Journal of Hazardous Materials 440 (2022) 129860

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

# Sedimentary records and stable lead isotopes reveal increasing anthropogenic impacts on heavy metal accumulation in a plateau lake of China over the last 100 years

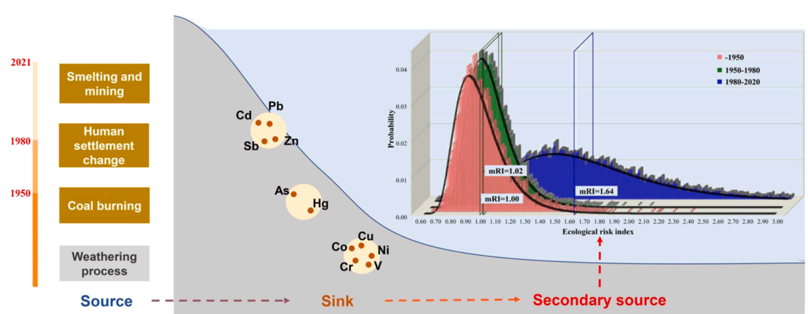
Xiang Gu[a], Xiaotong Han[b,c], Yixuan Han[b], Wenlei Luo[b,d], Muhua Feng[b], Di Xu[b], Peng Xing[b,*], Qinglong L. Wu[b,d]

[a] State Key Laboratory of Water Environment Simulation, School of Environment, Beijing Normal University, Beijing 100875, China
[b] State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China
[c] College of Life Sciences, Hebei University, Baoding 071002, China
[d] The Fuxianhu Station of Deep Lake Research, Chinese Academy of Sciences, Chengjiang 652500, China

## HIGHLIGHTS

- Correlations between heavy metals changed in reverse among three different periods.
- Anthropogenic sources obviously accelerated the accumulation of Cd, Pb, Sb, and Zn.
- Smelting and mining of galena was the main source of Pb in the lake sediments.
- A moderate ecological risk of metals might happen, with the possibility of 90 %.
- Cd was the most sensitive heavy metal in evaluating the combined ecological risk.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

Editor: <Jörg Rinklebe>

*Keywords:*
Heavy metal
Stable isotope
Source apportionment
Ecological risk
Plateau lake

## ABSTRACT

Plateau lakes in China are faced with heavy metal contamination. The present study investigated the concentrations, sources, and ecological risks of 11 heavy metals in sediments of Lake Fuxian over the last 100 years. The concentrations of V (124–244 mg/kg), Cr (69.3–127 mg/kg), Cu (59.4–105 mg/kg), Ni (36.0–66.3 mg/kg), and Co (15.4–25.5 mg/kg) significantly decreased in the last 100 years while Cd, Sb, Pb, and Zn had contrary trends. In the last 100 years, correlations between the heavy metals changed in reverse due to the combined influences of lithogenic and anthropogenic sources. In the last 40 years, the lithogenic sources mainly contributed to the concentrations of As, Hg, V, Cr, Co, Ni, and Cu, with the rates of 55 %– 85 %. Moreover, Cd, Sb, Pb, and Zn were mainly affected by anthropogenic sources (e.g., mining activities and human settlement change), with the contribution rates of 53 %– 80 %. The change of stable Pb isotope in the sediment cores indicated that galena mining was the main source of Pb. With increasing anthropogenic activities, there was a possibility of 90 % that a moderate ecological risk might appear in the last 40 years. Among the heavy metals, Cd should attract attention because it contributed 96 % to the combined ecological risk.

* Corresponding author.
*E-mail address:* pxing@niglas.ac.cn (P. Xing).

https://doi.org/10.1016/j.jhazmat.2022.129860
Received 13 July 2022; Received in revised form 21 August 2022; Accepted 24 August 2022
Available online 28 August 2022

**Environmental Implication**

The plateau lakes in China are faced with heavy metal contamination with increasing anthropogenic influences in the past decades. This is the first time to quantify the anthropogenic impacts on 11 heavy metal concentrations in sediments of Lake Fuxian, the third deepest lake in China, in three historical periods (−1950, 1950–1980, and 1980–2020) based on high-resolution sedimentary records. The change of staple lead isotope in the 16 sediment cores also reflected the increasing influences of mining activities on heavy metal accumulation in the lake. Increasing ecological risks of heavy metals are happening in the plateau lakes of China.

## 1. Introduction

Lakes account for almost 90 % of surface freshwater in the planet, playing an important role in supporting remarkable biodiversity with hundreds of thousands of species and providing key ecosystem services for human beings (Reynaud and Lanzanova, 2017; Huang et al., 2022). However, global lake ecosystems have been threatened by various environmental contaminants released from multiple anthropogenic activities with industrial development and increasing population (Sarkar et al., 2021; Varol et al., 2022). Among the contaminants, heavy metal (loid)s are considered as important lake sediment pollutants due to their ubiquity, high toxicity, non-biodegradability, and bioavailability for aquatic organisms (Rossi et al., 2017). In the last decade, heavy metal concentrations exceeding the sediment quality guidelines (SQG) have been observed in Lake Bafa of Turkey (Algül and Beyhan, 2020), Lake Illawarra of Australia (Schneider et al., 2015), Lake Geneva of Switzerland (Gascón Díez et al., 2017), and Lake Taihu of China (Li et al., 2018). Nowadays, heavy metals have been important ecological indicators of lakes or reservoirs and attracted increasing attention worldwide.

Sediments usually serve as final recipients of heavy metals that have been discharged into lakes (Dang et al., 2021). Meanwhile, the buried heavy metals might be released into overlying water when hydrodynamic, physical, or chemical conditions change in lakes, posing potential threats to aquatic organisms and drinking water safety (Geng et al., 2022). As credible environmental archives, sediment cores are often used to reconstruct past ecosystem changes in lakes (Sanchez-Cabeza and Ruiz-Fernández, 2012; Lintern et al., 2016). Combined with sedimentary chronology, vertical distribution characteristics of heavy metals in profiles of sediment cores can be used to investigate influences of historical human activities or climate change on metal accumulation in lakes (Gascón Díez et al., 2017). Moreover, sediment cores can provide background concentrations of heavy metals in lakes that experienced no or little anthropogenic impacts, which is essential for assessing contamination or ecological risks of heavy metals in sediments (Lintern et al., 2016; Gu et al., 2022a). Since the inception of industrial revolution, anthropogenic activities including burning fossil fuels, improper disposal of industrial and domestic wastes, overuse of agrochemical, mining and smelting operations, and traffic expansion are the most common sources of heavy metals entering into aquatic systems (Jacob et al., 2018; Rinklebe et al., 2019; Palansooriya et al., 2020; Ustaoğlu and Islam, 2020). However, it was difficult to quantify the human impacts on heavy metal concentrations in lake sediments in different historical periods due to the lack of environmental monitoring and records.

Plateau areas are widely distributed in China. Due to the high background concentrations of trace elements, lakes on the Yunnan-Guizhou Plateau are faced with severe heavy metal contamination (Wang et al., 2021a). Moreover, increasing concentrations of heavy metals were also observed in the plateau lakes caused by denser anthropogenic activities (e.g., mining activities, traffic emission, and disposal of industrial and domestic wastes) in these regions (Zhao et al., 2011; Lin et al., 2018; Liu et al., 2022). Lake Fuxian is the third deepest lake in China, located on the Yunnan-Guizhou Plateau (Xing et al., 2020). A dramatic elevation of lead (Pb) concentration since 1985 was observed in sediments of southern Fuxian Lake mainly due to the huge non-ferrous metal production in Yunnan Province over the past century (Liu et al., 2013). Moreover, the industrial and agricultural development, urbanization, and tourism have also increased heavy metal discharges into Lake Fuxian (Zhang et al., 2015; Li et al., 2017). However, a comprehensive investigation of historical records of concentrations, sources, and ecological risks of heavy metals in the whole Lake Fuxian is lacking considering the important ecological and economic values of the lake in China. In the present study, Lake Fuxian was selected to (1) investigate the historical concentration distribution of 11 heavy metals based on sedimentary records, (2) understand the sources of heavy metals in the plateau lake based on the correlation analysis of metal concentrations and stable Pb isotopes, (3) quantify the anthropogenic impacts on heavy metal in different historical periods, and (4) comprehensively evaluate the ecological risks of heavy metals in the lake during the last hundred years.

## 2. Materials and methods

### 2.1. Study area and sample collection

Lake Fuxian (24°35′N, 102°50′E, 1788.5 m a.s.l.) is a plateau lake in Yuxi City, Yunnan Province (Fig. 1). With a surface area of 212 $km^2$ and an average depth of 95 m, the lake has a water volume of nearly 20.6 billion cubic meters, ranking first among the freshwater lakes in China. Wang et al. (2011) found the sedimentation rate ranged from 0.096 cm $yr^{-1}$ to 0.192 cm $yr^{-1}$ in seven sediment cores of Lake Fuxian (Table S1). Satellite remote sensing revealed that human settlements are widely distributed in the north and southwest of Lake Fuxian since the 1990s (Fig. 1) (Gong et al., 2019). In December 2021, 16 sediment cores were collected from Lake Fuxian using a gravity corer equipped with a PVC tube of 50 cm in length and 11 cm in outer diameter (Fig. 1). Generally, they were uniformly distributed in the lake and detailed coordinates were listed in Table S2. Each core was sectioned into consecutive 1-cm increments on-site with a plastic ware. A total of 262 subsamples were obtained for further analysis. The sedimentation rates of the 16 sediment cores were extracted from the grid of sedimentation rates of the whole lake using ArcGIS 10.2, which were then used to recalculate the deposition ages of these sediment cores (Table S2). As a whole, the profiles of the sediment cores at different depth represented three historical periods, which were − 1950 (bottom−12.5 cm), 1950–1980 (12.5 cm − 7.5 cm), and 1980–2021 (7.5 cm − 0 cm) (Fig. 1).

### 2.2. Sample pretreatment and analysis

In laboratory, the sediment samples were freeze dried, uniformly ground, and sieved through a 2 mm mesh. Approximately 0.12 g of the freeze-dried sediment sample was digested with $HCl$-$HNO_3$-$HF$-$HClO_4$ in a Teflon beaker modified by Liu et al. (2013). The contents of Al, Fe, Mn, P, V, and Zn were analyzed by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES, Teledyne Leeman Labs, Prodigy, USA) and As, Cd, Co, Cr, Cu, Hg, Ni, Sb, and Pb were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent 7700x, USA). The standard reference material (GSD-9) was employed to ensure the reliability of the analytical methods. As a result, the recoveries of the elements of the GSD-9 ranged from 92 % to 106 %. The limit of detection (LOD) for each analyte was calculated as three times the standard deviation of the six blank samples and listed in Table S3. Besides, the coefficient of variation for the repeated samples was below 10 %. According to Chen et al. (2018), the total nitrogen (TN) and total organic carbon (TOC) of the sediment samples were measured by a CHNS elemental analyzer (EA3000, EuroVector, Italy). Additionally, the grain size of the sediment samples was measured using a laser particle analyzer after removing organic matter and carbonates using 30 % $H_2O_2$ and 10 % HCl (Wang et al., 2021b).

### 2.3. Stable Pb isotope analysis

Stable Pb isotopes have been a powerful tool for discriminating between natural and industrial sources of Pb (Shotyk et al., 2015; Wang et al., 2021c). Lead isotopes ($^{208}Pb$, $^{207}Pb$, and $^{206}Pb$) in sediment samples were measured by ICP-MS (Agilent 7700x, USA) following $HCl$-$HNO_3$-$HF$-$HClO_4$ digestion, which was described by Liu et al. (2013). A standard reference material (SRM981-NIST) was placed at every 10 samples interval for further calibration and analytical control during the measurement. The standard deviations of the $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios related to the repeated measurements of the standard were $< 0.05$ % and $< 0.10$ %, respectively.

### 2.4. Multiple linear regression of absolute principal component scores (APCS-MLR)

The principal component analysis (PCA) is often applied for source identification of heavy metals in sediments (Micó et al., 2006; Dang et al., 2021). Based on the hypothesis that heavy metal concentrations are equal to the gross contributions from all potential sources, the multiple linear regression of absolute principal component scores (APCS-MLR) can estimate individual contribution of each source (Haji Gholizadeh et al., 2016; Jiao et al., 2019). The APCS-MLR model was described as follows:

$$C_i = X_{0i} + \sum_{j=1}^{P} X_{ji} \times APCS_j \quad (1)$$

where $C_i$ is the concentration of heavy metal $i$ in sediments; $APCS_j$ is the absolute principal component score of potential source $j$; $P$ is the number of principal components; $X_{ji}$ is the regression coefficient between concentration and $APCS_j$ of heavy metal $i$; and $X_{0i}$ is the constant term of the MLR, representing the unknown source (Jiao et al., 2019).

Subsequently, the contributions of each source to heavy metal concentrations were calculated as follows (Haji Gholizadeh et al., 2016):

$$PC_{ji} = \frac{\left|X_{ji} \times \overline{APCS_j}\right|}{|X_{0i}| + \left|\sum_{j=1}^{P} X_{ji} \times \overline{APCS_j}\right|} \times 100\% \quad (2)$$

$$PC_{unknowni} = \frac{|X_{0i}|}{|X_{0i}| + \left|\sum_{j=1}^{P} X_{ji} \times \overline{APCS_j}\right|} \times 100\% \quad (3)$$

where $PC_j$ is the contribution of principal component $j$ and $PC_{unknowni}$ is the contribution of the unknown source to heavy metal $i$.

### 2.5. Probabilistic ecological risk assessment based on Monte Carlo simulation

The ecological risk index (RI) proposed by Hakanson (1980) has been widely used to assess ecological risks of heavy metals in sediments and soils (Kumar et al., 2019; Gu et al., 2022b). In the present study, a modified ecological risk index (mRI) was employed to assess the ecological risk of 11 toxic heavy metals in sediments of Lake Fuxian, which was calculated as follows:

$$Er_i = T_r \times \frac{C_i}{B_i} \quad (4)$$

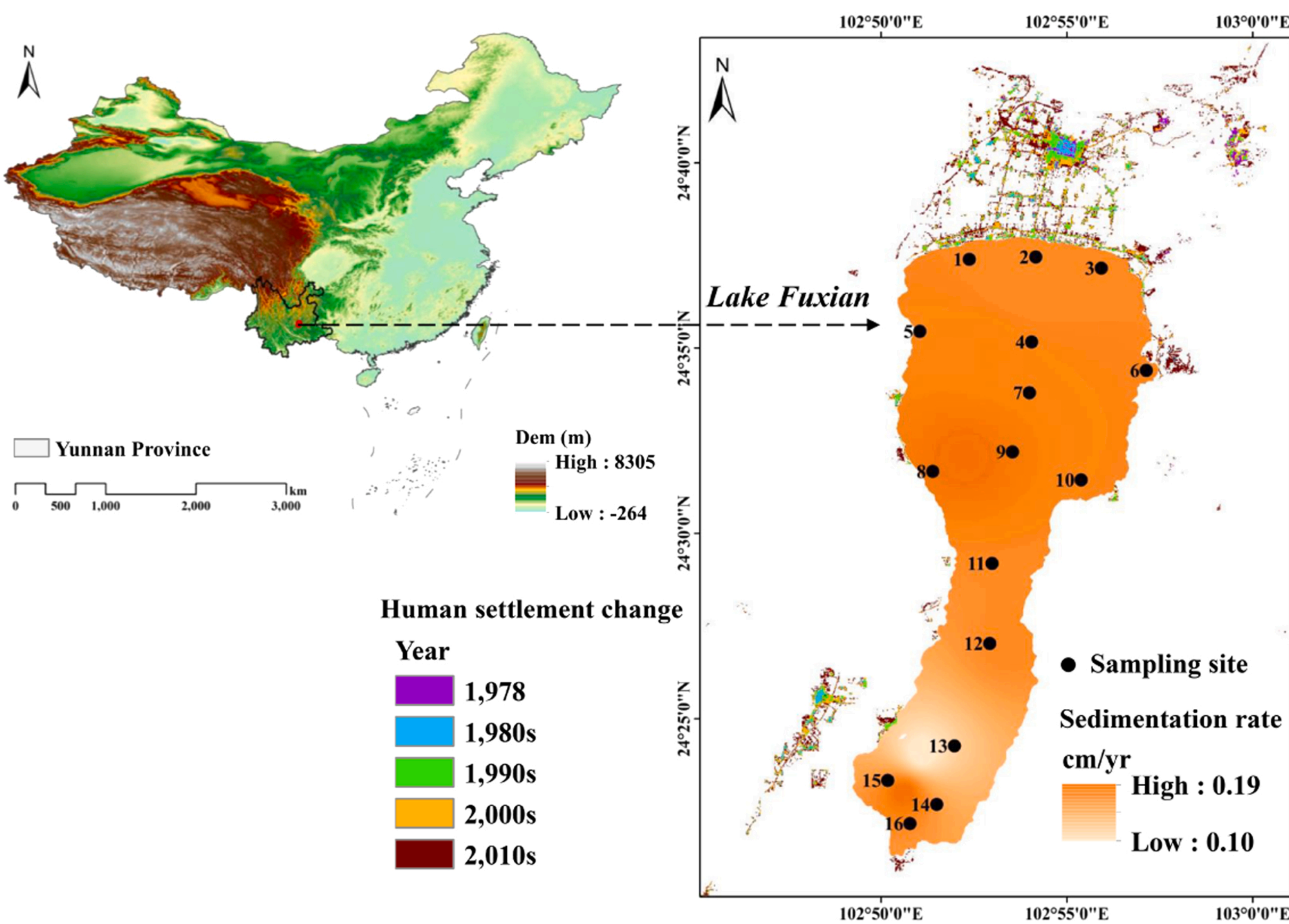



**Fig. 1.** Location of study area and sampling sites. Data of human settlement changes around Lake Fuxian was abstracted from the national database developed by Gong et al. (2019).

$$\text{mRI} = \frac{\sum_{i=1}^{n} Er_i}{\sum_{i=1}^{n} Tr_i} \quad (5)$$

where $Er_i$ is the ecological risk of heavy metal $i$; $Tr$ is the toxic-response factor of heavy metals, which is 40, 30, 10, 5, 5, 5, 5, 5, 2, 2, and 1 for Hg, Cd, As, Sb, Co, Ni, Cu, Pb, Cr, V, and Zn, respectively (Dang et al., 2021); $B_i$ is the background concentration of heavy metal $i$ in sediments of Lake Fuxian, which is the average concentration of heavy metal $i$ in sediment cores before 1950 when anthropogenic activities were rare around the lake. Five ecological risk levels are classified as follows: low (mRI $< 1.1$), moderate ($1.1 \leq$ mRI $< 2.3$), considerable ($2.3 \leq$ mRI $<$ 4.5), and very high (mRI $\geq 4.5$) risk (Gu et al., 2022b).

The probabilistic risk method, namely Monte Carlo simulation, allows for incorporating the uncertainty and variability related to the input parameters for risk assessment and estimate a full range of potential risks (Hosseini Koupaie and Eskicioglu, 2015). Recently, the Monte Carlo simulation has been successfully used in assessing probabilistic ecological and health risks of heavy metals in natural medium (Yang et al., 2019; Gu et al., 2022b). In summary, the $Er_i$ values of heavy metals in sediments of Lake Fuxian fit logistic or normal logistic distributions. Hence, the $Er_i$ values were defined as assumptions and the *mRI* values were defined as forecasts for the Monte Carlo simulation. Moreover, the sensitivity analysis was performed to determine the sensitivity of each heavy metal to the combined ecological risk assessment.

### 2.6. Statistical analysis

One-way ANOVA was performed to evaluate differences in heavy metals and other parameters of sediments in different periods at the level of $p < 0.05$. Spearman correlation analysis and APCS-MLR were performed to identify the relationships between heavy metals using SPSS Statistics 20 and Microsoft Excel 2019. The Monte Carlo simulations were executed for $10^5$ iterations to obtain reliable outputs using the Oracle Crystal Ball v11.1.24. Besides, the sensitivity analysis was carried out based on the coefficient of rank correlation (CRC) between each assumption and forecast. ArcGIS 10.2 was used to generate the sampling map and OriginPro 2020b was used for data visualization.

## 3. Results

### 3.1. Basic characteristics of sediment cores in Lake Fuxian

In the last 40 years, the average concentrations of TN and TOC in sediments of Lake Fuxian were 3.03 g/kg and 27.8 g/kg, respectively, significantly higher than those before 1980 ($p < 0.05$, Table 1). Silt particles were dominant in the sediment cores, with the average proportion of 72.5 %, 75.6 %, and 76.4 % in the periods of − 1950, 1950–1980, and 1980–2021, respectively (Table 1). Moreover, proportions of clay and sand separately ranged between 12.6 %– 34.4 % and 0.003 %– 17.2 %, showing no significant variation among different periods (Table 1).

In the sediment cores, the highest average concentrations of Al and Fe were 98.0 g/kg and 64.6 g/kg in the period of − 1950, significantly higher than those in 1950–1980 and 1980–2020 ($p < 0.05$, Table 1). The average concentrations of Mn ranged from 0.97 g/kg to 1.05 g/kg in different periods, with higher concentrations in the last 40 years. Moreover, it was observed that the average concentrations of P were 1.69 g/kg, 1.63 g/kg, and 1.63 g/kg in periods of − 1950, 1950–1980, and 1980–2021, respectively, showing no significant variations ($p > 0.05$, Table 1).

### 3.2. Vertical concentrations of heavy metals in sediment cores of Lake Fuxian

In the 16 sediment cores of Lake Fuxian, the concentrations of Co, Ni, V, Cr, and Cu generally had a decreasing trend from the bottom to the top layers, with the obvious fluctuations at the depth of 20.5 cm (Fig. 2a). Among them, V had the highest concentrations (124–244 mg/kg), followed by Cr (69.3–127 mg/kg), Cu (59.4–105 mg/kg), Ni (36.0–66.3 mg/kg), and Co (15.4–25.5 mg/kg). Specifically, concentrations of V, Cu, Ni, and Co were significantly higher in the periods of 1980–2020 and 1950–1980 than those before 1950 ($p < 0.05$, Fig. 2b). Moreover, differences of Cr concentrations were more significant among the three periods, with the average concentrations of 87.9 mg/kg, 98.6 mg/kg, and 105 mg/kg in 1980–2020, 1950–1980, and − 1950, respectively (Fig. 2b).

Furthermore, vertical distribution patterns of Hg, As, Sb, Cd, Pb, and Zn were different from the aforementioned heavy metals (Fig. 2c and e). Among them, concentrations of Hg ranged from 86.7 μg/kg to 120 μg/kg in the sediment cores of Lake Fuxian, showing no significant variations in different periods ($p > 0.05$, Fig. 2d). Arsenic was another heavy metal that had no significant variations among different periods, with the concentrations of 18.9–20.9 mg/kg (Fig. 2f). Concentrations of Cd, Pb, Sb, and Zn had similar vertical distribution characteristics, with an increasing trend since 1950 (Fig. 2c and e). Moreover, the average concentrations of Cd, Pb, Sb, and Zn in the sediment during 1980–2020 were 1.68 mg/kg, 75.9 mg/kg, 2.48 mg/kg, and 173 mg/kg, respectively significantly higher than those during − 1950 and 1950–1980 (Fig. 2d and f).

### 3.3. Correlations between heavy metals in sediment cores of Lake Fuxian in different periods

The correlation of 11 heavy metal in the sediment cores of Lake Fuxian varied among different periods (Fig. 3). In the period before 1950, all heavy metals except Pb were positively correlative with each other with a significance at 0.05 level (Fig. 3a). Lead was only positively correlative with As, Hg, Sb, Cd, and Zn, showing sno significant correlations with others. In the period of 1950–1980, more poor correlations appeared between heavy metals, such as Cd vs. V, Cr, and Ni, as well as

**Table 1**
Concentrations of total nitrogen (TN, g/kg), total organic carbon (TOC, g/kg), proportions of clay (%), silt (%), and sand (%), and Al (g/kg), Fe (g/kg), Mn (g/kg), and P (g/kg) in different periods of sediment cores of Lake Fuxian.

| | TN | TOC | Clay | Silt | Sand | Al | Fe | Mn | P |
|---|---|---|---|---|---|---|---|---|---|
| 1980–2021 | | | | | | | | | |
| Average | $3.03^a$ | $27.8^a$ | $20.7^a$ | $76.4^a$ | $2.97^a$ | $84.9^b$ | $56.9^b$ | $1.05^a$ | $1.63^a$ |
| Std. | 1.88 | 18.9 | 4.40 | 3.64 | 3.05 | 13.3 | 10.5 | 0.24 | 0.37 |
| 1950–1980 | | | | | | | | | |
| Average | $1.32^b$ | $10.7^b$ | $20.1^a$ | $75.6^{ab}$ | $4.29^a$ | $95.4^b$ | $62.3^b$ | $0.99^{ab}$ | $1.63^a$ |
| Std. | 0.32 | 2.96 | 5.88 | 4.78 | 4.49 | 6.36 | 7.65 | 0.16 | 0.25 |
| − 1950 | | | | | | | | | |
| Average | $1.16^b$ | $9.19^b$ | $21.9^a$ | $72.5^b$ | $5.58^a$ | $98.0^a$ | $64.6^a$ | $0.97^b$ | $1.69^a$ |
| Std. | 0.26 | 2.38 | 7.68 | 4.82 | 6.03 | 6.39 | 8.00 | 0.15 | 0.29 |

Note: Lower-case letters indicate significant differences among different periods ($p < 0.05$).

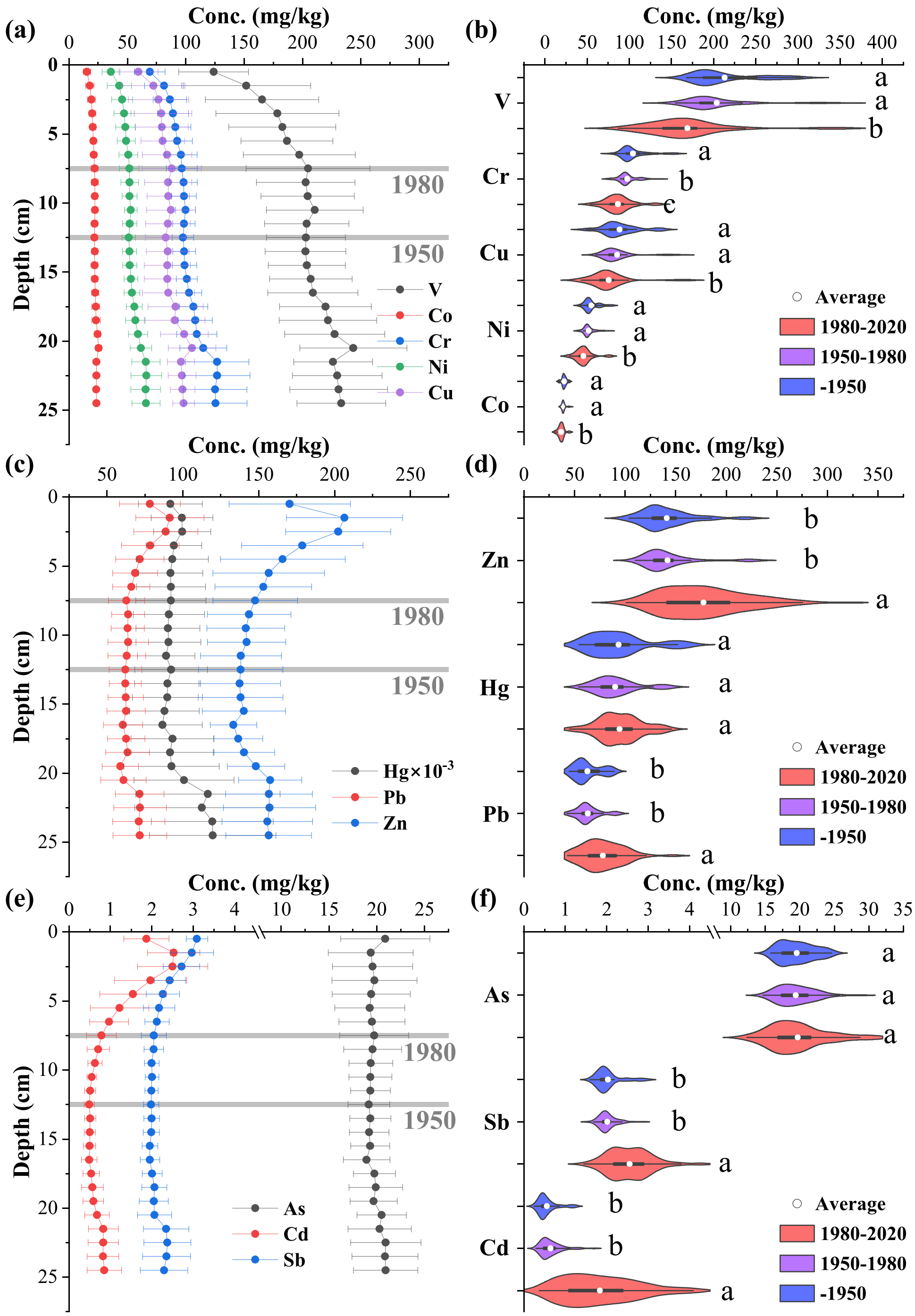



**Fig. 2.** Vertical distribution of heavy metals in sediment cores of Lake Fuxian (a, c, and e). Violin plots of heavy metal concentrations during – 1950, 1950–1980, and 1980–2020 (b, d, and f). Lower-case letters indicate significant differences among different periods ($p < 0.05$).

Zn vs. Ni (Fig. 3c). A remarkable shift of correlation matrix appeared in the period of 1980–2020, that was, significant negative correlations were observed between metals including Sb, Cd, and Pb and several others including V, Cr, Co, Ni, and Cu ($p < 0.05$, Fig. 3e). Moreover, Zn was also negatively correlative with V, Co, Ni, and Cu at a significance > 0.05 (Fig. 3e).

In each historical period, two principal components (PCs) of the 11 heavy metals in sediments were extracted, whereas the compositions of PCs varied among different periods (Fig. 3). In the period before 1950, PC2 was mainly loaded with Pb while PC1 was loaded with other heavy metals, explaining 87.4 % of the total variance (Fig. 3b). During 1950–1980, Pb, Cd, and Zn had higher loads in the PC2, which was different from that in the period before 1950 (Fig. 3d). In the last 40 years, the PC2 was characterized by Cd, Pb, Sb, and Zn and accounted for 29.8 % of the total variance, showing obvious different with that before 1950 (Fig. 3f).

### 3.4. *Quantitative source identification of heavy metals in sediment cores of Lake Fuxian in different periods*

Results of APCS-MLR revealed the contributions of three factors (i.e., lithogenic, anthropogenic, and unknown sources) to heavy metal concentrations in the sediment cores of Lake Fuxian (Fig. 4). Among the heavy metals, concentrations of V, Co, Ni, and Cu were mainly affected

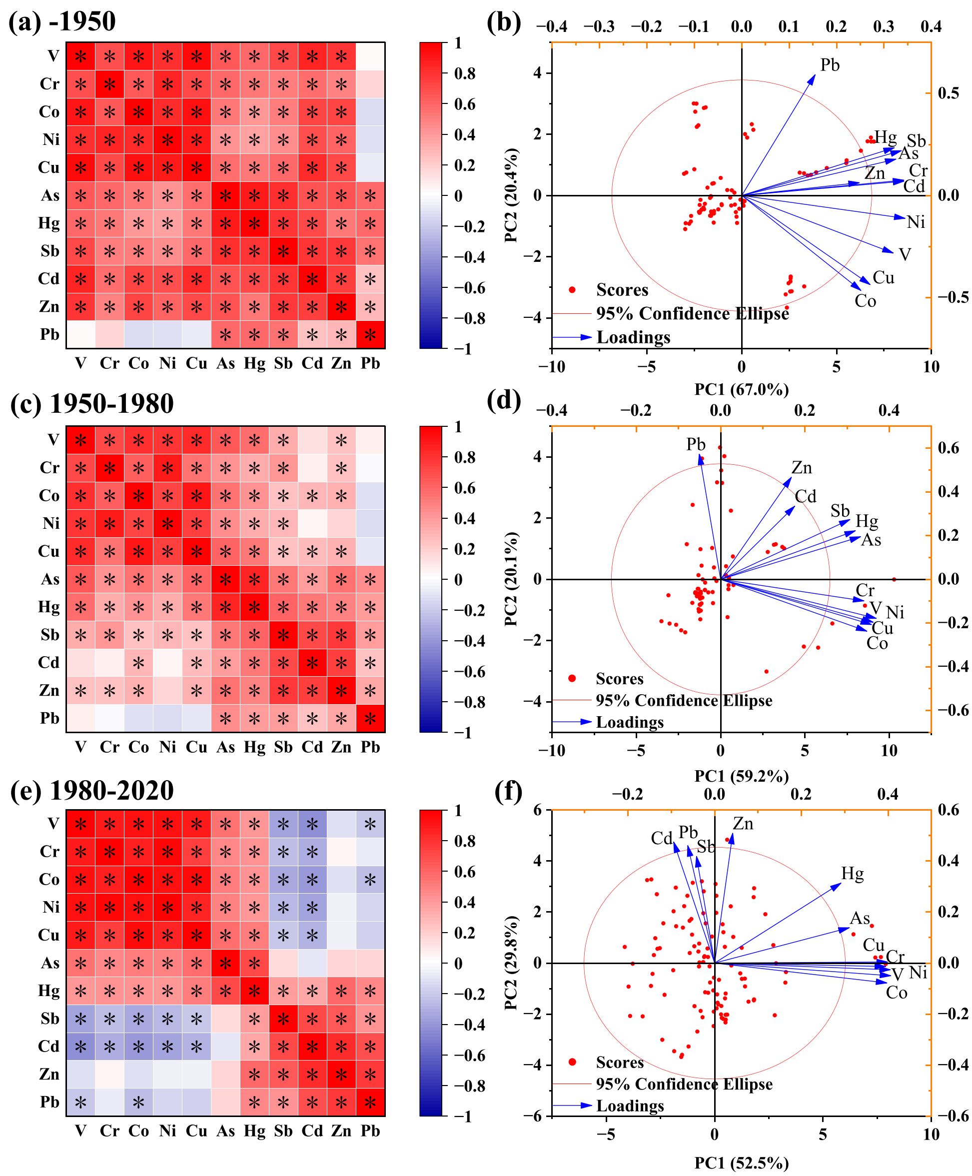



**Fig. 3.** Spearman correlation analysis (a, c, and e) and principal component analysis (b, d, and f) of heavy metal concentrations in sediments of Lake Fuxian during − 1950, 1950–1980, and 1980–2020. * represents a significance at 0.05 level.

by lithogenic sources over the last one hundred years, and the contributions of lithogenic sources generally increased from 1950 to 2020. Before 1950, Cr, As, and Hg in sediments were mainly influenced by anthropogenic sources. However, after 1950, especially in the last 40 years, the lithogenic sources turned into the major factors that influenced the concentrations of Cr, As, and Hg, with the contribution rates of 65 %, 67 %, and 55 %, respectively (Fig. 4). Differently, heavy metals including Sb, Cd, Pb, and Zn were mostly originated from anthropogenic sources in the three periods. It was estimated that anthropogenic sources contributed 53 %, 34 %, and 53 % to the concentrations of Sb in the periods of − 1950, 1950–1980, and 1980–2020, respectively (Fig. 4). Moreover, influences of anthropogenic sources on the concentrations of Cd, Pb, and Zn in sediments all increased during the last one hundred years, with the contribution rates of 73 %, 67 %, and 80 % in the last 40 years, respectively.

### 3.5. Stable lead isotopic ratios in sediment cores of Lake Fuxian in different periods

The ratios of $^{207}Pb/^{206}Pb$ in the sediment cores of Lake Fuxian ranged from 0.83 to 0.85, with an increasing trend from the bottom to top layers. Similar vertical distribution characteristics were observed for the ratios of $^{208}Pb/^{206}Pb$ whose values were between 2.06 and 2.09 (Fig. 5a). Moreover, the average ratios of $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ in the last 40 years were both significantly higher than those in the earlier two periods ($p < 0.05$, Fig. S3). Most the ratios of $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ in sediment cores of Lake Fuxian were in the range of that of coal in Yunnan (Fig. 5c). Moreover, it was observed that ratios of stable Pb isotopes in the sediment cores of Lake Fuxian in the period of 1980–2020 were closer to that of the galena in Yunnan. However, the ratios of stable Pb isotopes in the sediment cores were far less than that of the leaded gasoline once used in China (Fig. 5c).

The province of Yunnan is abundant with non-ferrous metal resources. The annual output of ten non-ferrous metals (i.e., Al, Mg, Cu, Pb, Zn, Ni, Co, Ti, Sn, and Hg) has dramatically increased from $7.48 \times 10^4$ tons in 1978–$5.11 \times 10^6$ tons in 2020 (Fig. 5b). Accompanied with the industrial development, the coal consumption increased from $1.10 \times 10^6$ tons in 1957–$5.10 \times 10^7$ tons in 2013. However, a decreasing trend of coal consumption has been observed since 2013 (Fig. 5b).

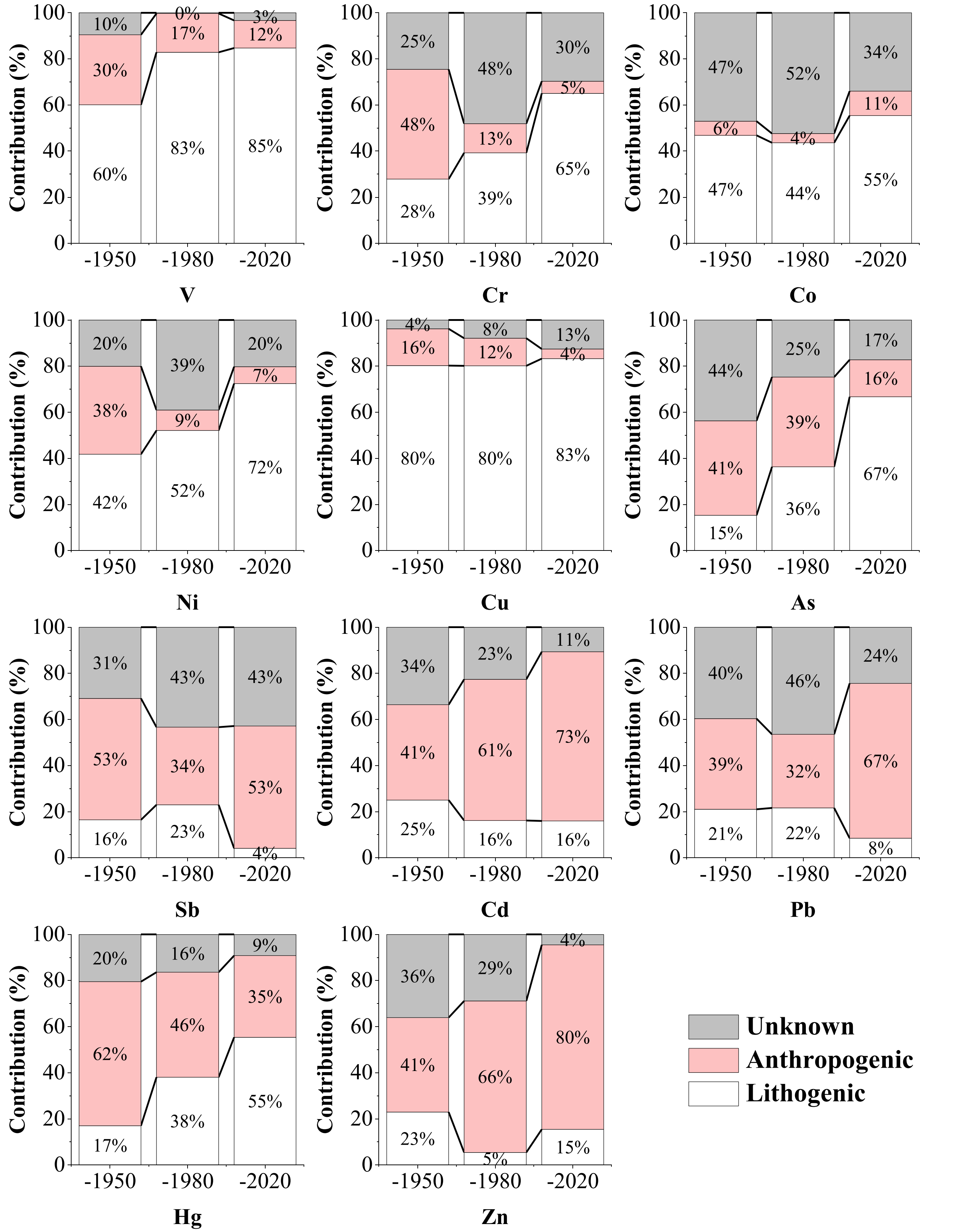



**Fig. 4.** Contribution rates of lithogenic, anthropogenic, and unknown sources to heavy metal concentrations in sediments of Lake Fuxian during – 1950, 1950–1980, and 1980–2020.

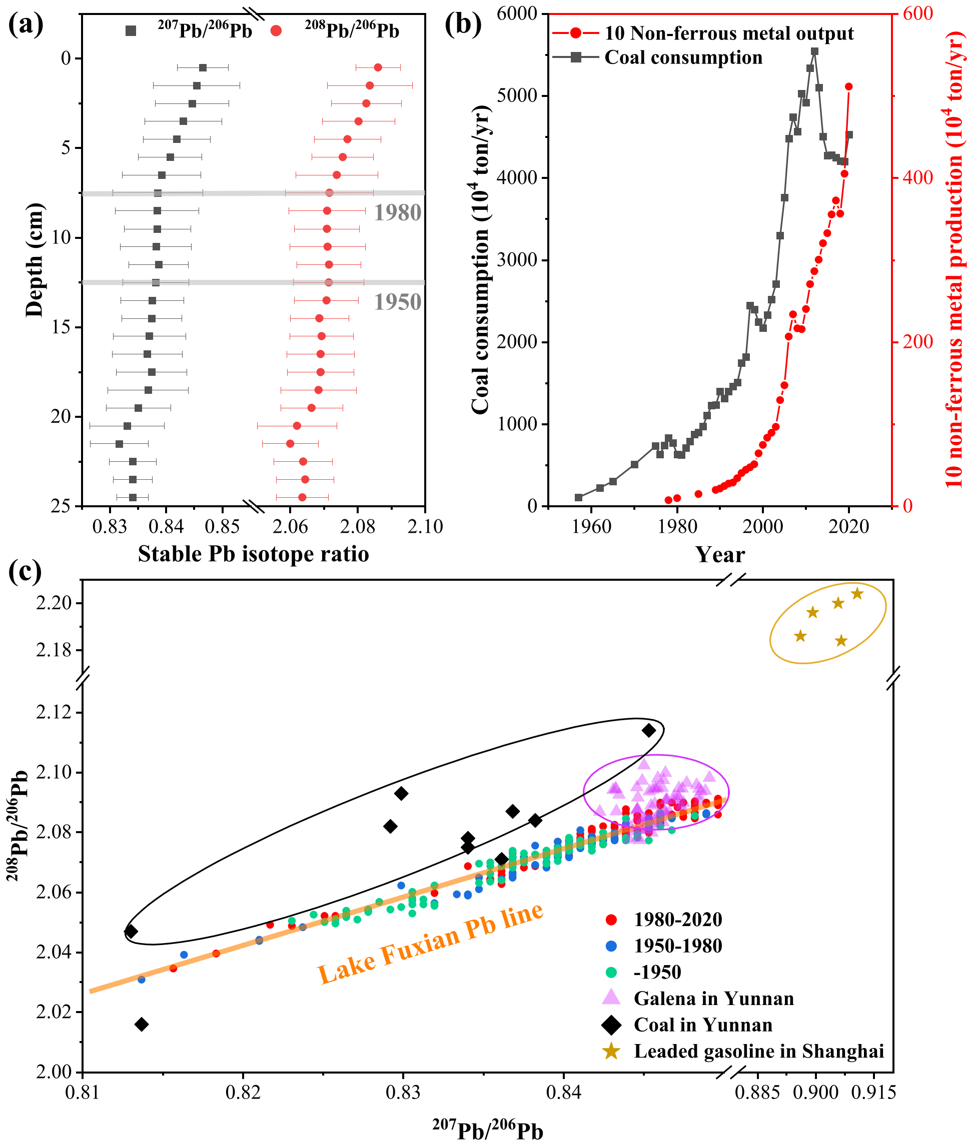



**Fig. 5.** Vertical distribution of stable Pb isotope compositions in sediment cores of Lake Fuxian (a), coal consumption and 10 non-ferrous metal production in Yunnan during 1957–2020 (Yunnan Provincial Bureau of Statistics, 2021) (b), and dispersion plot of stable Pb isotope compositions in sediment cores of Lake Fuxian, galena and coal in Yunnan, and leaded gasoline in Shanghai (Xue et al., 2007; Bi et al., 2017a) (c).

### *3.6. Probabilistic ecological risks of heavy metals in Lake Fuxian in different periods*

The mRI values of the heavy metals in Lake Fuxian ranged from 0.70 to 2.78 in the last one hundred years. Probability estimation indicated that the mRI values of heavy metals were significantly higher in the period of 1980–2020 than those in the other two periods (Fig. 6a). The simulated results showed that a moderate ecological risk of heavy metals in sediments of Lake Fuxian might appear in the last 40 years, with the possibility of 90 %. However, the possibility of a moderate ecological caused by heavy metals in the sediments was merely 20 % in the periods of – 1950 and 1950–1980 (Fig. 6a). Moreover, in the period of 1980–2020, a considerable risk and a very high risk might happen in Lake Fuxian, with the possibility of 10 %, which was not observed in the earlier two periods (Fig. 6a). The sensitive analysis of Monte Carlo simulation revealed that Hg and Cd were the predominant factors affecting the ecological risks in Lake Fuxian in the periods of – 1950 and 1950–1980, with the sum contribution of 97 % and 98 %, respectively (Fig. 6b). However, in the recent 40 years, Cd became the most sensitive factor that contributed 96 % to the combined ecological risk (Fig. 6b). An obvious transfer of spatial distribution of ecological risks in Lake Fuxian was also observed among the three periods (Fig. 7). Specifically, higher ecological risks initially appeared in the south bank of Lake Fuxian before 1950 (Fig. 7a). Since 1950, sediments in the northern part of Lake Fuxian began to experience high ecological risks of heavy metals and eventually held the highest ecological risk in the last 40 years (Fig. 7b and c).

## 4. Discussion

### *4.1. Potential sources of heavy metals in sediments of Lake Fuxian*

According to Wang et al. (2021a), the average concentrations of Cd, Hg, As, Pb, Cr, Cu, Zn, and Ni were 0.23 mg/kg, 0.07 mg/kg, 11.8 mg/kg, 31.4 mg/kg, 78.8 mg/kg, 32.6 mg/kg, 82.5 mg/kg, and 35.4 mg/kg, respectively in 1797 sediment samples from Chinese lakes. Obviously, the concentrations of the aforementioned heavy metals in sediments of Lake Fuxian were much higher than the average values in China's lakes (Fig. 2). Hence, understanding the potential sources of heavy metals is essential for protecting the ecosystems in plateau lakes

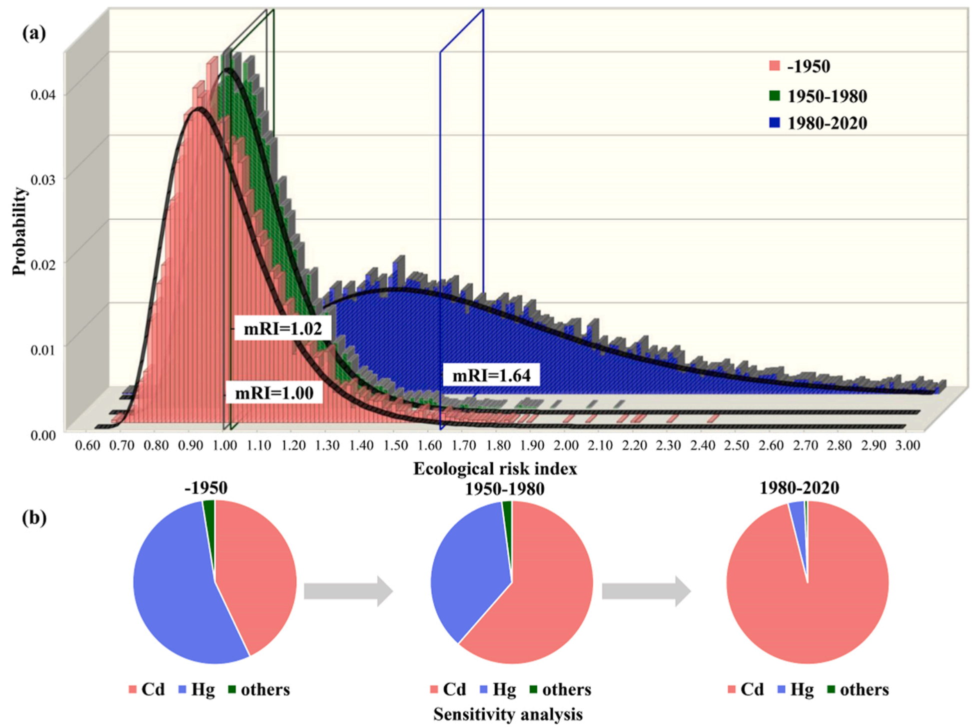



**Fig. 6.** Probabilistic ecological risk distribution of heavy metals (a) and sensitivity of Cd, Hg, and other heavy metals in risk assessment (b) in sediments of Lake Fuxian during – 1950, 1950–1980, and 1980–2020.

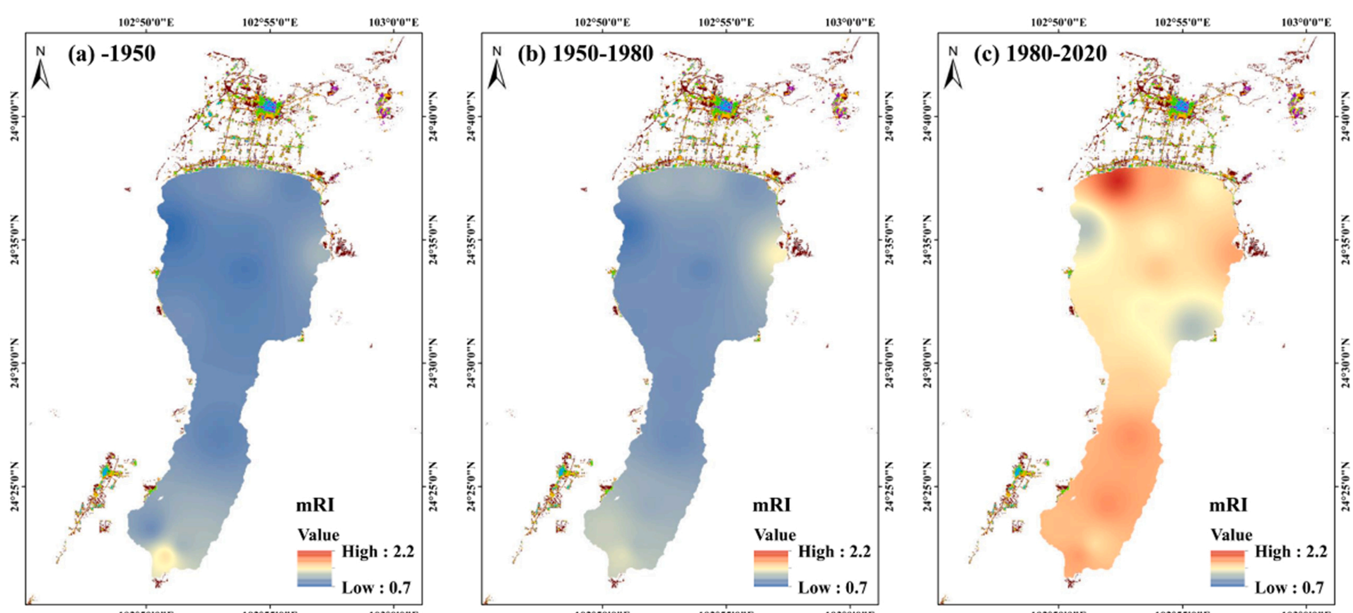



**Fig. 7.** Spatial distribution of ecological risk index of heavy metals in sediments of Lake Fuxian during – 1950 (a), 1950–1980 (b), and 1980–2020 (c).

(Guo et al., 2018). Based on the high-resolution sedimentary records in Lake Fuxian, the 11 heavy metals were divided into three groups. The first group consisted of 4 siderophile transition metals (i.e., V, Cr, Co, and Ni) and Cu, the second group contained As and Hg, and the third group comprised Cd, Pb, Sb, and Zn. On one hand, these three groups were characterized by different, even contrast vertical distribution patterns (Fig. 2). On the other, the correlations between heavy metals in the third group and those in other groups changed in reverse during the last 100 years (Fig. 3).

Previous studies have shown that the heavy metals belonging to the first group in river or lake sediments mainly originated from lithogenic sources, such as mineral weathering and atmospheric precipitation (Ustaoğlu and Islam, 2020; Dang et al., 2021). On the Yunnan-Guizhou Plateau, the weathering and erosion of parent rocks is common and considered as an important natural source of the siderophile transition metals (e.g., Fe, Ni, and Cr) in sediments of lakes including Qilu Lake and Erhai Lake (Yang et al., 2020; Liu et al., 2022). The second and third groups were characterized by chalcophile metals, which were often deemed to mainly come from various anthropogenic sources (Rinklebe et al., 2019). Due to the developed non-ferrous metal production industry in Yunnan Province, smelting and mining activities might be the major anthropogenic sources of these chalcophile metals in Lake Fuxian and other plateau lakes in this region, such as Lake Hongfeng (Zhao et al., 2011), Lake Qingshui (Liu et al., 2013), Lake Lugu (Lin et al., 2018), and Lake Erhai (Liu et al., 2022). Apart from mining and smelting, coal burning was considered as one major source of Pb in sediments after removing the leaded gasoline since 1998 in China (McComb et al., 2015; Bi et al., 2017a). In the present study, evidence from the stable Pb isotopes indicated that mining the galena in Yunnan rather than the coal combustion mainly contributed to the Pb accumulation in sediments of Lake Fuxian, especially in the last 40 years (Fig. 5). Moreover, the leaded gasoline seemed had little contribution to the Pb concentrations in the last hundred years, which was in accordance with a previous investigation in the lake (Liu et al., 2013). In fact, due to the high background concentrations in the top soils and parent rocks on the Yunnan-Guizhou Plateau, the concentrations of the chalcophile metals in Lake Fuxian also experienced different degrees of lithogenic influences (Fig. 5) (Wang et al., 2021a). Previous investigations showed that lake sediments and surrounding soils and minerals were characterized by high concentrations of heavy metals on the Yunnan-Guizhou Plateau (Wu et al., 1991; Wang et al., 2021a). Furthermore, the frequent weathering and soil erosion processes also accelerated the accumulation of heavy metals in lake sediments, which accounting for the lithogenic influences on concentrations of heavy metals in sediments of Lake Fuxian.

### 4.2. Increasing anthropogenic influences on heavy metal concentrations in plateau lakes

As Smol (2019) concluded, various anthropogenic activities have caused significant responses for global aquatic ecosystems, especially in the last 40 years. Many studies have reported the increasing trends of heavy metals (especially chalcophile metals) in sediment cores of lakes that are characterized by highly urbanization or intensive human activities, such as Lake Taihu of China (Li et al., 2018), Lake Geneva of Switzerland (Thevenon et al., 2011), and Lake Illawarra of Australia (Schneider et al., 2015). The plateau lakes in China used to be rarely disturbed by anthropogenic activities, and most of them are essential ecosystems for regional development and human needs in the plateau regions (Liu et al., 2022). Since the 1980s, increasing trends of heavy metals (e.g., As, Cd, Hg, Pb, and Zn) in sediments have been observed in lakes of the Yunnan-Guizhou Plateau (Yang et al., 2020; Liu et al., 2022). In Lake Fuxian, the change of correlations between lithogenic and anthropogenic heavy metals appeared since the 1950s and became apparent in the period of 1980–2020 (Fig. 3). This transformation attributed to the accumulation of these anthropogenic heavy metals (i.e., Cd, Sb, Pb, and Zn) in sediments of the lake due to the increasing impacts of human activities, especially the mining and smelting activities in Yunnan Province (Fig. 5b). As reported, industrial and domestic wastewater usually contain toxic heavy metals, such as Cd, Pb, and Zn (Ranjbar Jafarabadi et al., 2021; Gu et al., 2022a). Since the 1990s, the rapid development of population and industries around Lake Fuxian has accelerated the heavy metal enrichment in the lake and caused higher concentrations in the northern part (Fig. 7).

In sediments, organic matter can greatly influence the adsorption of heavy metals (e.g., Cd, Pb, and Zn) (Gao and Chen, 2012). In the present study, contents of TOC had similar increasing trends with those of Cd, Pb, Sb, and Zn in sediment cores of Lake Fuxian and the further analysis revealed significant correlations among them ($p < 0.05$, Table S4). Due to the increasing urbanization and industrialization, a mass of organic matter has been discharged into aquatic ecosystems worldwide and resulted in increasing concentrations of them in sediment cores (Gascón Díez et al., 2017; Gu et al., 2022a). Since the 1980s, the anthropogenic nutrient inputs, such as sewage and fertilizers, has increased the abundance of dinoflagellate and green algae in Lake Fuxian (Zhang et al., 2015). The anoxic conditions and the rise of organic matters in the sedimentary environments can further limit the migration of heavy metals and trap them in sediments (Ren et al., 2019), which likely accelerated the heavy metal accumulation in Lake Fuxian. Moreover, silt and clay were dominant compositions of sediments in Lake Fuxian, which also contributed to absorbing heavy metals (e.g., As, Cd, Pb, Sb, and Zn) due to their larger specific area and cation exchange ability in comparison with sand (Dang et al., 2021). Previous studies showed that the land use changes could induce severe soil erosion in the catchments (Balascio et al., 2019; Liu et al., 2022). With the frequent human settlement changes around Lake Fuxian (Fig. 1), a significant rise of silt proportion was observed in the last 40 years (Table 1). More importantly, the contents of silt were significantly negative with the lithogenic heavy metals in sediments of Lake Fuxian (Table S4), indicating that the terrestrial material inputs might be main reason for the smaller-size sediment particles in the lake. Hence, the indirect effects from anthropogenic activities (i.e., nutrient input and soil erosion) on heavy metal accumulation in sediments should also be noticed for the protection of plateau lakes.

### 4.3. Implication for ecological risk control of heavy metals in plateau lakes of China

Due to the high background concentrations of heavy metals in lake sediments of the Yunnan-Guizhou Plateua, the concentrations of As, Cr, and Ni in sediments of Lake Fuxian exceeded their probable effect levels (PELs), indicating that harmful effects of the three heavy metals on aquatic organisms might frequently occur (Table S5) (MacDonald et al., 2000). The harmful effects of other heavy metals (e.g., Cd, Cu, Hg, Pb, and Zn) might happen in Lake Fuxian because their concentrations were higher than the threshold effect levels (TELs) or lowest effect levels (LELs), but lower than the PELs (Table S5) (MacDonald et al., 2000). To eliminate the interference of high background concentrations for risk assessment, the average concentrations of heavy metals in sediments of Lake Fuxian before the 1950s were used as the baselines to calculate the ecological risk indexes to avoid the overestimation. The temporal and spatial distribution characteristics of the mRI values indicated that the anthropogenic activities were the primary cause of increasing ecological risks in Lake Fuxian (Figs. 6 and 7). Recently, high ecological risks caused by heavy metals have been reported in sediments of several plateau lakes, such as Qilu Lake in the Yunnan-Guizhou Plateua and Lake Bierruoze Co on the Tibetan Plateau (Guo et al., 2018; Yang et al., 2020). Moreover, the high ecological risks caused by Cd and Hg should attract attention because the excessive concentrations of Cd and Hg in the drinking water and aquatic products might cause environmental disasters (e.g., *itai-itai* and Minamata diseases) (Bi et al., 2017b).

The United Nations has set 17 Sustainable Development Goals (SDGs) to be reached by 2030, among which SDG 6 (clean water and sanitation) and SDG 14 (life below water) rely on a healthy aquatic environment (Gusmão Caiado et al., 2018). The present study first quantified the contributions of lithogenic and anthropogenic influences to the heavy metal concentrations in sediments of Lake Fuxian during different historical periods, which was the first step in developing an ecologically realistic mitigation strategy for plateau lakes (Smol, 2019). To lower the ecological risks of highly toxic metals (e.g., Hg, Cd, As, Sb, and Pb) in Lake Fuxian, the external inputs of heavy metals should be strictly limited. Proper remedial options need to be applied in the mine tailing disposal sites to reduce the anthropogenic influences on the increasing concentrations of heavy metals in the lake sediments. Firstly, the mining technology should be improved and the reuse of waste rocks need to be strengthened (Johnson et al., 2016). Secondly, it is essential to introduce the sustainable remedial strategies, such as the phytoremediation and biochar remediation, to reduce the residuals of heavy metal in contaminated soils (Karaca et al., 2018; Gao et al., 2022). Thirdly, the heavy metal accumulation in sediments of the plateau lakes caused by soil erosion cannot be overlooked (Liu et al., 2019), which demonstrating the need for efficient management of soil erosion and heavy metal discharge to protect fragile aquatic ecosystems.

## 5. Conclusions

The study of Lake Fuxian on the Yunnan-Guizhou plateau, China, showed that the lake sediments were characterized by increasing concentrations of Cd, Pb, Sb, and Zn in the last 100 years. Inversely, the concentrations of Co, Cr, Cu, Ni, and V generally decreased in the lake sediments while the concentrations of As and Hg showed little fluctuations. There was an obvious fractionation of heavy metals in different historical periods (i.e., −1950, 1950–1980, and 1980–2020), which was attributed to the combined influence of lithogenic and anthropogenic sources. Based on the APCS-MLR results, the lithogenic sources (e.g., mineral weathering, soil erosion, and atmospheric precipitation) mainly affected the concentrations of As, Hg, V, Cr, Co, Ni, and Cu in the last 40 years, with the rates of 55 %− 85 % and the anthropogenic sources (e.g., mining activities and human settlement change) accelerated the concentrations of Cd, Sb, Pb, and Zn in Lake Fuxian. The change of stable Pb isotope in the sediment cores among different periods further indicated that the smelting and mining of galena was the main source of Pb in Lake Fuxian. It was estimated that a moderate ecological risk of the 11 heavy metals might appear in Lake Fuxian in the last 40 years, with the possibility of 90 %. Moreover, the risk of Cd should attract attention considering that it contributed 96 % to the combined ecological risk. The study first quantified the source contributions to heavy metals and subsequent ecological risks in Lake Fuxian during different historical periods, which was of great significance in developing an ecologically realistic mitigation strategy for protecting plateau lakes.

## CRediT authorship contribution statement

**Xiang Gu**: Methodology, Writing – original draft. **Xiaotong Han:** Methodology, Conceptualization, **Yixuan Han:** Methodology, Conceptualization, **Wenlei Luo**: Conceptualization, Validation, **Muhua Feng**: Conceptualization, Validation, **Di Xu**: Conceptualization, Validation, **Peng Xing:** Funding acquisition, Writing – Review & Editing, **Qinglong L.** Wu: Conceptualization, Project administration.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

## Acknowledgments

We would like to thank Mr. Shu Tang for his help in the field sampling. We appreciated the contribution of Mrs. Yanjie Cai in Institutional Center for Shared Technologies and Facilities of NIGLAS, CAS for doing the sediment pre-treatment. This work was supported by National Natural Science Foundation of China (No. U2102216, 31722008), the Second Tibetan Plateau Scientific Expedition and Research (STEP) program (Grant No. 2019QZKK0503), and the Youth Innovation Promotion Association of CAS (No. 2014273).

## Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2022.129860.

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