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A Historical Sedimentary Record of Mercury in a Shallow Eutrophic Lake: Impacts of Human Activities and Climate Change



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ABSTRACT

Mercury and its derivatives are hazardous environmental pollutants and could affect the aquatic ecosystems and human health by biomagnification. Lake sediments can provide important historical information regarding changes in pollution levels and thus trace anthropogenic or natural influences. This research investigates the 100-year history of mercury (Hg) deposition in sediments from Chao Lake, a shallow eutrophic lake in China. The results indicate that the Hg deposition history can be separated into three stages (pre-1960s, 1960s–1980s, and post-1980s) over the last 100 years. Before the 1960s, Hg concentrations in the sediment cores varied little and had no spatial difference. Since the 1960s, the concentration of Hg began to increase gradually, and showed a higher concentration of contamination in the western half of the lake region than in the eastern half of the lake region due to all kinds of centralized human-input sources. The influences of anthropogenic factors and hydrological change are revealed by analyzing correlations between Hg and heavy metals (Fe, Co, Cr, Cu, Mn, Pb, and Zn), stable carbon and nitrogen isotopes ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$), nutrients, particle sizes, and meteorological factors. The results show that Hg pollution intensified after the 1960s, mainly due to hydrological change, rapid regional development and urbanization, and the proliferation of anthropogenic Hg sources. Furthermore, the temperature, wind speed, and evaporation are found to interactively influence the environmental behaviors and environmental fate of Hg.

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1. Introduction

Mercury (Hg) and its derivatives, including in particular the most toxic form of mercury, methylmercury (CH_3Hg^+), are hazardous environmental pollutants that can exist in the atmosphere and in aquatic food chains for extended periods of time, primarily because of their persistence and tendency toward bioaccumulation [1]. Mercury is discharged to the environment from natural sources and from human activities, such as ore refining and cement production and reemission [2]. With global industrial and economic development, increasing amounts of Hg have been discharged into the environment. Mercury has been identified in lakes, wetlands, reservoirs, rivers, and estuaries worldwide [3,4], and Hg pollution has become recognized as a global environmental problem. The United Nations Environment Program (UNEP) recently adopted the “Minamata Convention,” which is aimed at controlling and

decreasing global Hg emissions in order to reduce the damage caused by Hg to the environment and human health.

Direct long-term water quality monitoring was not initiated in most of the lakes in China until the 1980s, due to the limitations of monitoring technology and cost constraints [5]. In the absence of long-term water quality monitoring data, sediment cores can serve as an important archive, as they record metal fluxes in aquatic systems and can be an effective tool for reconstructing pollutant histories and environmental fates [6]. Mercury concentration in the sediment cores of lakes has increased significantly over the last century [7]. Mercury contamination of lake sediments has been investigated in Lake Ballinger and Lake Whittington, in the United States [4,8]. Recently, Lin et al. [9] investigated the influences of climate change on Hg accumulation in a remote and shrinking lake located on the southern Tibetan Plateau in China. Most of the existing research has been focused on the use of Hg isotopes in lake sediments to reliably trace historical, anthropogenic Hg contamination or to evaluate Hg cycling [10,11].

Chao Lake, a semi-enclosed shallow eutrophic lake in China, has been impacted by anthropogenic activities over the past three

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decades, and has been a target of wide societal concern since the 1980s, due to anthropogenic eutrophication [12]. A considerable amount of research has been performed on Chao Lake in order to resolve its historical records of multiple heavy metals [13], nutrients [12], polychlorinated biphenyls [14], polycyclic aromatic hydrocarbons, and organochlorine pesticides [15], but few studies have focused on Hg. Research on Chao Lake can also help in understanding the Hg profiles of the same type of lakes in the Yangtze River Basin. To address this research gap, the objectives of this study are as follows: to examine variability in the Hg supply to the lake and in the Hg distribution within the lake over approximately the last 100 years using sediment core chronology; and to resolve important anthropogenic factors by applying statistical analysis using indicators including heavy metals (iron (Fe), cobalt (Co), chromium (Cr), copper (Cu), manganese (Mn), lead (Pb), zinc (Zn), and Hg), nutrients (total nitrogen (TN), total phosphorus (TP), total organic carbon (TOC), $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$), and grain size. This study also examines this lacustrine environment in the context of climate variability by exploring correlations between Hg concentrations and temperature, precipitation, wind speed, and evaporation in the Chao Lake basin.

2. Materials and methods

2.1. Sampling and analysis

Chao Lake (117°16'–117°51' N, 31°25'–31°43' E) is a shallow natural lake in China with a surface area of 780 km² (Fig. 1). Intensified human activities have caused eutrophication and metal pollution in Chao Lake [12,13]. Since the construction of the dam in 1963, the water level of the lake has been controlled, and the natural hydrological cycles have been obstructed after its impoundment [12]. The two sample sites (CL1 and CL2) were selected due to differing levels of pollution in different regions. Moreover, the sample sites had to meet the requirements of likely long-term sediment accumulation and minimal effects of disturbances, including bottom-scouring or re-suspension.

The sediment cores were collected in 2009 using a columnar sampler (50 cm long and 8 cm in diameter). Both sediment cores were 30 cm in length, and were sectioned at 1 cm intervals. Each sample was frozen to below $-20\text{ }^{\circ}\text{C}$. The sample pretreatment procedure was as follows: The samples were ① freeze-dried,

② homogenized with an agate mortar and pestle, and ③ sieved using a 0.125 mm screen. The samples were then digested using an acid mixture of hydrochloric acid (HCl) and nitric acid (HNO₃) (3:1, v/v) [16]. TP was determined after heating the samples for 2 h at a temperature of 500 °C and performing 1 mol·L⁻¹ HCl extraction. TN was analyzed using an elemental analyzer (Vario EL III, Elementar Co., Germany). TOC was measured using a TOC analyzer (Multi N/C® 2100, Analytik Jena AG, Germany), after treatment with 1 mol·L⁻¹ HCl to remove inorganic carbon. The analytical precision was less than 3% for TN and 5% for TOC, using six replicates of a homogenized sample.

Heavy metals were detected by inductively coupled plasma mass spectrometry (iCAP Qc, Thermo Fisher Scientific, USA) after microwave digestion in a mixture of HNO₃, HCl, and hydrofluoric acid (HF) (1:3:6, v/v). The standard reference sediment SUD-1 (National Water Research Institute, Environment Canada, Canada) was used to estimate the accuracy of the heavy metal analyses. The recoveries (analytical value/certified value ratio, %) of Cr, Cu, Pb, Mn, Zn, and Co were between 85% and 110%. The total Hg in samples and blanks was measured by means of cold vapor atomic fluorescence spectrometry, using a Hg detector (F732-V, Shanghai Huaguang Instrument, China) [9,17]. The average recovery of Hg was 98.3%. Therefore, the certified values and analytical values were in good agreement.

2.2. Data processing

The background concentration of Hg in the Chao Lake sediments was determined using the paleoecological investigation of recent lake acidification (PIRLA) method [18]. The enrichment factor (EF) [5] is widely used to assess heavy metal pollution in sediments, as it aids in discriminating between anthropogenic and natural sources of heavy metals. The degree of contamination of Hg in the sediments of Chao Lake was evaluated using the EF. Moreover, the geoaccumulation index (I_{geo}) was used to evaluate heavy metals pollution caused by human activities [19].

The Hg flux was calculated using the average sediment accumulation rate. Zan et al. [13] calculated the average sediment accumulation rate at these locations in Chao Lake using unsupported or “excess” ²¹⁰Pb (²¹⁰Pb_{ex}) and the constant flux sedimentation (CFS) model [20]. The average sediment accumulation rate of core CL1 was found to be 0.224 g·cm⁻²·a⁻¹, and the average sediment

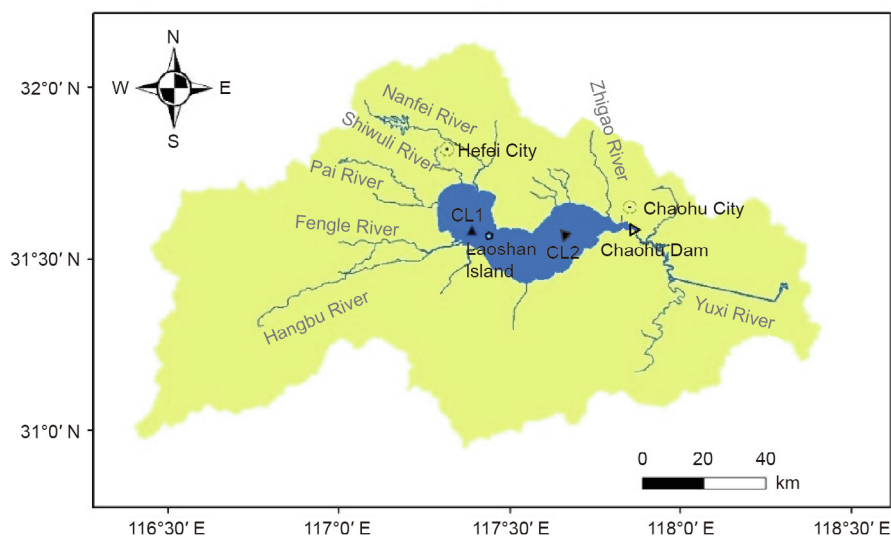


Fig. 1. Location of sampling sites.

accumulation rate of core LC2 was found to be $0.242 \text{ g}\cdot\text{cm}^{-2}\cdot\text{a}^{-1}$ [13].

Meteorology data came from the CN05.1 grid observation dataset, which was constructed by the anomaly approach during interpolation with the horizontal resolution of $0.25^\circ \times 0.25^\circ$ [21]. The data normality and Pearson correlations were tested using the SPSS 18.0 software (SPSS Statistics, IBM Corporation, USA). The normality distribution of data was determined by the Kolmogorov–Smirnov test. The data on the Hg and heavy metals concentration, nutrients, and grain size aligned with the normal distribution at the 0.05 significance level.

3. Results and discussion

3.1. Vertical profiles of Hg

The concentrations of total Hg deposited in the two sediment cores indicated a consistent change with a concentration increasing from the bottom layers to the surface or subsurface layers and reaching peak values (Fig. 2(a)). The Hg concentrations accumulated in cores CL1 and CL2 were divided into three different periods: a relatively stable stage before the 1960s, a slow increase from the 1960s to the 1980s, and a sharp increase from the 1980s to the present.

Before the 1960s, the concentrations of total Hg ranged from 52.4 to $69.9 \text{ ng}\cdot\text{g}^{-1}$ and from 43.5 to $68.8 \text{ ng}\cdot\text{g}^{-1}$ dry sediment for CL1 and CL2, respectively. Based on the PIRLA procedure, the Hg background values between two sample sites showed little difference; the Hg background values for the lake sediments are shown by the mean value of the two sample sites. The Hg background

concentration in the Chao Lake sediments is $(52.29 \pm 5.15) \text{ ng}\cdot\text{g}^{-1}$. The results indicate that the Hg concentrations in the lake sediment cores during the first stage were in the background period (pre-1960s). Chemical fertilizers were not used in the traditional agriculture of the whole watershed [12]. No significant increasing trends were found in the two sediment cores for Hg concentration before the 1950s; the only slight increase observed in the 1930s may have been induced by the First Chinese Civil Revolutionary War (1924–1927) or the Second Sino-Japanese War (1937–1945) [22,23]. The almost constant concentrations of Hg existing in the two sediment cores show the succession of a natural lake system with few human-input impacts before the 1960s, which is consistent with the nutrient, heavy metal, and persistent organic compounds concentrations in the sediment cores [12–16].

During the second stage, the Hg concentrations rose slowly from 76.3 and $71.6 \text{ ng}\cdot\text{g}^{-1}$ in the 1960s to 95.4 and $80.9 \text{ ng}\cdot\text{g}^{-1}$ in the early 1980s for CL1 and CL2, respectively. Mercury input began to increase slightly in the 1960s, which was consistent with the first immigration settlement of China in the studied watershed [24]. During the third stage, the CL1 and CL2 sediment cores exhibited a sharp increase in Hg concentrations, from respective levels of 95.4 and $80.9 \text{ ng}\cdot\text{g}^{-1}$ in the early 1980s to the present levels of 169.0 and $102.0 \text{ ng}\cdot\text{g}^{-1}$. These increasing trends in Hg concentration may be attributed to the Hg emissions and discharge results from coal combustion and industrial effluent during the industrialization of China [25]. With China's reform and opening-up policy initiated in 1978, the rapid economic development in China resulted in a huge consumption of energy [23]. The growing use of fossil fuels—especially coal and oil as dominant sources of energy—led to high levels of Hg emission [26].

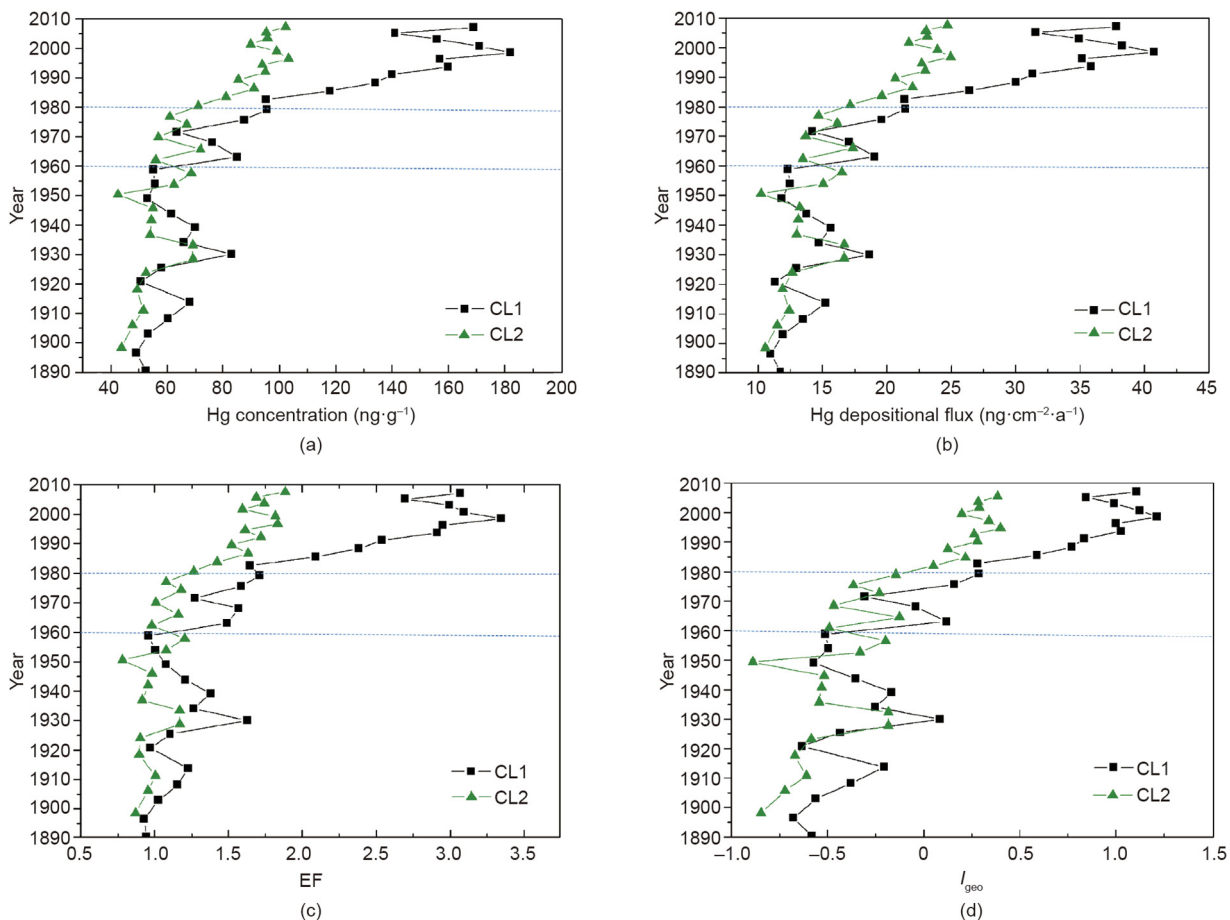


Fig. 2. Vertical profiles of (a) Hg concentration in the dry sediment, (b) Hg deposition flux, (c) EF, (d) I_{geo} in the two cores.

The Hg content in the CL1 core is much higher at different historical stages than in the CL2 core. Most of the population and industry in the catchment is distributed in the city of Hefei, the capital of Anhui Province; this has resulted in a heavier Hg input in the western lake region (core CL1), which is influenced by Hefei, than in the eastern lake region (core CL2), which is surrounded by the smaller city of Chaohu. As such, human sources of Hg including agricultural runoff, urban sources, and industrial sources have had a greater impact at the CL1 sample site than at the CL2 site. A number of studies have shown that the contamination trends recorded in sediment cores are often significantly influenced by the degree of local/regional urbanization [27].

3.2. Deposition fluxes and mass inventories of Hg

Hg emissions in the lake catchment, as well as the sedimentary rates and sedimentological conditions of the lake, influence Hg accumulation in sediment cores. Hence, Hg depositional fluxes are more appropriate than Hg concentration to reflect the accumulation of Hg in sediments [28]. The Hg depositional fluxes, *F*, were calculated using the following equation:

$$F = C_i \rho_i \gamma_i \tag{1}$$

where *C_i* is the Hg concentration of slice *i* (ng·g⁻¹), *ρ_i* is the mass density of the dry sediment for slice *i* (g·m⁻³), and *γ_i* is the sedimentation rate of slice *i* (cm·a⁻¹). The depositional fluxes of each core (Fig. 2(b)) are somewhat similar to the Hg content profiles for the different stages in the western and eastern regions. High emission intensity resulted in the higher depositional fluxes observed in the western lake core [25,26]. Sedimentary flux records were helpful in calculating the gradient rate of sediment flux change in Chao Lake. In the western lake region (CL1), the slow accumulation of low Hg flux (Fig. 2(b)) was displayed at the beginning of the founding of China. Before the 1960s, a low zero-order rate of increase in Hg flux, *k*, was found, with rates of increase of 0.0348 and 0.0520 a⁻¹ for CL1 and CL2, respectively (Table 1). The Hg depositional flux then increased between the 1960s and the 1980s, with rising rates of 0.3842 and 0.1358 a⁻¹ for CL1 and CL2, respectively. The Hg depositional flux sharply increased after the 1980s, when China began its rapid industrialization process. The sedimentary flux in this period increased 2–3 times more than before the 1960s (Table 1).

The mass inventories of the Hg in the sediment cores were calculated according to the three stages (Table 1). Compared with those in the western region, the mass inventories in the eastern region were relatively low, indicating heavier pollution in the western region due to the imbalance of economic and industrial development in the catchment. Furthermore, the mass inventories in CL1 and CL2 after the 1960s contributed 69%–74% of the total Hg inventories in the past century, which indicates that the Hg pollution can be mainly attributed to the recent increase in input related to anthropological activities in Chao Lake.

3.3. Intensity of Hg pollution

In these two sediment cores, the Hg EF was lower in the deeper sediments (before the 1980s, EF < 1.5), followed by an increase upwards into the shallower sediments (after the 1980s, EF > 1.5); the highest values were exhibited at the near surface (Fig. 2(c)). Based on the work of Haris and Aris [29], an EF value between 0.5 and 1.5 indicates that the metal may be entirely derived from the weathering and transport of crustal materials, while an EF value above 1.5 suggests that some fraction of the metal is derived from anthropogenic sources. Therefore, Chao Lake had a substantial increase in human Hg input after the 1980s, when the reform and opening-up policy was implemented in China.

Before the 1980s, the *I_{geo}* in these two cores was below 0 (pollution free), except in 1970 (15 cm) and in 1936 (23 cm) in core LC1 (Fig. 2(d)). After the 1980s, the *I_{geo}* of core CL2 was between 0 and 0.5, which indicates some level of Hg pollution. The *I_{geo}* of core CL1 increased rapidly since the 1980s and reached above 1 in 1990, reflecting moderate levels of pollution. The results of the geoaccumulation index indicate that although the Hg contamination of Chao Lake is slight, the trend of pollution is significantly increasing.

Anthropogenic sources of Hg in China can be divided into unintentional emissions of Hg by fuel combustion, industrial processes that utilize Hg (i.e., the calcium carbide method–PVC process), Hg-containing products (e.g., medical products, fluorescent lighting, and batteries), and Hg mining and smelting. However, unintentional emission sources of Hg are the main source of atmospheric Hg, and these emissions are produced by three main industries. The coal industry is a major source of atmospheric Hg emissions, due to the large production volumes and the high Hg content and low halogen characteristics of the coal [25,26]. Another source of atmospheric Hg emissions is the process of non-ferrous metal smelting, which includes the smelting of Zn, Pb, and Cu, since these ores often contain Hg [30]. And finally, the cement industry is a major source of atmospheric Hg emissions. China is the largest cement producer in the world (> 80% of global cement production), and Hg is associated with both the limestone raw materials, and the coal that is used for fuel [31]. Based on the Anhui Statistical Yearbook 2016 [32], the coal mining, non-ferrous metal smelting, and cement-production industries have continued to develop over the past ten years. The coal-fired energy-production industry is the main source of Hg emissions in Asia [31,33].

Although human activities (and especially fossil fuel combustion) have caused massive atmospheric emissions of Hg, atmospheric deposition is probably not the main source of Hg in Chao Lake sediments. The highly positive correlations between multiple metals and Hg (Figs. 3 and 4) indicate that they have the same or similar sources from anthropogenic activities. Industries are closely related to water pollution in the lake; for example, the production and supply of electric power and thermal power, non-ferrous metal smelting, and rolling processing industries will produce wastewater and aerosols with different concentrations

Table 1
Fluxes and inventories of Hg deposition in the dated sediment cores at different stages.

Core site	Time period	Linear regression				Fluxes (ng·cm ⁻² ·a ⁻¹)	Inventories (ng·cm ⁻²)
		<i>k</i> (a ⁻¹)	<i>R</i> ²	<i>p</i>	<i>n</i>		
LC1 in western lake region	Before 1960s	0.0348	0.0895	0.321	13	11.0–18.6 (13.6)	218.0
	1960s–1980s	0.3842	0.9715	0.020	5	13.4–19.6 (16.3)	130.6
	1980s–present	0.6759	0.8520	0	12	21.4–38.3 (31.8)	477.0
LC2 in eastern lake region	Before 1960s	0.0520	0.7589	0	13	10.5–15.0 (12.7)	203.2
	1960s–1980s	0.1358	0.5273	0.165	5	13.5–16.1 (14.5)	115.7
	1980s–present	0.2137	0.7547	0	13	17.1–24.7 (21.7)	325.7

“()”: mean value of deposition fluxes.

	Fe	Co	Cr	Cu	Mn	Pb	Zn	Hg
Fe	1							
Co	0.745 ^a	1						
Cr	0.806 ^a	0.937 ^a	1					
Cu	0.820 ^a	0.864 ^a	0.917 ^a	1				
Mn	0.736 ^a	0.819 ^a	0.888 ^a	0.862 ^a	1			
Pb	0.795 ^a	0.739 ^a	0.876 ^a	0.877 ^a	0.829 ^a	1		
Zn	0.818 ^a	0.753 ^a	0.841 ^a	0.912 ^a	0.817 ^a	0.851 ^a	1	
Hg	0.749 ^a	0.636 ^a	0.747 ^a	0.831 ^a	0.736 ^a	0.773 ^a	0.926 ^a	1

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

Fig. 3. Pearson correlation matrix for Hg and other metals from cores LC1 and LC2.

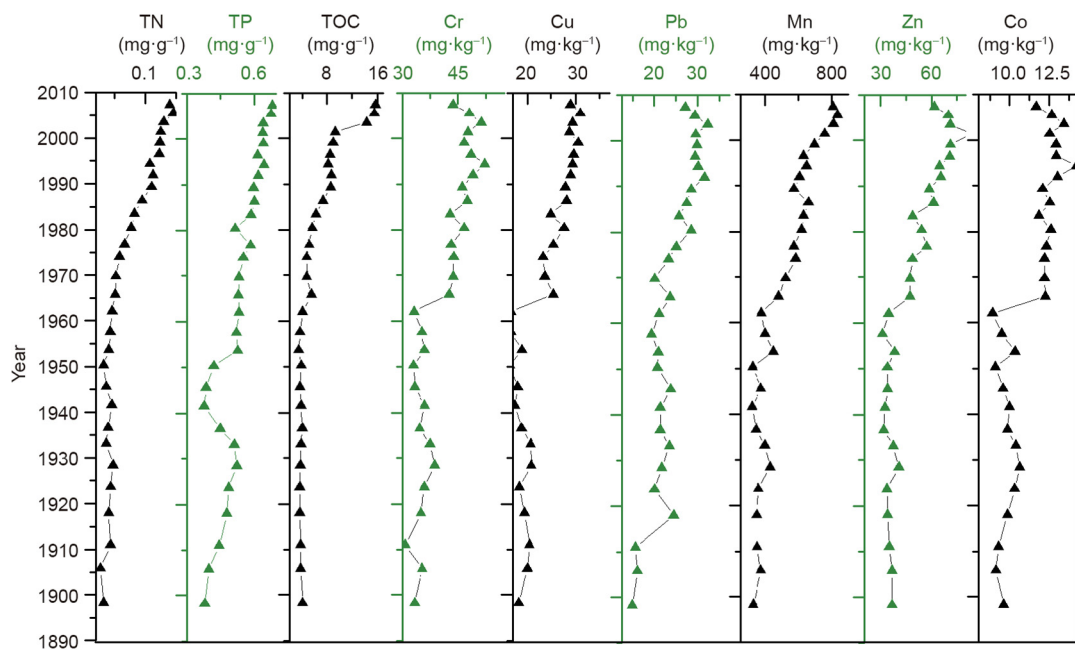
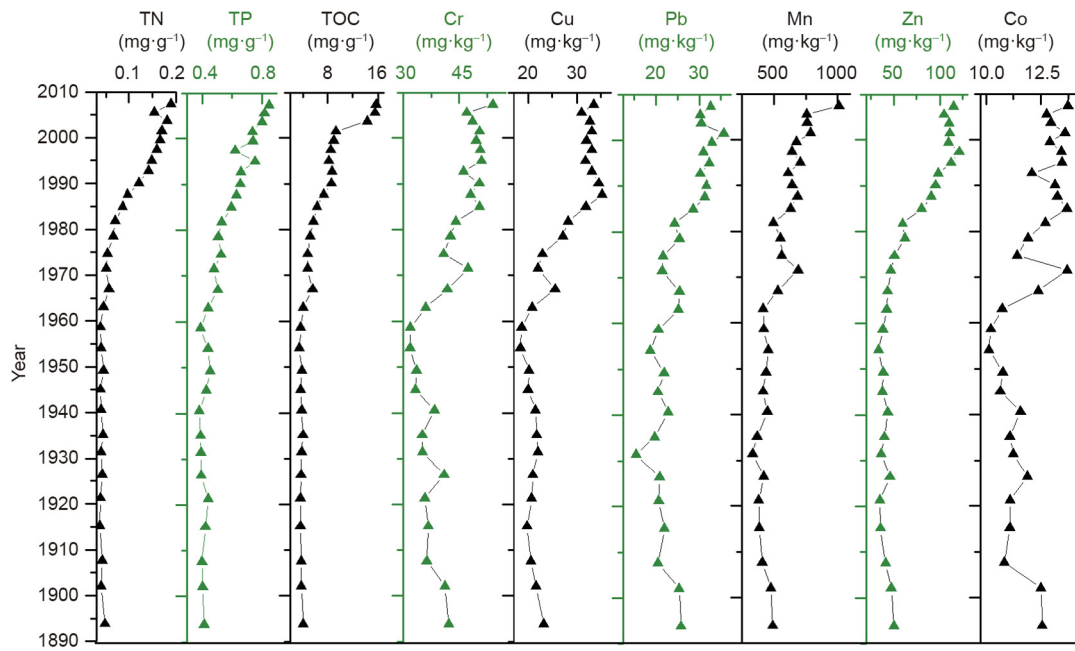


Fig. 4. Vertical profiles of nutrients and heavy metals in the (a) core CL1 and (b) core CL2.

of heavy metals in the production process [14]. Industrial wastewater discharge is the most direct and significant source of heavy metal pollution in the lake [15].

3.4. Relationships between Hg and other factors

A Pearson correlation analysis of the nutrients and Hg from the two cores showed them to be significantly correlated ($P < 0.01$). Strong positive correlations between TN, $\delta^{15}\text{N}$, TP, TOC, and Hg were observed in both cores (Fig. 5, $p = 0.000$ for all). However, $\delta^{13}\text{C}$ and carbon nitrogen ratio (C/N) had negative correlations with Hg. Since the 1970s, a large amount of industrial, agricultural, and domestic sewage has been discharged into Chao Lake, resulting in an increase in nutrient load [12]. For example, nitrogen (N) and phosphorus (P) fertilizer use in the Anhui Province intensified in the 1980s, with application rates of up to $750 \text{ kg}\cdot\text{hm}^{-2}$, and is still increasing [34]. Mercury-containing pesticides and fungicides have been widely used in agriculture for a long time, resulting in high concentrations of mercury in intensive farming soils [31]. In the Chao Lake catchment, paddy soil is the main soil type; this is a kind of soil with high fertility that is formed by human cultivation [35], which has a strong adsorption capacity for Hg [36]. Therefore, fertilizers (containing N and P) and soil organic matter adsorb the Hg discharged into the lake from surface runoff. Furthermore, Hg can easily be adsorbed by inorganic sediments in natural water [37]. Hydrosulfonyl, amine, carboxyl, and other functional groups contained in sediments can bind to Hg to form stable organic complexes [31,38].

Variables including the C/N ratio, $\delta^{15}\text{N}$, and $\delta^{13}\text{C}$ can be used as proxies to identify sources of sedimentary organic matter (SOM) [39], which can record changes in lacustrine primary productivity [40]. Increases in lacustrine primary productivity (including algal production) lead to increases in the deposition of SOM, along with changes in $\delta^{13}\text{C}$ signatures [41]. Organic matter has a dual effect on methylmercury [42], as it can promote Hg methylation by stimulating microbial activity [43,44], but also inhibit Hg methylation by combining with inorganic Hg to reduce methylation efficiency [45]. Remarkably, labile carbon sources have the greatest impact on Hg^0 production [38,46]. Mercury methylation and demethylation are dynamic processes that operate at the interfaces of aqueous and particulate phases, so the effects of organic matter (quantity and types) on Hg deposition are complex.

The contents and distributions of various elements found in sediments are often closely related to the sedimentological condition changes [47]. Grain size in sediment samples can provide a record of hydrological change, because hydrodynamic intensity determines the ability to transport different sizes of grains [14]. As expected, the correlation analysis (Fig. 6) showed that Hg was positively correlated with small particles (grain size $< 8 \mu\text{m}$), and negatively correlated with larger particles (grain size $> 8 \mu\text{m}$). The deposition and accumulation of fine-grained sediments in Chao Lake has been enhanced since the 1963 construction of the Chaohu Dam on the Yuxi River [12]. With the semi-closed water body subject to artificial regulation, its natural water-exchange capacity has been lost, and its sedimentary environment tends to be stable. Stable depositional conditions result in the accumulation of fine particles, SOM, and nutrients, and Hg was deposited along with the deposition of a large amount of fine grains.

3.5. Climate change and Hg deposition

An examination of climate factor statistics shows that the annual average air temperature and annual precipitation increased by $0.98 \text{ }^\circ\text{C}$ and 148.1 mm , respectively, from 1961 to 2017 in the Chao Lake catchment (Fig. 7(a,b)). The annual average wind speed and annual evaporation decreased by $1.128 \text{ m}\cdot\text{s}^{-1}$ and $267.9 \text{ mm}\cdot\text{a}^{-1}$, respectively, from 1961 to 2017 in the catchment (Fig. 7(c,d)). The average annual temperature in the Chao Lake catchment has increased (Fig. 7(a)), which is probably due to the increase in greenhouse gas emissions caused by the rapid development of industrialization and urbanization after 1980; this warming is known to be altering terrestrial and aquatic ecosystems, including lakes [44,48,49]. The influence of climate change on Hg deposition was analyzed by comparing it to meteorological factor variation in the catchment [49]. The direct emissions of Hg by human activities are regarded as “original emissions,” while the process of Hg evaporating from surficial media due to ambient temperature increases is considered as “secondary emissions” [50]. The correlation analysis showed that Hg was positively and moderately correlated with the annual average air temperature (Fig. 7). Obviously, climate warming will change the reemissions of Hg, which should lead to increased Hg emissions on the whole. Furthermore, numerous studies have documented rising water temperatures in aquatic environments around the world [51,52],

	TP	TN	TOC	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	C/N	Hg
TP	1						
TN	0.929 ^a	1					
TOC	0.775 ^a	0.861 ^a	1				
$\delta^{15}\text{N}$	0.924 ^a	0.927 ^a	0.845 ^a	1			
$\delta^{13}\text{C}$	-0.866 ^a	-0.898 ^a	-0.775 ^a	-0.914 ^a	1		
C/N	-0.632 ^a	-0.648 ^a	-0.226 ^b	-0.647 ^a	0.667 ^a	1	
Hg	0.829 ^a	0.895 ^b	0.786 ^a	0.891 ^a	-0.734 ^a	-0.516 ^a	1

^aCorrelations are significant at $p < 0.01$ (two-tailed).

^bCorrelations are significant at $p < 0.05$ (two-tailed).

Fig. 5. Pearson correlation coefficients between nutrients and Hg in cores LC1 and LC2.

	$< 4 \mu\text{m}$	$4-8 \mu\text{m}$	$8-16 \mu\text{m}$	$16-64 \mu\text{m}$	Hg
$< 4 \mu\text{m}$	1				
$4-8 \mu\text{m}$	0.917 ^a	1			
$8-16 \mu\text{m}$	-0.578 ^b	-0.245	1		
$16-64 \mu\text{m}$	-0.912 ^a	-0.991 ^a	0.242	1	
Hg	0.838 ^a	0.758 ^a	-0.540 ^a	-0.787 ^a	1

^aCorrelations are significant at $p < 0.01$ (two-tailed).

^bCorrelations are significant at $p < 0.05$ (two-tailed).

Fig. 6. Pearson correlation coefficients between Hg and different frequency distributions of particle size from cores LC1 and LC2.

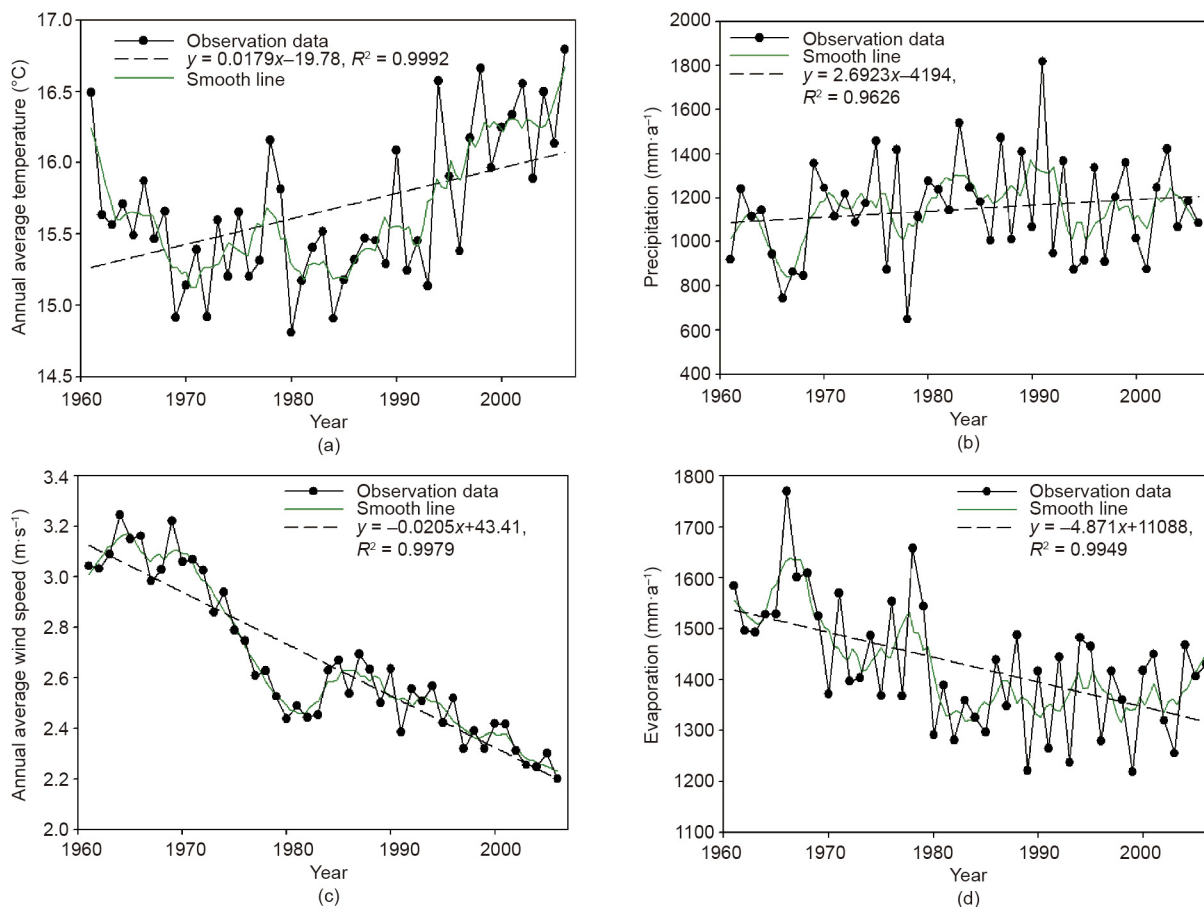


Fig. 7. Variation of meteorological factor in the Chao Lake catchment from 1961 to 2016.

	Hg	TP	TN	TOC	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	C/N
Temperature	0.524 ^a	0.516 ^a	0.86 ^a	0.927 ^a	-0.898 ^a	-0.648 ^a	0.895 ^b
Precipitation	0.081	0.113	0.099	0.845 ^a	-0.775 ^a	-0.226 ^b	0.786 ^a
Wind speed	-0.799 ^a	-0.826 ^a	-0.825 ^a	-0.628 ^a	-0.840 ^a	0.821 ^a	0.640 ^a
Evaporation	-0.472 ^a	-0.462 ^a	-0.825 ^a	-0.395 ^a	-0.477 ^a	-0.477 ^a	0.350 ^b

^a Correlations are significant at the 0.01 level (two-tailed).

^b Correlations are significant at the 0.05 level (two-tailed).

Fig. 8. Pearson correlation coefficients between Hg and meteorological factor and nutrients in cores LC1 and LC2.

which influence a very wide range of biological, chemical, and physical processes [53,54]. Therefore, the rise in air temperatures leads to an increase in water temperatures, which may have an effect on the transformation and deposition of Hg.

However, the relationship between precipitation and Hg deposition is not corrected (Fig. 8), mainly because this change in meteorological conditions has a combined effect on Hg transport. Indeed, the variation of precipitation intensity and frequency affects soil erosion intensity and the input of Hg pollutant sources [9]. Most anthropogenic emissions of Hg to the atmosphere are in the form of elemental mercury vapor (more than 95%), and the rest consists of divalent Hg compounds, or Hg adsorbed to particulate matter [55]. The residence time of elemental Hg vapor in the atmosphere is approximately several months to one year, which facilitates long-distance transport [56]. The correlation with Hg deposition is strongly negative for wind speed, and moderate for annual evaporation (Fig. 8). Change in wind speed affects the migration and deposition of Hg by regulating water circulation and water flow rate [24]. The decrease of the average wind speed

in the Chao Lake catchment has contributed to the relatively stable hydrodynamic conditions, which are in favor of Hg deposition. In addition, the volatile amount of Hg in the water body decreases as the annual evaporation decreases, resulting in more Hg deposition in the sediment [49]. Ongoing climate change can lead to changes in the amounts and distributions of precipitation, temperature, wind speeds and directions, and evaporation, which will likely impact Hg transport intensity and pathways [49].

4. Conclusions

This study reports that the natural background Hg concentration is $(52.29 \pm 5.15) \text{ ng}\cdot\text{g}^{-1}$ in the sediments of Chao Lake, and that there are three identifiable periods of Hg deposition: pre-1960s, 1960s–1980s, and post-1980s. There is a relatively stable stage before the 1960s, a slow increase from the 1960s to the 1980s, and a sharp increase from the 1980s to the present. The Hg deposition was highly correlated with variables including the concentrations of heavy metals and nutrients, and sediment grain size.

As the impacts of global warming are many and complex, the effects of increasing air temperature on Hg deposition are difficult to fully separate and quantify, and warrant additional investigation.

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Compliance with ethics guidelines

Hanxiao Zhang, Shouliang Huo, Kevin M. Yeager, Beidou Xi, Jingtian Zhang, and Fengchang Wu declare that they have no conflict of interest or financial conflicts to disclose.

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