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Arsenic fractionation and contamination assessment in sediments of thirteen lakes from the East Plain and Yungui Plateau Ecoregions, China

Fengyu Zan[1,2], *Shouliang Huo*[1,*], *Jingtian Zhang*[1], *Li Zhang*[1], *Beidou Xi*[1,*], *Lieyu Zhang*[1]

1. *State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Science, Beijing 100012, China. E-mail:* zanfengyu@126.com
2. *College of Environmental Science and Engineering, Anhui Normal University, Anhui 241000, China*

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ABSTRACT

Arsenic (As) fractions in the sediments of seven lakes from East Plain Ecoregion and six lakes from Yungui Plateau Ecoregion, China, were investigated. Results indicated that the total As concentrations in sediment samples of lakes of the East Plain Lake Ecoregion are higher than those of Yungui Plateau Lake Ecoregion. Residual As is the main fraction in sediment samples of lakes from both ecoregions, followed by reducible As and soluble or oxidizable As. The total As is correlated to oxidizable As and residual As in sediment samples from both lake ecoregions. As distribution in sediment samples of lakes of the East Plain Ecoregion appears to be affected by human activity, while the As origin mainly comes from natural sources in sediment samples of lakes in the Yungui Plateau Ecoregion. The potential ecological risk index and geoaccumulation index values suggest "low to moderate" risk degree and "unpolluted to moderately polluted" for As in the studied lake sediments.

Introduction

Arsenic (As) occurs as a metalloid element in a variety of environmental media, including minerals, rocks, soils, sediments, surface water and plants. As contamination in sediments may originate from geochemical background sources (e.g., As-bearing minerals) and human sources (e.g., coal combustion, copper smelting, arsenical pesticides and herbicides production) (Giacomino et al., 2010). The levels of As in lake sediments are typically lower than the content in soils, and As concentration varies all over the world (Mandal and Suzuki, 2002). As accumulation in sediments is of increasing concern due to its mobility, toxicity and bioavailability (Nikolaidis et al., 2004; Kamala-Kannan et al., 2008).

The potential ecological risk of As in sediments is determined by its total content and speciation. Low levels of As reported in some sediments are relatively stable and are less toxic to benthos (Garnaga et al., 2006), whereas high As level found in some sediments and porewaters (e.g., Lake Balmer, Meikong River, Antofagasta region, Lake Sehwan Sindh) (Hoang et al., 2010; Martin and Pederson, 2004; Flynn et al., 2002; Arain et al., 2009) may be transferred to surface water through desorption and dissolution, and may be toxic to benthos and other organisms by bioaccumulation and ingestion (Arain et al., 2008). As adsorbed or co-precipitated with metal oxides in sediments may undergo complex migration and transformation between sediments and surface water, so even As in buried lake sediments can pose a threat for ecological and human health (Charlet et al., 2011). Although studies on mechanism of As mobilization from sediments to surface water have been developed, pathways for As movement vary in different regions due to variation of sorption and redox conditions (Barringer et al., 2010; Schaller et al., 2011) and is necessary to clarify the release mechanism of As fractions

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[*] *Corresponding authors.* E-mail: huoshouliang@126.com (Shouliang Huo); xibeidou@263.net (Beidou Xi).

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from polluted sediments. In general, bioavailability, toxicity, and mobility of heavy metals including As in sediments, are largely influenced by their speciation (Baig et al., 2009). The determination of the As species is thus important for accurate assessments of environmental impact and human health risk. A modified three-step sequential extraction method proposed by the Community Bureau of Reference (now Standards, Measurements and Testing Programme) has been widely adopted to fractionate the forms of heavy metal (Nemati et al., 2011).

China now faces a serious problem of contamination by As, especially drinking water contamination (He and Charlet, 2013). As concentrations average 9.1 mg/kg in river sediments of China, notably higher than average content of 5 mg/kg in world river sediments (Luo et al., 2010). Although there have been investigations on As distribution and contamination in a single lake (Yuan et al., 2011; Wang et al., 2012), or areas polluted by As (Wang et al., 2013), few researches have focused on the spatial distributions of As forms in sediments and pollution assessment at a regional scale. Descriptions of As compounds in different types of lakes in different lake ecoregions of China are required from an ecological and human health perspective to protect water quality. It is necessary to differentiate natural sources and anthropogenic As contamination, for example, by using sequential extraction method, to provide guidance for As contamination control and policy. The objectives of this article are: (1) to elucidate the distribution of As fractions in the surface sediments of lakes from East Plain and Yungui Plateau lake Ecoregions, China; (2) to investigate the geochemical relationships between the As chemical forms and nutrient indicators; and (3) to evaluate the environmental risks of As in the studied lakes using the potential ecological risk index and geoaccumulation index (I_{geo}).

1. Materials and methods

1.1. Study area

Thirteen lakes were chosen based on regional differences between the East Plain and Yungui Plateau Lake Ecoregions. East Plain Ecoregion has the largest number of freshwater lakes, most of which are intensively affected by anthropogenic activities. Four lakes were selected in the middle and lower reaches of the Yangtze River (i.e., Lake Chaohu, Lake Wuchang, Lake Baidang and Lake Shengjin), and three lakes were studied in the middle reach of Huaihe River (i.e., Lake Chengdong, Lake Chengxi and Lake Huayuan). Lake Chaohu, one of the five biggest freshwater lakes in China, has experienced an accelerated eutrophication process since the 1950s due to municipal and industrial wastewater discharge from Hefei and Chaohu cities, Anhui Province, and heavy agricultural pollution in the watershed (Zan et al., 2011; Huo et al., 2011). Lake Wuchang and Lake Baidang are two reservoir-style eutrophic lakes affected by human activities including aquaculture, flood storage and irrigation (Wang and Dou, 1998). Lake Shengjin is also a eutrophic lake located in lower reach of the Yangtze River, and it is a national waterfowl nature reserve region. Lake Chengdong and Lake Chengxi are two mesotrophic lakes and Lake Huayuan is a eutrophic lake in the middle reach of the Huaihe River.

Yungui Plateau Lake Ecoregion, located in the southwest of China with high altitude in the north and low altitude in the south, is a cluster lake region. There are more than 30 lakes with a water depth from 10 to 200 m in this area. A subtropical, highland monsoon climate prevails throughout the lake region. Lake Qilu (eutrophic lake), Lake Chenghai (mesotrophic lake), Lake Lugu (oligotrophic lake), Lake Mahu (oligotrophic lake), Lake Qionghai (eutrophic lake) and Lake Yihai (oligotrophic lake) were selected in this region because of their different depths and trophic conditions. The geographic and limnological features of all studied lakes are shown in Appendix A Table S1.

1.2. Sediment sampling

Surface sediment samples were collected from seven lakes of the East Plain Lake Ecoregion in May 2010 and six lakes of Yungui Plateau Ecoregion in July and August 2009, using a grab sampler. The samples were retrieved from two to twelve locations at each lake and sealed in polyethylene bags and temporarily kept in iceboxes at 4°C. Immediately after being transferred to the laboratory, the samples were stored below −20°C and then freeze-dried under −50°C. Dried samples were ground with an agate mortar and pestle to homogenize before being passed through a 100 mesh sieve and stored at 4°C for analysis.

1.3. As fractionation

The determination of total nitrogen (TN) was done using the persulfate digestion and ultraviolet spectrophotometry method. Total phosphorus (TP) was measured by phosphomolybdate blue spectrophotometry after treating at 500°C (2 hr), followed by 1 mol/L HCl extraction (Huo et al., 2011). Total organic carbon (TOC) was measured using a TOC analyzer (HT1300, Analytik Jena, Germany) after samples were treated with 1 mol/L HCl to remove carbonates.

Sequential selective extraction procedures were applied to estimate As fractions in sediments for explanation of geochemical prospects and anthropogenic contributions to the sediments. The Community Bureau of Reference three-step sequential extraction procedure was used to obtain chemical speciation distribution data for metals in the sediment samples (Farkas et al., 2007; Baig et al., 2009; Nemati et al., 2011). It produced four chemical phases including: carbonates/exchangeable ions (soluble fraction), Fe/Mn oxides (reducible fraction), organic matter/sulfides (oxidizable fraction), and residual metal forms (residual fraction). The concentrations of As fractions were determined directly by hydride generation atomic fluorescence spectrometry (AFS-9800, Haiguang Instrument, China). The concentration of total As was determined by HG-AFS after digestion with aqua regia.

1.4. Quality control

Accuracy of the metal analyses was evaluated by using eight replicates of the standard reference sediment SUD-1 (Environment Canada, National Water Research Institute, Canada). The measured As contents for eight replicates were: 34.1, 31.6, 33.8, 34.4, 31.2, 29.5, 30.4, and 30.8 mg/kg with average 32.0 mg/kg, certified value 31.1 ± 5.0 mg/kg and RSD 5.8%. All the values in the range of the standard certified values and RSD were less than 8% for the analyzed As. A quality control procedure was applied throughout the different steps from sampling preparation to analysis. Reagent blanks were regularly run within each series of analyses to validate the data. Every sediment sample was determined by using three replicates, and a reagent blank was incorporated into the analytical procedure for every twelve samples. All reagents used during the experimental process were certified and all vessels were soaked in 25% HNO_3 for at least 48 hr prior to use.

1.5. *Data analysis*

1.5.1. *Potential ecological risk index*

The potential ecological risk index is used to evaluate the pollution of heavy metals in sediments (Hakanson, 1980). The formula for potential ecological risk index for a single heavy metal pollutant is (E_r^i):

$$E_r^i = T_r^i \times C_f^{\ i} = T_r^i \times C_s^i / C_n^i \tag{1}$$

where, $T_r^{\ i}$ is the toxic-response factor for a given substance (e.g., As = 10, Yu et al., 2012); $C_f^{\ i}$ is the pollution coefficient for As, which can reflect the pollution character of the investigated region but cannot reveal the ecological effects and hazards; $C_s^{\ i}$ is the measured As concentration in surface sediments, and $C_n^{\ i}$ is the reference values of As. $C_n^{\ i}$ in East Plain and Yungui Plateau Lake Ecoregions is considered as 8.8 mg/kg and 8.0 mg/kg, respectively (Yu et al., 2012; Li and Wang, 2008).

1.5.2. *Geoaccumulation index*

The I_{geo} has been widely employed in trace metal pollution studies caused by anthropogenic activities since the late 1960s (Müller, 1969). The index of I_{geo} can be used to assess pollution by comparing current concentrations with pre-industrial concentrations. It can be calculated by the following Eq. (2):

$$I_{geo} = \log_2\left(\frac{C_n}{K \times C_b}\right) \tag{2}$$

where, C_n is the measured As concentration in surface sediments and C_b is the geochemical background concentration of As. The factor of $K = 1.5$ is used because of the possible variations in background values due to lithological variability (Farkas et al., 2007).

2. Results and discussion

2.1. *Total As distribution*

Total concentrations of As in the studied sediments from two lake ecoregions are presented in Table 1. Total As concentrations in sediments of seven lakes of East Plain Lake Ecoregion ranged from 9.24 to 22.16 mg/kg with an average value of 14.45 mg/kg, suggesting different As pollution levels by human activities comparing with the background concentration of 8.8 mg/kg (Yu et al., 2012). The results indicate that similar As sources are present in East Plain Lake Ecoregion, including arsenical herbicide and fertilizer application, coal combustion and As compound production along the watersheds (Tang et al., 2012). It was reported that the total amounts of herbicide and fertilizer application in Anhui Province were up to 8.5×10^4 and 3.05×10^6 t respectively. Thus, large amounts of As compounds in these sources entered into the sediments by atmospheric deposition, and industrial and agricultural development, resulting in the accumulation of As in sediments.

Total As concentrations ranged from 6.98 to 41.05 mg/kg, averaging 14.16 mg/kg in the sediment samples of lakes in the Yungui Plateau Lake Ecoregion. The maximum As concentration was observed in the sediment samples of Lake Lugu, which was a deep, oligotrophic lake. Lake Lugu is a typical rift lake affected by karstification with high total dissolved solid of 188.02 mg/L (Wang and Dou, 1998). It is possible that large amounts of As compounds permeated into sediments from carbonate aquifer (Kim et al., 2000). The tectonic structure of Lake Lugu is composed of several fault zones formed at different geological times (e.g., Yunning faulting, Anjia faulting and Ganzi fold system). Bedrock in this region contains volcanic rocks (e.g., whinstone, lava breccia) that may be another potential source of As in sediments. Lake Mahu had the lowest total As in the sediment samples among all lakes and was the only one where the total As concentration was lower than the background concentration of 8.0 mg/kg (Li and Wang, 2008), demonstrating less As input from natural enrichments. Lake Mahu was born in a fault basin of the boundary region of Sichuan–Yunnan active block, and a natural As source may be attributed to several natural geochemical processes, including As input from Jinshajiang and release from geothermal water underground. Although Lake Qilu is a seriously polluted lake, the total As concentration in the sediment samples was not as high as that in Lake Lugu, indicating the low natural and anthropogenic source contribution.

The total As concentrations in all lakes except Lake Lugu are uniformly distributed, due to the complicated variable geological conditions of different areas in this lake. The variations and concentrations of the total As in East Plain Lake Ecoregion were relatively higher than those in Yungui Plateau Lake Ecoregion (only averaging 8.78 mg/kg except Lake Lugu, close to the background value). The results support the inference of different pollution levels and higher anthropogenic sources in East Plain Lake Ecoregion.

2.2. *As fractionation*

Concentrations and the recovery of different As fractions are shown in Table 1. The recovery of As ranged from 63% to 109.34%, with an average of 90.77% ± 9.99%. This suggests that total As in sediments was satisfactorily extracted with this procedure. The As was composed of soluble, reducible, oxidizable and residual fractions ranging from 0.22 to 1.59 (mean 0.77), 0.38 to 2.11 (mean 1.27), 0.22 to 1.41 (mean 0.65), and 7.42 to 13.78 (mean 14.45) mg/kg with the relative contribution of 6.29%, 10.38%, 5.14% and 78.19% to total As in East Plain Lake Ecoregion, respectively. The As concentration of soluble, reducible, oxidizable and residual fractions varied from 0.35 to 1.64, 0.53 to 6.71, 0.25 to 2.35, and 4.50 to 31.30 mg/kg in Yungui Plateau Lake Ecoregion, respectively, averaging 0.77, 2.52, 0.96 and 10.08 mg/kg. The relative contribution of soluble, reducible, oxidizable and residual fractions to the total As was 7.35%, 15.36%, 7.73% and 69.48%, respectively. Among the sequentially extracted As forms in the sediment samples of two lake ecoregions, the order of the As fractions

Table 1 – Concentrations and the recovery of different As fractions in the sediment samples (unit: mg/kg).

Lake region	Lake	Fraction				Total concentration	Total recovery (%)
		Soluble	Reducible	Oxidizable	Residual		
East Plain Ecoregion	Lake Chaohu	1.59 ± 0.48 (13.11 ± 2.43)	1.21 ± 0.42 (9.88 ± 2.48)	0.42 ± 0.29 (3.41 ± 2.06)	8.92 ± 1.73 (73.59 ± 3.26)	13.45 ± 2.57	91.14 ± 12.47
	Lake Chengdong	0.83 ± 0.09 (7.55 ± 1.69)	0.67 ± 0.12 (6.29 ± 2.46)	0.64 ± 0.40 (5.17 ± 2.76)	9.39 ± 2.56 (80.99 ± 2.73)	12.42 ± 2.37	92.29 ± 10.41
	Lake Wuchang	0.66 ± 0.21 (5.06 ± 2.27)	1.44 ± 0.21 (10.68 ± 2.62)	0.22 ± 0.12 (1.69 ± 0.96)	11.93 ± 4.37 (82.58 ± 5.29)	17.54 ± 4.89	80.67 ± 6.65
	Lake Shengjin	0.68 ± 0.35 (3.84 ± 1.79)	2.11 ± 0.45 (11.85 ± 1.72)	1.19 ± 0.44 (6.55 ± 1.90)	13.78 ± 1.29 (77.76 ± 3.62)	22.16 ± 1.43	80.12 ± 6.97
	Lake Chengxi	0.22 ± 0.09 (2.23 ± 0.45)	1.58 ± 0.22 (17.14 ± 5.86)	0.34 ± 0.04 (3.70 ± 1.16)	7.42 ± 2.15 (76.93 ± 6.57)	12.12 ± 2.10	78.60 ± 2.75
	Lake Baidang	0.32 ± 0.07 (3.50 ± 0.68)	0.38 ± 0.06 (4.25 ± 0.72)	0.33 ± 0.15 (3.69 ± 1.73)	7.99 ± 0.29 (88.55 ± 2.48)	9.24 ± 0.32	97.75 ± 3.78
	Lake Huayuan	1.06 ± 0.31 (8.75 ± 1.95)	1.48 ± 0.07 (12.52 ± 1.60)	1.41 ± 0.27 (11.77 ± 1.56)	8.04 ± 1.31 (66.96 ± 2.89)	14.21 ± 3.00	85.74 ± 9.72
Yungui Plateau Ecoregion	Lake Qilu	0.35 ± 0.54 (3.59 ± 5.90)	1.67 ± 1.20 (14.24 ± 9.47)	1.96 ± 0.98 (18.59 ± 12.49)	7.91 ± 3.85 (63.57 ± 20.21)	13.43 ± 3.29	88.93 ± 3.41
	Lake Chenghai	0.50 ± 0.09 (6.41 ± 1.23)	0.82 ± 0.25 (10.28 ± 2.43)	0.27 ± 0.23 (3.40 ± 2.81)	6.47 ± 1.84 (79.91 ± 5.81)	8.30 ± 2.30	97.79 ± 8.69
	Lake Mahu	0.51 ± 0.29 (8.69 ± 4.77)	0.53 ± 0.11 (9.48 ± 4.05)	0.32 ± 0.16 (5.09 ± 2.49)	4.58 ± 0.92 (76.73 ± 5.62)	6.98 ± 1.44	86.07 ± 5.90
	Lake Yihai	0.61 ± 0.23 (7.99 ± 2.62)	0.88 ± 0.28 (11.28 ± 1.53)	0.61 ± 0.34 (8.86 ± 5.99)	5.72 ± 2.04 (71.86 ± 6.90)	8.05 ± 2.41	97.94 ± 8.18
	Lake Lugu	1.64 ± 1.26 (4.04 ± 2.20)	6.71 ± 2.91 (17.91 ± 4.92)	2.35 ± 0.71 (7.48 ± 3.79)	31.30 ± 24.65 (70.11 ± 7.63)	41.05 ± 25.24	100.26 ± 5.14
	Lake Qionghai	1.07 ± 0.19 (13.36 ± 2.85)	4.53 ± 0.34 (28.98 ± 3.29)	0.25 ± 0.24 (2.96 ± 2.79)	4.50 ± 1.12 (54.69 ± 5.83)	9.02 ± 1.76	91.03 ± 8.49

Data in brackets represent the relative distributions of different As fractions in the sediment samples (%).

is: residual > reducible > soluble or oxidizable, which is similar to the previous study of Sahuquillo et al. (2003). The residual fraction is the dominant fraction in all the samples and accounted for 55%–89% of total As. This residual fraction is relatively stable and not easily released. About 11%–46% of total As composed of soluble, reducible, oxidizable fractions can be remobilized depending on external conditions (i.e., pH, DOM, reducing or acidic conditions) (Anawar et al., 2004; Bauer and Blodau, 2006; Xu et al., 2011).

2.2.1. Soluble As distribution

The soluble As is composed of the exchangeable phase adsorbed by surface sediments (e.g., clay, humus) and the acid-soluble phase that may be precipitated or co-precipitated with carbonates (Ko et al., 2005). This fraction is loosely bound and transferable to surface water under certain environmental conditions (Baig et al., 2009). The soluble As fraction is low, ranging from 0.22 to 1.64 mg/kg and accounting for 2.23%–13.36% of total As in all sediment samples (Table 1), suggesting low mobility and toxicity to aquatic life (Baig et al., 2009). However, the average concentrations of the soluble As in Lakes Chaohu, Huayuan, Lugu and Qionghai were higher than 1.0 mg/kg. Although the soluble As originated from different natural and anthropogenic sources, the potential toxicity to organisms in water was relatively high considering the As content of 2.0 and 1.5 mg/kg in igneous rocks and sandstone, respectively. Furthermore, uneven soluble As distribution occurs in the sediments of Lakes Chaohu and Lugu. It has been reported that lake trophic status is significantly correlated with As distribution and mobility in water and sediments (Martin and Pederson, 2004). Lake Chaohu is a eutrophic lake affected by serious pollution in its western half due to industrial activities and municipal wastewater input from Hefei City (Zan et al., 2011; Huo et al., 2011). And the spatial distribution of current-use insecticides in the sediments of Lake Chaohu is similar with the trophic status in western and eastern half lake, which is an important As source from agriculture. The soluble As in the sediments of Lake Lugu was mainly bound to carbonates originating from volcanic rocks formed in different geological ages, which resulted in varied distribution of the soluble As.

The relative content of the soluble As fraction in sediments of Lakes Chaohu and Qionghai, located adjacent to a city, reached to approximately 13% and was higher than other lakes located away from the city. This suggests that city development may have influenced soluble As levels in these sediments.

2.2.2. Reducible As distribution

The reducible As is bound to Fe and Mn (hydro) oxides with the relatively strong ionic binding and could not be released until the matrix is subjected to reducing conditions (Baig et al., 2009; Wang et al., 2010). The strength of metal-binding by Fe and Mn (hydro) oxides is sensitive to pH that may result in an increase or decrease in solubility of sediment-associated heavy metals and As (Ho et al., 2012). In all lakes, the average reducible As concentration was much higher than the soluble or oxidizable As except in Lake Qilu (Table 1). The higher oxidizable As concentration than the reducible As in Lake Qilu might result from eutrophication as a result of industrial and agricultural activities (Ko et al., 2005; Baig et al., 2009), indicating stronger binding capacity of Fe and Mn (hydro) oxides than carbonates, organic matters and sulfides.

The reducible As concentration varied greatly in Lake Lugu and Lake Qilu, ranging from 4.02 to 12.79 mg/kg and 0.69 to 3.02 mg/kg, respectively. High reducible As concentrations of 11.14 and 12.79 mg/kg are found in samples from two sites (i.e., L3 and L6) of Lake Lugu; these sites are located next to the villages. As from anthropogenic sources may first bind to Fe oxides and oxyhydroxides in lake sediment (Wang et al., 2010). The relative standard deviation of nutrient (e.g., NH_4^+-N, TP and Chl-*a*) and heavy metal (e.g., As, Cr, Pb and Cd) parameters in Lake Qilu were higher than 50% according to the monitored data of Liu et al. (2010).

2.2.3. Oxidizable As distribution

The oxidizable fraction is related to organics and sulfides and not considered to be mobile or bioavailable until decomposition under acidic conditions (Baig et al., 2009). The concentration of the oxidizable fraction of As is lower than reducible and the residual fractions in all lakes except Lake Qilu (Table 1), reflecting a weak combining ability between As and organics. Inorganic As exists as arsenite and arsenate in water, so the charge repulsion with organics may inhibit As adsorption or increase As leaching from mineral surfaces in sediments (Yu et al., 2012). Unlike other lakes (e.g., Lakes Yihai and Lugu) in which the source of sediment organic matter is mainly from natural sources, Lake Qilu is heavily polluted by organic materials from an anthropogenic source. The oxidizable As content are higher than the soluble As in some sediments, which may be due to the relative high contribution of organoarsenic compounds (e.g., monomethylarsonic, dimethylarsinic) (Sanchez-Rodas et al., 2005).

The distribution of the oxidizable As in sediments of every lake varied greatly with standard deviation ranging from 0.04 to 0.98 mg/kg, particularly in Lake Qilu and Lake Lugu. The oxidizable As content in Lake Qilu varied greatly ranging from 0.70 to 3.02 mg/kg, consistent with the variation of reducible As content. The inhomogeneous distribution of oxidizable As content in Lake Lugu may be related to the complex geological features and the presence of an anthropogenic source (Wang et al., 2010).

2.2.4. Residual As distribution

Unlike the previous reports for As fractions in sediments (Brook and Moore, 1988; Wang et al., 2010), 54.69%–88.55% of total As content remained in the residual fraction in all sediments (Table 1). These results indicate that As adsorbed into the crystal lattice of primary and secondary silicate minerals represents a large proportion of the total As in the sediments (Yu et al., 2012; Sahuquillo et al., 2003). The residual As content in Lake Lugu is much higher than those in other lakes, indicating a stable As source from natural environment. The residual phase is relatively stable with low mobility and bioavailability that is not easily transferred to overlying water (Teasdale et al., 2003). Thus, the residual phase is not expected to release significant amounts of As to the water column (Krysiak and Karczewska, 2007).

2.3. Relationships between As fractions and other factors

Correlation analysis between As fractions and other indicators in the sediment samples of East Plain and Yungui Plateau Ecoregions is shown in Table 2. Oxidizable As, residual As and total As are significantly correlated with each other in the two regions ($p < 0.01$). Residual As is the main fraction present in the studied lake sediments. The oxidizable fraction, presented as diagenetic or detrital sulfides and chemically bound with organic material was an important As source of sediments (Brook and Moore, 1988).

TOC is significantly correlated with TN, TP, the reducible As fraction, oxidizable As fraction and total As, but not significantly correlated with soluble As and residual As in the sediment samples of East Plain Ecoregion. Sediment organic matter, especially humic substances, are known to be adsorbents for As (Pikaray et al., 2005), and organic matter-bound As had a high stability in the sediment (Wang et al., 2010). The results indicate that organic matter plays a significant role in controlling As transport in the sediments of East Plain Ecoregion. The significant correlation between TN and total As may suggest that As pollution is related to human impacts. Zan et al. (2011) reported that increasing nutrient concentration in sediments was consistent with the observation of the appearance of anthropogenic-derived heavy metal enrichment. Furthermore, the loading of nutrients to shallow eutrophic lakes has the potential to further amplify the release of sediment-derived As *via* alteration of redox conditions at the water–sediment interface (Martin and Pederson, 2004).

In Yungui Plateau Ecoregion, total As and different As fractions are significantly correlated with each other ($p < 0.01$), suggesting that As in the sediment samples is similarly distributed in most lakes with mesotrophic or oligotrophic status that are less affected by human activities. The reducible fraction bound to Fe oxides is mainly adsorbed into organic matters, explaining the good correlation between TOC and reducible fraction. There are large amounts of low temperature deposits containing As in Yungui Plateau Ecoregion, such as coal and black shale. As compounds in the sediments of studied lakes of Yungui Plateau Ecoregion may have come from these low temperature deposits by groundwater migration. The behavior of As in lake sediments is largely governed by the redox cycling of Fe, organic matter characteristics, and especially the redox state of sediment–water interface (Martin and Pederson, 2004). The difference of the redox state of sediment–water interface between the shallow lakes of East Plain Ecoregion and deep lakes of Yungui Plateau Lake Ecoregion may influence the As species and the relationship between the As fractions and other factors.

2.4. Contamination assessment of As

The potential ecological risk indices (E_r^i) of As in surface sediments of the studied lakes are listed in Table 3. The E_r^i values of As in all studied lakes except Lake Lugu were below 30, which indicates low risk, while Lake Lugu poses a potentially moderate risk to the local ecosystem. This result is consistent with the total As concentration of those studied lakes in general. The E_r^i values of 10.50–25.18 in the sediment

Table 2 – Correlation coefficient matrix of different As fractions and other indicators in sediments of East Plain Ecoregion and Yungui Plateau Ecoregion.

		Soluble	Reducible	Oxidizable	Residual	Total	TOC	TN	TP
East Plain Ecoregion	Soluble	1							
	Reducible	0.055	1						
	Oxidizable	0.273	0.307	1					
	Residual	0.020	0.162	0.418^{**}	1				
	Total	0.136	0.296	0.732^{**}	0.850^{**}	1			
	TOC	0.093	0.624^{**}	0.602^{**}	0.065	0.363^{*}	1		
	TN	0.286	0.466^{**}	0.703^{**}	0.324^{*}	0.637^{**}	0.816^{**}	1	
	TP	0.280	0.459^{**}	0.332^{*}	−0.082	0.163	0.562^{**}	0.564^{**}	1
Yungui Plateau Ecoregion	Soluble	1							
	Reducible	0.262	1						
	Oxidizable	0.562^{**}	0.573^{**}	1					
	Residual	0.668^{**}	0.410^{**}	0.876^{**}	1				
	Total	0.676^{**}	0.500^{**}	0.919^{**}	0.991^{**}	1			
	TOC	−0.160	0.518^{**}	−0.002	0.023	0.045	1		
	TN	−0.099	0.685^{**}	0.135	0.120	0.167	0.918^{**}	1	
	TP	0.400^{**}	0.584^{**}	0.824^{**}	0.769^{**}	0.793^{**}	0.236	0.302	1

TOC: Total Organic Carbon; TN: Total Nitrogen; TP: Total Phorphous.

* Correlations are significant at $p < 0.05$ (two-tailed).

** Correlations are significant at $p < 0.01$ (two-tailed).

samples of East Plain Ecoregion show different ecological risk levels from anthropogenic sources. Generally, most anthropogenic releases of As are from nonferrous metal mining and smelting, fossil fuel processing and combustion, wood preserving, pesticide production and application, and disposal and incineration of municipal and industrial wastes (Smedley and Kinniburgh, 2002). Thus, As discharged from various industrial and agricultural activities around the lake basin may be accumulated in the sediments. The average E_r^i value of Lakes Qilu, Chenghai, Mahu, Yihai and Qionghai in Yungui Plateau Ecoregion is 11.45, indicating the lower ecological risk of As in this region. The result is consistent with the trophic status of the two lake regions, suggesting that the distribution of As in sediments was influenced by eutrophication (Hasegawa et al., 2010).

Table 3 – Potential ecological risk index (E_r^i) of As in the studied sediments and the geoaccumulation index (I_{geo}) of heavy metals in sediments of lake areas.

Lake	E_r^i	Risk degree	I_{geo}	Pollution status
Chaohu	15.28	Low	0.03	Unpolluted to moderately polluted
Chengdong	14.11	Low	−0.09	Unpolluted
Wuchang	19.93	Low	0.41	Unpolluted to moderately polluted
Shengjin	25.18	Low	0.75	Unpolluted to moderately polluted
Chengxi	13.77	Low	−0.12	Unpolluted
Baidang	10.50	Low	−0.51	Unpolluted
Huayuan	16.15	Low	0.11	Unpolluted to moderately polluted
Qilu	16.79	Low	0.16	Unpolluted to moderately polluted
Chenghai	10.38	Low	−0.53	Unpolluted
Mahu	8.73	Low	−0.78	Unpolluted
Yihai	10.06	Low	−0.58	Unpolluted
Lugu	51.31	Moderate	1.77	Moderately polluted
Qionghai	11.28	Low	−0.41	Unpolluted

The geoaccumulation indices (I_{geo}) can be used as a reference to estimate the extent of heavy metal pollution in sediments. As shown in Table 3, Lakes Chengdong, Chengxi, Baidang, Chenghai, Mahu, Yihai and Qionghai are in an unpolluted status for As with $I_{geo} < 0$, while Lakes Chaohu, Wuchang, Shengjin, Huayuan and Qilu are in unpolluted to moderately polluted status with $0 < I_{geo} < 1$. Lake Lugu is moderately polluted by As with the I_{geo} value of 1.77. The results are not apparently consistent with the E_r^i method, but the variations of I_{geo} and E_r^i in the studied lakes are nearly similar. This is because more detailed classification is obtained by I_{geo} method.

3. Conclusions

The accumulation of As in lake sediments in East Plain Ecoregion is mainly caused by anthropogenic factors, but from natural origins in Yungui Plateau Ecoregion. In the sediment samples of the two lake ecoregions, fractions of As are ranked in the following order: residual As > reducible As > soluble or oxidizable As. The oxidizable As, residual As and total As are significantly correlated with each other in the sediment samples. TOC is significantly correlated with TN, TP, reducible As, oxidizable As and total As in East Plain Ecoregion, which is consistent with the higher trophic status in these lakes relative to Yungui Plateau Lake Ecoregion. In Yungui Plateau Ecoregion, total As and the As fractions are significantly correlated with each other, suggesting that the As origin is less affected by human input in those lakes. The I_{geo} and E_r^i values suggested “low to moderate” risk degree and “unpolluted to moderately polluted” for As in the studied lake sediments.

Supporting materials

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jes.2014.07.010.

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