Journal of
Applied Sciences

ISSN 1812-5654

Journal of Applied Sciences 15 (4): 682-688, 2015
ISSN 1812-5654

ansinet
Asian Network for Scientific Information

RESEARCH ARTICLE OPEN ACCESS

DOI: 10.3923/jas.2015.682.688

Distribution Features and Ecological Risk Assessment of Heavy Metals in Superficial Sediments of Hulun Lake

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ARTICLE INFO
Article History:
Received: April 07, 2014
Accepted: February 23, 2015

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ABSTRACT
Concentrations, distribution and accumulation of heavy metals in the sediments of Hulun Lake were investigated. The highest value for heavy metals concentration in sediments before the modern industrialization of the world and grade criteria of the National Standard for soil environmental quality were used to analyze the accumulation coefficient and Potential Ecological Risk (PER) coefficients for heavy metal. The PER index for each sample location was also discussed. The method for evaluation PER index presented by Hakanson was applied to assess the ecological risk. The results showed that the distribution of seven kinds of heavy metals content didn't reveal the regional distributive characteristics and the differential of space distribution was small. However, Zn, Cr and Cu have the same trend, that the concentrations are higher in northeast and southwest parts of Hulun Lake than those at the entrance of lake for Xinkai River, Wuerxun River and Kerulen River. When the highest value for heavy metals concentrations in sediments before the modern industrialization of the world were taken as reference, the accumulating order of these heavy metals is: Pb>Zn>Cd>Cu>As>Cr>Hg and the order of pollution level is Cd>As>Pb>Cu>Hg>Cr>Zn but most of them were in the low risk states. When the grade criteria of the National Standard for Soil Environmental Quality worked as reference, the accumulating order is Cd> Zn > Cu >As>Pb>Cr>Hg and the order of pollution level is Cd>As> Hg >Cu> Pb >Cr>Zn. Cadmium (Cd) was the potential impact element of the ecological environment of Hulun Lake.

Key words: Hulun lake sediment heavy metals ecological risk assessment

INTRODUCTION

The lake sediment is the main accumulation place of the lake water body pollution and is also the potential pollution sources of lake (Milenkovic *et al*., 2005; Liu *et al*., 2006). The heavy metals were adsorbed by suspended solids in water and eventually deposited to sediments what on the lake surface water body (Fan *et al*., 2002). The pollutants can re-enter overlying water again, because of the water-sediment interface under the action of a series of biogeochemical process (Zhou *et al*., 2005). The heavy metals in sediment are considered as sensitive indicator of water pollution and had the significance of reflection the status of water system (Yang *et al*., 2005). Through bioaccumulation and amplification effect, it becomes a direct and an indirect threat to the ecosystem. Therefore, according to the research of the content of heavy metals in sediment and its distribution, we could evaluate the harmful of heavy metals to the potential ecological and determine the main pollutant, to understand the impact of heavy metals on water quality, that have the important practical significance and at the same time can provide the basis for water pollution control.

Hulun lake, also known as Dalai lake, located in the west of Hulunbeier Grassland, between Xin Barag Right Banner, Xin Barag Left Banner and Manchuria, east longitude 117 °00 ‘10"~117° 41 ’40”, latitude 48° 30 ‘40”~49 °20 ‘40”. The lake shows irregular oblique rectangle, shaft to the northeast to southwest, length of 93 km, maximum width is 41 km, circumference of the lake is 447 km, the lake covers an area of 2339 km^2, the average water depth is 5.7 m, the maximum depth is 10 m, the total water content is 13.85 billion cubic metres. In 1992, Hulun Lake was approved as the national wetland nature reserve, its water and wetlands play an irreplaceable role in Hulunbeier Grasslands' ecological protection and economic development (Yue *et al*., 2008). In recent 40 years, due to the impact of climate change and human activities, the lake had to face a series environment problems: the water level decreased, the water area reduced year by year, wetlands declined, the surrounding ecological environment and serious water quality deterioration, the lake total salt content and pH value increased year by year, large area of reed lake disappeared, fishery resources dried up and a large number of rare birds migrated. Currently, the water quality of Hulun lake is in a medium eutrophication level (Han and Chi, 2002), wetland ecological environment is deteriorated sharply, this has become a serious threat to the security of northeast and north China region's ecological.

However, little information has been done on the study of Hulun lake. The researchers only focus on the investigation or analysis the lake water quality, water quantity and water level and their aims at the following aspect: How the climate change impact on regional wetland ecological status and the environmental governance (Li *et al*., 2006; Wang, 2005; Zhao *et al*., 2008). Based on Hulun Lake's sediments, the researchers studied the heavy metal elements, such as Cu, Zn, Pb, Cr, Cd, As, Hg, including their distribution and enrichment characteristics. According to their studies, we could know that the lake's sediment pollution status and the characteristics of the pollutant distribution. Besides, the study using the potential ecological harm index evaluated the heavy metals' harmful. This thesis aims at providing a scientific basis for Hulun lake basin water environment quality comprehensive evaluation and water pollution control.

MATERIALS AND METHODS

Sampling points set: According to the characters of the lake area size, shape, water flow direction and the river into the lake, setting up 11 sediment sampling points in lake. The points' locations are showed in Fig. 1. The study, since December 2008, using the Global Positioning System (GPS) to fixed position and the columnar sampler, collected 21 samples of the surface sediment above 15 cm. The samples were collected in winter in the frozen lake by ice drill and then sealed into the polyethylene plastic bags, cryopreservation at once after back to the lab.

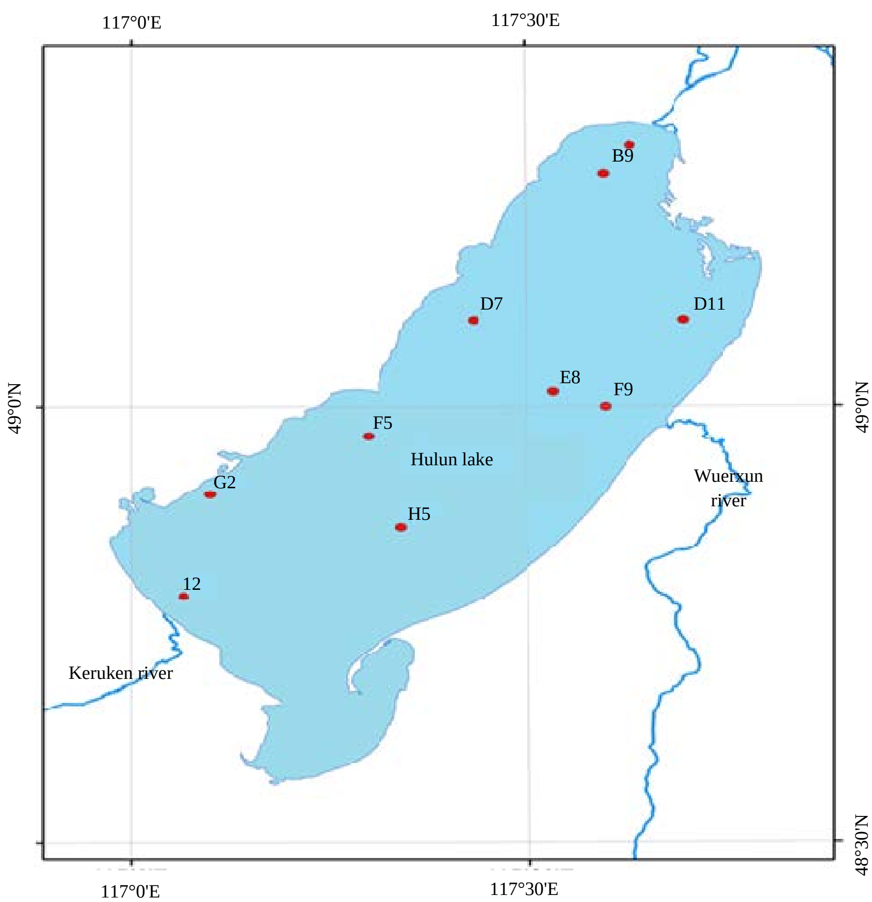

Fig. 1: Distribution of sampling sites for the sediment of Hulun lake

Analysis method: Samples were crushed by the glass rod after natural drying in the laboratory, removing gravel, shells, animal and plant residues and other impurities, then through 100 mesh sieve mortar after grinding, analytic determination the heavy metal elements: Cu, Zn, Pb, Cd, Cr, As and Hg. The contents of the heavy metals element in the sediment samples meter aged after the samples digest by HNO_3-HF-$HClO_4$, then Cu, Zn, Pb, Cd, Cr and As use the flame atomic absorption spectrophotometry to analysis determination, Hg use the cold atomic absorption spectrophotometry. All samples menstruated by Z-8000 atomic absorption spectrophotometer.

Potential ecological risk assessment of heavy metals: There are many methods to evaluate the heavy metals pollution in sediments, this paper adopts the ecological hazard index method which was proposed by Swedish scientists Hakanson. This method using relative to the heavy metals in sediments and sediment before the industrialization the highest concentration of heavy metals pollution degree and the corresponding values of ecological toxicity coefficient weighted sum, getting the ecological hazard index. The characteristic of this method is starting from the biological toxicity of heavy metals, not only reflects each pollutant's influence of a particular environment but also reflects the synthesis influence of a variety pollutants. And from using the method of quantitative we could differentiate the harmful degree to potential ecological caused by heavy metals pollution in sediments (Jiang *et al*., 2008; Xiang *et al*., 2006).

RESULTS AND DISCUSSION

Content and distribution of heavy metals in sediment

Difference of heavy metals' content in space: From the statistical results of several kinds of heavy metal elements in Table 1, it can be concluded that: The content value of Zn is the highest, the mean value is 68.80 mg kg^{-1}, Cr, Cu, Pb lower than Zn and the mean content of each is 36.37, 23.44 and 22.34 mg kg^{-1}, the content value of As is in a middle level with a value of 10.36 mg kg^{-1}, the content value of Cd is relatively small, only 0.41 mg kg^{-1}, the content value of Hg is the lowest with a value of 0.019 mg kg^{-1}. From the variation coefficient about the seven kinds of heavy metal, Cd has the largest space variation coefficient, at 68.30%, because of the content of Cd in sediment on B9 point which is near the entrance of the northeast Xingkai River (0.78 mg kg^{-1}) is much higher than other sample points. At the same time, As and Hg have a larger space variation coefficient (except Cd), at 57.84%, 56.39% respectively, the main reason is that the content of As (24.18 mg kg^{-1}) at the E8 points is much higher than other points and the content of Hg (0.002 mg kg^{-1}) in Kerulen estuary (I2 point) is far lower than the other points. Followed by Pb, Zn, Cu and Cr, the variation coefficient was 30-44%. In a word, the spatial distribution difference of seven heavy metals in lake sediment is relatively small and besides Cd, the six other kinds of heavy metals content is below the grade criteria of the National Standard for Soil Environmental Quality.

Heavy metals content characteristics of regional distribution: From Table 2, the correlation coefficient between heavy metals for the sediment of Hulun lake, we can conclude that the seven different heavy metals content in sediment, Cu, Zn, Cr has extremely significant correlation relationship between the two. So, the Hulun Lake's all sampling points has a consistent content change trend of Zn, Cr, Cu. Namely, the northeast end of the lake (B9, D7, D11 point) and southwest (F5, G2, the G8, H5 point) content on the high side while the Xinkai River (A10 point), Wuerxun River (F9 point) and Kerulen (I2 point) at the entrance to the lake content is on the low side but the regional distribution is not obvious. In addition, As and Hg also has a significant correlation relation but the content of the As is far higher than that of Hg. The content of Hg is rarely, so it could be negligible. According to the Fig. 2, the content order of six kinds heavy metals in the surface of sediment in Hulun Lake from more to less is: Zn> Cr > Cu > Pb > As > Cd, at B9 point the Cr, Zn and Cd has the highest level, at F5 point Pb has the highest level, at E8 point the As has the highest level, Cu at G2 point has the highest level.

Enrichment of heavy metals in sediment: Usually adopt the enrichment coefficient to measure the enrichment degree of single heavy metal, the enrichment coefficient can be represented as Eq. 1:

$$C_f^i = C_m^i / C_n^i \tag{1}$$

where, C_m^i is the measured content value of heavy metal i in sediment and C_n^i is the required ratio for calculating (environmental background value).

This study uses the highest background value of heavy metal content in normal particles sediment before the modern industrialization, the high background values of w (Hg),

Table 1: Statistic values of heavy metals for the sediment of Hulun lake (n = 21)

Metallic elements	Minimum value	Maximal value	Mean value	Standard deviation	Coefficient of variation (%)
	(mg kg^{-1})				
Cu	3.390	31.920	23.440	7.42	31.67
Pb	4.450	55.290	22.340	9.78	43.77
Cr	9.330	50.760	36.370	10.92	30.02
Zn	10.200	105.600	68.800	24.56	35.69
Cd	<0.010	0.780	0.410	0.28	68.30
As	4.260	31.550	10.360	5.99	57.84
Hg	0.002	0.039	0.019	0.01	56.39

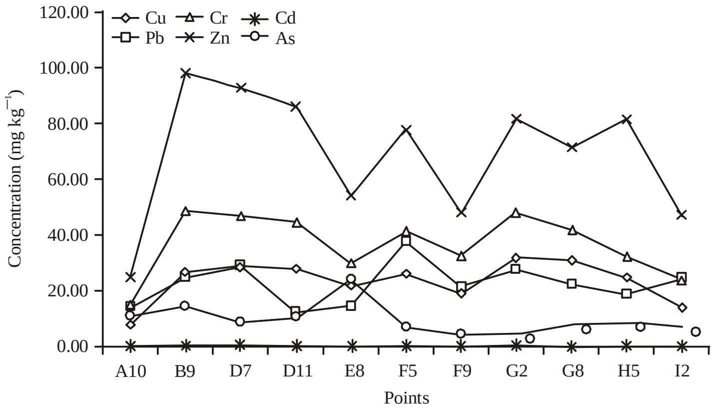

Fig. 2: Distribution of heavy metals concentrations in each sample for the sediment

Table 2: Correlation coefficient between heavy metals for the sediment of Hulun lake(1)

Elements	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	0.030	0.191	0.086	0.492*

Significant correlation *$p<0.05$, Very significant correlation **$p<0.01$. (Two-tailed test)

w (As), w (Cu), w (zn), w (Pb), w (Cd), w (Cr) were 0.25, 0.25, 30.00, 80.00, 25.00, 80.00 and 25.00 mg kg^{-1}, respectively. According to the high background values we could know the real pollution level of the lake. Due to lack of the lake surrounding soil environment background values of the seven kinds of heavy metal, Hulun Lake belongs to the national wetland nature reserve, so, reference the grade criteria of the National Standard for Soil Environmental Quality, the natural background values of w (Hg), w (As), w (Cu), w (zn), w (Pb), w (Cd), w (Cr) were 0.15, 0.15, 35.00, 100.00, 35.00, 100.00 and 35.00 mg kg^{-1}, respectively ,they reflected the relative pollution degree of Hulun Lake. Combined the two can better response the lake's potential ecological damage. According to the Eq. 1 we could calculate the heavy metal enrichment coefficient of each sample point can be calculated, the results are shown in Table 3 and 4.

From the enrichment coefficient and reference the heavy metal content about the highest background values in normal granular sediment of modern pre-industrial, we could conclude that: Pb enrichment degree is the highest at an average enrichment coefficient of 0.90, followed by Zn, Cd, Cu, As, Cr and Hg is the lowest with an enrichment coefficient only 0.073 on average. If reference the grade criteria of the National Standard for Soil Environmental Quality, Cd enrichment degree is the highest at an average enrichment coefficient of 0.90, followed by Zn, Cu, As, Pb and Hg is the lowest. In terms of the sampling points, Zn, Cr, Cd have the highest accumulation degree at the B9 point (the northeast end of the lake), Pb and Hg have the highest accumulation degree at the F5 point (the northwest edge of the lake), As has the highest accumulation degree at the E8 point (close to the lake central), Cu has the highest accumulation degree at the G2 point (the southwest edge of the lake). In brief, the concentration degree is generally low at the three river estuary place, namely at A10, F9 and I2 points.

Table 3: Enrichment coefficients of heavy metals in sediment of Hulun Lake(1)

Sampling poin	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

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

Table 4: Enrichment coefficients of heavy metals in sediment of Hulun Lake(2)

Sampling poin	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

2 Grade I criteria of the National Standard for Soil Environmental Quality worked as references

Assessment of potential ecological risk caused by heavy metals: In 1980, the Swedish scientists Hakanson proposed the potential ecological risk index method to assess the ecological risk assessment of heavy metals (Caeiro *et al*., 2005). The potential ecological risk assessment is based on the principle of element abundance and releasing ability, the hypothesis of the evaluation consist of the following conditions: (1) The response of the element abundance, namely, the potential ecological risk index (risk index, RI) increases with metal pollution in sediments, (2) The synergy of many pollutants is that the metals in sediment obey the additive property in ecological harm. A variety of metals pollution has a bigger potential ecological risk, the heavy metal such as Cu, Zn, Pb, Cd, Cr is preferred and (3) The heavy metals has different toxicity response, the metal of strong biological toxicity has higher weights to the RI. The potential ecological harm index (RI) can reflect the four aspects: (1) The concentration of the metal in surface sediment, (2) Species number of metal pollutants, (3) The level of metals toxicity and (4) The sensitivity of the water body to the metal contamination (Huang *et al*., 2008).

According to this method, E_r^i represents the individual potential ecological harm coefficient of the No. i heavy metal in sediment of an area and it can be expressed by Eq. 2, RI represents the composite index of a variety of heavy metals in sediments and it can be expressed by Eq. 3:

$$E_r^i = T_r^i C_f^i = T_r^i C_s^i / C_n^i \tag{2}$$

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i C_r^i = \sum_{i=1}^{n} T_r^i C_s^i / C_n^i \tag{3}$$

Where:
- C_f^i = Enrichment coefficient of No.i heavy metal
- C_s^i = Measured concentrations of No.i heavy metal
- C_n^i = Reference value of No.i heavy metal, using the pre-industrial background values of heavy metals in sediment
- T_r^i Toxicity coefficient of the No.i heavy metal. It mainly reflects the heavy metal toxicity levels and biological sensitive degree of heavy metal pollution, according to the relevant data (Gong *et al*., 2006; Yang *et al*., 2005) and the pollution characteristics of heavy metals, so, set up the numerical order of biological toxicity response factor of 7 kinds of heavy metal: Hg (40) > Cd (30) > As (10) > Cu (5) = Pb (5) > Cr (2) > Zn (1). Table 5 shows the potential ecological risk assessment index caused by heavy metals in sediment pollution and its ecological risk classification (Chen and Zhou, 1992).

According to Eq. 3 and based on the high background reference values of heavy metals in sediment before the modern industrialization, we can calculate the results and showed in Table 6. If based on the grade criteria of "the National Standard for Soil Environmental Quality", we can calculate the results and showed in Table 7.

From Table 6 and reference the heavy metal content about the highest background value in normal granular sediments of modern pre-industrial, we could know that the E_r^i of a single heavy metal is smaller than the average value 40, belong to slight pollution level, the potential ecological risk of Cd pollution is relatively serious and its average value is 25.55, besides, the value of Cd at B9 point and G2 point were 45.90 and 44.40, belong to the medium pollution level. Considering seven kinds of heavy metals, using their potential ecological risk index to evaluate, the index value range from 24.30-71.42, all belong to low pollution level.

From Table 7 and reference the grade criteria of the National Standard for Soil Environmental Quality, the enrichment of seven kinds of heavy metals obey this order: Cd> Zn > Cu >As>Pb>Cr>Hg, Cd is the most serious heavy

Table 5: Correlation between index of potentially ecological risk and grade

Potential ecological risk factor		Potential ecological risk index	
Threshold range of single metal	Level of risk factors	Threshold of the 6 metals	Level of risk index
E_r^i <40	I Ecological risk slightly	R I<150	A low
40≤ E_r^i <80	II Ecological risk secondary	150≤R I<300	B secondary
80≤ E_r^i <160	III Ecological risk		
160≤ E_r^i <320	IV Ecological risk is strong	300≤RI<600	C high
E_r^i ≥320	V Ecological risk is very strong	R I≥600	D very high

Table 6: Potential ecological risk coefficients and indices of heavy metals in the sediment of Hulun Lake(1)

	E_r^i							
Sampling point	Cu	Zn	Pb	Cr	Cd	As	Hg	Ri
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

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

Table 7: Potential ecological risk coefficients and indices of heavy metals in the sediment of Hulun Lake[2]

	E_r^i							
Sampling point	Cu	Zn	Pb	Cr	Cd	As	Hg	Ri
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

2 Grade I criteria of the National Standard for Soil Environmental Quality worked as references

metal risk to the potential ecological, the mean value is 63.89, belong to secondary pollution level. Especially at B9 point and G2 point, the E_r^i value of Cd is more than 80,belong to strong ecological damage level and other heavy metals value are far less than 40. In terms of ecological damage index, the RI value of each sample point is less than 150, in a low degree of risk, the risk of all sampling points to the ecology obey the following order: B9>G2>D7>E8>F5>A10>D11>H5>F9>I2>G8.

CONCLUSION

The pollution levels order size of the Cu, Zn, Pb, Cd, Cr, Hg at the surface of sediment is: Zn > Cr > Cu > Pb > Cd > Hg but on the whole, the content of the seven kinds of heavy metals were lower than the normal value content in sediment particles which is the heavy metal high background value before modern industrialized, in addition to the Cd, the content of other heavy metals are below the grade criteria of the National Standard for Soil Environmental Quality.

The heavy metals in surface sediment distribution have no obvious characteristics but the content of Zn, Cr and Cu is variation coincidently and their distribution presents certain regularity. Namely, the northeast end of the lake (B9, D7, D11 point) and southwest (F5, G2, the G8, H5 point) content on the high side while the Xinkai River(A10 point), Wuerxun River (F9 point) and Kerulen (I2 point) at the entrance to the lake content is on the low side.

The individual potential ecological risk coefficient and the evaluation comprehensive index results showed that the heavy metals in surface sediment in Hulun Lake’s potential ecological damage are mild and moderate. Only Cd has the potential risk, especially in the northeast end of the lake (B9 point) and the lakeside (G2 point).

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