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Heavy Metal Contamination in the Surface Sediments of Representative Limnetic Ecosystems in Eastern China

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A comprehensive analysis of heavy metal pollution was conducted in the representative limnetic ecosystems of eastern China, which are subject to rapid economic development and population growth. The results demonstrated that the average contents with standard deviations of Cd, Cr, Cu, Ni, Pb and Zn in the surface sediments were 0.925 ± 0.936 , 142 ± 46.8 , 54.7 ± 29.1 , 60.5 ± 21.6 , 61.9 ± 36.0 and 192 ± 120 mg/kg dry wt., respectively, and that higher values were mainly observed in the southern portion of the study area, especially in the basins of Southeast Coastal Rivers (SCRB) and the Zhu River (ZRB). The six heavy metals in the surface sediments all had anthropogenic origins. In addition, the limnetic ecosystems, especially in the southern portion of the study area were found to be polluted by heavy metals, especially Cd. Overall, two hotspots of heavy metal pollution in the limnetic ecosystems of eastern China were found, one that consisted of the heavy pollution regions, SCRB and ZRB, and another composed of Cd pollution. These results indicate that heavy metal contamination, especially Cd, should be taken into account during development of management strategies to protect the aquatic environment in the limnetic ecosystems of eastern China, especially in the two aforementioned basins.

Heavy metal pollution in aquatic ecosystems is a worldwide environmental problem that has received increasing attention over the last few decades because of its adverse effects^{1–5}. The contamination of aquatic systems by heavy metals, especially in sediments, has become one of the most challenging pollution issues owing to the toxicity, abundance, persistence, and subsequent bio-accumulation of these materials^{6,7}. When discharged into aquatic ecosystems, heavy metals can be absorbed by suspended solids, then strongly accumulated in sediments and biomagnified along aquatic food chains^{8,9}. Moreover, these sediments act as sinks, and may in turn act as sources of heavy metals^{10,11}. Thus, heavy metal pollution in the sediments of aquatic ecosystems has recently been extensively investigated to effectively manage these ecosystems^{6,12–14}.

In China, environmental problems owing to heavy metal pollution of sediments received little attention from researchers and governments prior to 2000^{4,15}. Recently, anthropogenic activities for economic development, especially by industry have intensified continuously and rapidly, particularly in eastern China. Consequently, one-sixth of all arable lands in China have been contaminated by heavy metals¹⁶, and heavy metal pollution is becoming increasingly serious and has been extensively investigated in the sediments of limnetic ecosystems (river, lake, wetland, etc.) in eastern China^{4,6,8,17}. Although extensive investigations of heavy metals in sediments have been carried out in the study area, these studies have mainly focused on individual ecosystems, and there have been few investigations of whole limnetic ecosystems in this area.

Therefore, this study was conducted to evaluate heavy metal pollution status in the sediments of limnetic ecosystems located in eastern China, including rivers, lakes and wetlands. The specific goals of this study were: 1) to investigate heavy metal contents (Cd, Cr, Cu, Ni, Pb and Zn) in the surface sediments; 2) to determine if they were of natural or anthropogenic origin based on enrichment factor (EF), Pearson correlation analysis (CA), hierarchical cluster analysis (HACA), and principal components analysis (PCA); and 3) to assess the risk associated with the six heavy metals by consensus-based sediment quality guidelines (SQGs), potential ecological risk (PER) index, geo-accumulation index (I_{geo}), and pollution load index (PLI).



Results and Discussion

Properties and nutrient contents of surface sediments. As shown in Figure S1, the pH of surface sediments in the studied ecosystems ranged from 5.23 to 8.94 with an average of 7.73, higher than the soil background value (pH = 6.70)¹⁸. Additionally, the OM ranged from 0.551% to 16.6% with an average of 5.33, while the TC, TN, TS and TP contents varied from 0.124%, 0.009%, 0.010% and 0.018% to 13.5%, 0.967%, 0.994% and 0.293%, respectively, with an average of 1.87%, 0.143%, 0.123% and 0.057%. The TC and TN contents had basically the same spatial distribution patterns, with the highest values being observed in the surface sediments from HaRB, HuRB and YaRB (TC: 2.85%, 2.44% and 1.84%; TN: 0.230%, 0.162% and 0.140%, respectively). The TS and TP contents were highest in HaRB (0.196% and 0.084%, respectively).

Heavy metal contents of surface sediments. The heavy metal contents of surface sediments are shown in Figure 1, and comparative analyses of the threshold effect concentration (TEC), probable effect concentration (PEC), and background value (BV) are presented in Table S1. The average contents with standard deviations of Cd, Cr, Cu, Ni, Pb and Zn across the basins were 0.925 ± 0.936 , 142 ± 46.8 , 54.7 ± 29.1 , 60.5 ± 21.6 , 61.9 ± 36.0 and 192 ± 120 mg/kg dry wt., respectively. In general, the contents of these metals in the sediments of the study area were at the medium level compared with those reported from other areas^{12,19,20}. Spatially, heavy metal contents were higher in the southern portion of the study area than in the north. The average Cd contents were highest in the surface sediments of SCRB (2.57 mg/kg dry wt.), ZRB (1.48 mg/kg dry wt.) and YaRB (1.38 mg/kg dry wt.), while the average Cr and Zn contents were highest in ZRB (161 and 417 mg/kg dry wt., respectively), the Cu and Pb contents were highest in SCRB (113 and 147 mg/kg dry wt., respectively) and the average Ni contents were highest in HuRB (70.7 mg/kg dry wt.).

Upon analysis of all sediment samples, the percentages of Cd, Cr, Cu, Ni, Pb and Zn contents exceeding the PEC values were 0.0%, 79.1%, 1.1%, 68.6%, 3.5% and 7.0%, respectively, indicating that adverse effects caused by Cr and Ni are expected. When compared with the BV, the percentages exceeding the values were all over 90.0% (Table S1), indicating that the heavy metals all had different degrees of accumulation in the sediments of the study area.

Heavy metal sources of surface sediments. The EF values of all analyzed metals were calculated to evaluate anthropogenic influences on heavy metals in the surface sediments of the study area. The spatial distribution and basic statistics for the EF values are shown in Figure 2 and Figure S2. An EF value between 0.5 and 1.5 suggests that the metal may be entirely from crustal materials or natural weathering processes. However, an EF greater than 1.5 suggests that a significant portion of the metal originated from anthropogenic processes^{6,21–23}. The EF values of Cd were highest in this study, reaching 7.92 on average, indicating a high degree of anthropogenic contamination by this metal, mainly from industrial activities. The mean values of Zn, Pb, Cu, Cr and Ni were 2.25, 2.06, 2.04, 2.02 and 1.95, respectively, indicating that they also originated from anthropogenic sources in most samples.

Various multivariate techniques such as CA, HACA and PCA have been shown to be useful for identification of sources of heavy metals⁶. In the present study, a high positive correlation ($p \leq 0.01$) was observed among heavy metals (except Ni and Zn, $p \leq 0.05$) (Table S2), as well as among C, N and S. However, no significant correlation was observed between P and other elements. Similar to the results of CA, the studied elements could be roughly clustered into three primary groups obtained by HACA (Figure 3). The first cluster consisted of C, N and S, the second included the six heavy metals and the third contained only P. In addition, three major components representing different levels of anthropogenic input were extracted from the six heavy metals. The first component

included Cr and Ni, the second Cd, Pb and Cu, and the last one Zn. PCA was also applied to identify sources of these anthropogenic components. The relationships among the studied elements based on the three principal components were illustrated in three-dimensional (3D) space (Figure S3), and the results agreed with those of CA and HACA. Overall, these findings indicate that the six heavy metals in the surface sediments of the study area all had anthropogenic origins. Moreover, the degrees of enrichment of the heavy metals were as follows: Cd > Zn > Pb > Cu > Cr > Ni, and these metals could be divided into three sources.

Risk associated with heavy metals. The risks associated with the studied metals in the surface sediments of the study area were assessed by consensus-based SQGs, PER index, I_{geo} and PLI. With the exception of the HeRB and LRB, the mean PEC quotient (Q_{m-PEC}) of the surface sediments from HaRB, YeRB, HuRB, YaRB, SCRB and ZRB were larger than 0.5 (0.662, 0.528, 0.666, 0.741, 0.879 and 0.846, respectively), indicating that these sediments were toxic and highly contaminated by heavy metals (Figure 4).

PER represents the sensitivity of the biological community to a given substance and illustrates the risk posed by contamination^{4,6}. In the surface sediments of the studied limnetic ecosystems, the sequence of E'_r was Cd > Zn > Cu > Pb > Ni > Cr, and the studied metals all showed low PER with the exception of Cd, Cu, Pb and Zn, which were present in 87.2%, 1.2%, 2.3% and 2.3% of the sediment samples, respectively (Table S3). The E'_r values of Cd ranged from 13.9 to 1389 with an average of 286, indicating high Cd contamination of the sediments in the study area. These findings were consistent with the results obtained from EF, as well as those of different aquatic ecosystems in this area²⁴. The mean RI values of the surface sediments were 175, 122, 244, 168, 205, 487, 879 and 534 in HeRB, LRB, HaRB, YeRB, HuRB, YaRB, SCRB and ZRB, respectively, with an overall average of 339 (Figure 4). According to the PER index (Table S4), many stations in the SCRB and ZRB had high and serious contamination, primarily as a result of Cd pollution (Figure 4). This may have been owing to mining and metallurgical industry development in these basins. The distribution plots of RI developed in the present study could be useful in identifying stations that need the most attention in the study area.

To further examine the heavy metal pollution in the surface sediments, the I_{geo} of each heavy metal was determined (Figure S4 and Figure S5). The mean I_{geo} values of Cd, Cr, Cu, Ni, Pb and Zn were 1.97, 0.559, 0.497, 0.483, 0.482 and 0.578, respectively. These findings indicate that the sediments were moderately polluted with Cd, while they were unpolluted to moderately polluted with the other heavy metals (Table S4). Spatially, the degree of Cd pollution in the surface sediments of the SCRB and ZRB were all strongly polluted ($I_{geo} = 4.00$ and 2.78, respectively), which is in accordance with the Q_{m-PEC} and RI values. In addition, the results were further confirmed by the PLI, and the values exceeded one at over 95.0% of the stations indicating that heavy metal pollution existed in the study area, especially in the SCRB and ZRB (Figure 4 and Table S4).

Heavy metals are widespread and persistent in aquatic ecosystems, potentially toxic, and can become incorporated into food webs^{11,25}. Sediments contaminated with heavy metals have been found to be via several pathways, including atmospheric deposition, industrial activities and agricultural activities^{26,27}. In the current study, heavy metals, especially Cd, all showed different degrees of enrichment in the surface sediments (Figure 2), which has resulted in toxicity, PER, and pollution (Figure 4 and Figure S4). Moreover, economic development and population growth in the study area have influenced the distribution of heavy metals in the sediments, especially in the HaRB, which is the most developed region and has the highest population density in eastern China (Figure 1 and Figure S6)¹⁶. However, mining and metallurgical industries were found to be the dominant factors responsible for heavy metal enrichment, especially

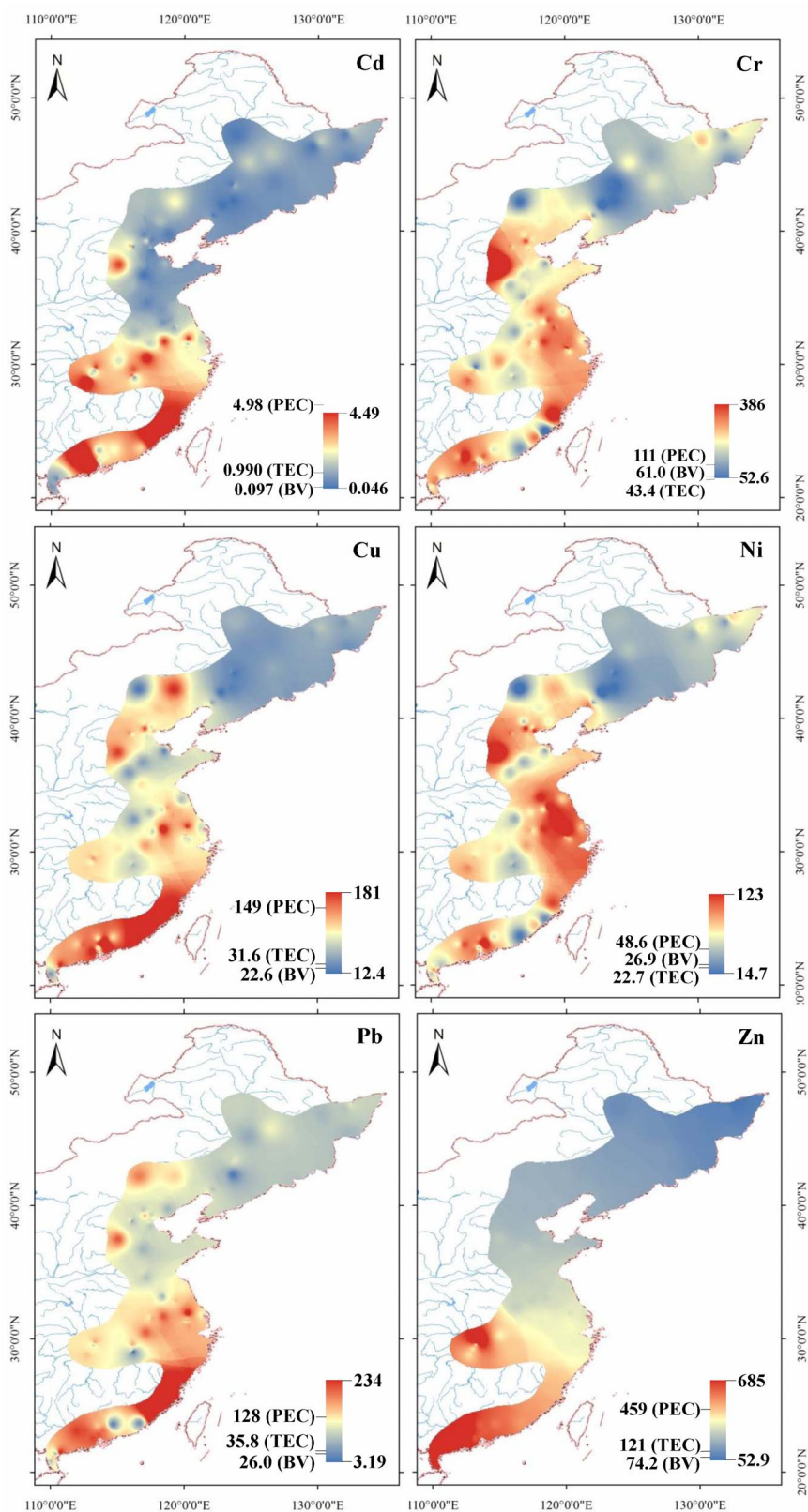


Figure 1 | Heavy metal contents (mg/kg) of surface sediments (ArcGIS 10.0). PEC, probable effect concentration; TEC, threshold effect concentration; BV, background value.

in the SCRB and ZRB. Taken together, these results indicate that heavy metal pollution of sediments, particularly Cd, has become a major environmental problem in the limnetic ecosystems of eastern China, which is consistent with the results of other studies^{4,14,16,21}. Moreover, sediment contaminants may move into the food chain,

and heavy metals in the sediments can accumulate in relatively large amounts in aquatic organisms without any apparent effects, resulting in a threat to human health^{28–30}. Accordingly, it is important that heavy metal contaminants, especially Cd, be carefully monitored in the study area. The information obtained in this study could be useful

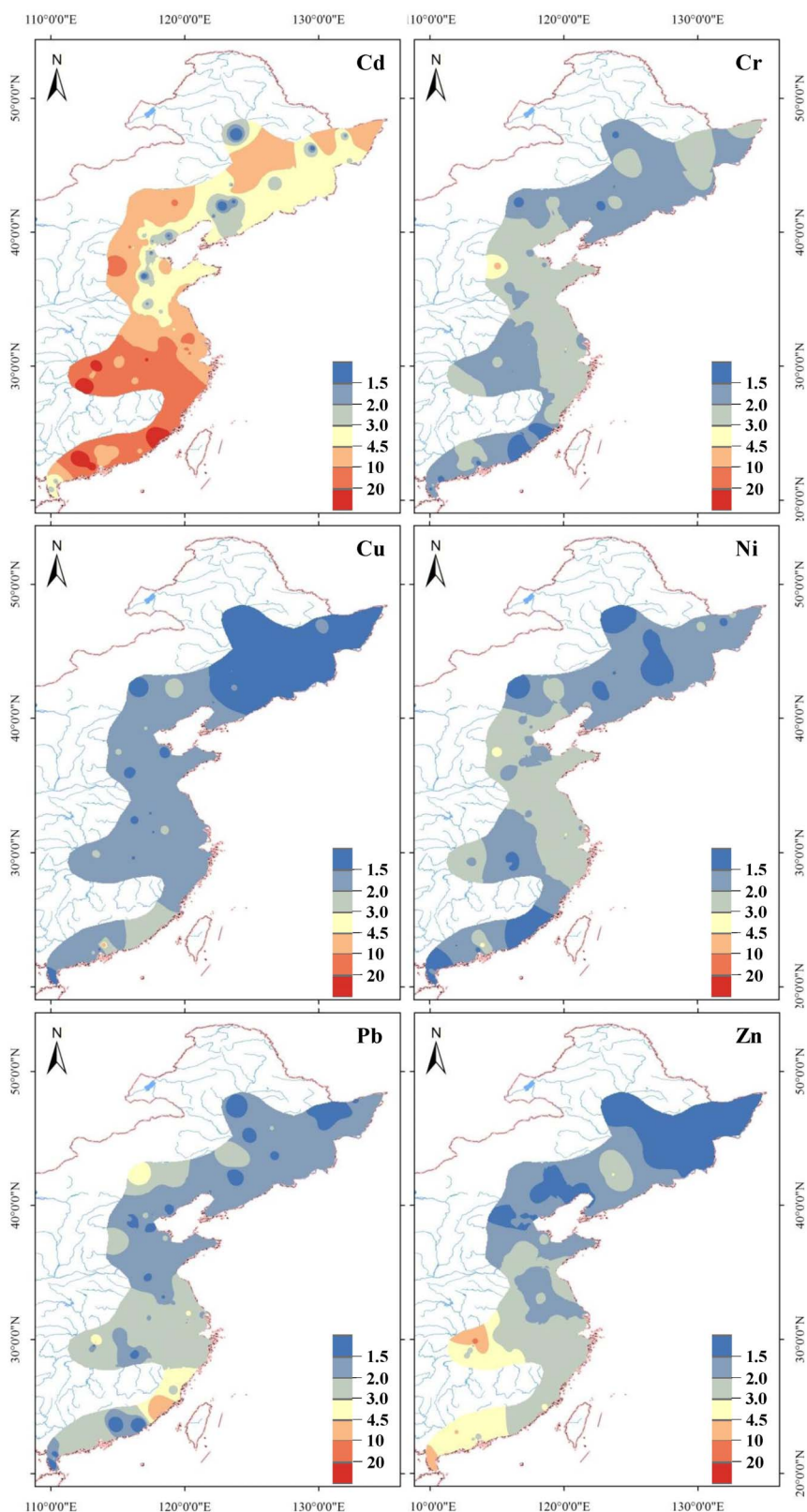


Figure 2 | Heavy metal enrichment factor distribution of surface sediments (ArcGIS 10.0).

in the development of effective management strategies for control of heavy metal pollution in the limnetic ecosystems of eastern China.

Methods

Study area. Eastern China is the most developed region of China and has the highest population density. Rapid economic development and population growth have

caused significant pollution of aquatic ecosystems in this area. The representative limnetic ecosystems investigated in this study include lakes, wetlands and rivers located in the basins of the Heilong River (HeRB), Liao River (LRB), Hai River (HaRB), Yellow River (YeRB), Huai River (HuRB), Yangtze River (YaRB), Southeast Coastal Rivers (SCRB) and Zhu River (ZRB), within the provinces of Heilongjiang, Liaoning, Hebei, Jiangsu, Anhui, Hubei and Guangdong, and the municipalities of Beijing, Tianjin and Shanghai (Figure 5). In addition, point (mainly

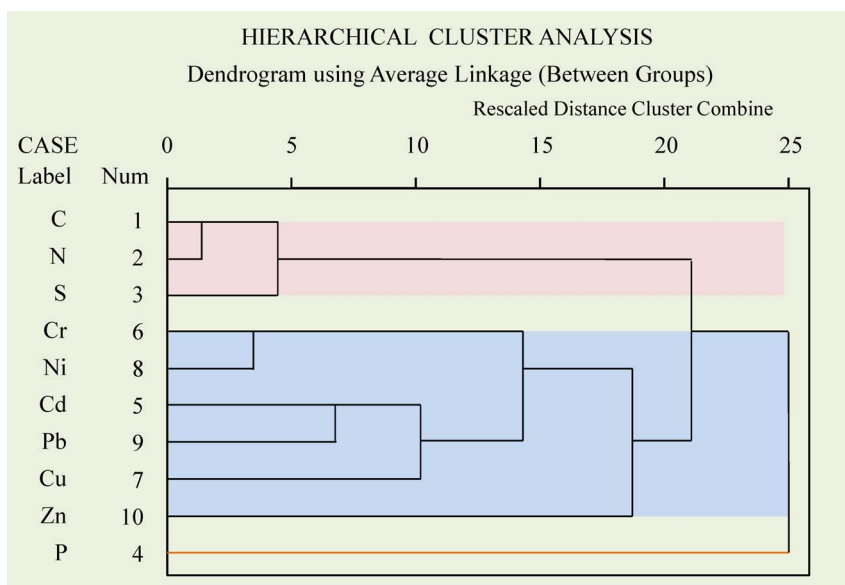


Figure 3 | Hierarchical dendrogram of metal elements of surface sediments (SPSS Statistics 17.0).

due to industrial development) and non-point (mainly agricultural development and urbanization) sources have caused severe heavy metal pollution in the aquatic ecosystems of eastern China (mainly plains)^{14,24}.

Sample collection and analysis. Surface sediments (0–10 cm) were collected from August to October 2013 at eighty-six sampling stations (Figure 5 and Table S5) using a Peterson grab sampler. Five parallel samples were collected from each station (Figure S7). Samples ($n = 430$) were air-dried, then dried in an oven at 40°C, after which they were ground and passed through a 100-mesh sieve prior to analysis.

Sediment pH was determined after mixing samples with water at a sediment: water ratio of 1 : 2.5³¹. The organic matter (OM) content was determined by residual titration of $K_2Cr_2O_7$ ³². The total carbon (TC), nitrogen (TN) and sulfur (TS) were measured using an element analyzer (Vario EL III, Elementar, Germany), while the total phosphorus (TP) of the sediment samples was analyzed by the $HClO_4-H_2SO_4$ digestion method³². For total heavy metal analysis, sediment samples (0.100 g) were digested with a 5 : 1 mixture of hydrofluoric: perchloric acid³³ in a microwave digestion system (MARS Xpress; CEM, Matthews, NC, USA) under the conditions presented in Table S6. All of the above solutions were stored at 4°C prior to analysis. The concentrations of P, Cd, Cr, Cu, Ni, Pb, Zn and Fe were measured by inductively coupled plasma optical emission spectrometry (ICP-OES) (Optima 2000DV; Perkin Elmer, Waltham, MA, USA) (detection limit 0.001–0.030 mg/L) and inductively coupled

plasma-mass spectrometry (ICP-MS) (7500a; Agilent Technologies, Santa Clara, CA, USA) (detection limit 0.015–0.120 µg/L). Laboratory quality control consisted of analysis of sediment reference material (GBW07302a; National Institute of Metrology, Beijing, China) and triplicate samples. The recoveries all fell within the range of 90%–110%, and the relative standard deviation was less than 5%.

Enrichment factor and risk assessment. To obtain information regarding the sources and factors influencing metals in the studied sediments, EF values of metals were calculated according to the following equation^{34,35}.

$$EF = (C_n/C_{Fe}) / (B_n/B_{Fe}) \quad (1)$$

where C_n is the metal content in the sediments, B_n is the background content of the metal, C_{Fe} is the Fe content in the sediments, and B_{Fe} is the background content of Fe. In this study, the soil background values of the studied regions were adopted as the baseline values¹⁸. In addition, the risk associated with heavy metals in the surface sediments were assessed using consensus-based SQGs³⁶, the PER index^{10,37}, I_{geo} ³⁸ and PLI³⁹ as described in (Table S4).

Statistical analysis. The experimental data were analyzed by SPSS Statistics 17.0 for Windows. CA was used to assess the relationship of heavy metal contents and

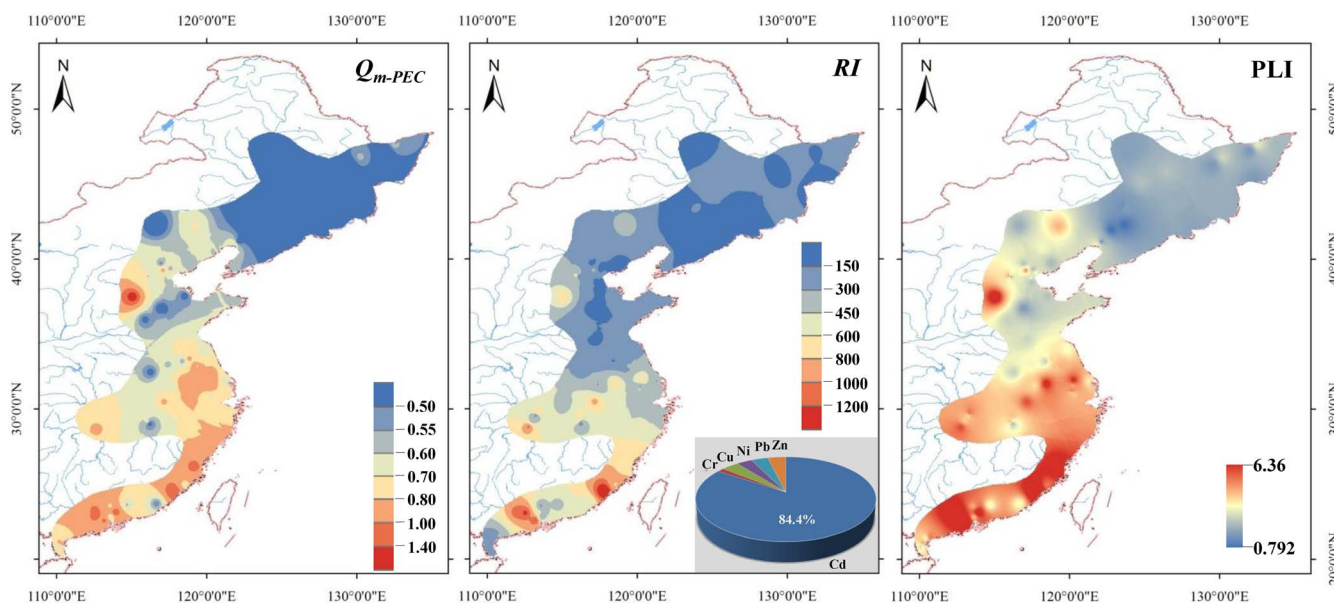


Figure 4 | Risk assessments of heavy metals in the surface sediments (ArcGIS 10.0). Q_{m-PEC} , mean probable effect concentration quotient; RI, comprehensive potential ecological risk index; PLI, pollution load index.

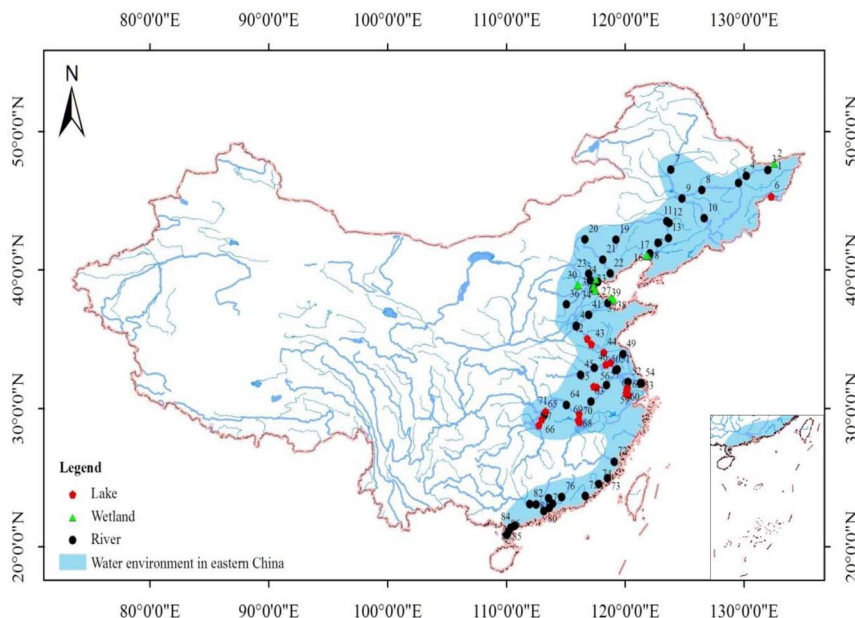


Figure 5 | Map showing the sampling stations (ArcGIS 10.0).

sediment properties, while HACA and PCA were carried out to identify potential sources of the metals⁴⁰. In addition, ArcGIS 10.0 and Origin Pro 8.0 were used to plot the experimental data.

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Author contributions

W.T. and B.S. designed and coordinated the study. W.T., W.Z., Y.D., N.R. and X.Z. performed the experiments. W.T., B.S. and H.Z. were involved in analyzing the data. W.T. and Y.Z. contributed analysis tools. W.T. and B.S. wrote the paper and all the authors participated in revising. All authors have read and approved the final manuscript.

Additional information

Supplementary information accompanies this paper at <http://www.nature.com/scientificreports>

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