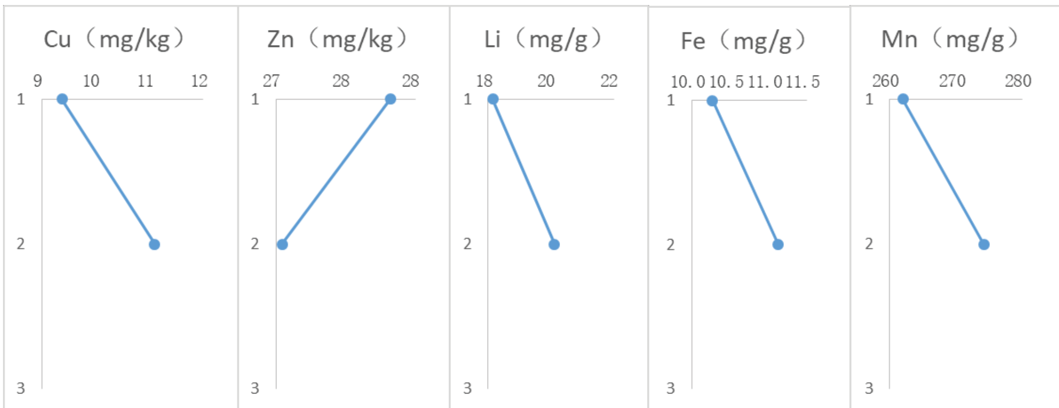
Fig. 6. Vertical distribution of the nutrients at different sampling sites in Qinghai Lake ((a) Site 2, (b) Site 6, and (c) Site 7).

capacity of the lake. TN content remains basically stable at other profiles, ranging from 1.24 to 1.46 mg/L.

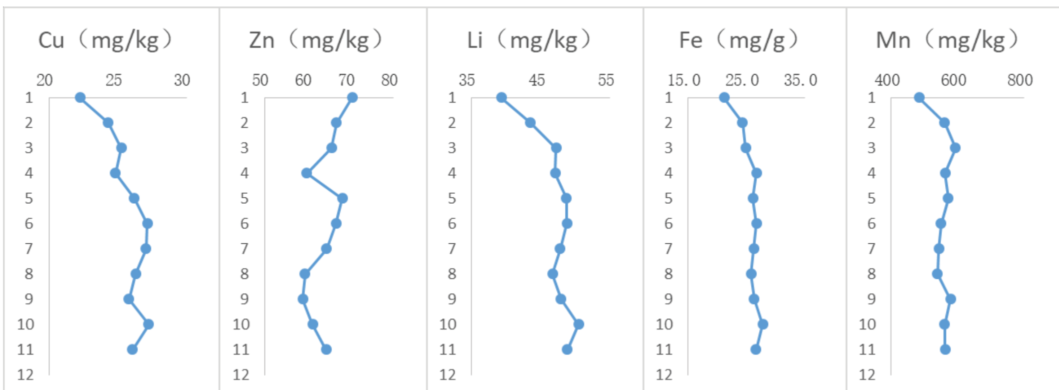
Sediment columns at three sampling sites (Site 2, Site 6, and Site 7) have different vertical distributions of nutritive salt. The vertical distribution of nutritive salt at Site 2 fluctuates gently. TN and TOC at Site 6 present similar vertical distribution, reaching peak at 2 cm, but TP at Site 6 distributes relative evenly. Nutritive salt elements in sediment column at Site 7 show similar vertical distribution, increasing gradually from bottom to top (Fig. 7).



(a)



(b)



(c)

Fig. 7. Vertical distribution of the heavy metals at different sampling sites in Qinghai Lake ((a) Site 2, (b) Site 6, and (c) Site 7).

Take the sediment column at Site 2 for instance. Cu content increases from 20 to 13 cm, but decreases dramatically from 13 to 10 cm and then maintains stable from 10 to 0 cm. The lowest Cu content occurred within 3–2 cm. Cr content fluctuates significantly. Zn content fluctuates without clear trend in more than 10 cm deep, but increases significantly from 10 to 3 cm and then decreases from 3 to 0 cm. Al, Fe, and Mn contents show similar variation trend. They fluctuate gently from 20 to 6 cm, increases at 6 cm, maintains stable from 6 to 3 cm, decreases slightly at 3 cm, and then maintains stable again from 3 to 0 cm. Heavy metals in sediments in Qinghai Lake have relative stable natural source and most heavy metal elements enjoy higher homology, which can be accumulated in sediments through coprecipitation or absorption. Human activities may be the major cause of the fluctuating vertical distribution of heavy metals. In addition, sedimentary environmental changes (e.g., diagenesis, physical and chemical properties of sediments, and biological effect) may affect the vertical distribution of heavy metals.

3.3. Analysis of sediment source in Qinghai Lake

Table 1 demonstrates high correlation among three nutrients in sediment column in Qinghai Lake. A significant positive correlation ($P < 0.01$) is observed between TOC and TN as well as TN and TP. TOC is significantly correlated with TP ($P < 0.05$). Except for Mg, all of rest seven heavy metals show a significant positive correlation ($P < 0.01$). However, three nutrients are slightly correlated with heavy metals. Except for the significant positive correlation between TP and Zn, no significant correlation is observed between other nutritive salts and heavy metals. Specifically, Mg has no significant correlation with both three nutritive salts and other heavy metals.

According to the research results (Table 2), the mean contents of Zn, Cu, Pb, and Cr in collected sediment samples are significantly higher than corresponding soil background value in Qinghai Province, valuing 1.37, 1.23, 1.19, and 1.14 times of the soil background value, respectively. This indicates the different accumulation extents of Zn, Cu, Pb, and Cr in lake sediments. The heavy metal content at eight sampling site is significantly lower than the soil background value and the mean content is only 45% of the soil background value. Different sampling sites have different heavy metal contents.

A factor load analysis based on factors extracted from three principal component tables of heavy metals in sediments in Qinghai Lake was conducted through varimax

Table 1
The Pearson correlation coefficients of the nutrient elements in surface sediment of Qinghai Lake

Item	TOC	TN	TP	Al	Cr	Cu	Fe	Li	Mg	Mn	Zn
TOC											
TN	0.979**										
TP	0.790*	0.877**									
Al	−0.186	−0.066	0.386								
Cr	−0.110	0.021	0.459	0.993**							
Cu	0.128	0.255	0.647	0.944**	0.969**						
Fe	−0.145	−0.021	0.423	0.998**	0.997**	0.960**					
Li	0.039	0.155	0.550	0.963**	0.978**	0.989**	0.977**				
Mg	0.612	0.527	0.134	−0.661	−0.603	−0.454	−0.631	−0.482			
Mn	−0.160	−0.040	0.402	0.998**	0.996**	0.954**	0.999**	0.974**	−0.619		
Zn	0.285	0.396	0.732*	0.881**	0.917**	0.981**	0.902**	0.959**	−0.359	0.894**	

*$P < 0.05$, **$P < 0.01$.

Table 2
Component matrix of the heavy metals

Heavy metal	Unrotated component matrix			Rotated component matrix		
	1	2	3	1	2	3
Fe	0.997	−0.036	0.072	0.880	0.428	0.206
Cr	0.997	0.002	0.056	0.897	0.397	0.187
Mn	0.993	−0.029	0.110	0.876	0.416	0.242
Al	0.992	−0.082	0.092	0.854	0.465	0.229
Li	0.984	0.155	0.008	0.954	0.259	0.125
Cu	0.976	0.196	−0.078	0.971	0.230	0.034
Zn	0.931	0.300	−0.205	0.984	0.135	−0.105
Mg	−0.613	0.782	0.114	−0.236	−0.971	−0.037

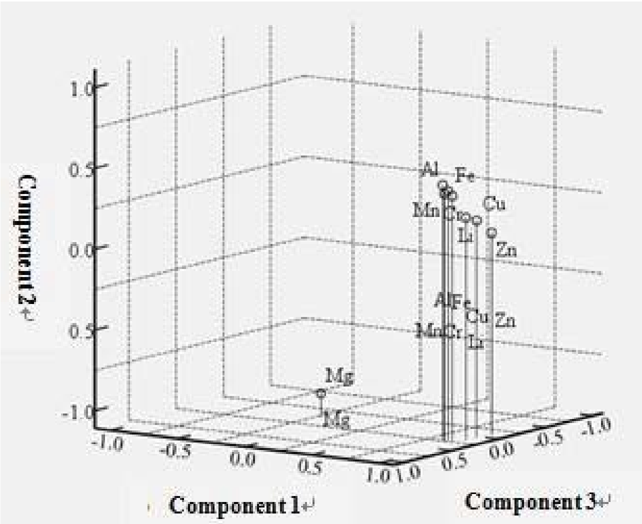


Fig. 8. Rotated component matrix of the heavy metals.

rotation (Fig. 8). The analysis results disclosed certain effect of organics in lake sediments on the absorption and desorption of heavy metals. Such effect is mainly realized by the syngenetic sedimentation of humic acid with heavy metals through absorption and "exchange" complexation. On the other hand, organics in natural lake sediments are mainly generated from sedimentation of dead animals, plants, planktons, and micro-organism in the lake. As a result, sources of heavy metals highly correlated with organics in sediments may be related with the sedimentary environment. Since Qinghai Lake lies in high-altitude cold regions, heavy metals in sediments are mainly affected by the climatic environment and have strong correlation with each other. This implies the similar source of these heavy metals: release of subsided parent materials. This agrees with the factor analysis results (Table 2) and larger fraction of some heavy metals in endogenous factors.

3.4. Discussion

Based on the background concentration of typical soil heavy metal in Qinghai (Table 3), it can be seen that surface sediments were slightly polluted by Zn and Cu. In Buha River, surface sediments were slightly polluted by Cr. The river inflow water to the lake also played an important role in heavy metal input. Both Pb and Zn had weak correlations with the other heavy metals, which represented the transferring and transformation pattern of agriculture activity and tourist and traffic emission. The Cu accumulation in surface sediments was related with endogenous, agricultural, and tourism emission factor. The Ni distribution was mainly affected by endogenous factors and tourist.

Table 3
Background concentration of typical soil heavy metal in Qinghai

Heavy metals	Background values
Zn	80.30
Cu	22.20
Pb	20.90
Ni	29.60
Cr	70.10
Fe	2.85

The unit for Zn, Cu, Pb, Ni and Cr is mg/kg, the unit for Fe is %.

Mn and Fe in Qinghai Lake have the strongest correlation. Their source is mainly influenced by the sedimentation of parent materials. Meanwhile, enrichment of heavy metals in surface sediments in some inflowing river mouth is influenced by exogenous input. No significant correlation has been found among Pb or Hg and other heavy metals. Pb and Hg may be brought by human factors, such as tourist communications and agricultural activities in the Qinghai Lake. Generally, exogenous inputs of heavy metals from human activities and rapid economic development are the main sources of heavy metals in lake sediments. For example, the total volume of wastewater discharged inadequately increases year by year and reached 71.6 billion tons in 2015, which may cause metals to discharge into lakes. Heavy metals in concentrations beyond food safety standards have appeared in aquatic products that are still sold as edible foods [24]. Cu and Zn in sediments have many complicated sources, including not only differentiation of subsided parent materials, but also human factors like tourist, communication, and agricultural activities. The heavy metal pollution and its potential ecological risk of sediments in Qinghai Lake were slight and controllable.

4. Conclusions

- The distribution of TN differs significantly at different positions (4.4 times difference between the highest content and the lowest content). Such spatial distribution is similar with that of TP. The lowest TN content lies near the inflowing mouth of Buha River. The TN contents in the open water area and regions near the outflowing mouth are 1–1.5 times of the lowest content. This highest TN content is in the north arm of Qinghai Lake, about two times of the lowest content. TOC in surface sediments distributed relatively evenly (2.5 times difference between the highest content and the lowest content).
- Different sediment columns in the Qinghai Lake have different vertical distributions of nutritive salts. The TOC and TP contents in sediment column near the estuary fluctuate due to the great impacts from inflowing river basin and strong scouring effect of water flow. Due to the industrial sewage discharge at upstream, TN has a small accumulation peak at 6 cm of the sediment column, followed a sharp increase at 8–6 cm profile. Heavy metals in lake sediments have relative stable natural source and most heavy metal elements enjoy higher homology, which can be accumulated in sediments through coprecipitation or absorption.
- A significant positive correlation ($P < 0.01$) is observed between TOC and TN as well as TN and TP. TOC is significantly correlated with TP ($P < 0.05$). Except for Mg, all of rest seven heavy metals show a significant positive correlation ($P < 0.01$). Human activities may be the major cause of the fluctuating vertical distribution of heavy metals. In addition, sedimentary environmental changes

(e.g., diagenesis, physical and chemical properties of sediments, and biological effect) may affect the vertical distribution of heavy metals.

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