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Historical Change and Ecological Risk of Potentially Toxic Elements in the Lake Sediments from North Aral Sea, Central Asia

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Abstract: The Aral Sea has received worldwide attention for the deterioration of its biological and chemical status. The accumulation of potentially toxic elements (PTEs) in the lake sediments reflects changes in the surrounding watershed and represents a potential hazard for the lake ecosystem. In conjunction with existing environmental records from the Aral Sea basin, sedimentary records of PTEs in North Aral Sea covering a short time scale, anno Domini (AD) 1950–2018, were used to reveal historical changes in PTE concentrations and potential risks to lake functioning. The results suggested that the levels of PTEs in lake sediments from North Aral Sea changed abruptly around 1970 AD, which is concurrent with the intensification of human activities within the basin. After 1970 AD, with the exception of As, which remained at unpolluted-to-moderately polluted levels, the geo-accumulation indices of the remaining PTEs studied (V, Cr, Zn, Co, Pb, Ni, Cu and Cd) inferred a moderately polluted status. Before 1970 AD, the total ecological risk was low, but since 1970, the total ecological risk index has exceeded 150, indicating moderate risk. Historical changes in PTE levels of lake sediments from North Aral Sea and their potential ecological risks are reported for the first time. The conclusions provide an important reference for the protection of lake ecosystems and will provide data for regional/global comparisons of environmental change during the Anthropocene.

Keywords: potentially toxic elements; potential ecological risk; lake sediments; Aral Sea; Kazakhstan

1. Introduction

The arid region of Central Asia with sparse precipitation and very fragile ecosystems [1]. Climate change and human activities are very likely to cause changes in ecosystems, even major ecological disasters [2,3]. Environmental problems caused by excessive development and utilization of water and ecological resources have become increasingly prominent, resulting in reduced water reserves, deteriorated water quality, salt dust storms, and impacted ecosystems. They have seriously restricted the social development and stability of countries within arid regions. The “Aral Sea Crisis” is the most prominent environmental problems in Central Asia [4–7]. Excessive and unreasonable development and water uses in the Aral Sea area have resulted in a large amount of industrial and domestic wastewater, pesticides, chemical fertilizers, saline and alkaline water entering the Aral Sea, with residual irrigation water and agricultural discharges also becoming important sources of pollution [4–8]. The dependence of agricultural activities on chemical supplies for decades has resulted in chemical pollutions in lakes and watersheds [9], e.g., from 1980 to 1992, the pesticides used in the Karakalpakstan area reached 29,000 tons, and the usage per unit area reached 72 kg/ha, much higher than in the United

States (1.6 kg/ha) and Russia (4 kg/ha) during the same period [10]. Former studies have suggested that pesticide use in Aral Sea basin is as high as 54 kg/ha, which is much higher than the average value of 3 kg/ha in the former Soviet Union [11]. In addition, wastewater containing high-concentrations of agricultural chemicals and salt generated by leaching and discharging from farmland, which eventually resulted in the enrichment of the brackish seawater and sediments in organic phosphorus compounds, dichloro-diphenyl-trichloroethane (DDT), lindane and other pesticides and fertilizers [5,7]. With an increase in lake water salinity, pollutants and salt deposit onto the lake bottom, and are swept away in frequent dust storms when dried out. Harmful chemicals such as pesticides and heavy metals thus disperse, posing a serious health hazard to residents in the local area and areas traversed by the chemical dust storms [9,12,13]. The most widely reported health effects in the area of the Aral Sea include anemia, cancer, respiratory diseases and birth defects. These conditions are directly or indirectly related to regional environmental degradation [7,9,14,15].

Lakes in arid areas provide a record of ecological responses to the interaction between humans and nature. Research examining the environmental evolution of lake sediments in the Aral Sea has mainly been focused on the systematic study of changes in the historic climate, vegetation, and other environmental components over long-term scales since the Quaternary [16–19]. In view of research findings, more remote sensing techniques were used to assess the impact of human activities on the Aral Sea and its watersheds, to explore the causes of lake shrinkage and to predict the lake's future destiny [20–22]. The current environmental status of water, soil and sediment within the Aral Sea basin was determined [23]. Schettler et al. [24,25] conducted related research on the evolution of different hydrochemical types of surface water, lake water and groundwater. Törnqvist et al. [26] found the concentrations of harmful substances such as copper, arsenic, nitrite and DDT in the water system of the basin to exceed applicable health standards. Rosen et al. [27] used the characteristics of pesticides (DDT, Hexachlorocyclohexane) in the sediments of 13 shallow lakes in Uzbekistan to study the impact of human activity on lakes. Rzymiski et al. [28] studied the status of pollutants of trace elements in the surface waters of the North Aral Sea and Syr Darya River. Heavy metals occurring in sediments are potentially toxic to lake functioning [29–31], but once the lake dries up, these elements are transferred to the atmosphere through salt dust storms, with the potential to impact the broader environment [32–34]. However, no research has been conducted on historical changes in the levels of potentially toxic elements (PTEs) occurring in the lake sediments. Limited water resources, destruction of ecosystems and pollution have severely affected the development of the region's socioeconomics. A balance between the sustainable use of natural resources by humans and the protection of the ecological environment needs to be established. Coping with complex developmental challenges through innovations in technology and cooperation between the international scientific and technology communities have become common appeals of all countries in Central Asia.

How to coordinate the relationship between lake-basin resources, and sustainable environmental, social and economic development needs to be studied urgently. In this paper, sedimentary records of PTEs and magnetic susceptibility from North Aral Sea combined with the evaluation of potential ecological hazards, the process of historical change and ecological risks of PTEs were revealed over a short time scale. It will provide a scientific basis for assessing the influences of the anthropogenic factors on the lake environment of Aral Sea in Central Asia.

2. Materials and Methods

2.1. Geographic Background

Aral Sea is a closed inland lake, formerly the largest Lake in Central Asia, with the runoff of the largest rivers, Amu Darya and the Syr Darya in Central Asia (Figure 1). Total surface area of Aral Sea Basin is about $154.9 \times 10^4 \text{ km}^2$ [35]. Before 1960, the entire Aral Sea remained at $6.8 \times 10^4 \text{ km}^2$ with a water salinity of about 10 g/L [35]. The records show that although the irrigated area of the basin increased from 1911 to 1960, human activities did not significantly affect the lake area [7]. Before 1960,

the inflow into the entire Aral Sea was about 63 km^3 . After the 1960s, the cultivation of crops resulted in a large increase in agricultural irrigation and domestic water consumption. During the period from 2006 to 2010, the inflow to the lake was reduced to only 4.1 km^3 [7,36]. In 1988, the North Aral Sea (small Aral) became separated from the South Aral Sea; however, a channel continued to connect them. In 1988, the water level of the lake was the lowest, with an elevation of 39.7 m (above the Baltic Sea level) [37–39]. From the sampling position (Figure 1), it ensured that our sampling point was always within the range of the lake. Kokaral Dam was built in 1992, which prevent the outflow of water from the North Aral to the South Aral [40]. The flow of the Syr Darya into the North Aral Sea induced the stability and increasement of water level of North Aral Sea [37–39]. Different from the North Aral Sea, the water quality of the South Aral continues to deteriorate, not only in terms of the continuous decline in lake water level [37,39]. The current maximum salinity of the South Aral Sea is estimated to be 200 g/L [41]. Field surveys in South Aral Sea conducted between 1990 and 2002 found that many species of phytoplankton (216 species), zooplankton (38 species) and benthic fauna (57 species) had disappeared, and only two of the original 20 fish species were observed [6].

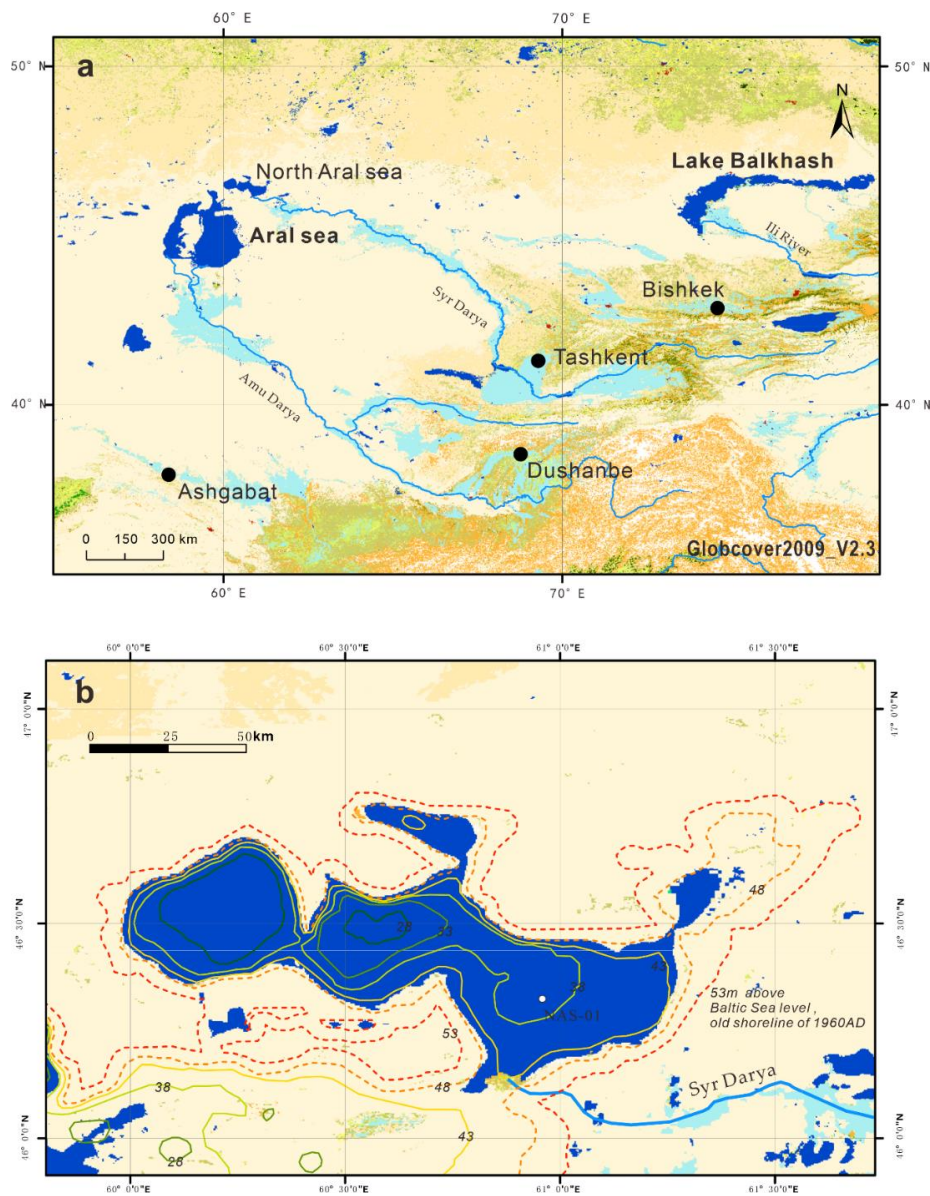


Figure 1. (a) Location of the Aral Sea in Central Asia; and (b) topographic map of North Aral Sea and the sampling location of the sediment core (NAS-01) denoted by the white cycle.

2.2. Sampling and Laboratory Methods

On 9 June 2018, a sediment core (NAS-01) was collected from North Aral Sea (60.96052° E, 43.32531° N). The core was 30 cm in length and was sampled in situ in the field at 1 cm intervals for a total of 30 samples.

The specific activity of ¹³⁷Cs is typically used to determine the chronological sequence of lake sediment cores. ¹³⁷Cs analysis was performed by γ-spectrometry using an ORTEC HPGe GWL germanium detector (EG&G ORTEC, Oak Ridge, TN, USA).

Samples were ground to 200 mesh, digested with nitric acid (HNO₃), hydrofluoric acid (HF) and perchloric acid (HClO₄), and analyzed by ICP-AES (Prodigy, Teledyne Leeman Labs, Hudson NH, USA) to determine the V concentration (limit of detection: LOD = 2 mg/kg). ICP-MS (7700x, Agilent Technologies, Palo Alto, USA) was used to measure PTE concentrations Cr (LOD = 0.1 mg/kg), Co (LOD = 0.01 mg/kg), Ni (LOD = 0.05 mg/kg), Cu (LOD = 0.02 mg/kg), Zn (LOD = 0.2 mg/kg), As (LOD = 0.1 mg/kg), Cd (LOD = 0.01 mg/kg) and Pb (LOD = 0.02 mg/kg). The standard reference material GBW07311 [42] was used to ensure the reliability of the analytical data. The relative error was determined to be less than 5%.

To measure the magnetic susceptibility (MS) with low-frequency (470 Hz) and high-frequency (4700 Hz) with a Bartington MS2 fitted with a MS2B dual-frequency sensor (Bartington Instruments, UK), a freeze-dried sediment sample was loaded into a special box with a volume of 8 mm³, and the box containing the sample was weighed and measured. Each sample was measured three times and the mean value was taken.

2.3. Potential Ecological Risk Index (PERI)

The PERI [30,43–46] for PTEs was calculated as follows

$$C_f^i = (C_n^i / B_0^i) \tag{1}$$

$$E_r^i = \sum_{i=1}^n T_r^i \times C_f^i \tag{2}$$

$$RI = \sum_{i=1}^n E_r^i \tag{3}$$

where RI: total risk index; E_r: individual PTE PERI; T_r: toxic-response factor, V = 2, Zn = 2, Cr = 2, Cu = 5, Co = 5, Ni = 5, Pb = 5, As = 10, Cd = 30 [47]; C_n: the content of PTEs; B₀: background PTE concentration. Potential ecological risk index classification criteria are shown in Table 1 [29].

Table 1. PERI classification standards for PTEs.

E _r	Classification	RI	Classification
E _r < 40	Low	RI < 150	Low risk
40 ≤ E _r < 80	Moderate	150 ≤ RI < 300	Moderate
80 ≤ E _r < 160	Considerable	300 ≤ RI < 600	Considerable
160 ≤ E _r < 320	High	600 ≤ RI	high
320 ≤ E _r	Very high	-	-

2.4. Geo-Accumulation Index (I_{geo})

The index of I_{geo} [29,48,49] can determine the extent of PTE pollution in the natural environment of the North Aral Sea.

$$I_{geo} = \log_2 \left[\frac{C_n}{1.5B_n} \right] \tag{4}$$

where C_n : PTE concentration; B_n : PTE background concentration. The classification standards for I_{geo} are as follows: practically unpolluted ($I_{geo} < 0$); $0 < I_{geo} < 1$, suggested unpolluted to moderately polluted; $1 < I_{geo} < 2$, reflected moderately polluted, etc. [29].

3. Results

3.1. Summary Statistics for 12 Environmental Parameters

Summary statistics for 12 environmental parameters measured in the sediment core from the North Aral Sea was shown in Table 2. The mean value for the potentially toxic element V was 91.6 mg/kg, with maximum and minimum values of 127.5 and 29.5 mg/kg, respectively. The mean, maximum and minimum concentrations for Cr were 59.3, 80.4 and 18.2 mg/kg, respectively. The average Co concentration was 10.2 mg/kg (minimum: 3.18 mg/kg, maximum: 14.0 mg/kg). The minimum, maximum and average concentrations of Ni were 7.72, 40.9 and 33.2 mg/kg, respectively. The maximum value of Cu concentration was 37.5 mg/kg, the minimum was 5.16 mg/kg and the mean value was 22.8 mg/kg. The mean value of Zn concentration was 61.2 mg/kg, with maximum and minimum values of 90.0 and 17.4 mg/kg, respectively. The mean, maximum and minimum values for as were 11.4, 18.3 and 3.78 mg/kg, respectively. Cd levels were the lowest among the nine PTEs with an average concentration of 0.192 mg/kg. The minimum, maximum and mean Pb concentrations were 4.64, 22.8 and 15.4 mg/kg, respectively. The maximum value of low-frequency MS was $15.9 \times 10^{-8} \text{ m}^3/\text{kg}$, and the minimum was $1.91 \times 10^{-8} \text{ m}^3/\text{kg}$ with average of $10.7 \times 10^{-8} \text{ m}^3/\text{kg}$. The mean high-frequency measured in North Aral Sea sediment was $10.3 \times 10^{-8} \text{ m}^3/\text{kg}$.

Table 2. Summary statistics for 12 environmental parameters: ^{137}Cs , magnetic susceptibility (MS) with high (HF) and low frequency (LF) and potentially toxic elements (PTEs) in the North Aral Sea sediment core.

Parameters	Range	Minimum	Maximum	Mean		Std. Deviation
				Statistic	Std. Error	
V (mg/kg)	97.9	29.5	127	91.6	6.94	38.0
Cr (mg/kg)	62.2	18.2	80.4	59.3	4.53	24.8
Co (mg/kg)	10.8	3.18	14.0	10.2	0.772	4.23
Ni (mg/kg)	33.2	7.72	40.9	27.4	2.28	12.5
Cu (mg/kg)	32.3	5.16	37.5	22.8	2.11	11.5
Zn (mg/kg)	72.6	17.4	90.0	61.2	5.00	27.4
As (mg/kg)	14.6	3.78	18.3	11.4	0.797	4.37
Cd (mg/kg)	0.317	0.0453	0.362	0.192	0.0176	0.0966
Pb (mg/kg)	18.2	4.64	22.8	15.4	1.23	6.74
LF (m^3/kg)	14.0	1.90	15.9	10.7	0.960	5.26
HF (m^3/kg)	13.4	2.00	15.4	10.3	0.929	5.09
^{137}Cs (Bq/kg)	29.3	0	29.3	13.0	1.17	6.42

3.2. Vertical Distribution of Environmental Parameters

The specific activity of ^{137}Cs in top layer of the lake sediments from North Aral Sea was 13.6 Bq/kg, corresponding to the sampling time of 2018. The main accumulation peak (29.3 Bq/kg) appeared at 18 cm, which may correspond to the Chernobyl nuclear leak that occurred in the former Soviet Union in 1986 [50,51]. The peak at 28 cm (2.29 Bq/kg) corresponds to year 1954 [52], while the peak at 24 cm (13.3 Bq/kg) may correspond to year 1963 [53,54]. The age of the sediment core was interpolated from the ^{137}Cs time-markers. The linear fitting curve of depth (x) and age (y) was $Y = -2.37X + 2022.4$ ($r^2 = 0.98$, $p = 0.01$).

Interestingly, the most striking result to emerge from Figure 2, showing the vertical distributions of environmental parameters, is the occurrence of three obvious discontinuities that divide the PTE profiles into four relatively obvious stages. Below 21 cm, the PTE concentration is low, and after a rapid

increase to a depth of 9 cm, another spike in PTE concentration occurs. From 9 to 2 cm, the increase in PTE concentration is gradual. A significant change in PTE concentration is noted again in the surface sediments, but the trend is opposite to that of the first two mutation points, with the PTE concentration starting to decrease.

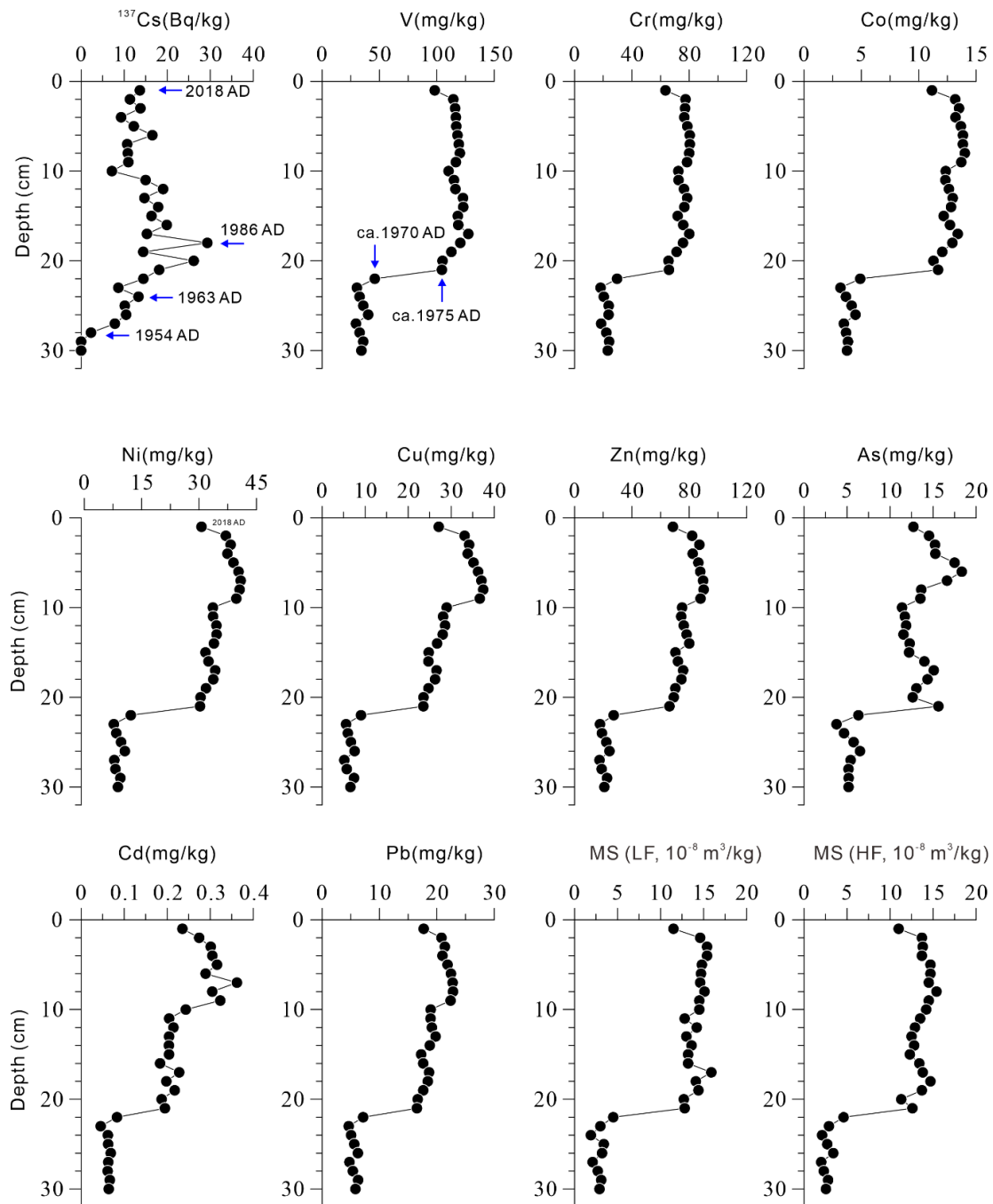


Figure 2. Vertical variations of ^{137}Cs , PTEs and magnetic susceptibility (MS) in lake sediments from North Aral Sea.

4. Discussion

Although the statistical data (Table 2) and vertical distributions of heavy metals (Figure 2) can reflect changes in PTEs as a whole, clustering analysis can provide an objective grouping method for PTEs [55,56]. In general, most studies apply cluster analysis to PTEs examine the relationship between

variables. However, Figure 3 shows the two-way dendrogram for the clustering of PTEs and sample layers generated with two-way cluster analysis.

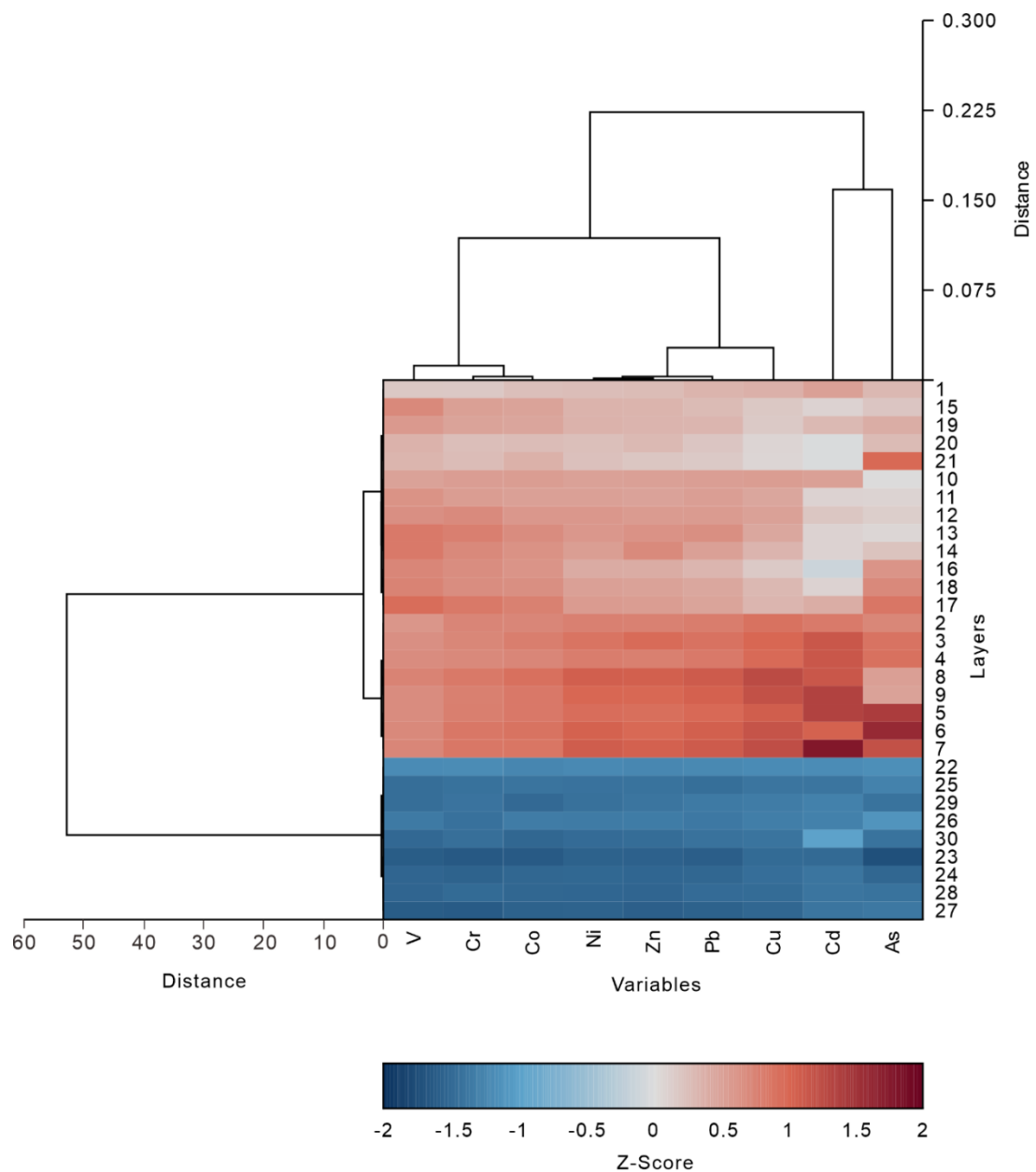


Figure 3. Cluster dendrogram for samples and PTE concentrations.

The connection distance shown on the graph is a standardized Euclidean distance and represents the relative similarity between the sample layers and the PTEs. Differences between the clusters can be used to distinguish sample layers and PTEs. Judging from the distances between the sample horizons, there is a huge difference between the first layer (0–21 cm) and the second layer (22–30 cm). If the first layer (0–21 cm) were further divided, it could be divided into three secondary layers (0–1 cm), (2–9 cm) and (10–21 cm), which are consistent with the stages noted in the vertical distribution plots (Figure 2). The cluster analysis showed that separation of the sediment core into layers based on the characteristics of PTEs achieves good results. In addition to sample horizons, the cluster analysis also allows for easier determination of the affinity between PTEs. Clustering between PTEs indicated that the nine PTEs could be divided into three clusters. The first PTE cluster includes Cd and As, which are

also the elements with the largest toxicity coefficients among the nine PTEs studied. The second group includes Ni, Zn, Pb and Cu, and the third group includes V, Cr and Co. The elements in the second and third groups are more closely related compared with the elements of the first group. The results show the usefulness of micro-geochemical features that may not be identifiable by comprehensive statistical data alone. Temporal changes in the geo-accumulation index of each PTE are depicted in Figure 4. The geo-accumulation index of all nine PTEs was less than 0 before 1970, indicating that the status of all PTEs was practically unpolluted. From 1970 to 1975, PTEs became significantly enriched. After 1975, with the exception of as (0–1), the geo-accumulation index of the PTEs was greater than 1, reflecting a moderately polluted status. What factors have caused this obvious change?

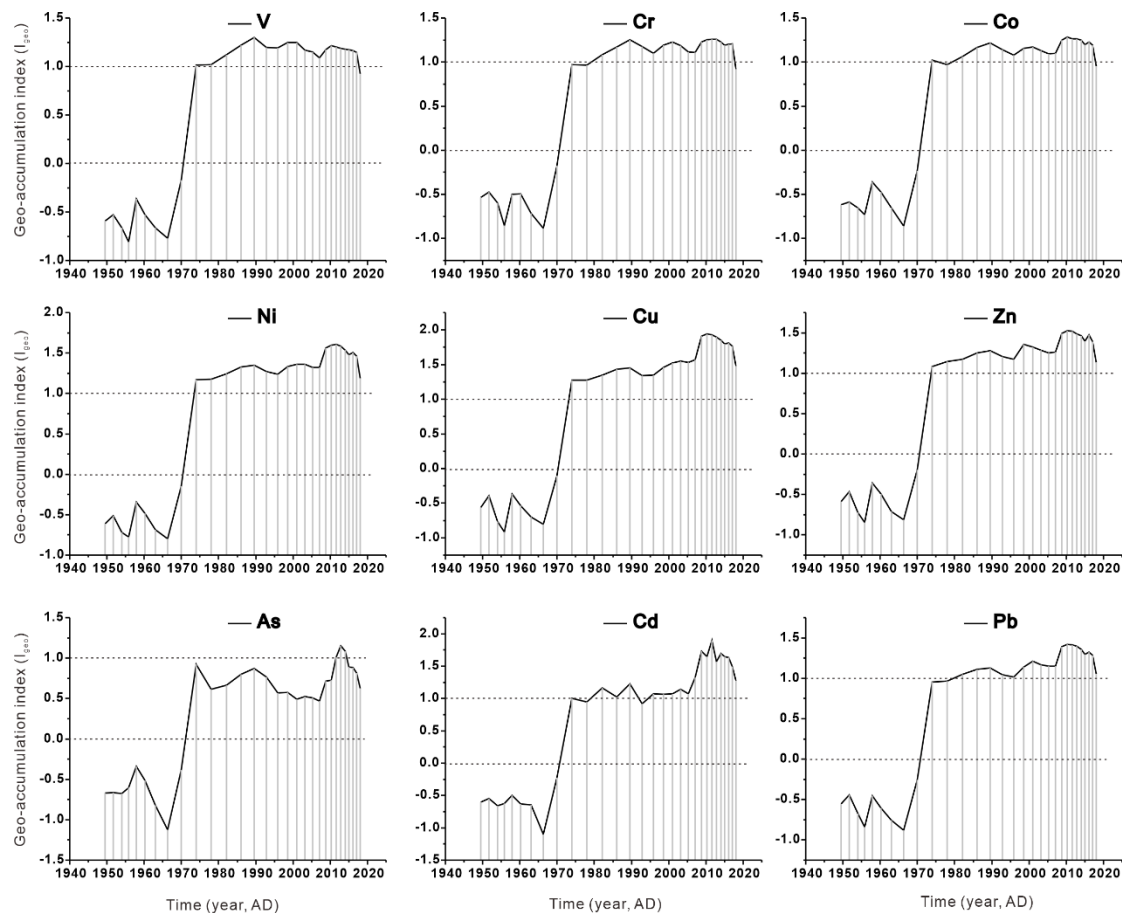


Figure 4. Time-varying curves of PTE geo-accumulation indexes (I_{geo}).

The high-frequency MS and low-frequency MS of the Aral Sea sediments are significantly correlated, which is a typical characteristic of lake sediments [57]. In lacustrine sediments, the correlation between magnetic susceptibilities measured at different frequencies is very good, and the correlation coefficient (r) can exceed 0.99 ($p < 0.001$), indicating that increasing magnetic susceptibility is related to an increase in the content of magnetic minerals. Previous studies have shown that MS can be used to reflect the variations in the intensity of human activities [58–60]. The intensification of human activities has led to the enrichment in ferromagnetic materials of surface materials entering the lake, which has led to a significant increase in magnetic susceptibility since 1970 AD. Figure 5 shows changes over time in magnetic susceptibility, temporal changes in surface area of irrigated Land in Aral Sea basin [61], and the volume of import and export trade between the former Soviet Union and the United States. Temporal changes in surface area of irrigated Land (million hectares) in the Aral Sea basin, to a certain extent, reflect changes in agricultural intensity. The area of irrigated Land in Aral Sea basin was slowly increasing before the 1970s but showed a drastic increase during the 1970s (Figure 5). A sudden change

The PERI inferred the polluted degree of PTEs in the Lake sediments of North Aral Sea (Table S1). The results showed that E_r values for the entire sediment core follow the ascending order of $V < Cr < Zn < Co < Pb < Ni < Cu < As < Cd$. With the exception of Cd, the ecological risk of individual elements was lower than 40, suggested the ecological risks for PTEs were low. Before 1970, the average ecological risk of Cd was 29.7, lower than 40. However, after 1970, the average Cd ecological risk index increased to 114.5, suggesting the Cd constitutes a considerable risk to the ecosystem of North Aral Sea (Figure 6).

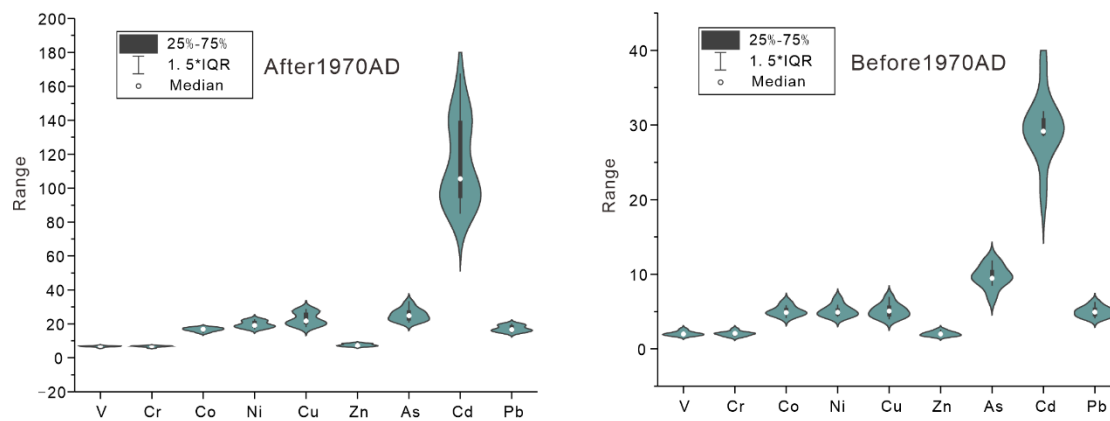


Figure 6. Violin plots for the PERI of PTEs (V, Cr, Co, Ni, Cu, Zn, As, Cd and Pb) from the lake sediments of North Aral Sea.

The RI values in the sediment core were all lower than 150 before 1970 AD, indicating that the integrated ecological risk of PTEs was at a low risk level. Since 1970, however, it is worth noting that the RI values have exceeded 150, which represents moderate risk. In particular, at a depth of 7 cm (ca. 2012 AD), the RI value reached 309.8, indicating a considerable risk (Figure 7). Overall, the PERI is used as an evaluation method for a large number of applications [72–74], and it will provide an important reference for the protection of lake ecosystems and will provide data for regional/global comparisons of environmental change during the Anthropocene. It must be mentioned that the grain size and the deposition rate of sediments will change with the variation in lake water level. On the other hand, the crystal precipitation of chemicals in lake water may also have an effect on the content of potentially toxic elements in sediments. Therefore, the relative contents of potentially toxic elements in core sediments may be quite different in different locations. Because of the difficulty of sampling in the Aral Sea, only one sediment core of the lake cannot fully reflect the overall enrichment law of lake sediments in the North Aral Sea, which is also one of the shortcomings of our paper.

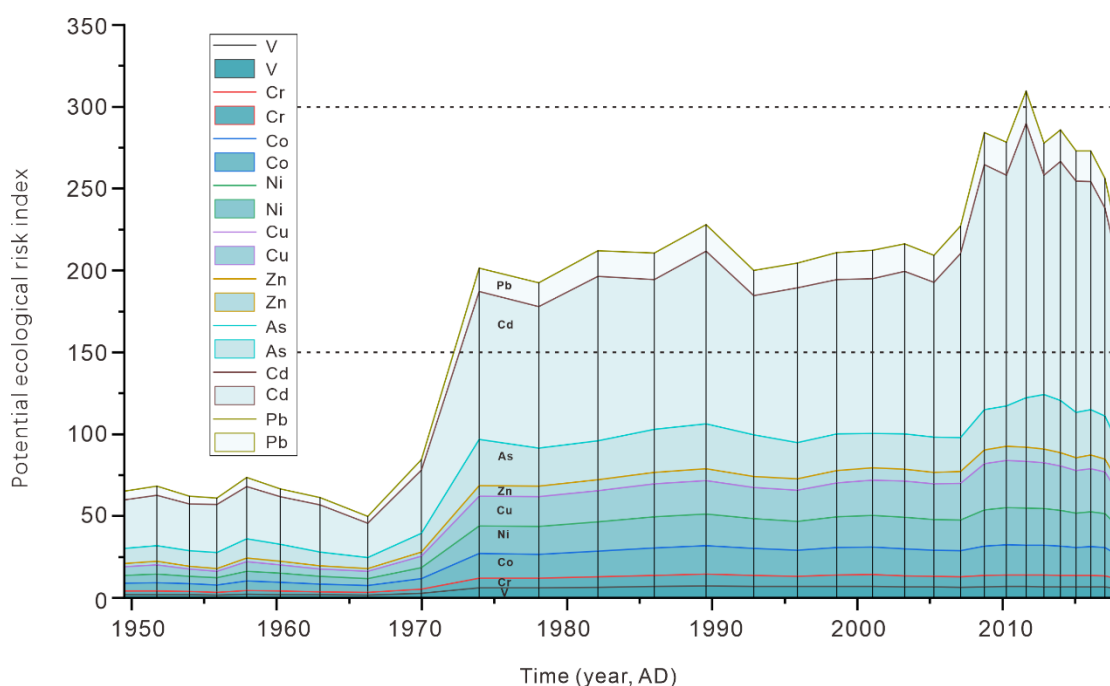


Figure 7. Stacked area chart shows how the PTEs have collectively changed over the past ca. 70 years.

5. Conclusions

This study evaluates historical changes in concentration and the potential ecological risks of PTEs in the lake sediments from North Aral Sea. The conclusions are as follows:

1. The contents of PTEs in the lake sediments from North Aral Sea changed abruptly around 1970 AD, which is associated with the anthropogenic intensification in the basin;
2. After 1975 AD, with the exception of As ($I_{geo} < 1$), the geo-accumulation index of PTEs (V, Cr, Zn, Co, Pb, Ni, C, and Cd) was greater than 1, reflecting a moderately polluted status;
3. Before 1970 AD, the total ecological risk index was lower than 150, indicating a low risk, but since 1970, the total ecological risk index has exceeded 150, which represents a moderate risk.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2076-3417/10/16/5623/s1>, Table S1: Potential ecological risk index (PERI) of Potentially Toxic Elements (V, Cr, Co, Ni, Cu, Zn, As, Cd and Pb) from the lake sediments of North Aral Sea.

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