



Statistical Distribution Features and Evaluation of Ecological Risk in Superficial Sediments of Hulun Lake

Rong Li* and Xiaohui Shu

Department of Economics, Huaihua University, Huaihua 418008, Hunan Province, P. R. China

*Corresponding Author: Rong Li

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ABSTRACT

The paper discusses concentrations, statistical distributions and ecological risks of seven heavy metals in superficial sediments of Hulun Lake. The highest values for heavy metal concentrations in sediments before the modern industrialization of the world and grade I criteria of the National Standard for Soil Environmental Quality were used to analyse the accumulation and potential ecological risk (PER) coefficients of heavy metals. The PER index for each sample location is also discussed. The PER index was evaluated using the method of Lars Hakanson to evaluate ecological risk. The distribution of content of heavy metal does not reveal regional distributive characteristics; the space distribution differential is small. However, three metals (i.e., Zn, Cr, Cu) exhibit the same trend. These heavy metals are highly concentrated in the northeast and southwest parts of Hulun Lake than at the entrance of the Xinkai, Wuerxun, and Kelulun rivers. Using the highest values for heavy metal concentrations in sediments before the modern industrialization of the world as reference, these heavy metals have an accumulation order of $Pb > Zn > Cd > Cu > As > Cr > Hg$ and pollution level order of $Cd > As > Pb > Cu > Hg > Cr > Zn$. However, most of these heavy metals are in low risk states. Using the grade I criteria of the National Standard for Soil Environmental Quality work as reference, these heavy metals have an accumulation order of $Cd > Zn > Cu > As > Pb > Cr > Hg$ and pollution level order of $Cd > As > Hg > Cu > Pb > Cr > Zn$. Cd is the potential impact element for the ecological environment in Hulun Lake.

INTRODUCTION

Lake water pollution mainly accumulates in lake sediments, which are the potential lake pollution sources (Milendovic et al. 2005, Liu et al. 2006). Heavy metals are adsorbed by suspended solids in water and eventually deposited to sediments in lakes (Fan et al. 2002). Pollutants could re-enter the overlying water because of the water-sediment interface under a series of biogeochemical processes (Zhou et al. 2005). Heavy metals in sediments are sensitive indicators of water pollution and significantly reflect the status of the water system (Yang et al. 2005). Heavy metals in sediments become direct and indirect threats to the ecosystem through the bioaccumulation and amplification effect. Therefore, heavy metal content in sediments and its distribution should be investigated to evaluate its potential to harm the ecosystem. Determining the main pollutants and understanding their impacts on water quality is important and provide the basis for water pollution control.

Hulun Lake, also known as Dalai Lake, is located west of Hulunbeier Grassland, in the middle of Xinba'erhuyouqi, Xinba'erhuzuoqi, and Manzhouli (east longitude: $117^{\circ}00'10''$ to $117^{\circ}41'40''$, north latitude: $48^{\circ}30'40''$ to $49^{\circ}20'40''$). The lake exhibits an irregular oblique rectangle shape, with a shaft from the northeast to southwest. It has a

length of 93 km, maximum width of 41 km, circumference of 447 km and an area of 2339 km². The average water depth of the lake is 5.7 m; the maximum depth is 10 m; and the total water content is 13.85 billion m³. Hulun Lake was approved as a national wetland nature reserve in 1992 by the Hulunbuir Grassland Ecological Protection and Economic Development, and its waters and wetlands are irreplaceable (Yue et al. 2008). The lake has faced a series of environmental problems caused by climate change and human activities in the last 40 years. Its water level and water area decreases annually. In addition, the wetlands are decreasing, and the surrounding ecological environment and water quality are deteriorating. The total salt content and pH value of the lake increases annually. A large area of reed lake has disappeared. Fish resources have dried up, and a large number of rare birds have migrated. The water quality of Hulun Lake is currently at a medium eutrophication level (Han & Chi 2002), and the wetland ecological environment has sharply deteriorated. This phenomenon has become a serious threat to the ecological security of northeast and north China.

However, studies on Hulun Lake are minimal. Most researchers only investigated and analysed the quality, quantity and water level of the lake and focused on the impact of climate change on regional wetland ecological status and

environmental governance (Li et al. 2006, Wang 2005, Zhao et al. 2008). In this study, we investigated the heavy metal elements (i.e., Cu, Zn, Pb, Cr, Cd, As and Hg) in the sediments of Hulun Lake, including their distribution and enrichment characteristics, to determine the sediment pollution status of the lake and the characteristics of pollutant distribution. We utilized the potential ecological harm index to evaluate the damage caused by heavy metals. This paper would provide a scientific basis for quality comprehensive evaluation and water pollution control of the Hulun Lake environment.

MATERIALS AND METHODS

Sampling points set: Eleven sediment sampling points in the lake were set-up according to area size, shape and direction (i.e., water flow). Twenty-one samples of surface sediment above 15 cm were collected since December 2008 through a columnar sampler at a fixed position from a specific location using the global positioning system. The samples were collected in winter from the frozen lake by ice drill, sealed into polyethylene plastic bags, and then immediately cryopreserved in the laboratory.

Analysis methods: The samples were crushed using a glass rod after natural drying in the laboratory to remove gravel, shells, animal and plant residues and other impurities. The samples were then processed through a 100-mesh sieve mortar after grinding and analytically determined the heavy metal elements, such as Cu, Zn, Pb, Cd, Cr, As and Hg. The heavy metal content in the sediment samples were measured after the samples were digested with $\text{HNO}_3\text{-HF-HClO}_4$. Flame atomic absorption spectrophotometry was utilized for analytic determination of Cu, Zn, Pb, Cd, Cr and As, whereas cold atomic absorption spectrophotometry was utilized for Hg.

Potential ecological risk (PER) assessment of heavy metals: Several methods are available to evaluate heavy metal pollution in sediments. This paper adopted the ecological hazard index method proposed by Swedish scientist Hakanson. The method employs the index relative to heavy metals in sediments and sediment before industrialization, highest concentration of heavy metals pollution degree, and

corresponding values of ecological toxicity coefficient weighted sum to obtain the ecological hazard index. The method starts at the biological toxicity of heavy metals, which reflects the influence of each pollutant on a particular environment and the synthesis of various pollutants. This method is used quantitatively to differentiate the harmful degree to potential ecological risk caused by heavy metal pollution in sediments (Jiang et al. 2008, Xiang et al. 2006).

RESULTS AND DISCUSSION

The Content and Distribution of Heavy Metals in Sediments

The difference of heavy metal content in space: Table 1 shows the statistical results of different heavy metal elements. Zn has the highest value, with a mean value of 68.80 mg/kg. Cr, Cu, and Pb are lower than Zn, with mean values of 36.37, 23.44 and 22.34 mg/kg, respectively. Arsenic has a mid-level value, with a content of 10.36 mg/kg. Cadmium content is relatively small at only 0.41 mg/kg and Hg content is the lowest at 0.019 mg/kg. Cd has the largest space variation coefficient at 68.30% because of Cd content in the sediment on B9, which is near the entrance of northeast of Xingkaihe; this value is higher (0.78 mg/kg) than the other sample points. Simultaneously, arsenic and Hg have large space variation coefficients (except Cd) at 57.84% and 56.39%, respectively. Arsenic content (24.18 mg/kg) at E8 is higher than the other points, whereas Hg content (0.002 mg/kg) at I2 is lower than the other points. The variation coefficients of Pb, Zn, Cu, and Cr are between 30% and 44%. Thus, the spatial distribution difference of seven heavy metals in lake sediments is relatively small. Except for Cd, the content of the other six heavy metals is below level-1 of the natural background value of soil environmental quality standards in China (GB 15618-1995).

Regional distribution of heavy metal content characteristics: Table 2 shows the correlation coefficients among the seven heavy metals found in the sediments of Hulun Lake. Copper, Zn and Cr exhibit an extremely significant correlation. All sampling points at the Hulun Lake have consistent content change trend of Zn, Cr and Cu metals. The heavy

Table 1: Statistical values of heavy metals in sediments at Hulun Lake (n=21).

Metallic element	Minimum value (mg/kg)	Maximal value (mg/kg)	Mean value (mg/kg)	Standard deviation value (mg/kg)	Coefficient of variation (%)
Cu	3.39	31.92	23.44	7.42	31.67
Pb	4.45	55.29	22.34	9.78	43.77
Cr	9.33	50.76	36.37	10.92	30.02
Zn	10.20	105.60	68.80	24.56	35.69
Cd	<0.01	0.78	0.41	0.28	68.30
As	4.26	31.55	10.36	5.99	57.84
Hg	0.002	0.039	0.019	0.01	56.39

Table 2: Correlation coefficient between heavy metals for the sediment of Hulun Lake.

Element	Cu	Zn	Pb	Cr	Cd	As
Zn	0.823**					
Pb	0.297	0.337				
Cr	0.934**	0.854**	0.352			
Cd	0.282	0.262	0.128	0.424		
As	-0.008	0.052	-0.373	-0.044	-0.016	
Hg	0.119	0.182	0.03	0.191	0.086	0.492*

Note: *(P<0.05) significant correlation, **(P<0.01) very significant correlation (two-tailed test).

metal content at the northeast (B9, D7, D11) and southwest (F5, G2, G8, H5) ends of the lake are high, whereas that at the Xinkai River (A10), Wuexun River (F9), and Kerulen (I2) at the entrance to the lake content are low. Regional distribution is not obvious. Arsenic and Hg also have a significant correlation. However, arsenic content is higher than that of Hg. Mercury content is extremely small, and thus, negligible. The content order of six heavy metals on the sediment surface in Hulun Lake from most to least is as follows: Zn > Cr > Cu > Pb > As > Cd. Chromium, Zn and Cd have the highest content in B9; Pb is the highest in F5; As is the highest in E8; and Cu is the highest in G2.

The Enrichment of Heavy Metals in Sediments

Several scholars have adopted the enrichment coefficient to measure the enrichment degree of single heavy metal, which is represented as formula (1):

$$C_f^i = C_m^i / C_n^i \quad \dots(1)$$

Where, C_m^i is the measured values of heavy metal i in the sediments and C_n^i is the required ratio for calculating the environmental background values.

This paper uses the highest background value of heavy metals contained in normal particle sediment before the modern industrialization proposed by Lars Hakanson; the high background of values w (Hg), w (As), w (Cu), w (zinc), w (Pb), w (Cd), w (Cr) were 0.25, 0.25, 30.00, 80.00, 25.00, 80.00, and 25.00 mg/kg, respectively (Lars Hakanson, 1988). The real pollution level of the lake can be determined according to the high background values. The surrounding soil environment of the lake lacked the background value of the seven heavy metals, and Hulun Lake belonged to the national wetland nature reserve. Thus, we chose the level-1 natural background value ratio of “the soil environment quality standard” (GB 15618-1995). The natural background value w (Hg), w (As), w (Cu), w (zinc), w (Pb), w (Cd), w (Cr) were 0.15, 0.15, 35.00, 100.00, 35.00, 100.00, and 35.00 mg/kg, respectively, which reflected the relative pollution degree of Hulun Lake. Combining the results, an

improved response to the potential ecological damage of the lake was observed. The heavy metal enrichment coefficient of each sample point can be calculated using formula (1); the results are given in Tables 3 and 4.

Lead has the highest enrichment degree, with an average enrichment coefficient of 0.90 followed by Zn, Cd, Cu, As and Cr and Hg has the lowest value, with an average enrichment coefficient of only 0.073. If the level-1 natural background value of the soil environment quality standard in our country is used as reference, Cd enrichment degree is the highest, with an average enrichment coefficient of 0.90 followed by Zn, Cu, As and Pb; Hg has the lowest value. In terms of sampling points, Zn, Cr and Cd have the highest accumulation degree at B9 (northeast end of the lake); Pb and Hg have the highest accumulation degree at F5 (northwest edge of the lake); As has the highest accumulation degree at E8 (close to centre of the lake); and Cu has the highest accumulation degree at G2 (southwest edge of the lake). In summary, the concentration degree at the three river estuaries (i.e., A10, F9, I2) is generally low.

PER Assessment Caused by Heavy Metals

Hakanson proposed the PER index method in 1980, which assesses the ecological risk assessment of heavy metals (Caeiro et al. 2005). PER assessment is based on the principle of element abundance and releasing ability. The evaluation hypothesis consists of the following conditions: (1) response of element abundance [i.e., potential ecological risk index (RI)] increases with metal pollution in sediments; (2) synergy of several pollutants (i.e., the metal in sediment obeys the additive property in ecological harm). A variety of metal pollution potential has a big ecological risk, and heavy metals such as Cu, Zn, Pb, Cd, and Cr are preferred; (3) heavy metals have different toxicity responses (i.e., the metal of strong biological toxicity has high weights than RI). The potential ecological harm index reflects four aspects: (1) metal concentration in the sediment surface; (2) species number of metal pollutants; (3) metal toxicity level; (4) sensitivity of water body to metal contamination (Huang et al. 2008).

According to this method, E_r^i represents the individual potential ecological harm coefficient of the i^{th} heavy metal in sediments of an area and is expressed by equation 2, and RI represents the composite index of various heavy metals in sediments and is expressed by equation 3.

$$E_r^i = T_r^i C_f^i = T_r^i C_s^i / C_n^i \quad \dots(2)$$

$$RI = \sum_{i=1}^n E_r^i = \sum_{i=1}^n T_r^i C_r^i = \sum_{i=1}^n T_r^i C_s^i / C_n^i \quad \dots(3)$$

Where, C_f^i is the enrichment coefficient of the i^{th} heavy

Table 3: Enrichment coefficients of heavy metals in sediment of Hulun Lake.

Sampling point	Cu	Zn	Pb	Cr	Cd	As	Hg
A10	0.26	0.31	0.58	0.25	0.77	0.74	0.08
B9	0.91	1.22	1.00	0.81	1.53	0.98	0.08
D7	0.97	1.16	1.17	0.78	1.26	0.59	0.10
D11	0.94	1.08	0.50	0.74	0.70	0.73	0.09
E8	0.73	0.68	0.58	0.50	0.71	1.61	0.13
F5	0.88	0.97	1.50	0.69	0.74	0.49	0.13
F9	0.65	0.60	0.86	0.55	0.68	0.32	0.08
G2	1.06	1.02	1.10	0.80	1.48	0.32	0.04
G8	1.04	0.90	0.90	0.70	0.18	0.55	0.04
H5	0.83	1.02	0.76	0.54	0.75	0.60	0.03
I2	0.48	0.59	0.99	0.40	0.57	0.48	0.02
Mean value	0.79	0.87	0.90	0.62	0.85	0.67	0.07

Note: The heavy metal content about the highest background value is normal granular sediments of modern pre-industrial time.

Table 4: Enrichment coefficients of heavy metals in sediment of Hulun Lake.

Sampling point	Cu	Zn	Pb	Cr	Cd	As	Hg
A10	0.23	0.25	0.41	0.17	1.93	0.74	0.13
B9	0.78	0.98	0.71	0.54	3.83	0.98	0.14
D7	0.83	0.93	0.84	0.52	3.15	0.59	0.16
D11	0.80	0.86	0.35	0.49	1.75	0.73	0.15
E8	0.62	0.54	0.41	0.33	1.78	1.61	0.22
F5	0.75	0.78	1.07	0.46	1.85	0.49	0.22
F9	0.55	0.48	0.61	0.36	1.70	0.32	0.13
G2	0.91	0.82	0.79	0.53	3.70	0.32	0.07
G8	0.89	0.72	0.64	0.47	0.45	0.55	0.06
H5	0.71	0.82	0.54	0.36	1.88	0.60	0.05
I2	0.41	0.47	0.71	0.26	1.43	0.48	0.03
Mean value	0.68	0.69	0.64	0.41	2.13	0.67	0.12

Note: Level-1 natural background value of soil environment quality standard is normal in China.

metal, C_s^i is the measured concentration of the i^{th} heavy metal, C_n^i is the reference value of the i^{th} heavy metal using the pre-industrial background values of heavy metals in sediments, and T_r^i is the toxicity coefficient of the i^{th} heavy metal. The heavy metal toxicity levels and biological sensitive degrees of heavy metal pollution are presented according to relevant data (Gong et al. 2006, Yang et al. 2005) and the pollution characteristics of heavy metals. Thus, we set up the numerical order of biological toxicity response factor of the seven heavy metals: Hg (40) > Cd (30) > As (10) > Cu (5) = Pb (5) > Cr (2) > Zn (1). Table 5 gives the PER assessment index caused by heavy metals in sediment pollution and its ecological risk classification (Chen & Zhou 1992).

We calculated the results according to equation 3 and based on the high background reference values of heavy metals in sediments before the modern industrialization (Table 6). We also calculated the results according to the level-1 natural background value of the "soil environment quality standard" (GB 15618-1995) in our country (Table 7).

We determined from Table 6 that E_r^i of a single heavy metal is smaller than the average value 40, which belongs

to the slight pollution level. The PER of Cd pollution is relatively serious with an average value of 25.55. Moreover, the Cd value at B9 and G2 were 45.90 and 44.40, which belong to the medium pollution level. Using the PER indices of the seven heavy metals for evaluation, we identify that the index value ranged from 24.30 to 71.42, which belong to the low pollution level.

Table 7 shows that the enrichment of the seven heavy metals was Cd > Zn > Cu > As > Pb > Cr > Hg. Cadmium has the most serious risk potential with a mean value of 63.89, which belongs to the secondary pollution level. The E_r^i value of Cd exceeds 80 at B9 and G2, which belong to the strong ecological damage level; the values of the other heavy metals were less than 40. In terms of ecological damage index, the RI value of each sample point was less than 150, which indicates a low degree of risk. The risk of all sampling points was B9 > G2 > D7 > E8 > F5 > A10 > D11 > H5 > F9 > I2 > G8.

CONCLUSIONS

The pollution level order size of Cu, Zn, Pb, Cd, Cr and Hg at the surface of the sediment was Zn > Cr > Cu > Pb > Cd >

Table 5: Correlation between index of PER and grade I.

Potential ecological risk factor		The potential ecological risk index	
The threshold range of single metal	The level of risk factors	The threshold of the 6 metals	The level of risk index
$E_r^i < 40$	I Ecological risk slightly	$RI < 150$	A, low
$40 \leq E_r^i < 80$	II Ecological risk secondary	$150 \leq RI < 300$	B, secondary
$80 \leq E_r^i < 160$	III Ecological risk		
$160 \leq E_r^i < 320$	IV Ecological risk is strong	$300 \leq RI < 600$	C, high
$E_r^i \geq 320$	V Ecological risk is very strong	$RI \geq 600$	D, very high

Table 6: Potential ecological risk (PER) coefficients and indices of heavy metals in the sediment of Hulun Lake.

Sampling point	E_r^i							Ri
	Cu	Zn	Pb	Cr	Cd	As	Hg	
A10	1.31	0.31	2.88	0.50	23.10	7.40	3.00	38.51
B9	4.55	1.22	5.00	1.62	45.90	9.79	3.34	71.42
D7	4.83	1.16	5.87	1.57	37.80	5.92	3.84	60.98
D11	4.68	1.08	2.48	1.48	21.00	7.26	3.62	41.60
E8	3.63	0.68	2.90	1.00	21.30	16.12	5.21	50.84
F5	4.38	0.97	7.51	1.39	22.20	4.94	5.39	46.77
F9	3.23	0.60	4.29	1.09	20.40	3.17	3.06	35.83
G2	5.32	1.02	5.51	1.60	44.40	3.23	1.57	62.66
G8	5.21	0.90	4.48	1.40	5.40	5.47	1.44	24.30
H5	4.14	1.02	3.80	1.07	22.50	5.97	1.14	39.65
I2	2.40	0.59	4.95	0.79	17.10	4.84	0.66	31.33
Mean value	3.97	0.87	4.51	1.23	25.55	6.74	2.93	45.81

Note: Reference of heavy metal content about the highest background value in normal granular sediments of modern pre-industrial.

Table 7: Potential ecological risk (PER) coefficients and indices of heavy metals in the sediment of Hulun Lake.

Sampling point	E_r^i							Ri
	Cu	Zn	Pb	Cr	Cd	As	Hg	
A10	1.13	0.25	2.06	0.33	57.75	7.40	5.00	73.92
B9	3.90	0.98	3.57	1.08	114.75	9.79	5.56	139.63
D7	4.14	0.93	4.19	1.04	94.50	5.92	6.40	117.13
D11	4.01	0.86	1.77	0.99	52.50	7.26	6.04	73.43
E8	3.12	0.54	2.07	0.67	53.25	16.12	8.69	84.45
F5	3.76	0.78	5.36	0.92	55.50	4.94	8.98	80.24
F9	2.77	0.48	3.06	0.73	51.00	3.17	5.09	66.30
G2	4.56	0.82	3.94	1.07	111.00	3.23	2.62	127.24
G8	4.47	0.72	3.20	0.94	13.50	5.47	2.40	30.69
H5	3.55	0.82	2.72	0.72	56.25	5.97	1.90	71.92
I2	2.06	0.47	3.54	0.53	42.75	4.84	1.10	55.29
Mean value	3.40	0.69	3.22	0.82	63.89	6.74	4.89	83.66

Note: Reference of level-1 natural background value of the soil environment quality standard in China.

Hg. However, the content of the seven heavy metals was generally lower than the normal value content in sediment particles, which is the heavy metal high background value before modern industrialized. Except for Cd, the content of the other heavy metals was below the level-1 natural background value of the soil environmental quality standards in our country.

The heavy metals in surface sediment distribution had no obvious characteristics. However, Zn, Cr and Cu contents varied coincidentally, whereas their distributions presented a certain regularity. The contents on the northeast (B9, D7, D11) and southwest (F5, G2, G8, H5) end of the lake were high, whereas the contents on the Xinkai River (A10), Wuerxun River (F9), and Kerulen (I2) at entrance of the lake were low.

The individual PER coefficient and evaluation comprehensive index results show that the potential ecological damage of heavy metals in the surface sediments in Hulun Lake were mild and moderate. Only Cd exhibited a potential risk, especially in the northeast end of the lake (B9) and the lakeside (G2).

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