Arsenic has attracted great interests from scientists and governments due to its high toxicity and ubiquitous occurrence in the environment. People who are chronically exposed to As through oral, dermal, or respiration may develop serious diseases (e.g., Black foot disease) and the risks of cancers may increase [1-3]. Arsenic is globally presented in the Earth’s crust, including soils, sediments, water bodies, and rocks [4-6]. Arsenic occurs in more than 200 types of minerals, which mostly are ore minerals or the alteration products of ore minerals [7]. Some sulfide minerals such as pyrite, marcasite, and sphalerite may even contain As with concentrations of higher than 10,000 mg·kg⁻¹ [7]. Arsenic occurrence in soils, sediments, and rocks may be largely caused by these As-rich minerals [8, 9].
other hand, the As-bearing minerals may also increase the As concentration in groundwater, and consequently cause many endemic diseases. For instance in Bangladesh, groundwater with high As content has threatened the health of millions of people, where the As was mainly derived from sediments in addition to human pollution [8, 10, 11].

The sources of As in the environment can be classified as anthropogenic inputs and natural releases. The anthropogenic sources include discharge of industrial wastewaters [12, 13], combustion of fossil fuels [14], and production and use of As-bearing chemicals such as herbicides, pesticides, and crop desiccants [15]. The natural inputs of As include weathering and erosion of As-bearing minerals and rocks, hydrothermal emanations, and volcanic eruptions [7, 16]. Arsenic will be released from As-bearing sulfides into groundwater under oxidizing conditions, and inversely As will precipitate as sulfide into sediment or clays under reduction conditions [8, 17]. In addition, the weathering and alteration of As-bearing minerals and rocks will also release As into the environment [7, 18, 19]. Therefore, it is essential to study As occurrence in soils, sediments, and so on to fully understand the contamination and source of As in a specific area.

Yangzonghai Lake is a typical fault-controlled plateau lake in Yunnan province, China. In June 2008, the As concentration of the lake water increased dramatically from less than 0.005 mg·L⁻¹ to higher than 0.1 mg·L⁻¹ [20]. Although the lake water has been treated and the As concentration has been decreased to orginal levels (<0.01 mg·L⁻¹), this lake is still particularly important for scientists and local governments to study the sources, behaviors, and fate of As in plateau lakes. For example, Wang et al. [20] studied the As concentrations in the lacustrine sediment, lake water, and aquatic organisms, and investigated the risk of As pollution in Yangzonghai Lake. Zhang et al. [21-23] studied the As concentrations in lake water and lake sediments, and assessed the pollution level by calculating the geoaccumulation indexes. Qi et al. [24] investigated the source and speciation of As in this lake, and suggested that As in the lake was mainly polluted by the chemical industry.

These studies, however, lacked in-depth assessment of As enrichment in the lake, and the As occurrence around the lake is still unknown. In this study we collected various samples, including altered volcanic rocks, lacustrine sediments, lakeside sediments, soils, slope deposits, and chemical wastes of a phosphate plant in and around Yangzonghai Lake. We analyzed the distributions of Arsenic and related trace metals in these samples, and studied the enrichments of these elements. In addition, we studied the correlation coefficients among these elements and classified these sam-

![Geographical position and geological setting of Yangzonghai Lake](image)

Fig. 1. Geographical position and geological setting of Yangzonghai Lake. The sample locations are shown in various symbols according to the types of obtained samples.
samples by hierarchical cluster analysis. We further suggested the potential factors that controlled the distribution and enrichment of As in and around the lake.

Field Descriptions

The studied area – Yangzonghai Lake – is a fault-controlled freshwater lake located in Yiliang County, Yunnan province, China (Fig. 1). The geographical location of the rod-shaped lake is between 102°55' and 103°02' longitude and 24°27' and 24°54' latitude, with a length of about 12 km and width of 3 km. Xiaojiang fault developed along the north-south direction of the lake. The major water supply is groundwater, which discharges from various springs that ubiquitously surround the lake. Important springs A, B, C, D, E, and F with large runoffs are marked on Fig. 1. In addition to meteoric water, the major surface water input is Yangzong River that flows through Yangzong city and recharges water to the south of the lake. The main output is Tangci Canal at the north end of the lake. A phosphate fertilizer plant is located along the southwest bank of the lake, but it has been closed by the local government who proposed that this plant polluted the lake water in 2008 by discharging a large amount of As-bearing wastewater into the lake.

Material and Methods

Sampling Methods

Rock, sinter, soil, and lakeside sediment samples were collected by bare hands with pre-cleaned gloves. These samples were conserved in plastic bags. The lacustrine sediment samples were collected by a self-made core sampler with an inner diameter of 52 mm and a length of 0.9 m. The sampling started from November 3 to November 10, 2009. The sample locations were recorded using a hand-held GPS (Fig. 1). The collected samples were separated into seven groups according to their characteristics and sample locations: altered volcanic rock (three samples), lacustrine sediment from the bottom of the lake (eight samples), lakeside sediment that near the lake bank (24 samples), sinter that formed in the springs (seven samples), slope deposit (seven samples), surface soil (17 samples), and chemical waste from the phosphate plant (four samples).

Determination of the Elements

The concentrations of As, Sb, Hg, and Bi in the samples were analyzed by atomic fluorescence spectroscopy (AFS) (model: AF-610A, Beijing Rayleigh Analytical Instrument Co., Beijing, China). The operating conditions were maintained as follows: negative high voltage of 270 V, atomizer height of 8 mm, air flow rate of 300 mL·min⁻¹, lamp current of 30 mA, atomization temperature of 170°C, and recording time of 18 s. The concentrations of Fe, Mn, and Al were determined using an x-ray fluorescence spectrometer (XRF) (model: SPECTRO XEPOS XEP01, Spectro Analytical instruments, Germany). The operating voltage and current were maintained at 50 kV and 50 mA, respectively. The contents of Pb, Zn, Ni, Cd, and Co were determined by inductively coupled plasma–mass spectrometry (ICP-MS) (model: 7500 CE, Agilent, USA) with operating conditions of 1,350 W in RF Power, 6.0 mm sampling depth, 1.02 L·min⁻¹ carrier gas (Ar), and 4.5 mL·min⁻¹ helium flow rate. Reagent blanks, duplicate samples, and reference materials (Dorm-2, Dogfish muscle, NRC, Canada) were used for quality control. The estimated error as a relative standard deviation was between 2% and 8%.

Data Analyses

The correlation and cluster analyses were conducted by software SPSS (version 19.0 for Windows). The bivariate correlations were calculated using Pearson correlation coefficients with a two-tailed test of significance. Prior to cluster analysis, the elements were reduced to three factors using factor analysis by extracting principal components. The obtained three factors were subsequently hierarchically clustered by Ward’s method with squared Euclidean distance as the interval.

Results and Discussion

Heavy Metal Distributions of the Samples

The collected samples were analyzed to study their heavy metal concentrations and distributions. The elemental concentrations varied greatly with the types of samples (Fig. 2). Lacustrine sediment, lakeside sediment, and soil samples showed comparable elemental distributions with average heavy metal concentrations of: As 20-70 mg·kg⁻¹, Sb 1.4-2.0 mg·kg⁻¹, Pb 32-48 mg·kg⁻¹, Fe 4.2-6.9 mg·kg⁻¹, Mn 420-660 mg·kg⁻¹, Zn 100-120 mg·kg⁻¹, Co 16-6 mg·kg⁻¹, Cd 0.6-0.8 mg·kg⁻¹, and Hg 0.16-0.25 mg·kg⁻¹. It is suggested that they might have originated from the same sources, because surface runoff may flush the soil into the lake bank and form lakeside sediment; subsequently the lakeside sediment may deposit onto the bottom of the lake and form lacustrine sediment. The sinter samples that deposited from the spring water presented high As (158 mg·kg⁻¹), Sb (~8 mg·kg⁻¹), Pb (~190 mg·kg⁻¹), Mn 420-660 mg·kg⁻¹, Zn 100-120 mg·kg⁻¹, Co 16-6 mg·kg⁻¹, Cd 0.6-0.8 mg·kg⁻¹, and Hg 0.16-0.25 mg·kg⁻¹. It is attributed to the fact that groundwater in the Yangzonghai area was rich in As, Sb, and Hg [20]. The altered volcanic rock showed similar elemental distributions to that of sinter except for the relatively lower As contents (43.7 mg·kg⁻¹). The geochemistry of metamorphic rock is greatly determined by its precursor and therefore the volcanic rocks in this area should contain high contents of chalcophile elements, which is common due to the vast presence of sulfide-rich magmas [25]. The As therein may be leached out by waster during metamorphism. The slope deposit and chemical waste samples showed similar chemical compositions with high contents of heavy metals, including As (175-370 mg·kg⁻¹), Sb (~8 mg·kg⁻¹), Pb (~190 mg·kg⁻¹), etc.
Zn (500-1,000 mg·kg⁻¹), Cd (4-7 mg·kg⁻¹), and Hg (1.2-4.2 mg·kg⁻¹). Because most slope deposit samples were collected near the phosphate plant where we also sampled the chemical wastes, the similarity between slope deposits and chemical waste samples should be attributed to the contamination of the phosphate plant. The extremely high concentrations of As, Pb, Zn, Cd, and Hg in the chemical waste confirmed the severe pollution of this chemical industry. Compared with sinter samples, the chemical wastes exhibited comparable concentrations of Sb and Hg, doubled As contents, and much higher concentrations of Pb, Zn, Co, and Cd – reflecting the different characteristics of heavy metals contributed by groundwater and the phosphate plant.

Table 1 shows the comparisons on the As concentrations in the sediment between Yangzonghai Lake and other lakes. Arsenic concentrations in the lake sediments of Yangzonghai are significantly higher than that of other Yungui plateau lakes, where As mainly originated from natural sources [26, 27]. It is indicated that either groundwater or human pollution elevated the As contents in Yangzonghai Lake sediments. The As contents in Yangzonghai are also higher than that in freshwater lakes of the East China plain, Lake Cildir in Turkey, and Lake Geneva [28-30]. The As concentrations in the sediments from Lake Dianchi, however, are generally higher than that of this study, because Dianchi is located near Kunming and was severely polluted [31].

<table>
<thead>
<tr>
<th>Source of As</th>
<th>Source</th>
</tr>
</thead>
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<tr>
<td>Natural source and human pollution</td>
<td>This study</td>
</tr>
<tr>
<td>Human pollution</td>
<td>[26]</td>
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<td>[27]</td>
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<td>[29]</td>
</tr>
<tr>
<td>Human pollution</td>
<td>[30]</td>
</tr>
</tbody>
</table>

Table 1. Comparisons on the As concentrations in the lake sediment between Yangzonghai and other lakes.
Correlations between As and Heavy Metals

In natural conditions, As is usually positively correlated with Sb, Hg, and Pb due to their analogous geochemical behaviors [26], and Fe and Mn ascribed to the adsorption of As by metal oxides [4, 7]. Therefore, it is important to study the correlations between As and these elements to investigate the behaviors and potential sources of As in the samples. As shown in Table 2, As is strongly positively correlated with Sb, Bi, Fe, Zn, Pb, and Cd, with significant correlation at the 0.01 level, consistent with the above statement.

However, these results were calculated based on the data of all samples, while the correlation coefficients may vary with the types of the samples. As shown in Fig. 3, As shows low correlations with all other heavy metals in both soil and sinter samples. In contrast, As shows strong positive correlations with almost all elements in the slope deposit and chemical waste samples. These phenomena were partly attributed to the low quantities of both types of samples, and partly ascribed to the fact that most slope deposit samples were polluted by the phosphate plant where the chemical waste (with relatively constant chemical compositions) was produced. Arsenic (As) shows moderate correlation (r2 value of 0.317) with Sb in the lakeside sediments, and the value was increased to 0.975 in the lacustrine sediments. In addition, As exhibits moderate correlations with Zn, Cd, and Fe in the lacustrine sediments and low correlation in the lakeside sediments. The enhanced correlation coefficients in the lacustrine sediments than those in the lake sediments can be explained as follows. First, the lakeside sediment was mainly formed from soil through flushing of surface water, while the lacustrine sediment was generated from lakeside sediment via sedimentation. During these processes, As and Sb acted coherently due to their analogous geochemical behaviors and their correlations were gradually increased in the sediments. Second, the groundwater and human pollution will discharge heavy metals (such as As, Zn, and Cd) into the lakeside sediment and lacustrine sediment. The correlations between As and these elements were enhanced as a result. Third, arsenic adsorption by hydrous metal oxides is only significant in aqueous systems. Therefore, the correlation between As and Fe was increased in the lacustrine sediments. Note that all samples except for the slope deposits and chemical wastes show extremely low correlation coefficients between As and Pb, indicating that the high correlation between them in Table 2 was probably merely a coincidence.

Enrichment Factors of Arsenic and Heavy Metals

Enrichment factor (EF) was widely used to assess the relative enrichment and contamination of heavy metals in sediments and soils [32]. Aluminum was commonly used as the reference element to calculate the EF values of heavy metals due to its conservative behavior during geochemical processes [33]. The EF value was calculated using the following equation:

\[
EF = \frac{(C_{d}/C_{b})_{\text{sample}}}{(C_{d}/C_{b})_{\text{background}}} \tag{1}
\]

where \(C_{d}\) and \(C_{b}\) are the contents of assessed metal and aluminum, respectively. The average elemental concentrations of soils of Yunnan province were selected as the background values, which are As concentration of 10.8 mg·kg⁻¹, Sb 1.75 mg·kg⁻¹, Bi 0.52 mg·kg⁻¹, Fe 4.55%, Al 8.56%, Zn 80.5 mg·kg⁻¹, Pb 36 mg·kg⁻¹, and Cd 0.1035 mg·kg⁻¹.

Fig. 4 shows the EF values of As, Sb, Pb, and Cd in the Yangzonghai samples. Although the EF values may be not suitable to assess the sinter, rock, and chemical waste sam-

<table>
<thead>
<tr>
<th></th>
<th>As</th>
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<th>Fe</th>
<th>Mn</th>
<th>Ni</th>
<th>Zn</th>
<th>Pb</th>
<th>Co</th>
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<tr>
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<tr>
<td>Bi</td>
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<td>0.465**</td>
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<td></td>
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<tr>
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<td>0.182</td>
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</tr>
<tr>
<td>Fe</td>
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<td>0.234*</td>
<td>0.409**</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Mn</td>
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<td>-0.239*</td>
<td>-0.101</td>
<td>0.465**</td>
<td>0.326**</td>
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<td></td>
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</tr>
<tr>
<td>Zn</td>
<td>0.695**</td>
<td>0.490**</td>
<td>-0.013</td>
<td>0.763**</td>
<td>0.291*</td>
<td>0.308**</td>
<td>-0.025</td>
<td>0.115</td>
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<tr>
<td>Pb</td>
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<td>Co</td>
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<td>-0.191</td>
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<td>0.926**</td>
<td>0.389**</td>
<td>0.954**</td>
<td>0.047</td>
<td>0.145</td>
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<td>Cd</td>
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<td>0.491**</td>
<td>-0.007</td>
<td>0.796**</td>
<td>0.263*</td>
<td>0.290**</td>
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<td>0.108</td>
<td>0.982**</td>
<td>0.959**</td>
<td>0.035</td>
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</tbody>
</table>

The number of value (n) for each data set is 76.
**Correlation is significant at the 0.01 level.
*Correlation is significant at the 0.05 level.
Bold means strong correlation between As and other elements.
ples, their relative EF values can reflect the accumulation of heavy metals in these samples. Arsenic was significantly enriched (EF>2) in most of the samples, including all altered volcanic rocks, 80% lakeside sediments, all sinter, 5/7 slope deposit samples, and all of the chemical waste samples from the phosphate plant. The lakeside sediment and slope deposit samples showed comparable EF values of 1-55, while the chemical waste and sinter samples exhibited extremely high As enrichments with maximum EF values of higher than 200. Note that the sinters that deposited from spring water presented 30 times higher EF values than those of chemical wastes, suggesting that groundwater may even possess higher capacity of As pollution. Only 25% of lacustrine sediment samples and nearly 2/3 soil samples exhibited EF values of >2, and the maximum EF values were less than 10 – much lower than that of other samples. The EF values of As in the soil samples were close to those of reference soils, suggesting that the soil in the Yangzonghai area was relatively unpolluted. The groundwater or chemical pollution subsequently increased the As enrichment in the lakeside sediments. The As enrichment in the lacustrine sediments was recovered to less than five, because As in the sediment may be released into lake water under reduction conditions [4, 8].

Fig. 3. Correlations between As and Fe, Sb, Bi, Cd, Zn, and Pb in: (a) lacustrine sediment, (b) lakeside sediment, (c) soil, (d) slope deposit, (e) sinter, and (f) chemical waste samples. The horizontal coordinate, left vertical coordinate, and right vertical coordinate for all of the patterns are As concentration (mg·kg⁻¹), concentrations of heavy metals (mg·kg⁻¹), and concentrations of Pb and Zn (mg·kg⁻¹), respectively.
Sb and Pb were dramatically enriched in sinter, slope deposit, and chemical waste samples, and the EF values of Sb in sinters were orders of magnitude higher than those in chemical wastes. Therefore, groundwater around the Yangzonghai area may also contribute a large amount of Sb into the ambient environment. Lakeside sediments that exhibited high As enrichments did not show significant enrichments of Sb and Pb, suggesting that As in the lakeside sediment may be additionally contaminated by groundwater or anthropogenic activities. Almost all samples presented EF values of Cd of higher than 10. The Cd enrichments were higher than any other heavy metals in the slope deposit and chemical waste samples, indicating that the phosphate plant produced products or wastes with high Cd contents. Note that the sinter samples also showed high Cd enrichments, suggesting that groundwater also possess the potential of Cd contamination. Therefore, the high EF values of Cd in the lakeside sediment samples were contaminated from groundwater and the chemical industry.

Cluster Analysis

Cluster analysis is a useful method to group samples according to their common characteristics, which may determine the potential sources of heavy metals [34, 35]. Fig. 5 shows the dendrogram of the samples calculated...
from the hierarchical cluster analysis. The samples can be classified into five groups. All lacustrine sediment samples, seven soil samples, and three lakeside sediment samples form group 1. Most lakeside sediments (12 samples), seven soil samples, and three slope deposit samples constitute the second group. The Euclidean distance between groups 1 and 2 is 3, suggesting that they have a close relationship. Thirty-three out of 40 samples in groups 1 and 2 were collected near springs, indicating that these samples were greatly affected by groundwater. Group 3 includes all sinter samples (7 samples), three soil samples, nine lakeside sediment samples, and two slope deposit samples. Only 11 of the total of 25 samples in group 3 were affected by springs/groundwater. The Euclidean distance between group 3 and the former two groups is enlarged to 9. Group 4 consists of two slope deposit samples that collected near both spring F and the phosphate plant, and two chemical waste samples from the plant. However, the internal distance between slope deposit and chemical waste samples is 3, which is much higher than that of groups 1, 2, and 3. The last cluster (group 5) includes all altered volcanic rock samples and two chemical waste samples. The internal distance is even higher – reaching a value of 16, suggesting that the relationship between altered volcanic rocks and chemical wastes is distant. All samples influenced by groundwater are included in groups 1, 2, and 3, while 80% of samples collected near the phosphate plant belong to groups 4 and 5. In addition, 80% of samples affected by both chemical industry and springs belong to groups 1, 2, and 3. These results indicate that groundwater and chemical pollution posed distinctive influences of heavy metals on the samples, and the influence of groundwater was more intensive than that of chemical pollution from the phosphate plant. The influence of the chemical industry was quite limited and localized.

**Conclusion**

In summary, we investigated the occurrence and behavior of Arsenic in and around a typical fault-controlled plateau lake – Yangzonghai in China. The elemental compositions, correlation coefficients, and cluster analyses on the samples indicate that the groundwater and human activities exhibited different influences on the samples. The samples affected by groundwater presented high concentrations of chalcophile elements, including As, Sb, and Hg, while that by the phosphate plant showed elevated concentrations of almost all heavy metals.

The As geochemistry in the soil and sediments were affected by both groundwater and the phosphate plant, and the groundwater induced higher As enrichment than that of the plant. In addition, the groundwater influence was more intensive than that of the plant. This study shows a good prototype of plateau lake, where As occurrence was affected by both natural and anthropogenic activities. Furthermore, this study indicates that groundwater is an important factor that should be considered when studying As pollution in fault-related plateau lakes.

**Supplementary Materials**

Original data. Elemental concentrations of the samples in and around the Yangzonghai Lake. All units are mg·kg⁻¹ except for Al and Fe.
<table>
<thead>
<tr>
<th>Location</th>
<th>Type</th>
<th>Al (%)</th>
<th>Fe (%)</th>
<th>Mn</th>
<th>Ni</th>
<th>Zn</th>
<th>Pb</th>
<th>As</th>
<th>Sb</th>
<th>Hg</th>
<th>Bi</th>
<th>Co</th>
<th>Cd</th>
</tr>
</thead>
<tbody>
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<td>7. Yongjin park near spring C N24.9689, E103.0369</td>
<td>sinter</td>
<td>2.46</td>
<td>0.98</td>
<td>26.86</td>
<td>7.19</td>
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<td>33.63</td>
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<td>3.27</td>
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<tr>
<td>8. Yongjin park near spring C N24.9608, E103.0367</td>
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<td>1.60</td>
<td>1.47</td>
<td>7.42</td>
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<td>16.75</td>
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<td>11. valley near spring E N24.9661, E103.0100</td>
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Acknowledgements

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