Postprint: Monitoring and Analysis of Atmospheric Heavy Metal Deposition Characteristics in Caohai, Guizhou Using Moss

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Date: 2019-08-26T00:00:00+00:00

Abstract

Caohai in Guizhou is a typical plateau wetland ecosystem that plays an important role in regulating regional climate and maintaining ecological balance. Investigating the flux and pollution characteristics of atmospherically deposited heavy metals is of profound significance for the stability and sustainable development of the Caohai wetland. The authors utilized the sensitivity and tolerance of bryophytes to heavy metals, employing them as bio-monitoring indicator plants, and applied the moss bag method (MossBag) to monitor atmospheric heavy metal deposition in the Caohai plateau wetland system, measuring seven heavy metals (Cu, Pb, Zn, Cd, Cr, As, Hg) to provide baseline data for Caohai' s ecological protection. The contents of the seven elements were determined using ICP-MS and atomic fluorescence spectrometry, deposition fluxes were calculated, principal component analysis and correlation analysis were employed to interpret pollution characteristics and sources, and the geoaccumulation index method was used for evaluation and analysis. The results indicate significant differences in atmospheric deposition fluxes among elements in Caohai, with Cu and Zn deposition fluxes (21.43 g・m-2・d-1 and 102.82 g・m-2・d-1, respectively) being substantially higher than other elements and accounting for a large proportion of total deposition. Correlation analysis revealed positive relationships between Cu and Zn, As and Cr, and Cd and Pb, suggesting common sources, while correlations among other elements were not significant. The geoaccumulation index method classified all seven heavy metals as severely polluted, with Cd being particularly severe. Based on analysis of pollution characteristics and sources, atmospheric heavy metal pollution in Caohai is primarily influenced by multiple factors, including industrial structure, production and lifestyle patterns, and even legacy effects from previously closed indigenous zinc smelting.

Full Text

Preamble

DOI: 10.11931/guihaia.gxzw201901014
Title: Monitoring and Analysis of Heavy Metal Atmospheric Deposition in Caohai Lake, Guizhou Using Bryophytes

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Abstract

Caohai Lake in Guizhou represents a typical plateau wetland ecosystem that plays a crucial role in regional climate regulation and ecological balance maintenance. Investigating the flux and pollution characteristics of heavy metals in atmospheric deposition holds profound significance for the stability and sustainable development of the Caohai wetland ecosystem. This study employed the moss bag method (MossBag) to monitor atmospheric heavy metal pollution in the Caohai plateau wetland lake ecosystem, utilizing bryophytes as bioindicator plants due to their sensitivity and tolerance to heavy metals. Seven heavy metals (Cu, Pb, Zn, Cd, Cr, As, Hg) were measured to provide baseline data for Caohai’ s ecological environmental protection. Elemental contents were determined using ICP-MS and atomic fluorescence spectrometry, deposition fluxes were calculated, and pollution characteristics and sources were analyzed through principal component analysis and correlation analysis. The geoaccumulation index method was applied for data evaluation.

The results demonstrated significant variations in atmospheric deposition fluxes among different elements. Cu and Zn exhibited substantially higher fluxes than other elements, reaching 21.43 $\mu g \cdot m^{2} \cdot d^{1}$ and 102.82 $\mu g \cdot m^{2} \cdot d^{1}$, respectively, and accounting for a major proportion of total deposition. Correlation analysis revealed positive correlations between Cu and Zn, As and Cr, and Cd and Pb, indicating common sources for these element pairs, while correlations among other elements were not significant. Assessment using the geoaccumulation index method classified all seven heavy metals as severely polluted, with Cd showing particularly severe contamination. Based on the pollution characteristics and source analysis, heavy metal pollution in Caohai’ s atmospheric deposition results from multiple combined factors, including industrial structure, production and lifestyle patterns, and even legacy contamination from previously closed primitive zinc smelting operations.

Keywords: Caohai, wetland, heavy metals, deposition flux, pollution characteristics, MossBag

0 Introduction

Atmospheric heavy metal pollution has become a critical environmental issue threatening human health. Heavy metals are typical atmospheric pollutants of global concern. Anthropogenic activities release heavy metals into the atmosphere, causing significant impacts on ecological environments and human health in source areas. Through atmospheric transport and continuous deposition, these pollutants also negatively affect remote regions' soils and water bodies (Dietz et al., 2009; Lee et al., 2005). Once released into the environment, heavy metals are not easily degraded and persist for extended periods. When inhaled by humans in particulate form, they similarly resist degradation, and toxic effects manifest once accumulation reaches certain thresholds (Tang et al., 2012a). Current research on atmospheric heavy metal deposition primarily focuses on rivers, oceans, and urban areas. Studies by Pekey et al. (1995) on the Black Sea and Zhan et al. (2012a) on the Yangtze River estuary have demonstrated that atmospheric deposition plays an extremely significant role in transporting heavy metals to water bodies, making it a major source of environmental heavy metals. Therefore, understanding the concentration and characteristic distribution of heavy metals in ambient air is of great importance.

Bryophytes have long been widely used for monitoring heavy metal pollution due to their unique morphological structures and physiological characteristics (Mendil et al., 2009; Rivera et al., 2011; Uyar et al., 2009), particularly in studying atmospheric dry and wet deposition of heavy metals, pollution sources, transport, and spatiotemporal distribution (Cao et al., 2008; Barandovski et al., 2007; Dragović et al., 2008; Fernández et al., 2001; Ge et al., 2013; Norouzi et al., 2016). Bryophytes possess strong cation exchange capacity, enabling them to effectively absorb metal ions from their surroundings and exhibit strong adsorption and retention capabilities for heavy metals (Büscher et al., 1990). As perennial plants, they can serve as long-term bioaccumulative monitoring materials for regional or source-specific atmospheric pollution, enhancing the stability and reliability of monitoring results (Carballeira et al., 2006). Research indicates that heavy metals in bryophytes primarily originate from atmospheric deposition, including precipitation and airborne dust particles (Rühling et al., 1969), a finding confirmed by recent experimental studies (Rühling, 2000; Stobart et al., 1985).

Current research on the Caohai wetland has primarily focused on biodiversity of plankton and benthic organisms, nutrient elements such as organic matter, and heavy metals in surface sediments, with results generally attributing contamination to anthropogenic activities and mineral coal combustion (Zhang et al., 2018a). However, atmospheric deposition as a potential pathway for heavy metal transport has been overlooked. Regionally, Caohai in Guizhou—characterized by low latitude, high altitude, and karst basin geology—has small soil pollution capacity, high mobility, wide impact range, and difficult remediation, making it an irreplaceable natural research site. Its rich biodiversity also holds important domestic and international significance. Methodologically, no studies have examined atmospheric deposition in the Caohai wetland ecosystem. Therefore, this study employed the moss bag method (MossBag), a passive sampling technique, to investigate atmospheric deposition of heavy metals (Cu, Zn, Pb, Cr, Cd, As, Hg) in the Caohai wetland lake ecosystem. This approach offers several advantages: controllable exposure time, ability to reflect relative deposition rates and pollution levels, clear background concentrations without root absorption interference, and simplicity, economy, and flexible site selection suitable for year-round monitoring (An et al., 2006; Cesa et al., 2006).

Due to the lack of corresponding environmental indicators, current assessments of atmospheric heavy metal deposition pollution primarily borrow methods from sediment heavy metal pollution evaluation (Hu et al., 2011a). The geoaccumulation index method considers not only anthropogenic pollution factors and environmental geochemical background values but also natural diagenetic processes that may cause background value fluctuations, compensating for limitations of other evaluation methods (Jia et al., 2000). This study aims to provide a baseline reference for environmental quality assessment, regulation, and management of the Caohai wetland lake ecosystem.

1.1 Study Area Overview

The Caohai National Nature Reserve is located on the southwestern side of Weining County, Guizhou Province, and represents the largest natural high-altitude freshwater lake in the world' s renowned karst region. With an average elevation of approximately 2,171.7 m, mean annual precipitation of about 950.9 mm, mean annual sunshine duration of approximately 1,805.4 h, and mean annual temperature of about 10.5 °C, the lake' s water supply primarily comes from atmospheric precipitation. It constitutes a complete, typical plateau wetland ecosystem and serves as an important wintering habitat for China' s endemic plateau crane species—the nationally first-class protected Black-necked Crane—and other rare birds, earning it the reputation of "Pearl of the Plateau, Kingdom of Birds." As a critical region for biodiversity conservation action plans (Zhang et al., 2018b), Caohai holds significant importance in regulating regional climate, maintaining regional ecological balance, and scientific research, attracting widespread attention from researchers and establishing it as a research base for subtropical wetland ecosystems in China (Li et al., 2007; Zhang et al., 2001). Located in a mountainous area with intensely developed karst landforms, Caohai has experienced multiple historical changes and severe human impacts, resulting in an extremely fragile ecological environment. In recent years, lake sediments, surrounding soils, and water bodies have shown varying degrees of heavy metal pollution risk, threatening the naturalness and stability of the Caohai wetland ecosystem and drawing broad attention to the environmental conditions around Caohai.

1.2 Experimental Materials

The monitoring material consisted of *Hypnum plumaeforme* Wills moss collected from Panlong Mountain, Wudang District, Guiyang City, Guizhou Province (106°51 40.063 E, 26°45 17.520 N), an area far from urban centers and pollution sources with excellent environmental quality. Plants longer than 6 cm were collected, with debris and dead stems/leaves removed. The moss was washed with tap water to remove soil and dust particles, soaked in 1% dilute nitric acid for 24 h, rinsed three times with deionized water, and dried. Nylon bags (mesh size 2.0 × 2.0 mm) were fabricated into 15.5 cm × 6.5 cm pockets. Each bag was filled with (3.0 ± 0.1) g of dried moss and sealed at the top, providing a moss bag surface area of 100 cm^2. Latex gloves were worn during all stages of moss bag preparation to avoid contamination, and completed moss bags were stored in sealed bags for later use.

1.3 Sample Collection

To accurately monitor and collect atmospheric deposition for estimating its input to the lake, sampling site selection followed these principles: avoiding local pollution from point and line sources such as chimneys and traffic arteries; placing bags on villagers' roofs to avoid ground dust contamination and uncontrollable anthropogenic interference; utilizing real-time collection to avoid repeated resuspension; and ensuring no obstructions around sampling points. Under the guidance of the Caohai Ecological Station, eight moss monitoring sites were established: S1-Yangguanshan, S2-Jiangjiawan Dock, S3-New Urban District, S4-Old Urban District, S5-Damachengcun, S6-Liujiaxiang, S7-Dongshan, and S8-Fuyelin. S1 is located at the lake' s outlet, S2 at Jiangjiawan Dock near an observation deck with high year-round pedestrian traffic and numerous commercial establishments, S3 features heavy traffic and rapid development of small enterprises, S4 is situated in the densely populated old urban district, S5 lies to the east, S7 belongs to a forested area, and both S8 and S6 are small residential clusters. These eight regions broadly cover all directions of Caohai.

[Figure 1: see original paper] Sampling sites in Caohai Lake

Moss bags were suspended at each monitoring site. Each site had eight bags: four covered with funnels to accumulate dry deposition only, and four fully exposed to the atmosphere to accumulate total deposition, all maintained at a certain height above ground. The monitoring period ran from November 2017 to November 2018 under normal climate conditions. Analysis of heavy metal content showed that weight loss of retrieved moss bags did not exceed 15%.

1.4 Sample Analysis

Exposed moss bags were labeled and stored in clean sealed bags until analysis. For analysis, 0.3 g samples were digested with a 5:2 volume ratio of HNO :H O in sealed polytetrafluoroethylene digestion tanks for 3 h at 140 °C. Reference samples and blanks (no sample but same amounts of HNO and H O , with identical digestion procedures) were included during digestion to avoid heavy metal introduction during pretreatment and analysis. Completely digested sample solutions were diluted to 25 ml for instrumental analysis. Background concentrations in moss bags were determined as the average of pretreated but unexposed samples. Cu, Zn, Pb, Cr, and Cd were measured using ICP-MS (7800, Agilent, USA/Japan) with multi-element solution standards, while Hg and As were analyzed using cold atomic absorption spectroscopy (AFS-230E, Beijing Haiguang Instrument Co., Ltd., Beijing). Average analytical error was 5%. All sample analyses were completed at the Guizhou Academy of Testing and Analysis.

For result accuracy, all reagents used were analytical grade, water was deionized, parallel samples were prepared, and analytical results are presented as averages to improve precision and reduce random error. Instrument blanks were automatically subtracted to ensure accuracy.

1.5 Calculation of Deposition Flux

Atmospheric deposition flux represents the mass of heavy metals deposited per unit area per unit time, calculated using the formula:

M—heavy metal enrichment in MossBags (µg)
S—deposition area (m^{2})
F—deposition flux (µg · m ^{2} · d ^{1})
D—sampling days (d)

1.6 Heavy Metal Evaluation Indices

The geoaccumulation index (Igeo) method was employed to evaluate pollution status of seven heavy metals in Caohai atmospheric deposition moss bags, using 1990 Guizhou Province soil values as background (Table 1) to reflect relative pollution levels and identify anthropogenic metal contamination. The Igeo calculation formula is:

where Ci is the measured value of element i, Bi is the soil background value of element i, and k is a parameter introduced to account for background value fluctuations caused by natural rock-forming processes (Hu et al., 2011b). Current geoaccumulation index studies typically adopt Muller' s classification standards, detailed in Table 2 .

Table 1 Background reference values for heavy metals

Table 2 Classification of Muller Geoaccumulation Index

Geoaccumulation Index (Igeo) Grade	Degree of Pollution
Igeo < 0	None
$0 < I$geo < 1	Slight
$1 < I$geo < 2	Medium
$2 < I$geo < 3	Strong
$3 < I$geo < 4	Strong to very strong
$4 < I$geo < 5	Very strong
$5 < I$geo < 10	Extremely strong

1.7 Data Analysis

This study used Microsoft Office Excel 2007 and SPSS 19.0 for data compilation and analysis. Pearson correlation analysis examined inter-element correlations, principal component analysis explored heavy metal sources, Origin 8.5 software generated figures, and the geoaccumulation pollution index method evaluated atmospheric quality status.

2.1 Atmospheric Deposition Flux of Metal Elements

Total atmospheric deposition around Caohai was calculated and presented in Table 3 .

Table 3 Heavy metals concentrations of total atmosphere deposition from Caohai Lake (µg · m ^{2} · d ^{1})

Analysis revealed significant differences in deposition fluxes among elements. Cu and Zn fluxes far exceeded other elements, reaching 21.43 µg · m ^{2} · d ^{1} and 102.82 µg · m ^{2} · d ^{1}, respectively, accounting for 14.68%-70.44% of total measured heavy metal deposition flux. Figure 2 [Figure 2: see original paper] shows variations among sampling sites, with Jiangjiawan Dock (S2) showing the most prominent values, followed by Damachengcun (S5), New Urban District (S3), Fuyelin (S8), Old Urban District (S4), Dongshan (S7), Yangguanshan (S1), and Liujiaxiang (S6). During the heating period, Zn dominated the entire deposition process, particularly prominent in dry deposition at the Old Urban District

(S4) and Jiangjiawan Dock (S2). These two sites experience high pedestrian and vehicular traffic. Due to industrial structure, heating relies primarily on coal combustion. While Zn mainly originates from metallurgical minerals and coal combustion, Weining County lacks related industries, suggesting combined effects from legacy primitive zinc smelting in Hezhang County, long-range atmospheric transport, and coal combustion. The Old Urban District (S4) also showed the highest Pb deposition, with the New Urban District (S3) showing similar levels. The developing New Urban District’ s increasing traffic and growing small enterprises elevate Pb content above other sites, though other elements showed no significant differences among sites. For total deposition, Yangguanshan (S1), Old Urban District (S4), and Dongshan (S7) showed equivalent deposition levels, while Liujiaxiang (S6) showed the lowest, likely due to its upwind location being less conducive to deposition. During non-heating periods, Zn and Cu deposition fluxes were highest, particularly at the New Urban District (S3) and Jiangjiawan Dock (S2). Temporally, deposition generally showed higher levels in winter-spring than summer-autumn, with heating periods exerting greater influence than non-heating periods. However, comparison of dry and wet deposition revealed wet deposition as the dominant factor. Due to Caohai’ s special geographical location along the southeast monsoon path toward the Tibetan Plateau, it easily becomes a heavy metal sink. The Tibetan Plateau to the west creates a special topographic barrier that condenses warm air masses from the southeast monsoon in summer, while cold Siberian air in winter is blocked by western mountains and cannot induce precipitation, resulting in distinct wet and dry seasons. This “high-altitude condensation” effect and rainfall influence heavy metal deposition in Weining’ s Caohai at low latitude and high altitude.

Compared with studies by Cong et al. (2008) on atmospheric annual deposition of Cd, Cr, Cu, Hg, Pb, Zn, As in Beijing’ s plain area (0.24, 11.86, 14.20, 0.024, 22.00, 54.49, 2.90 mg · m ^{2} · a ^{1}) and Tang et al. (2012b) on deposition fluxes in Daqing City (0.17, 17.85, 17.52, 0.03, 15.71, 78.81 mg · m ^{2} · a ^{1}), this study’ s Cd and Hg fluxes (0.65 and 0.04 mg · m ^{2} · a ^{1}) exceeded both studies, while other elements were lower. From geological and topographical perspectives, the southwestern karst region, centered on Guizhou, represents the world’ s largest contiguous exposed carbonate rock area, serving as an ecological barrier for the Yangtze and Pearl River basins and a major mineral resource province in China, particularly a low-temperature metallogenic center. Abundant mineral resources directly manifest as high background values of Cd, Hg, and Pb in karst areas, potentially explaining the elevated Cd and Hg levels. Zang et al. (2016) demonstrated that summer sandstorms contributed less to heavy metal pollution than winter heating and coal combustion in Lanzhou, showing lower heavy metal contributions during non-heating periods. Similar results were reported by Yu et al. (2015) for Urumqi dustfall. Significant differences also exist among monitoring sites due to climate, air currents, pollution emission sources, and long-distance transport.

2.2 Source Analysis of Atmospheric Deposition Heavy Metals

Pearson correlation analysis of seven heavy metal deposition fluxes in dry and wet deposition samples from Caohai showed correlation coefficients approaching 1 indicate significant or highly significant correlations, suggesting common sources or compound pollution (Bastami et al., 2014; Luo et al., 2011; Zhang et al., 2018c). Results are presented in Table 4 .

Table 4 Correlation Analysis of heavy metals in atmosphere deposition from Caohai Lake

Cu and Zn showed extremely significant positive correlation, As and Cr showed significant positive correlation, and Cd and Pb showed positive correlation, indicating common sources for these three element groups. Other element correlations were not significant, indicating complex heavy metal sources and severe anthropogenic interference. Pb generally originates from coal fly ash, industrial production, and vehicle exhaust, serving as an indicator element for vehicle emissions and the most characteristic element for road dust (Wong et al., 2003; Yu et al., 2009). Although unleaded gasoline has reduced vehicle exhaust impacts on atmospheric Pb, rapid economic development has increased vehicle ownership annually, and legacy Pb emissions have accumulated, making traffic pollution the primary Pb source (Mei et al., 2011). As and Hg are indicator elements for coal combustion (He et al., 2002; Taylor et al., 1982). Weining County lacks centralized heating, resulting in mostly unorganized coal smoke emissions without treatment. The special geographical location causes poor atmospheric diffusion, contributing to elevated Cd levels. The significant positive correlation between As and Cr and positive correlation between Cd and Pb indicate that atmospheric deposition of As, Cr, Cd, and Pb relates to coal combustion and road traffic. The extremely significant positive correlation between Cu and Zn reflects Zn as a characteristic element of metallurgical dust (Tang et al., 2007). Although Weining County lacks related enterprises, this may reflect legacy issues from closed primitive zinc smelting or long-range atmospheric transport. Cu primarily originates from tire wear and fungicides (Huang et al., 2009; Ötvos et al., 2003), indicating that Cu and Zn in atmospheric dust relate not only to coal combustion but also to industrial and mining enterprise emissions. Additionally, population density, long-distance transport, waste incineration, and wood/straw burning also influence atmospheric heavy metal deposition (Deng et al., 2012; Sun et al., 2009).

Overall, heavy metal pollution in atmospheric deposition primarily originates from four sources: direct vehicle exhaust emissions and secondary dust resuspension caused by exhaust; Pb-containing fuel additives and Zn/Cd lubricant additives; wheel and brake system wear; and industrial and mining enterprise emissions.

Principal component analysis further explored pollution sources (Mohammad et al., 2016; Simeonova et al., 2003), a method widely applied in analyzing pollution sources and comprehensive factor contributions (Chen et al., 2005;

Loska et al., 2003; Li et al., 2010; Zhang et al., 2009; Zhang et al., 2018d). Principal component analysis of heavy metals in Caohai wetland lake sediments typically selects eigenvalues >1. This study identified four components with cumulative variance contribution of 86.192%, indicating these four principal components reflect 86.192% of information for Caohai’ s seven heavy metals. After rotation, all four principal components maintained eigenvalues >1, with cumulative variance contribution remaining at 86.192% (Table 5).

Table 5 Total variance of heavy metals in atmosphere deposition explained using principal component analysis

Table 6 Loadings of heavy metals in atmosphere deposition based on principal component analysis

The first principal component accounted for 26.393% of total variance, representing the most important factor controlling heavy metal sources and distribution around Caohai. Table 5 shows minimal difference between pre- and post-rotation loadings, indicating Factor 1 primarily controls Cu and Zn distribution. The second principal component comprised Cd and Cr (22.665% contribution). The third principal component included Pb and As (20.77% contribution). The fourth principal component was Hg (16.364% contribution).

2.3 Pollution Assessment of Atmospheric Deposition Heavy Metals

The geoaccumulation index (*I*geo) method evaluated pollution status of seven heavy metals in Caohai atmospheric deposition to identify anthropogenic contamination (Figure 3 [Figure 3: see original paper]).

[Figure 3: see original paper] Geoaccumulation index of heavy metals in atmosphere deposition from Caohai Lake

Elemental *I*geo values ranked as: Cd > Zn > Hg > Cu > Pb > As > Cr. Comparing calculated *I*geo values with the classification table shows As, Cr, Cu, Hg, Pb, and Zn at severe pollution levels, while Cd reached an extremely high index of 10.67. Geoaccumulation index analysis confirms significant heavy metal pollution, primarily attributed to industrial activities. Even in areas far from industrial zones, heavy metal pollution can occur through long-distance transport via ocean currents and other mechanisms, representing another pollution cause. Weining County’ s special geographical location, slow urban development, backward industrial structure, and scarcity of high-tech industries make it a typical small city relying on self-heating coal combustion. Combined with six months of annual coal burning and increasing vehicle ownership, these factors severely impact the Caohai wetland ecosystem. Atmospheric deposition research reveals comprehensive influences from economic development, industrial layout, energy structure, and particularly regional differences and climatic conditions (Wang et al., 2014).

Heavy metals in Caohai atmospheric deposition show regional variations, with Jiangjiawan Dock most severely polluted, followed by Weining County’ s new urban district. Pollution is influenced by multiple factors with unclear distribution patterns. Principal component and correlation analyses identified Zn and Cu as primary pollutants with common sources, related to closed primitive zinc smelting, coal combustion, and industrial structure. This analysis of pollution characteristics and sources provides a theoretical basis for Caohai atmospheric heavy metal pollution control. Due to Caohai’ s special location, heavy metal pollution remediation will be a long-term challenge. Recommendations include optimizing energy consumption structure and production/lifestyle patterns, increasing clean energy use, appropriately controlling coal combustion, raising public pollution prevention awareness, and strengthening policy regulation. During evaluation, the absence of standardized detection methods and evaluation criteria limits comparative advantages across studies, and equipment/methodological differences cause substantial variations in results, hindering comprehensive synthesis and evaluation.

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Note: Figure translations are in progress. See original paper for figures.

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