

Article

Spatial Distribution and Ecological Risk Assessment of Potentially Harmful Trace Elements in Surface Sediments from Lake Dali, North China

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Abstract: Potentially harmful trace element (PHTE) pollution in lakes has important implications for regional management in North China, yet is seldom investigated. Surface sediments from 13 sites in Lake Dali were collected for PHTE analysis. Combined with the enrichment factor, potential ecological risk index, and multivariate statistical analysis, the spatial distribution and ecological risk of the pollutants were studied. The results showed that the contamination levels of As, Cd, Cr, Cu, Ni, Pb, and Zn were minor to moderate. Cd, Cr, Cu, Ni, Zn, and part of Pb pollution were mainly attributed to atmospheric deposition with the development of industry in North China, and As pollution resulted from the agricultural use of chemical fertilizers and As-containing pesticides. For the overall lake, the southwest part requires the most management, and targeted measures should be tailored to mitigate the ecological risk of PHTE pollution.

Keywords: remote lake; atmospheric pollution; Lake Dali; ecological risk; lake sediment

1. Introduction

Lakes are vitally important ecosystems, providing many social and ecological functions such as regional water supply, biodiversity preservation, and climate regulation. However, with increasing industrialization in recent decades, many lakes in China are at risk of potentially harmful trace element (PHTE) pollution [1–3]. In lacustrine environments, PHTEs primarily adsorb and accumulate on the sediments; hence, lake sediments can provide records for understanding the history and consequential risk of PHTE contamination [4,5]. Characterized by high toxicity, long persistence, and biological accumulation, the PHTE pollution of lake sediments has attracted increasing attention [6,7].

Metal mining and smelting, fossil fuel combustion, and agrochemicals are the main anthropogenic sources of PHTE pollution [8,9]. Studies have mainly concentrated on large lakes in populated areas, such as the lower Yangtze River Basin and Southwest China, which reported that PHTE pollution is increasing serious in these lakes [10–12]. Remote lakes are also experiencing considerable PHTE contamination due to atmospheric deposition [13–18]. However, the spatial heterogeneity of PHTE pollution may differently affect the contamination in these lakes, so each lake needs to be considered individually.

Recent studies have improved our understanding of PHTE pollution by characterizing spatial pollution levels, identifying dominant sources, and assessing ecological risks [1,2,10,19–21]. Among them, consensus-based sediment quality guidelines (SQGs) and the potential ecological risk index (Er) are comparative methods used for assessing the risk of contamination in aquatic ecosystems [22]. Lake Dali is a large lake located in the margin of semi-arid areas in North China. The hydrologically closed feature makes Lake Dali sensitive to external disturbances. Shallow groundwater in alluvial or

lacustrine sediment aquifers from arid or semi-arid areas usually have high As concentrations due to redox and desorption processes, high evaporation rate, and biologic activity, which consistently pose a risk to human health [1,23]. Better understanding of the ecological risk of PHTE in this lake has implications for regional management. In this study, by presenting PHTE concentrations in surface sediments of the lake, we aimed to: (1) describe the spatial distribution of PHTE in Lake Dali surface sediment, (2) assess the pollution level and identify potential source of the PHTEs, and (3) evaluate the potential risk of these PHTE by combining the SQGs and Er.

2. Material and Methods

2.1. Study Area

Lake Dali (43°13′–43°23′ N, 116°29′–116°45′ E) lies in the west of Hexigten Banner, Inner Mongolia, North China (Figure 1a). As the second largest lake in Inner Mongolia, the water area of Lake Dali is ~238 km², with a catchment area of ~4000 km² [24]. The water depth increases from the northeast to the southwest, with a maximum depth of ~13 m (Figure 1c) [25]. Four sources flow into Lake Dali, with two permanent sources (the Gongger and Shali Rivers) in the northeast and two intermittent sources (the Haolai and Liangzi Rivers) in the southwest. Basaltic rocks surround the lake, distributed in the north and west; lacustrine and alluvial sediments are exposed in the east [25]. At the south of Lake Dali, the Hunshandak Sandy Land is characterized by fixed and semi-fixed dunes; thus, the strong northwest and west winds create intensified dust storms in springtime. The concentrations of As reach up to 2.43 mg/L in mountain spring water, and range from 19.8 to 82.2 µg/L in the groundwater in Hexigten Banner [23]. Although large outcrops of lead–zinc ores are distributed in the southern Great Xing’an Mountain area around Hexigten Banner (Figure 1b), the local industry is dominated by stock farming; mining and related industries are less developed in the region.

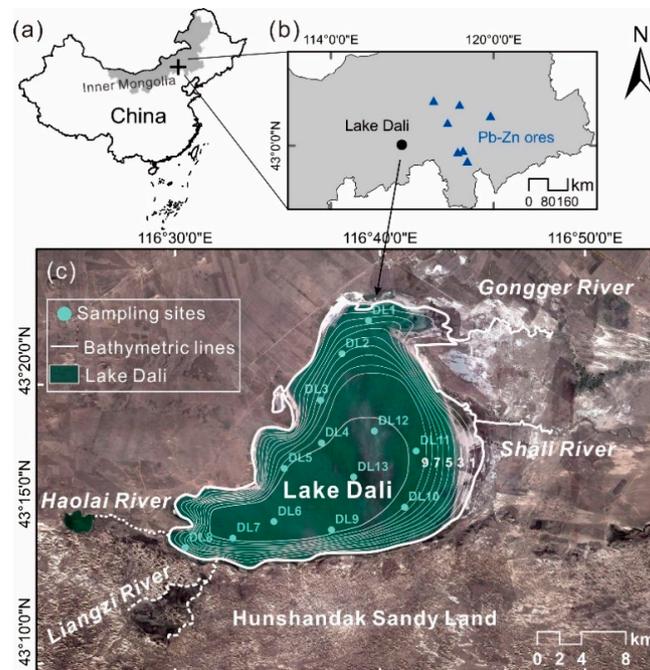


Figure 1. Location of Lake Dali and sampling sites. (a) The location of Lake Dali; the cross shows the lake’s location in China. (b) The sites of ores around the lakes. (c) The sites of 13 surface sediment samples (S1–S13) in Lake Dali.

2.2. Sample Collection

In September 2018, 13 surface sediment samples (0–2 cm) were collected using a Van Veen grabber in Lake Dali (Figure 1c). These sites were evenly distributed and covered the whole lake. The latitude

and longitude at the sampling sites were determined with a Garmin global positioning system (Garmin International, Olathe, KS, USA). All samples were freeze-dried and stored until laboratory analysis.

2.3. Laboratory Analysis

The freeze-dried samples were ground using an agate mortar, and then about 0.3 g of the sediment were completely digested by HCl, HNO₃, HF, and HClO₄ in Teflon beakers. The concentrations of Ti and Zn, and As, Cd, Cr, Cu, Ni, and Pb were determined using inductively coupled plasma-atomic emission spectrometry (Leeman Labs, Hudson, NH, USA), and inductively coupled plasma mass spectrometry (Agilent 7700x, Agilent Technologies, Palo Alto, CA, USA), respectively. The detection limits of the analytical procedure were 1 (Ti), 0.2 (Zn), 0.1 (As), 0.01 (Cd), 0.1 (Cr), 0.02 (Cu), 0.05 (Ni), and 0.01 (Pb) mg/kg. Data quality was ensured using blanks and standard reference materials (Chinese geological reference materials GBW07309). Accuracy, expressed as recovery of the reference material, was 92–106% for all the elements. The relative error was determined to be less than 5%.

2.4. Multivariate and Geostatistical Methods

Principal component analysis (PCA) was used to compress multivariable data for analyzing relationships among the variables and identify source of PHTE in sediments [10,21]. We conducted the PCA in Canoco 5.0 with ‘square root’ and ‘center and standardization’ transformation. The spatial distribution of PHTE in sediments from Lake Dali was analyzed in ArcMap 10.2 (Esri, Redlands, CA, USA). The inverse distance weighted method was used for interpolation; 2 and 12 were chosen as the power and the number of points in the search radius, respectively.

2.5. Pollution and Potential Ecological Risk Assessment

To reduce the influence of the physicochemical effects on PHTE contamination, the enrichment factor (EF) is referenced as concentration data normalized by a conservative element [6,10]. Mathematically, EF is calculated as

$$EF = (M_s/X_s)/(M_b/X_b) \quad (1)$$

where M_s and M_b are the elemental concentration measured in the samples and the background value, respectively, and X_s and X_b are the reference element concentration measured in the samples and the background value, respectively. Al and Ti are commonly used as reference elements; however, Ti concentration may be quite variable, whereas Al concentration is relatively constant among different types of rocks [26]. In this study, we selected Ti as the reference element, and PHTE concentrations of the Inner Mongolia soil were used as the geochemical background [1]. The contamination level was classified into five levels: minor contamination ($1 \leq EF < 2$), moderate contamination ($2 \leq EF < 5$), significant contamination ($5 \leq EF < 20$), very high contamination ($20 \leq EF < 40$), and extremely high contamination ($EF \geq 40$) [27].

SQGs are applied to evaluate the potential risk of the aquatic ecosystems [2,21,22]. The concentrations below the threshold effect concentration (TEC) represent a minimal effect, below which adverse effects are not expected (Table 1). Concentrations at or above the probable effect concentration (PEC) indicate adverse biological effects are likely to frequently occur.

Er is used to further comprehensive evaluate the ecological risk posed by PHTE in the lake sediments [28]. Er is calculated as

$$Er = Tr^n \times M_s/M_b \quad (2)$$

where Tr^n is the toxic-response factor for element n (Cd = 30, As = 10, Cu = Pb = 5, Cr = 2, Zn = 1). According to Hakanson [28], the grading standard of Er is classified into five categories: (1) $Er < 40$ (low risk); (2) $40 \leq Er < 80$ (moderate risk); (3) $80 \leq Er < 160$ (considerable risk); (4) $160 \leq Er < 320$ (high risk); and (5) $Er \geq 320$ (very high risk).

Table 1. Statistics of potentially harmful trace element (PHTE) concentrations in surface sediments from Lake Dali, comparisons with other selected lakes in China, background and the values of consensus-based sediment quality guidelines (SQGs, unit: mg/kg).

Location	As	Cd	Cr	Cu	Ni	Pb	Zn	Source
Lake Dali, China	13.87 ± 2.17	0.19 ± 0.05	46.90 ± 7.11	21.66 ± 5.58	25.55 ± 4.98	18.77 ± 0.89	56.67 ± 9.30	This study
Mongolia–Xinjiang Plateau, China	NA	0.13 ± 0.04	48.41 ± 15.19	22.19 ± 7.25	24.10 ± 7.54	19.42 ± 2.10	55.58 ± 14.73	[1]
Northeast China	NA	0.63 ± 0.88	92.95 ± 15.24	43.32 ± 30.97	41.4 ± 12.58	31.89 ± 13.90	133.27 ± 91.19	[1]
Qinghai–Tibet Plateau, China	NA	0.27 ± 0.23	59.89 ± 25.04	30.38 ± 15.53	33.17 ± 13.22	37.66 ± 29.40	90.54 ± 56.07	[1]
Yunnan–Guizhou Plateau, China	NA	0.78 ± 0.19	121.06 ± 48.82	76.80 ± 44.67	62.29 ± 28.82	43.62 ± 11.58	137.57 ± 18.94	[1]
Eastern Plain Region, China	NA	0.41 ± 0.17	84.42 ± 8.26	40.70 ± 7.14	43.81 ± 5.26	42.55 ± 8.67	116.79 ± 27.44	[1]
Songnen Plain, China	7.12 ± 2.31	0.25 ± 0.12	35.86 ± 12.53	14.43 ± 5.49	18.88 ± 7.28	20.78 ± 3.59	60.75 ± 19.96	[6]
Lake Dongping, China	25.30	0.29	89.30	52.00	NA	35.50	100.50	[21]
Lake Dongting, China	29.71 ± 27.70	4.65 ± 4.25	88.29 ± 12.88	47.48 ± 15.81	NA	60.99 ± 50.83	185.25 ± 78.84	[12]
Lake Erhai, China	26.90 ± 12.00	1.10 ± 0.50	103.80 ± 41.30	63.10 ± 27.30	52.20 ± 20.30	47.40 ± 18.80	109.00 ± 32.00	[10]
Lake Yilong, China	15.46 ± 4.46	0.76 ± 0.22	86.73 ± 25.18	31.40 ± 12.49	35.99 ± 12.10	53.19 ± 11.73	86.82 ± 30.05	[19]
Lake St. Clair, USA	5.9	<1	8.6	11.6	10.1	7.9	40.0	[30]
Lake Victoria, Tanzania	NA	2.5	11.0	21.6	NA	29.6	36.4	[31]
Lake Nasser, Egypt	NA	0.18	30.8	21.8	27.6	10.9	35.4	[32]
Background	7.50	0.06	41.40	14.40	19.50	17.20	59.10	[29]
Threshold effect concentration	9.79	0.99	43.4	31.6	22.7	35.8	121	[22]
Probable effect concentration	33	4.98	111	149	48.6	128	459	[22]

NA: not available.

3. Results

3.1. Spatial Distribution of PHTEs

The elemental concentrations in surface sediment of Lake Dali ranged from 10.52 to 19.16 mg/kg for As, 0.10 to 0.25 mg/kg for Cd, 28.24 to 53.16 mg/kg for Cr, 8.83 to 29.22 mg/kg for Cu, 12.84 to 30.24 mg/kg for Ni, 17.32 to 20.39 mg/kg for Pb, 33.34 to 66.34 mg/kg for Zn, and 2453 to 3047 mg/kg for Ti (Figure 2). The average concentrations for each PHTE follow the order Zn > Cr > Ni > Cu > Pb > As > Cd (Table 1). Among these PHTEs, the concentrations of As, Cd, and Pb in the surface sediments of Lake Dali were all higher than their corresponding background values; the values of Cr, Cu, Ni, and Zn in some sites were lower than their background values [29]. Compared with the published data from China and abroad (Table 1), we found the concentrations of PHTEs in Lake Dali were much lower than in the lakes in the Eastern Plain and Yunnan–Guizhou Plateau, but they were close to those of lakes in the Mongolia–Xinjiang Plateau and the Songnen Plain and in other countries [1,6,10,12,19–21].

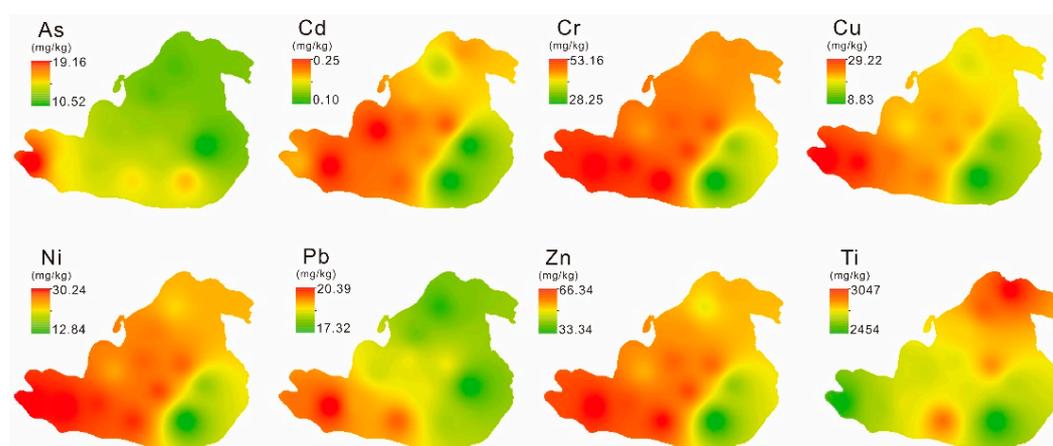


Figure 2. Spatial distribution patterns of As, Cd, Cr, Cu, Ni, Pb, Zn, and Ti in surface sediments from Lake Dali.

Spatially, As and Pb concentrations were higher in the southwestern inlets of Lake Dali. Cd, Cr, Cu, Ni, and Zn concentrations were higher in the southwestern part of the lake. Ti concentrations were primarily higher in the north part, with the lowest values in the southwestern and southeastern parts of Lake Dali.

3.2. Enrichment Factors of PHTEs

Figure 3 shows the PHTE EFs of the surface sediments from Lake Dali. The values of average EFs were: Cd (3.73) > As (2.16) > Cu (1.74) > Ni (1.52) > Cr (1.31) > Pb (1.27) > Zn (1.11). The EF for As, Cd, Cr, Cu, Ni, Pb, and Zn were 1.66–3.28, 1.96–4.90, 0.89–1.59, 0.80–2.61, 0.86–1.98, 1.11–1.47, and 0.74–1.36, respectively. According to the EF grading criteria [27], Cr, Cu, Ni, Pb, and Zn were associated with minor contamination, whereas moderate As and Cd contamination had occurred in Lake Dali. The sediments with high EFs of PHTEs mainly were found in the southwestern part of Lake Dali, indicating that this area was relatively highly contaminated.

3.3. Potential Ecological Risk Assessment of PHTEs

According to the SQGs, the As concentrations in all sites were above the TEC, but all were lower than the PEC. The concentrations of Cr and Ni were above the TEC in most (~85%) surface sediments, but all were lower than the PEC. The Cd, Cu, Pb, and Zn concentrations in all of the surface sediments were lower than the TEC (Table 1). The Er values of As, Cr, Cu, Ni, Pb, and Zn were all less than 40 in Lake Dali surface sediments, whereas Er values of Cd were 49.66–122.5, with a mean of 96.7 (Figure 4). According to Hakanson [28], moderate to considerable risk of Cd pollution exists in Lake Dali; however, other PHTEs were low-risk in this area.

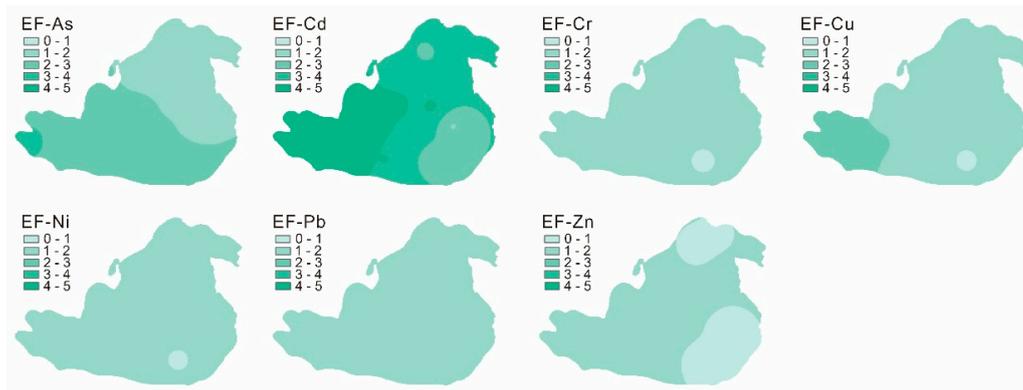


Figure 3. Distribution of the enrichment factors of As, Cd, Cr, Cu, Ni, Pb, and Zn in surface sediments from the Lake Dali.

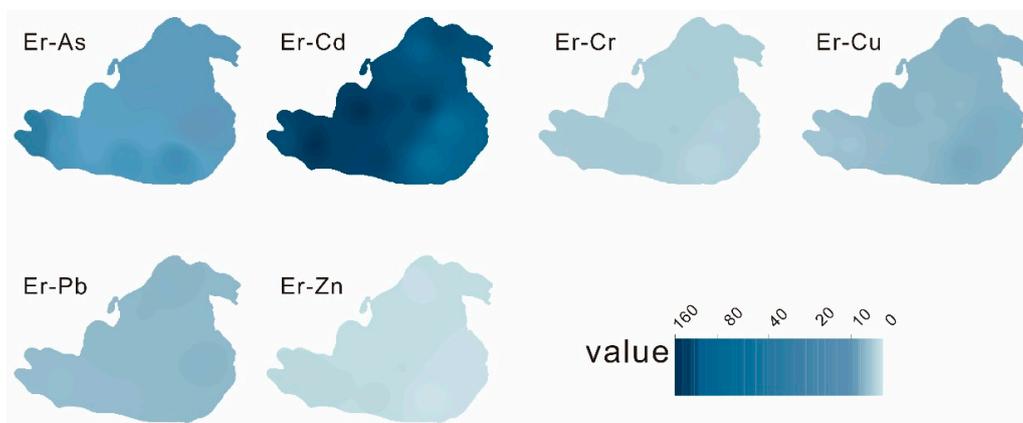


Figure 4. Distribution of the ecological risk index of As, Cd, Cr, Cu, Pb, and Zn in surface sediments from Lake Dali. Er: ecological risk index.

3.4. PCA of PHTEs

The PCA results (Figure 5) show that the first two axes, i.e., principal component 1 (PC1) and PC2, explained 94.9% of the data variance. PC1, loaded heavily by Zn, Cr, Ni, Cu, and Cd, accounted for 76.9% of the total variance, suggesting the same source of these PHTEs. PC2, with high positive loading on As, accounted for 18.0% of the total variance. Pb had both moderate loadings on PC1 and PC2, indicating a mixture Pb sources. The patterns of 13 samples along the ordination axes (PC1) were consistent with the northeast–southwest trend distribution along the lake, indicating the possible routes of PHTE mobilization within the lake.

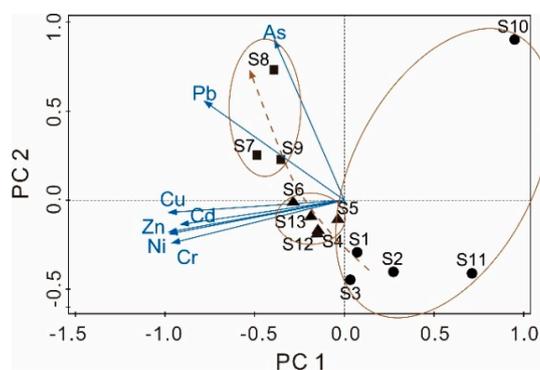


Figure 5. Principal component analysis (PCA) results of the PHTEs in surface sediments from Lake Dali.

4. Discussion

The EFs of the PHTEs were generally higher than one in surface sediments from Lake Dali, suggesting that in the whole lake, As, Cd, Cr, Cu, Ni, Pb, and Zn were influenced by human activities. Atmospheric deposition, catchment industrial and domestic sewage discharge, and agrochemical inputs are main pathways of anthropogenic input of PHTEs to lake sediments [11,15–17,33]. Lake Dali is located at the margin of Hunshandak Sandy Land, and no modern industry exists in the catchment. Thus, local industrial pollution can be ignored, and the pollution is most likely associated with atmospheric deposition and local agricultural use of chemicals.

Atmospheric deposition can be mainly identified as the source of Cd, Cr, Cu, Ni, and Zn, and part of Pb pollution. A sedimentary record of trace metal pollution from southern Lake Dali showed that the atmosphere pollution level of PHTEs in the region increased rapidly from about the 1980s [16]. Similar PHTe contamination can also be found in remote lakes in North China, such as Lake Sayram, Lake Qinghai, and Lake Gonghai [14,15,18]. Atmospheric Cd, Cr, Cu, Ni, and Zn emissions are mainly produced from coal combustion and non-ferrous metal smelting in North China [34]. Industry developed rapidly in North China after the ‘reform and opening-up’ in the late 1970s; however, the many high energy-consuming industries with poor awareness of environmental protection led to a sharp increase in atmospheric PHTe concentrations in this region [34]. This evidence indicates that regional atmospheric PHTe emissions could act as the most important factor affecting PHTe pollution in remote ecosystems.

The different PC loading and spatial distribution of As in the surface sediments from Lake Dali suggested that a large part of the anthropogenic As was not of atmospheric origin. The Lake Dali catchment is dominated by stock farming, and As compounds are components of fertilizers and pesticides. Chemical fertilizers could contain large amounts of As and other PHTEs as impurities. The As concentration in phosphate and compound fertilizers from China is ~16.0 mg/kg, suggesting that the application of chemical fertilizers is a likely source [35]. Although agricultural use of As-containing pesticides has been prohibited in China since 2002, the erosion of As previously stored in the catchment could act as an additional As source for Lake Dali surface sediments.

The results from the SQG assessment and those from the Er assessment were only partly consistent. Cu, Pb, and Zn likely posed low levels of ecological risk, and adverse effects are not expected to occur in Lake Dali. As, Cr, and Ni were recognized as the main concerning PHTEs in Lake Dali based on SQG assessment, whereas Cd likely poses high levels of ecological risk in the surface sediments from Lake Dali per the Er assessment. Cr and Ni exhibited minor contamination levels in the surface sediment, suggesting that these PHTEs were mainly present in residual forms, in agreement with reports from other lakes in China [2,6,11]. Ultramafic rocks generally exhibit Cr and Ni concentrations far higher than any other rock types. The shallow groundwater derived from late Pleistocene–Holocene alluvial–lacustrine aquifers in Hunshandak Sandy Land should have high As concentrations [33]. The minimal adverse effect of As, Cr, and Ni mainly resulted from high natural background values of these PHTEs. In contrast, Cd was most seriously influenced by human activities with the highest toxic response factor, resulting in Cd posing major ecological risk in the surface sediments from Lake Dali. Therefore, the potential adverse effects on aquatic organisms from As, Cr, and Ni pollution cannot be ignored, and different assessing methods must be synthesized to reduce the uncertainty among these assessing models in the future.

The external source and in-lake differential transport and sedimentation of PHTEs cannot be ignored. Sedimentary focusing can be the reason for the spatial heterogeneity of ecological risk. In Lake Dali, the sediment grain size is related to water depth; fine silt is mainly distributed in deep zones, whereas coarse silt and sand are found in shallow zones [36]. Due to sedimentary focusing, atmospheric PHTEs combined with fine sediments in the shallow zone can be gradually transferred to deeper parts of the lake, leading to larger pollutant accumulation in deeper parts of the lake [17]. Cd was mainly found in the carbonate-bound fraction in Lake Dali surface sediments, as Cd²⁺ can replace Ca²⁺ and Mg²⁺ in carbonates [5]. Inflows from the Gongger and Shali Rivers dilute the salinity, rendering the

concentration of carbonates in the northeastern part of the lake relatively low. These spatial distribution patterns along the lake are consistent with those of their bioavailable fractions [5], suggesting the southwestern part of Lake Dali is the most concerning region, and targeted management measures should be tailored to mitigate this risk.

5. Conclusions

In this study, PHTEs including As, Cd, Cr, Cu, Ni, Pb, and Zn in surface sediments from Lake Dali in Inner Mongolia, North China were analyzed. As, Cd and Pb concentrations in the surface sediments of Lake Dali all exceeded their corresponding background values. Based on the EF criteria, the surface sediments were moderately polluted by Cd and As on a whole-lake basis; other PHTEs were present in minor pollution level. PCA suggested that Cd, Cr, Cu, Ni, Zn, and part of the Pb pollution was of atmospheric origin, whereas As pollution was mainly attributed to agrochemicals. According to the consensus-based SQGs and the Er values, As, Cd, Cr, and Ni were identified as the priority pollutants, posing moderate to major ecological risk to the Lake Dali ecosystem. The high ecological risk in the southwestern part of Lake Dali indicates the necessity for management. Improved understanding of spatial distribution, source, and ecological risk assessment of PHTEs can provide comprehensive information for policy-makers.

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