Distribution and sources of pollutants in soils and sediments of different land-use types in degraded wetlands

Yangyang Han Equal first author, 1, 2, 3 , **Hongjie Wang** Equal first author, 1, 2, 3 , **Guangming Zhang** 4 , **Shengqi Zhang** 1, 2, 3 , **Xingchun Liu** 1, 2, 3 , **Ling Liu** Corresp. 1, 2, 3

1 Xiong'an Institute of Eco-Environment, Hebei University, Baoding, China

2 Institute of Life Science and Green Development, Hebei University, Baoding, China

3 College of Ecology and Environment, Hebei University, Baoding, China

4 School of Energy and Environmental Engineering, Hebei University of Technology, Tianjin, China

Corresponding Author: Ling Liu
Email address: liuling@hbu.edu.cn

Zaozhadian Lake (ZZDL) is a freshwater wetland in the North China Plain, which is facing a situation of ecosystem degradation and land-use types changes. Statistical description, spatial interpolation and multivariate visual statistical analysis were used to analyze and evaluate the pollutant distribution characteristics, and the pollution degree and sources at 25 sampling points of three land-use types of farmlands, lakes, and ditches in the ZZDL. The heavy metal contents in the farmland were higher than other areas. The contents of total nitrogen (TN) (3.71±1.03g/kg) and organic matter (OM) (57.17±15.16g/kg) in lake sediments were significantly higher than that in other regions. The geological accumulation index (I_{geo}) indicated that the primary heavy metal pollutants are Pb, Cu, and Ni, which are slightly polluted. The potential ecological risk index (*RI*) assessment of heavy metals showed that farmland, lakes, and ditches had low ecological risks, with the *RI* values of 84.21, 71.34, and 50.78, respectively. Furthermore, source analysis results showed that the Cu, As, Ni, Pb, and Zn primarily derived from agriculture pollution, and the source of Cd was the industrial pollution and Cr mainly originated from natural sources such as rock weathering. The nutrients primarily came from the decomposition of aquatic animal and plant and human activities. When the lake area was converted into farmland, the content of Hg, As, Cd, and Zn in the soils increased, and the TN and OM decreased. More attention should be paid to the impact of land-use types changes on wetland ecological environment.

Distribution and sources of pollutants in soils and sediments of different land-use types in degraded wetlands

Yangyang Han[1,2,3, &], Hongjie Wang[1,2,3, &], Guangming Zhang[4], Shengqi Zhang[1,2,3], Xingchun liu[1,2,3], Ling Liu[1,2,3]

[1] Xiong'an Institute of Eco-Environment, Hebei University, Baoding 071002, China
[2] Institute of Life Science and Green Development, Hebei University
[3] College of Ecology and Environment, Hebei University
[4] School of Energy and Environmental Engineering, Hebei University of Technology, Tianjin 300130, China

[&] Yangyang Han and Hongjie Wang contribute equally to the article.
Corresponding Author:
Ling Liu[1,2,3]
Wusi East Road, Baoding, Hebei, 071000, China
Email address: liuling@hbu.edu.cn

Abstract

Zaozhadian Lake (ZZDL) is a freshwater wetland in the North China Plain, which is facing a situation of ecosystem degradation and land-use types changes. Statistical description, spatial interpolation and multivariate visual statistical analysis were used to analyze and evaluate the pollutant distribution characteristics, and the pollution degree and sources at 25 sampling points of three land-use types of farmlands, lakes, and ditches in the ZZDL. The heavy metal contents in the farmland were higher than other areas. The content of total nitrogen (TN) (3.71±1.03g/kg) and organic matter (OM) (57.17±15.16g/kg) in lake sediments were significantly higher than that in other regions. The geological accumulation index (I_{geo}) indicated that the primary heavy metal pollutants are Pb, Cu, and Ni, which are slightly polluted. The potential ecological risk index (*RI*) assessment of heavy metals showed that farmland, lakes, and ditches had low ecological risks, with the *RI* values of 84.21, 71.34, and 50.78, respectively. Furthermore, source analysis results showed that the Cu, As, Ni, Pb, and Zn primarily derived from agriculture pollution, and the source of Cd was the industrial pollution and Cr mainly originated from natural sources such as rock weathering. The nutrients primarily came from the decomposition of aquatic animal and plant and human activities. When the lake area was converted into farmland, the content of Hg, As, Cd, and Zn in the soils increased, and the TN and OM decreased. More attention should be paid to the impact of land-use types changes on wetland ecological environment.

Introduction

Wetlands play an important role in improving water quality, protecting biodiversity, and regulating climate. People have realized the importance of wetlands for a long time, also understand that the large-scale reduction of wetlands will bring serious harm (*Maltby, 1986*). However, due to social and economic development, urbanization worldwide has made the expansion of arable land and construction land, and this has led to the degradation of wetlands, and the wetland ecosystem has been severely damaged. According to Ramsar Convention on Wetlands (*RCW, 2018*), 35% of the natural wetlands have been lost since 1970. Some researchers pointed out that since 2009, the world has reduced 33% of wetlands, most of which are natural wetlands, the main reason for the decrease of wetland is urbanization (*Hu et al., 2017*). As the population continues to grow and socioeconomic development, this trend of degradation and deterioration will be intensified (*Liu et al., 2020a*).

Both soils and sediments are important "sources" and "sinks" of pollutants in the wetland ecosystem. Pollutants enter the wetland due to industrial production, agricultural activities, transportation, etc., and eventually collect in soils and sediments of the wetland. This refers to soils and sediments as a "sink" of pollutants (*Harikrishnan et al., 2017; Wang et al., 2012*). On the other hand, under certain environmental conditions, nutrients in the sediments will be released to the overlying water, causing environmental pollution and eutrophication (*Wang et al., 2021*). Additionally, the accumulation of heavy metals and organic pollutants in sediments and soils also cause serious harm to animals, plant, and humans, leading to the destruction of wetland ecosystems (*Xia et al., 2020; Gopalakrishnan et al., 2020*). Therefore, it is essential for the restoration of wetland ecology to analyze the content of pollutants, evaluate pollution level, and find the source in wetland soils and sediments.

In recent years, researchers have conducted numerous studies on sediments pollution in rivers, lakes, and wetlands (*Liu et al., 2020b; Milacic et al., 2019; Rajeshkumar et al., 2018; Zhu et al., 2013b*). In order to efficiently describe the spatial distribution characteristics of pollutants, Geographic Information System (GIS) interpolation analysis and overlay analysis are used to visualize the pollutant content in the corresponding range. It is precisely because of the powerful spatial analysis capabilities that GIS is widely used in soils and sediments pollution research (*Facchinelli et al., 2001*). For pollution assessment purposes, various indexes were introduced. Among the many indexes, such as the I_{geo}, the Nemerow pollution index (*NPI*), pollution load index (*PLI*), the enrichment factor, and the *RI* were widely used in the evaluation of contaminant (*Peng et al., 2020; Ling et al., 2020; Xia et al., 2020; Xu et al., 2017; Yang et al., 2020a*). For the accuracy and effectiveness of the evaluation, a comparative assessment is needed, *RI*, I_{geo}, *PLI*, and the *NPI* have been employed in this study. Based on the long-term accumulation, sediments reflect the pollution input degree of natural and human activities in history, analyzing the man-made and natural sources of pollutants is of vital importance for pollution control, ecological restoration and policy making (*Singh et al., 2005*). Some methods and models have been introduced to explore the sources of pollutants, such as multivariate statistical analysis, chemical mass balance method, and positive matrix factorization (*Ma et al., 2016; Moghtaderi et al., 2020*). Among these methods, the multivariate statistical analysis can easily and accurately identify the pollution source by studying the correlation and statistical law of multiple pollution indicators, combined with the pollutant discharge characteristics of known pollution sources in the environment, and has been widely used in the field of hydrology, geochemistry, and chemistry in recent years. In this study, three multivariate statistical analysis methods were used: Pearson correlation analysis, Dual hierarchical cluster analysis (DHCA), and principal component analysis (PCA).

The ecosystem of ZZDL has been in a state of degradation in recent years, and the land-use types have undergone tremendous changes. From 1999 to 2018, the water area was reduced and the sediments was exposed. At the same time, the cultivated land, ditches, and construction land of ZZDL showed an overall increasing trend (*Lv et al., 2020*). Therefore, it is a representative area for this study. Moreover, ZZDL is part of the Baiyangdian Lake, which is the largest wetland in North China, and the study of ZZD is of great significance to the ecological restoration of lake wetlands in northern China. Some researchers have studied the nutrients and heavy metals in the sediments of ZZDL, mainly including the distribution, evaluation, and pollutant forms of pollutants (*Wang et al., 2020; Zhu et al., 2019*). However, there are few studies on the pollutants of different land-use types in ZZDL. In order to accurately understand the soils and sediments pollution in degraded wetlands, and explore the impact of land-use types changes on pollutants, the objectives of this study were to (1) study the spatial heterogeneity of various pollutants with different land-use types in wetlands by using statistical description and geostatistical methods; (2) assess the sediments nitrogen and phosphorus pollution, and evaluate the degree of heavy metal pollution and ecological risk in different regions through the I_{geo}, *PLI* and *RI*; (3) identify the main sources of pollutants. To our knowledge, this study is the first time to compare the pollutants of different land-use types in ZZDL. This study not only helps better understand the impact of land use change on pollutants, but also explains the pollution of soils and sediments in ZZDL. The results were beneficial to adjust land use planning in wetland (such as returning farmland to wetland) and improve the regional ecological environment.

Materials & Methods

Study area and sample collection

ZZDL located in the central region of the North China Plain, belonging to the Haihe River Basin. The average annual precipitation in this region is 563.9 mm, the distribution of precipitation during the whole year is extremely unbalanced, with an 80% in June to August, and most of these days have heavy rainfalls (*Wang et al., 2020; Yang et al., 2020b*). The water source of ZZDL mainly comes from rainfall and the supply of three rivers above it. The upstream urban rivers received the tail water from the sewage treatment station and the water body contains nutrients and heavy metals. The lack of precipitation and intensive human activities have led to the ecological degradation of the study area, mainly manifested as area shrinkage and eutrophication (*Zhang et al., 2020*).

Figure 1 shows that a total of 25 sampling sites were set up based on three land use types, i.e., farmland (10 sites, N1-10), lake (9 sites, D1-9), and ditches (6 sites, H1-6) (Note: Farmland and ditches were transformed from lake areas). The sampling time was September 2019, using a grab dredger to collect surface sediments in ditches and lake, and an original soil extractor to collect surface soil in farmland. Three sampling points were selected at each site, mixed into a complex and placed into polyethylene bags. After the samples were freeze-dried, the animal and plant residues and other remnants were removed, and the soil samples were sieved and divided into two parts. One part was used for the determination of TN, TP, OM, pH, cation exchange capacity (CEC), and the other part was for measurement of heavy metals, including Pb, Cd, Zn, Cu, Cr, As, Hg, Ni. The measurement methods of each index were based on relevant national testing standards (GB15618-1995, GB/T17141-1997, HJ491-2019).

Evaluation method

Nemerow pollution index. The *NPI* is used to evaluate nutrients in sediments, which is convenient to compare the overall pollution degree of the target area with other regions (*Pandit et al., 2020*). The method considers the average and maximum value of the pollution index of a single element. The formula (*Li et al., 2021*) is:

$$P_i = C_i/k_i, \quad (1)$$

$$P_n = \left(\sqrt{P_i^2 + [\max(P_i)]^2}\right)/2 \quad (2)$$

where, P_i is the single factor index, C_i is the measured value, and k_i is the standard value. The standard values of the two indicators TN and TP used in this study refer to the sediment evaluation guidelines issued by the Ministry of Environment and Energy of Ontario, Canada. Considering the content of the lowest level degree of ecotoxicity effect in the evaluation guidelines as the standard, the standard values of TN and TP were 0.55 and 0.60 g/kg, respectively (*Ye et al., 2019*). $\overline{P}_i$ is the average index of pollutants, max (P_i) is the maximum pollution index of a single pollutant. The pollution level classification is shown in Table S1.

Geological accumulation index. The I_{geo} was proposed by ~~the German scholar Muller~~ (*Wu et al., 2017*). This method uses the relationship between heavy metal content and geochemical background value to evaluate the degree of heavy metal pollution. The I_{geo} ~~value~~ is calculated using the following equation:

$$I_{geo} = \log_2(C_n/kB_n) \quad (3)$$

where, C_n is the measured content of the heavy metal, B_n is the geological background value of the heavy metal. ~~In this study,~~ the background value of soils elements in Hebei Province was adopted ~~in this study~~ (China Environmental Monitoring Station, 1990) (*Xiang et al., 2020*), the background values for As, Cd, Cu, Cr, Ni, Pb, Hg and Zn were 13.6, 0.094, 21.8, 68.3, 30.8, 21.5, 0.15, and 78.4 mg/kg, respectively. k is the correction coefficient, which is determined according to the change of the background value caused by the rock difference in various places, generally k= 1.5 (*Zhuang et al., 2021*). Also, it is divided into seven levels according to the I_{geo}. The specific pollution degree is shown in Table S2.

Pollution load index. *PLI* can identify the comprehensive pollution level of heavy metals (*Lu et al., 2014*). *PLI* is determined based on Eq (4). It is categorized into four levels, no pollution ($0 < PLI \leqslant 1$), slight pollution ($1 < PLI \leqslant 2$), moderate pollution ($2 < PLI \leqslant 3$), or highly pollution ($PLI > 3$).

$$PLI = \sqrt[n]{\frac{C_1}{B_1} \times \frac{C_2}{B_2} \times \frac{C_3}{B_3} \times \cdots \times \frac{C_i}{B_i} \times \cdots \times \frac{C_n}{B_n}} \quad (4)$$

where, C_i is the content of the metal i, B_i is the background value of the metal i, the background value is consistent with 2.2.2, n is the number of the elements.

Potential ecological risk index. *RI* is a comprehensive potential ecological hazard index. Compared with the I_{geo}, the *RI* method considers the toxicity of the pollutants (*Lars, 1980*), gives the biological toxicity coefficients of eight pollutants, and divides the potential ecological hazard levels quantitatively (*Men et al., 2018*). The calculation formula is:

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \times C_f^i, \quad (5)$$

$$E_r^i = T_r^i \times C_f^i, \quad (6)$$

$$C_f^i = C^i / C_n^i. \quad (7)$$

where, E_r^i is the potential ecological risk index of a single heavy metal, T_r^i is the heavy metal biological toxicity response coefficient (As= 10, Cd= 30, Cr=2, Cu= Ni= Pb= 5, Zn= 1, Hg= 40). C_n^i is the background reference value of the heavy metal, and C^i is the actual content of the heavy metal. The evaluation criteria for potential ecological hazards are shown in Table S3.

Statistical analysis

~~Google Earth and handheld GPS were used to arrange sampling points and record latitude and longitude information.~~ The pollutant Kriging interpolation and spatial heterogeneity analysis in the study area were conducted by using ArcGIS 10.4. SPSS Statistics 24, Excel, Origin Pro and

R software were applied to perform Variance analysis, Correlation analysis, PCA, and DHCA on the data.

Results

Distribution of nitrogen, phosphorus

Figure 2 shows the spatial distribution of TN, TP, OM, and pH in the sediments and soils. It can be seen that the content of TN and OM in lake sediments is higher than that in farmland soil and ditches sediments. Studies have shown that one of the main factors for the changes in OM and TN content in the wetland was the types of vegetation (*Zhao et al., 2020*). The TN content in sediments was related to the vegetation type and biomass, indicating that the plant litter was the main source of nitrogen element (*Campbell et al., 1989*). In this study, the spatial distribution of TN and OM showed a relationship with the types of land use, the area with the highest content was the water area where reeds were the dominant vegetation in the wetland with high yield and strong reproduction ability. The C, N and other elements were absorbed by the reeds during the growth and they entered the sediments through plant litter and residues. The organic matter returned to the wetland by the action of microorganisms, so the content of TN and OM in the wetland where reeds grow were high. In the farmland area, crops were planted all-year-round, destroying the aggregate structure of the surface soil, and the speed of respiration and decomposition of microorganisms was accelerated, which can reduce the soils organic matter content in the farmland area (*Chambers et al., 2014*). The area with the higher TP content located in the southeast of the study area, corresponding to two types of land use: farmland and lake area. Point source pollution can directly affect the distribution of pollutants (*Yang et al., 2018*). The nearby villages in the middle of the study area had the highest TP content. Here, most of the poultry was raised on the nearby water surface, and the feces were directly discharged into the lake. Moreover, the villagers directly discharged domestic sewage into the water area, which also resulted in higher TP content. Table 1 showed the statistical analysis of nutrients in soils with different land-use types. The land use types showed significant effects on TN, and OM (p_{TN} = 0.00, p_{OM} = 0.00). The average content of TN and OM was in the order of lake>farmland>ditch.

Spatial distribution of heavy metals and arsenic (As)

Figure 3 shows the spatial distribution of heavy metals and As in soils and sediments. There was a regional difference in heavy metals. The area with higher heavy metal content was the farmland, and compound accumulation of multiple heavy metals in farmland. The heavy metals Cr and Ni have similar distributions in the study area. The content of Ni and Cr gradually decreased from west to east. The content of Cu, Zn, As and Hg were generally higher in the south and lower in the north. These four pollutants were mainly concentrated in farmland areas. Pb and Cd were mainly concentrated in the middle of the study area near the southern area, and their contents diffused to the surroundings and decreased. The heavy metal pollution of surface sediments in the study area showed spatial differences, which was consistent with the research of previous studies (*Ji et al., 2019a; Ji et al., 2019b*). The impact of human activities (agricultural pollution, discharge of domestic waste) and the migration of heavy metals in rivers might lead to the accumulation of heavy metals (*Rajeshkumar et al., 2018*), thereby affecting the spatial distribution, migration, and transformation of heavy metals. The heavy metals Pb, Cu and Ni exceed the background value in three land-use types (Table 1). The content of Zn in farmland and lake area was higher, which exceeded the background value by 1.22 and 1.13 times, respectively. The Cd in the farmland exceeded the background value by 1.06 times. Moreover, the content of Cr, As and Hg were lower than the background value in all regions. The analysis of variance showed that the Pb, Cr, Cu and Ni in the samples of different types of land showed no significant difference (p_{Pb} = 0.01, p_{Cr} = 0.23, p_{Cu} = 0.15, p_{Ni} = 0.38). The land use types showed significant effects on Zn, Cd, As, and Hg (p_{Zn} = 0.03, p_{Cd} = 0.003, p_{As}=0.05, p_{Hg}=0.02). Among them, the content of As and Zn in farmland areas was significantly higher than that in other areas. The average content of Cd and Hg was in the order of farmland＞ lakes＞ ditch.

Evaluation of TN and TP

~~The *NPI* was used to evaluate the TN and TP pollution of sediments, H1-H6, D1-D9~~ (Table S4). The pollution index of TN and TP at each sampling point was greater than 1, indicating that there were nitrogen and phosphorus pollution in sediments, and nitrogen pollution in sediments is more serious than phosphorus (1.32<P_{TN}<8.69， 1.18<P_{TP}<2.35). Nitrogen pollution of sediments in the study area showed spatial differences. The P_{TN} ranges of ditches and lake areas were 1.32-5.56 and 2.91-8.69, respectively. Except for sampling point D9, all points in the lake area were heavily polluted. The degree of TP pollution varied among different regions, except for sampling points N6 and D8, which were moderately polluted, and other points were slightly polluted. Nitrogen and phosphorus pollution was high in the lake areas and neighboring villages in the northeast. In recent years, effective external pollution control in the study area has reduced the concentration of nutrients in the water body. However, the internal pollution was still relatively serious due to the comprehensive pollution of multiple factors such as rural life, agriculture, fishery and aquaculture, and some adverse factors such as extremely poor mobility of water. The comprehensive pollution index showed that more than 90% of the lake areas were seriously polluted.

Because the nutrients in farmland soil have little impact on wetland water bodies, in this study we used the method of evaluating soil fertility to evaluate the nitrogen and phosphorus in soils. According to the nutrient classification standard of the 2nd National Agricultural Census (*Luo et al., 2017*) (Table S5), the TN and TP content of soils samples were evaluated (Table S6). The TN content in the soils surface was between the medium and extremely rich levels, in which 40% of the soil samples were very rich in TN. More than 60% of soils TP reached a very rich level.

Pollution evaluation and risk assessment of Heavy metals and As

~~Pollution degree of heavy metals and As was evaluated by I_{geo} and the results are shown in Fig. 4.~~ The sequence of I_{geo} values of all metals in the study area was Pb > Cu > Ni > Zn > Cd > As > Cr > Hg, and the I_{geo} values of the eight contaminants were 0.49, 0.28, 0.18, -0.45, -0.99, -1.06, -1.17, -3.03, respectively. Among them, the Pb, Cu, Ni pollution was lower with the I_{geo} value range of 0 to 1. The I_{geo} of other heavy metals was less than 0, indicating that the concentration did not exceed the regional threshold value and the pollution degree was clean. The metal pollution levels of the three land types were consistent with the overall pollution levels. Fig. 4d shows that the difference in I_{geo} at each sampling point of the lake area was small, and there were no other abnormal values except Hg. Therefore, heavy metals in lake have homology. However, the I_{geo} of farmland and ditches showed a difference at each sampling point, suggesting that the source and accumulation of heavy metals may be different in the same land use type.

~~The *PLI* value of each sampling point is shown in~~ Fig. 5. The *PLI* values of farmland points (N1-N10) varied from 0.66 to 1.27 with an average of 1.06, indicating unpolluted to slight polluted. N1, N2, N6 points have the highest *PLI* values. N1, N2, and N6 are close to Caohe River, Baohe River and Fuhe River, respectively. All three rivers flow through towns, and domestic sewage and industrial waste water were discharged into the rivers. The migration of heavy metals in the rivers may be responsible for the high *PLI* values of these three samples. The average value of *PLI* in the ditch area is 0.82, except for H6, other points in ditches are not polluted by heavy metals. It may be that more human activities (i.e., fertilization, irrigation, and pesticide) have led to the accumulation of heavy metals at point H6. The *PLI* values of lake points varied from 0.78 to 1.04 with an average of 0.89, most areas of the lake were not polluted. The *PLI* results showed that the pollution level of heavy metals in the farmland area was lightly polluted, and most of the ditches and lake areas were not polluted.

~~The results of *RI* are shown in~~ Fig. 6. The order of the single heavy metal potential ecological risk index E_r^i was Cd > Pb > Cu > Ni > As > Hg > Cr > Zn, and the E_r^i of the eight elements were all lower than 40, indicating low ecological risks. Therefore, Cd was the dominant factor affecting *RI*, which represents the possible ecological risk of heavy metals in the sediments to the biological community (*Ali et al., 2015*). The E_r^i values of Cd in farmland, ditches and lakes were 31.28, 14.36, 27.30, respectively, which were quite different and indicated a higher potential ecological risk of Cd in farmland. ~~Fig. 6a shows that the heavy metals in the study area generally had low ecological risk.~~ The *RI* values of the three land-use types in the study area were in the order of farmland>lake>ditch, with the value of 84.21, 71.34, 50.78, respectively. Moreover, the potential ecological risk of heavy metals in the farmland was higher than that in other regions.

The comprehensive analysis showed that the average rankings of the I_{geo} and E_r^i were different. Still, both indicated that Pb, Cu and Ni ranked higher and represented the main heavy metal risk factors. The evaluation result of Cd I_{geo} is less than 1, which was different from the results of previous studies, on the contrary, the E_r^i value of Cd is the highest in this study. According to the results of Ji et al. (2019a), the average Cd concentration in the study area was 0.44 mg/kg, which was 4.7 times the local threshold value (0.094 mg/kg), but the analysis mentioned that this might be related to sampling point selection. Different ecological conditions and the surrounding environment may be the reason for the large difference in Cd evaluation results. In the study area, the surface soil or sediment of the three land types were not polluted by As and Zn, Cr, Hg. However, the heavy metals Ni, Cu, Pb, and Cd had varying degrees of harm to the ecosystem. Cd is highly toxic and highly soluble, and is widely distributed in aquatic ecosystems. Also, Cd has genetic and toxicological effects on biological systems and animals (*Geng et al., 2019; Marettová et al., 2015*). Therefore, Cd was the main ecological risk factor in this study. Pb is a toxic heavy metal enriched in the organism and can damage the liver, kidney, and gonads of the human body (*Khanam et al., 2020*). Although the Pb concentration in the study area is lower than the soils pollution risk control value (Soil Environmental Quality GB 15618-2018), the Pb concentration in farmland (49.25±9.94 mg/kg) reached 2.29 times of the soil threshold value (21.5 mg/kg). Both Cu and Ni are essential trace elements for plant growth and are related to the physiological and biochemical processes of plants (*Shabbir et al., 2020; Shahzad et al., 2018*). However, when Cu and Ni exceed a certain concentration, it can be toxic to plants and damage the normal metabolic system (*Ameh et al., 2019; Shahzad et al., 2018*). Therefore, Pb, Cu, Cd and Ni in the soils should be considered as the main control factors in the process of wetland ecological restoration.

Mutivariate ~~statisistical~~ analysis

~~The correlation of pollutants can be used to reveal that different pollutants may come from similar pollution sources (*Zhang et al., 2018*).~~ Some studies have shown that the positive correlation coefficient between heavy metals indicates a common source or similar behavior such as enrichment and migration (*Bastami et al., 2015; Yang et al., 2020a*). A Pearson's correlation matrix was conducted, Fig. 7 shows the correlation analyses of heavy metals, TN, OM, and TP. There was a high correlation among As, Zn, Ni and Cu, illustrating that the four elements have similar source or behavior. In addition, Cd was significantly positively correlated with Zn (p = 0.001) and the correlation coefficient was 0.76. Futhermore, there was a high correlation between TN and OM (r = 0.98, p = 0.001), while they have low correlation with other elements except Cd. The correlation coefficients of TN-Cd and OM-Cd are all 0.45. This result demonstrated that TN and OM were from same source, part of Cd also came from these sources.

Dual hierarchical clustering analysis is widely used in the analysis of environmental factors (*Li et al., 2015; Zhang, 2006*). In this study, DHCA was used to group the similar points and identify specific areas of contamination (Fig. 8). All elements were divided into two major categories, the first category was TN, TP, OM and the second category was As and heavy metals, indicating that the distribution and sources of the two categories were different. The indicators were further clustered and divided into four categories. ~~In DHCA~~, Cr was a single cluster, and the main pollution sites were N1, N2, and N3. Cd was a single cluster, and the concentration at N5, N6, N7, and N10 was significantly higher than other sites. Pb, Zn, Cu, Ni, As and Hg were classified into one category, indicating that the distribution of these elements was similar. In addition, TN, OM, and TP were classified into one category. This result implied that TN, OM, and TP came from similar source, but were different from As and heavy metals. The clustering of sampling points showed that all points were divided into 5 clusters. The farmland sampling points were mainly found in the fifth cluster, lake points were in the fourth cluster, and ditch points were in the first, third, fourth, and fifth clusters. This clustering analysis showed that the pollution characteristics of lake sediments and farmland soils were different. This finding also proves that the entryway of the pollutants into each land type is different. ~~In this study,~~ when the lake ecosystem was degraded and transformed into farmland, the metal pollutants concentrations increased significantly. The heatmap shows that the accumulation of metals in farmland areas is heavy, and the surface sediments in lake areas are rich in nutrients.

The principal component analysis method to perform dimensionality reduction and normalization analysis of data is an effective method to study pollution sources (*Xu et al., 2014*). The principal component analysis was performed to further analyze the impact of land type changes on pollutants (Fig. 9). The KMO (0.619) and Bartlett test showed that the results were valid. PCA results showed that the eigenvalues of the four principal components greater than 1 explained 84.062% of the total variance. The PC1 accounted for 39.8% of the total variance, of which Cu, Ni, As, Pb and Zn had loads greater than 0.7. The PC2 accounted for 22.125% of the total variance, which was related to TN and OM. Moreover, the PC3 was related to Cr, and the PC4 was associated with Cd. The sampling points in the lake area were mainly found in the first and second quadrants, TN and OM were the main pollutants, and Cd, Zn, and TP also contributed to the first quadrant. The farmland sampling points were in the fourth quadrant, these sampling points were mainly affected by Ni, As, Cu, Cr, Pb and Zn. Additionally, the points of the ditches were relatively scattered, probably due to the surrounding environment has a greater impact on it. Lake water was introduced into ditches for farmland irrigation, and collected rainwater from the farmland discharged into the lake through ditches. Therefore, ditches might serve as the crucial channels for the migration of pollutants from farmland and lakes.

Discussion

Pollution source identification

The results of PCA, DHCA and correlation analysis show that Cu, Ni, As, Pb and Zn in the study aera have similar sources. Except for As, the average values of Cu, Ni, Pb and Zn all exceed the background value. According to the previous analysis, compared with lake and ditch sediments, farmland soils was more polluted. Long-term use of fertilizers is the main cause of As and heavy metal pollution in farmland soils (*Yuan et al.,2021*). Studies have shown that As is very low in China and its growth rate is relatively fast, especially in farmland soils (*Yang et al., 2018*). Also, other study has shown that excessive phosphate fertilizer application is the main source of As in soils (*Gupta et al., 2014*). In the process of producing phosphate fertilizer, other heavy metals may also be brought into the fertilizer (*Zhang et al., 2016*). Since the breeding industry uses copper-containing feed, the Cu content in livestock manure cannot be ignored (*Blavi et al., 2021*). Long-term application of manure will cause Cu accumulation in the soil. In addition, some pesticides (organic arsenic fungicides, lead arsenate pesticides, etc.) contain heavy metals, and the use of pesticides also aggravates pollution by heavy metals (*Fallah et al., 2021*). Therefore, PC1 may be identified as an agricultural source.

In PCA and DHCA analysis, Cr is a group of its own, and in Pearson correlation analysis, Cr has no significant correlation with other elements. The Cr content in the three land use types was lower than the background value, and its I_{geo} value was less than 0, indicating that the sediments or soils were uncontaminated by Cr. Previous studies have shown that the content of Cr is higher in basic and ultrabasic rocks, and the weathering of rocks may be the main source of Cr, so PC4 is considered a natural source. This finding is consistent with the previous studies. The analysis result of Cd shows that it has different pollution sources from other elements. The content of Cd in farmland soil exceeded the background value, while in other land use types it was lower than the background value. The main sources of Cd are mining, industry, and automobile exhaust pollution (*Das et al. 1997*). Previous studies have shown that Cd in ZZDL originates from surrounding industrial sources (metal production plants, printing, and chemical fiber factory, etc.). Industrial wastewater was discharged into the river, and heavy metals entered the wetland soils and sediments through the river. Thus, PC1 represents the industrial source.

In the PCA and DHCA analysis, the sources of TN and OM were similar, and the correlation coefficient of TN-OM was 0.98. According to the previous analysis, the content of TN and OM in lake sediments is higher than that of farmland and ditches. Plants, algae, and mollusks grow in the lake, after these organisms die, they accumulate in the bottom of the lake and form organic sediments together with the clay silt (*Xing et al., 2021*). The poor water flow of ZZDL is more likely to cause a reduction environment at the bottom of the lake, promoting the accumulation of TN and OM in sediments. Therefore, TN and OM in sediments mainly come from animal and plant residues.

The impact of land-use type changes on the pollutants

The content distribution showed that TN, TP were quite different among the wetland soils with the three land use types. According to Table 1, the TN content in farmland (1.95±0.50 g/kg) is much lower than that in lake area sediments (3.71±1.03 g/kg), which indicates that after the lake area is converted into farmland, the sediment nitrogen content decreases. This may be due to planting crops, which consumes nutrients in the soil. With the heavy metal and As analysis from three types of land use conversions, it was found that all seven heavy metal and As concentrations of farmland were the highest among three land-use types, which supported our hypotheses regarding the impact of land-use conversion on heavy metals. Heavy metals can increase after the wetland being transformed into paddy fields and dry land (*Zheng et al., 2016*). Studies have also shown that wetland reclamation will lead to the release of heavy metals (*Bai et al., 2010*). When the soils were in a degrading state, the form of heavy metals changes, and the toxicity and mobility increase (*Han et al., 2000*). In addition, changes in land-use types will greatly change the soils moisture status and other physical and chemical properties, which will also greatly affect the solubility and mobility of pollutants, leading to the redistribution of pollutants in the land (*Han et al., 2007*). In this study, the conversion of lake area to farmland will increase the way for heavy metals to enter the wetland, which will damage the wetland ecosystem.

Implications for land-use planning and water quality maintenance

The results of this study provide useful information for the ZZDL wetland that can be used to raise managers awareness of potential long-term impacts of land-use type change so that policy can be developed to mitigate or minimize negative impacts.

In the agro-ecosystem, more of the heavy metals in the soil have originated from man-made sources more than natural sources. A greater area of farmland results in a greater risk to the regional environmental system. The increased non-point source pollution that will threaten wetland ecosystems and water quality should receive more attention, since the control of non-point source pollution is more difficult than point source pollution.

~~We know~~ land use influence wetlands in various ways (*Castelle et al., 1994*). In this study, we divided wetlands into three types of land-use, and analyzed the impact of land-use types on pollutants through the relationship between pollutants and the relationship between pollutants and land-use types. Regardless, we were unable to demonstrate any relationship between soil properties and the migration and distribution of pollutants. Differences in water content, exposure to the air, crops grown and methods of fertilization create a complex scenario that influences pollutant distribution. Therefore, we suggest that the adjustment of wetland land-use types should be based on clarifying the factors affecting the migration and transformation of pollutants and changes in toxicity.

Conclusions

Spatially, there were differences among TN, OM, As and heavy metals (Zn, Cd, and Hg) under different land-use types of ZZDL. The sediments in the lake area have high TN and OM content, and the farmland soils have a higher content of heavy metals. Based on the assessment results of I_{geo}, *PLI*, and *RI*, the surface sediments and soils can be described as low polluted by heavy metals. Pb shows the highest degree of pollution and Cd shows the highest ecological risk. The pollution level of heavy metals in the farmland area was lightly polluted, and most of the ditches and lake areas were not polluted. Furthermore, the results of multi-variate visual statistical methods proved that the Cu, As, Ni, Pb, and Zn primarily derived from agriculture pollution, and the source of Cd was the industrial pollution and Cr mainly originated from natural sources such as rock weathering. The nutrients primarily came from the decomposition of aquatic animal and plant and human activities. In this study, changes in wetland land-use types will affect the distribution and sources of pollutants. The change of lake to farmland decreased the concentrations of TN and OM, and increased the concentrations of Hg, As, Cd, and Zn. However, when the lake was transformed into trench, the content of pollutants was reduced except for Cr and Ni. Managers should pay more attention to the impact of changes in land-use types on wetland pollutants.

Acknowledgements

We are ~~thanks~~ to our all colleagues and research staff members for their constructive advice and help.

References

Ali, Z., Malik, R.N., Shinwari, Z.K., Qadir, A., 2015. Enrichment, risk assessment, and statistical apportionment of heavy metals in tannery-affected areas. Int. J. Environ. Sci. Technol. 12(2), 537-550. https://doi.org/10.1007/s13762-013-0428-4.

Ameh, T., Sayes, C.M., 2019. The potential exposure and hazards of copper nanoparticles: A review. Environ. Toxicol. Pharmacol. 71, 103220. https://doi.org/10.1016/j.etap.2019.103220.

Bai, J., Yang, Z., Cui, B., Gao, H., Ding, Q., 2010. Some heavy metals distribution in wetland soils under different land use types along a typical plateau lake, China. Soil Tillage Res. 106(2), 344-348. https://doi.org/10.1016/j.still.2009.11.003.

Bastami, K.D., Neyestani, M.R., Shemirani, F., Soltani, F., Haghparast, S., Akbari, A., 2015. Heavy metal pollution assessment in relation to sediment properties in the coastal sediments of the southern Caspian Sea. Mar. Pollut. Bull. 92(1-2), 237-243. https://doi.org/10.1016/j.marpolbul.2014.12.035.

Laia, B., David, S., Alessandra, M., J Francisco, P., Hans H, S., 2021. Inclusion of dicopper oxide instead of copper sulfate in diets for growing-finishing pigs results in greater final body weight and bone mineralization, but reduced accumulation of copper in the liver. J. Anim. Sci. 99(6), 127. https://doi.org/10.1093/jas/skab127.

Campbell, B.D., Grime, J.P., 1989. A Comparative Study of Plant Responsiveness to the Duration of Episodes of Mineral Nutrient Enrichment. New Phytol. 112(2), 261-267.

Castelle, A.J., Johnson, A.W., Conolly, C., 1994. Wetland and stream buffer size requirements-a review. J. Environ. Qual. 23, 878-882.

Chambers, L.G., Davis, S.E., Troxler, T., Boyer, J.N., Downey-Wall, A., Scinto, L.J., 2014. Biogeochemical effects of simulated sea level rise on carbon loss in an Everglades mangrove peat soil. Hydrobiologia 726(1), 195-211. https://doi.org/10.1007/s10750-013-1764-6.

Das, P., Samantaray, S., Rout, G.R., 1997. Studies on cadmium toxicity in plants: A review. Environ. Pollut. 98(1), 29-36. https://doi.org/10.1016/S0269-7491(97)00110-3.

Facchinelli, A., Sacchi, E., Mallen, L., 2001. Multivariate statistical and GIS-based approach to identify heavy metal sources in soils. Environ. Pollut. 114, 313–324. https://doi.org/10.1016/S0269-7491(00)00243-8.

Fallah, Z., Zare, E.N., Ghomi, M., Ahmadijokani, F., Amini, M., Tajbakhsh, M., Arjmand, M., Sharma, G., Ali, H., Hmad, A., Makvandi, P., Lichtfouse, E., Sillanpaa, M., Varma, R.S., 2021. Toxicity and remediation of pharmaceuticals and pesticides using metal oxides and carbon nanomaterials. Chemosphere 275, 130055. https://doi.org/10.1016/j.chemosphere.2021.130055.

Geng, H., Wang, L., 2019. Cadmium: Toxic effects on placental and embryonic development. Environ. Toxicol. Pharmacol. 67, 102-107. https://doi.org/10.1016/j.etap.2019.02.006.

Gopalakrishnan, G., Wang, S., Mo, L., Zou, J., Zhou, Y., 2020. Distribution determination, risk assessment, and source identification of heavy metals in mangrove wetland sediments from Qi'ao Island, South China. Regional Studies in Marine Science 133, 100961. https://doi.org/10.1016/j.rsma.2019.100961.

Gupta, D.K., Chatterjee, S., Datta, S., Veer, V., Walther, C., 2014. Role of phosphate fertilizers in heavy metal uptake and detoxification of toxic metals. Chemosphere 108, 134-144. https://doi.org/10.1016/j.chemosphere.2014.01.030.

Han, F.X., Banin, A., 2000. Long-term transformations of cadmium, cobalt, copper, nickel, zinc, vanadium, manganese, and iron in arid-zone soils under saturated condition. Commun. Soil Sci. Plant Anal. 31(7-8), 943-957. https://doi.org/10.1080/00103620009370489.

Han, F.X., Kingery, W.L., Hargreaves, J.E., Walker, T.W., 2007. Effects of land uses on solid-phase distribution of micronutrients in selected vertisols of the Mississippi River Delta. Geoderma 142(1-2), 96-103. https://doi.org/10.1016/j.geoderma.2007.08.006.
Harikrishnan, N., Ravisankar, R., Gandhi, M., Kanagasabapathy, K., Prasad, M.V.R., Satapathy, K., 2017. Heavy metal assessment in sediments of east coast of Tamil Nadu using energy dispersive X-ray fluorescence spectroscopy. Radiat. Prot. Environ. 40(1), 21-26. https://doi.org/10.4103/rpe.RPE_67_16.
Hu, S., Niu, Z., Chen, Y., Li, L., Zhang, H., 2017. Global wetlands: potential distribution, wetland loss, and status. Sci. Total Environ. 586, 319–327. https://doi.org/10.1016/j.scitotenv.2017.02.001.
Ji, Z., Zhang, H., Zhang, Y., Chen, T., Long, Z., Li, M., Pei, Y., 2019a. Distribution, ecological risk and source identification of heavy metals in sediments from the Baiyangdian Lake, Northern China. Chemosphere 237, 124425. https://doi.org/10.1016/j.chemosphere.2019.124425.
Ji, Z., Zhang, Y., Zhang, H., Huang, C., Pei, Y., 2019b. Fraction spatial distributions and ecological risk assessment of heavy metals in the sediments of Baiyangdian Lake. Ecotoxicol. Environ. Saf. 174, 417-428. https://doi.org/10.1016/j.ecoenv.2019.02.062.
Khanam, R., Kumar, A., Nayak, A.K., Shahid, M., Tripathi, R., Vijayakumar, S., Bhaduri, D., Kumar, U., Mohanty, S., Panneerselvam, P., Chatterjee, D., Satapathy, B.S., Pathak, H., 2020. Metal(loid)s (As, Hg, Se, Pb and Cd) in paddy soil: Bioavailability and potential risk to human health. Sci. Total Environ. 699, 134330. https://doi.org/10.1016/j.scitotenv.2019.134330.
Lars, H., 1980. An ecological risk index for aquatic pollution control.a sedimentological approach. Water Res. 14(8), 975-1001. https://doi.org/10.1016/0043-1354(80)90143-8.
Li, H., Zhang, Y., Wu, L., Jin, Y., Gong, Y., Li, A., Li, J., Li, F., 2021. Recycled aggregates from construction and demolition waste as wetland substrates for pollutant removal. J. Cleaner Prod. 311, 127766. https://doi.org/10.1016/j.jclepro.2021.127766.
Li, J., Jia, C., Lu, Y., Tang, S., Shim, H., 2015. Multivariate analysis of heavy metal leaching from urban soils following simulated acid rain. Microchem. J. 122, 89-95. https://doi.org/10.1016/j.microc.2015.04.015.
Ling, Z., Shi, Z., Xu, S., Dong, M., 2020. Temporal and spatial distributions of nutrients and pollution risk assessment of taxodium 'Zhongshanshan' wetland of Dianchi lakeside, southwest of China. Appl ecol env res. 18(1), 1161-1174. https://doi.org/10.15666/aeer/1801_11611174.
Liu, W., Guo, Z., Jiang, B., Lu, F., Wang, H., Wang, D., Zhang, M., Cui, L., 2020a. Improving wetland ecosystem health in China. Ecol. Indic. 113, 106184. https://doi.org/10.1016/j.ecolind.2020.106184.
Liu, Q., Sheng, Y., Jiang, M., Zhao, G., Li, C., 2020b. Attempt of basin-scale sediment quality standard establishment for heavy metals in coastal rivers. Chemosphere 245, 125596. https://doi.org/10.1016/j.chemosphere.2019.125596.
Lu, X., Zhang, X., Li, L.Y., Chen, H., 2014. Assessment of metals pollution and health risk in dust from nursery schools in Xi'an, China. Environ. Res. 128, 27-34. https://doi.org/10.1016/j.envres.2013.11.007.

Luo, H., Wu, J., Bianba, D., Gama, Qunzong., Zhu, X., 2017. Characterization of Soil Nutrient Status of Areas to be Afforested in Tibet, China. Acta Pedologica Sinica 54(02), 421-433. https://doi.org/10.11766/trxb201605250124.

Lv, T., Li, L., Zhu, S., Yang, G., Ma, H., 2020. Analysis on land use variation law and its driving force of Baiyangdian Lake since recent 30years. Water Resources and Hydropower Engineering 51(3), 37-44. https://doi.org/10.13928/j.cnki.wrahe.2020.03.005.

Maltby E., 1986. Waterlogged wealth: why wast the world's wet places. UK: An Earthscan Paperback. https://doi.org/10.4324/9781315066776.

Ma, L., Yang, Z., Li, L., Wang, L., 2016. Source identification and risk assessment of heavy metal contaminations in urban soils of Changsha, a mine-impacted city in Southern China. Environ. Sci. Pollut. Res. 23(17), 17058-17066. https://doi.org/10.1007/s11356-016-6890-z.

Marettová, E., Maretta, M., Legáth, J., 2015. Toxic effects of cadmium on testis of birds and mammals: A review. Anim. Reprod. Sci. 155, 1-10. https://doi.org/10.1016/j.anireprosci.2015.01.007.

Men, C., Liu, R., Xu, F., Wang, Q., Guo, L., Shen, Z., 2018. Pollution characteristics, risk assessment, and source apportionment of heavy metals in road dust in Beijing, China. Sci. Total Environ. 612, 138-147. https://doi.org/10.1016/j.scitotenv.2017.08.123.

Milacic, R., Zuliani, T., Vidmar, J., Bergant, M., Kalogianni, E., Smeti, E., Skoulikidis, N., Scancar, J., 2019. Potentially toxic elements in water, sediments and fish of the Evrotas River under variable water discharges. Sci. Total Environ. 648, 1087-1096. https://doi.org/10.1016/j.scitotenv.2018.08.123.

Moghtaderi, T., Alamdar, R., Rodriguez-Seijo, A., Naghibi, S.J., Kumar, V., 2020. Ecological risk assessment and source apportionment of heavy metal contamination in urban soils in Shiraz, Southwest Iran. Arabian J. Geosci. 13(16), 797. https://doi.org/10.1007/s12517-020-05787-9.

Pandit, P., Mangala, P., Saini, A., Bangotra, P., Kumar, V., Mehra, R., Ghosh, D., 2020. Radiological and pollution risk assessments of terrestrial radionuclides and heavy metals in a mineralized zone of the siwalik region (India). Chemosphere 254, 126857. https://doi.org/10.1016/j.chemosphere.2020.126857.

Peng, C., Shen, Y., Wu, X., Yuan, P., Jiang, L., Chen, S., Ze, S., Wang, X., Song, X., 2020. Heavy metals, nitrogen, and phosphorus in sediments from the first drinking water reservoir supplied by Yangtze River in Shanghai, China: spatial distribution characteristics and pollution risk assessment. Water, Air, Soil Pollut. 231(6), 298. https://doi.org/10.1007/s11270-020-04651-6.

Rajeshkumar, S., Li, X., 2018. Bioaccumulation of heavy metals in fish species from the Meiliang Bay, Taihu Lake, China. Toxicol. Rep. 5, 288-295. https://doi.org/10.1016/j.toxrep.2018.01.007.

Rajeshkumar, S., Liu, Y., Zhang, X., Ravikumar, B., Bai, G., Li, X., 2018. Studies on seasonal pollution of heavy metals in water, sediment, fish and oyster from the Meiliang Bay of Taihu Lake in China. Chemosphere 191, 626-638. https://doi.org/10.1016/j.chemosphere.2017.10.078.

Ramsar Convention on Wetlands (RCW), 2018. Global Wetland Outlook: State of the World's Wetlands and their Services to People. Ramsar Convention Secretariat, Gland, Switzerland. https://ssrn.com/abstract=3261606.

Shabbir, Z., Sardar, A., Shabbir, A., Abbas, G., Shamshad, S., Khalid, S., Natasha, Murtaza, G., Dumat, C., Shahid, M., 2020. Copper uptake, essentiality, toxicity, detoxification and risk assessment in soil-plant environment. Chemosphere 259, 127436. https://doi.org/10.1016/j.chemosphere.2020.127436.

Shahzad, B., Tanveer, M., Rehman, A., Cheema, S.A., Fahad, S., Rehman, S., Sharma, A., 2018. Nickel; whether toxic or essential for plants and environment-A review. Plant Physiol. Biochem. 132, 641-651. https://doi.org/10.1016/j.plaphy.2018.10.014.

Singh, K.P., Malik, A., Sinha, S., Singh, V.K., Murthy, R.C., 2005. Estimation of source of heavy metal contamination in sediments of Gomti River (India) using principal component analysis. Water, Air, Soil Pollut. 166(1-4), 321-341. https://doi.org/10.1007/s11270-005-5268-5.

Wang, C., Liu, S., Zhao, Q., Deng, L., Dong, S., 2012a. Spatial variation and contamination assessment of heavy metals in sediments in the Manwan Reservoir, Lancang River. Ecotoxicol. Environ. Saf. 82, 32-39. https://doi.org/10.1016/j.ecoenv.2012.05.006.

Wang, J., Zeng, X., Xu, D., Gao, L., Li, Y., Gao, B., 2020. Chemical fractions, diffusion flux and risk assessment of potentially toxic elements in sediments of Baiyangdian Lake, China. Sci. Total Environ. 724, 138046. https://doi.org/10.1016/j.scitotenv.2020.138046.

Wang, Y.T., Zhang, T.Q., Zhao, Y.C., Ciborowski, J.J.H., Zhao, Y.M., O'Halloran, I.P., Qi, Z.M., Tan, C.S., 2021. Characterization of sedimentary phosphorus in Lake Erie and on-site quantification of internal phosphorus loading. Water Res. 118, 116525. https://doi.org/10.1016/j.watres.2020.116525.

Wu, Q., Qi, J., Xia, X., 2017. Long-term variations in sediment heavy metals of a reservoir with changing trophic states: Implications for the impact of climate change. Sci. Total Environ. 609, 242-250. https://doi.org/10.1016/j.scitotenv.2017.04.041.

Xia, P., Ma, L., Sun, R., Yang, Y., Tang, X., Yan, D., Lin, T., Zhang, Y., Yi, Y., 2020. Evaluation of potential ecological risk, possible sources and controlling factors of heavy metals in surface sediment of Caohai Wetland, China. Sci. Total Environ. 740, 140231. https://doi.org/10.1016/j.scitotenv.2020.140231.

Xiang, Y., Wang, X., Shan, B., Zhao, Y., Tang, W., Shu, L., Jiang, S., Cao, Y., 2020. Spatial distribution, fractionation and ecological risk of heavy metals in surface sediments from Baiyangdian Lake. Huanjing Kexue Xuebao/Acta Scientiae Circumstantiae 40(6), 2237-2246. https://doi.org/10.13671/j.hjkxxb.2020.0060.

Xing, X., Chen, M., Wu, Y., Tang, Y., Li, C., 2021. The decomposition of macrozoobenthos induces large releases of phosphorus from sediments. Environ. Pollut. 283, 117104. https://doi.org/10.1016/j.envpol.2021.117104.

Xu, F., Liu, Z., Cao, Y., Qiu, L., Feng, J., Xu, F., Tian, X., 2017. Assessment of heavy metal contamination in urban river sediments in the Jiaozhou Bay catchment, Qingdao, China. Catena 150, 9-16. https://doi.org/10.1016/j.catena.2016.11.004.

Xu, J., Guo, J., Liu, G., Shi, G., Guo, C., Zhang, Y., Feng, Y., 2014. Historical trends of concentrations, source contributions and toxicities for PAHs in dated sediment cores from five lakes in western China. Sci. Total Environ. 470, 519-526. https://doi.org/10.1016/j.scitotenv.2013.10.022.

Yang, H.J., Jeong, H.J., Bong, K.M., Jin, D.R., Kang, T., Ryu, H., Han, J.H., Yang, W.J., Jung, H., Hwang, S.H., Na, E.H., 2020a. Organic matter and heavy metal in river sediments of southwestern coastal Korea: Spatial distributions, pollution, and ecological risk assessment. Mar. Pollut. Bull. 159, 111466. https://doi.org/10.1016/j.marpolbul.2020.111466.

Yang, Q., Li, Z., Lu, X., Duan, Q., Huang, L., Bi, J., 2018. A review of soil heavy metal pollution from industrial and agricultural regions in China: Pollution and risk assessment. Sci. Total Environ. 642, 690-700. https://doi.org/10.1016/j.scitotenv.2018.06.068.

Yang, W., Yan, J., Wang, Y., Zhang, B., Wang, H., 2020b. Seasonal variation of aquatic macrophytes and its relationship with environmental factors in Baiyangdian Lake, China. Sci. Total Environ. 708, 135112. https://doi.org/10.1016/j.scitotenv.2019.135112.

Ye, H., Yang, H., Han, N., Huang, C., Huang, T., Li, G.,Yuan, X., Wang, H., 2019. Risk assessment based on nitrogen and phosphorus forms in watershed sediments: a case study of the upper reaches of the minjiang watershed. Sustainability 11(20), 5565. https://doi.org/10.3390/su11205565.

Yuan, X., Xue, N., Han, Z., 2021. A meta-analysis of heavy metals pollution in farmland and urban soils in China over the past 20 years. Journal of Environmental Sciences 101, 217-226. https://doi.org/10.1016/j.jes.2020.08.01.3.

Zhang, C., 2006. Using multivariate analyses and GIS to identify pollutants and their spatial patterns in urban soils in Galway, Ireland. Environ. Pollut. 142(3), 501-511. https://doi.org/10.1016/j.envpol.2005.10.028.

Zhang, C., Shan, B., Zhao, Y., Song, Z., Tang, W., 2018. Spatial distribution, fractionation, toxicity and risk assessment of surface sediments from the Baiyangdian Lake in northern China. Ecol. Indic. 90, 633-642. https://doi.org/10.1016/j.ecolind.2018.03.078.

Zhang, Z., Lei, L., He, Z., Su, Y., Li, L., Wang, X., Guo, X., 2020. Tracking Changing Evidences of Water in Wetland Using the Satellite Long-Term Observations from 1984 to 2017. Water-Sui 12(6), 1602. https://doi.org/10.3390/w12061602.

Zhao, Q., Ding, S., Liu, Q., Wang, S., Jing, Y., Lu, M., 2020. Vegetation influences soil properties along riparian zones of the Beijiang River in Southern China. Peerj 8, e9699. https://doi.org/10.7717/peerj.9699.

Zhang, Q., Xiang, R., Liu, Z., Wan, Y., Zhong, Z., You, X., Qi, Y., 2016. Content and morphology characteristics of heavy metals in phosphatefertilizers in Hunan province. Nonferrous Met. Sci. Eng. 7(5), 1674-9669. https://doi.org/10.13264/j.cnki.ysjskx.2016.05.022.

Zheng, R., Zhao, J., Zhou, X., Ma, C., Wang, L., Gao, X., 2016. Land Use Effects on the Distribution and Speciation of Heavy Metals and Arsenic in Coastal Soils on Chongming Island in the Yangtze River Estuary, China. Pedosphere 26(1), 74-84. https://doi.org/10.1016/S1002-0160(15)60024-8.

Zhu, X., Ji, H., Chen, Y., Qiao, M., Tang, L., 2013b. Assessment and sources of heavy metals in surface sediments of Miyun Reservoir, Beijing. Environ Monit Assess. 185(7), 6049-6062. https://doi.org/10.1007/s10661-012-3005-2.

Zhu, Y., Jin, X., Tang W., Meng, X., Shan, B., 2019. Comprehensive analysis of nitrogen distributions and ammonia nitrogen release fluxes in the sediments of Baiyangdian Lake, China. J. Environ. Sci. 76, 319-328. https://doi.org/10.1016/j.jes.2018.05.024.

Zhuang, S., Lu, X., Yu, B., Fan, X., Yang, Y., 2021. Ascertaining the pollution, ecological risk and source of metal(loid)s in the upstream sediment of Danjiang River, China. Ecol. Indic. 125(3), 107502. https://doi.org/10.1016/j.ecolind.2021.107502.

Figure 1

Sampling site location

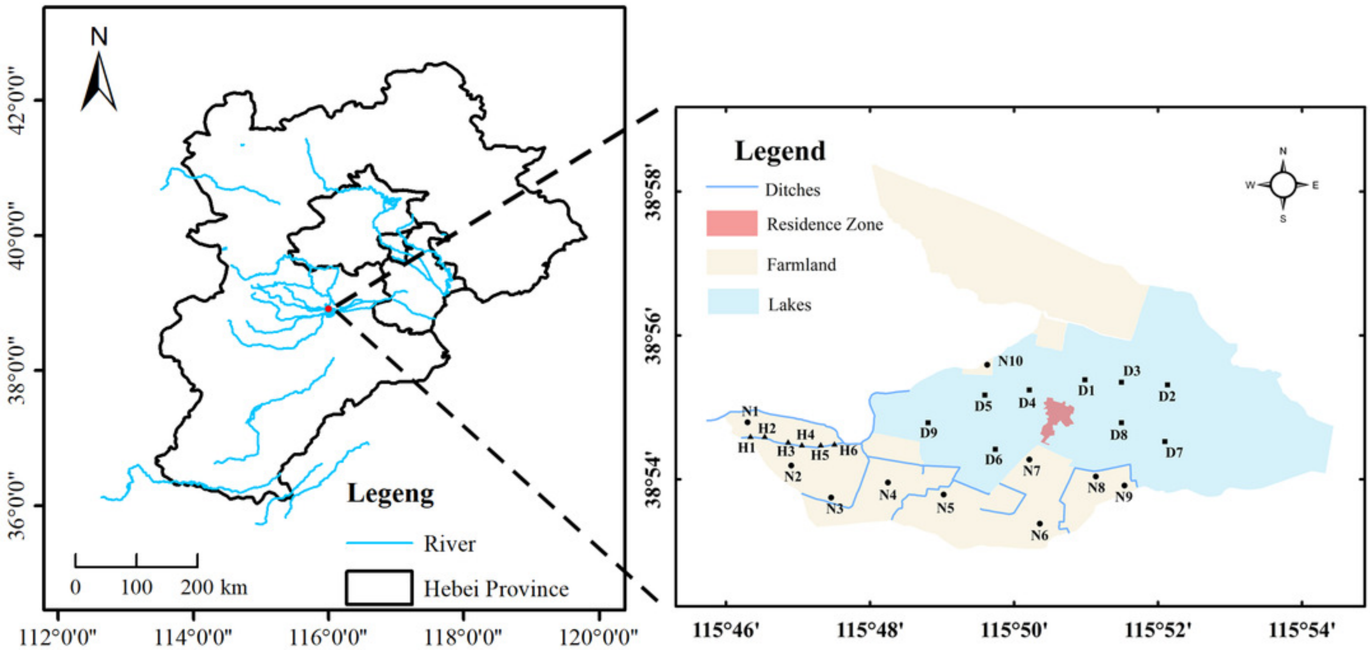

Figure 2

Distribution of nutrients and pH

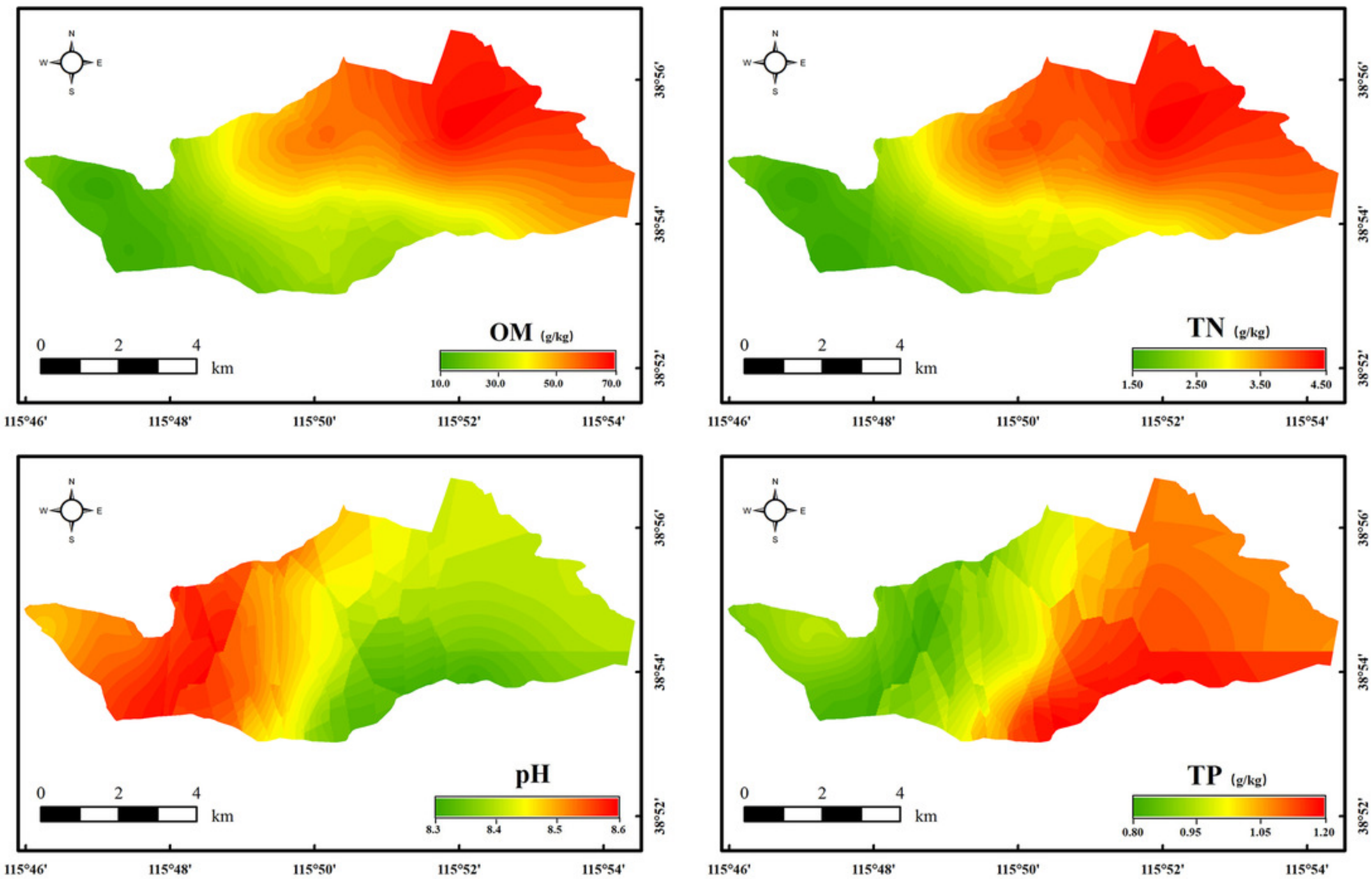

Figure 3

Distribution map of heavy metals and As

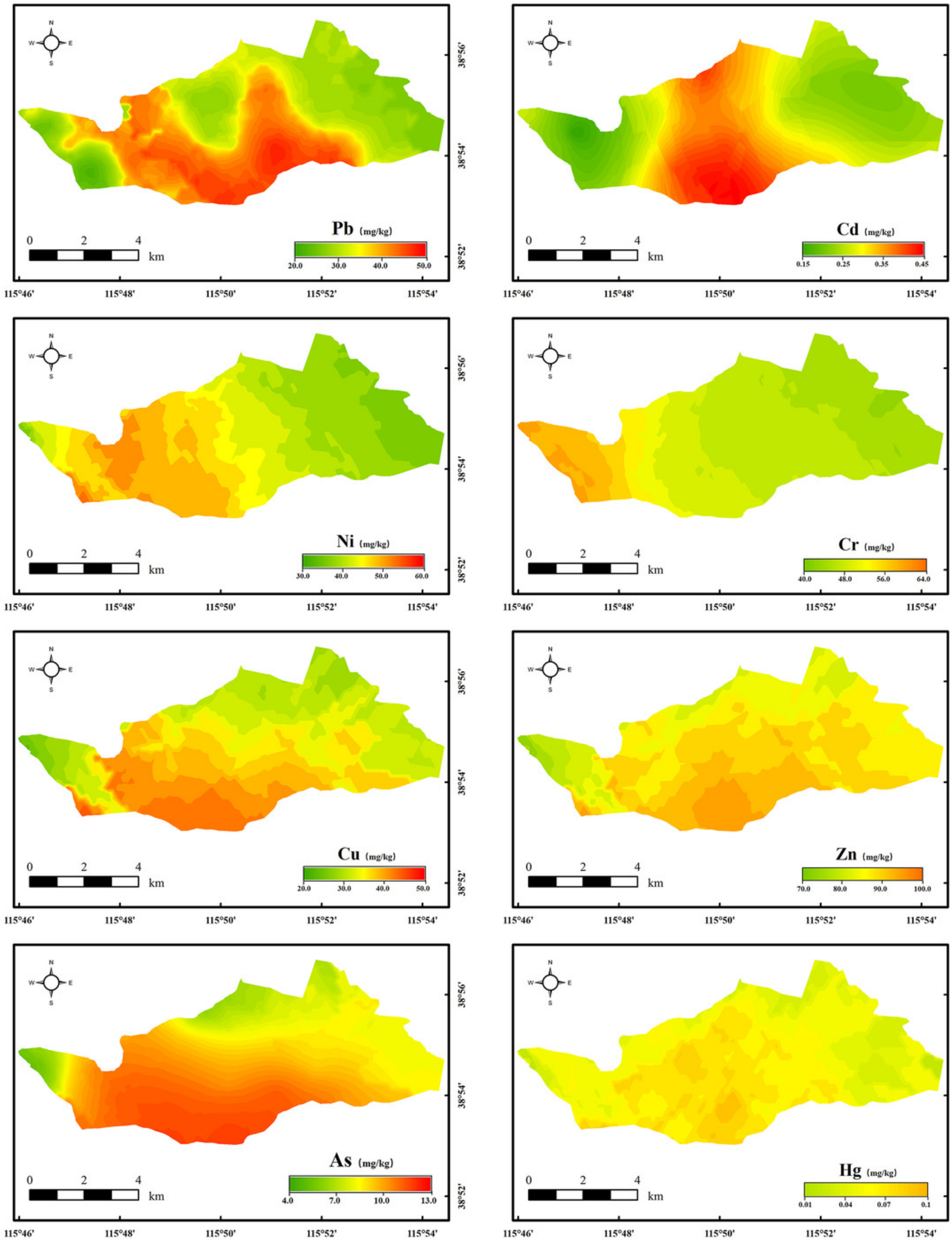
N
S
W
E
0 2 4 km
Pb (mg/kg)
20.0 30.0 40.0 50.0
Cd (mg/kg)
0.15 0.25 0.35 0.45
Ni (mg/kg)
30.0 40.0 50.0 60.0
Cr (mg/kg)
40.0 48.0 56.0 64.0
Cu (mg/kg)
20.0 30.0 40.0 50.0
Zn (mg/kg)
70.0 80.0 90.0 100.0
As (mg/kg)
4.0 7.0 10.0 13.0
Hg (mg/kg)
0.01 0.04 0.07 0.1
38°56'
38°54'
38°52'
115°46'
115°48'
115°50'
115°52'
115°54'

Figure 4

I_{geo} in sediments of different regions (a~d represent the I_{geo} value of the whole study area, farmland, ditch, and lake area, respectively)

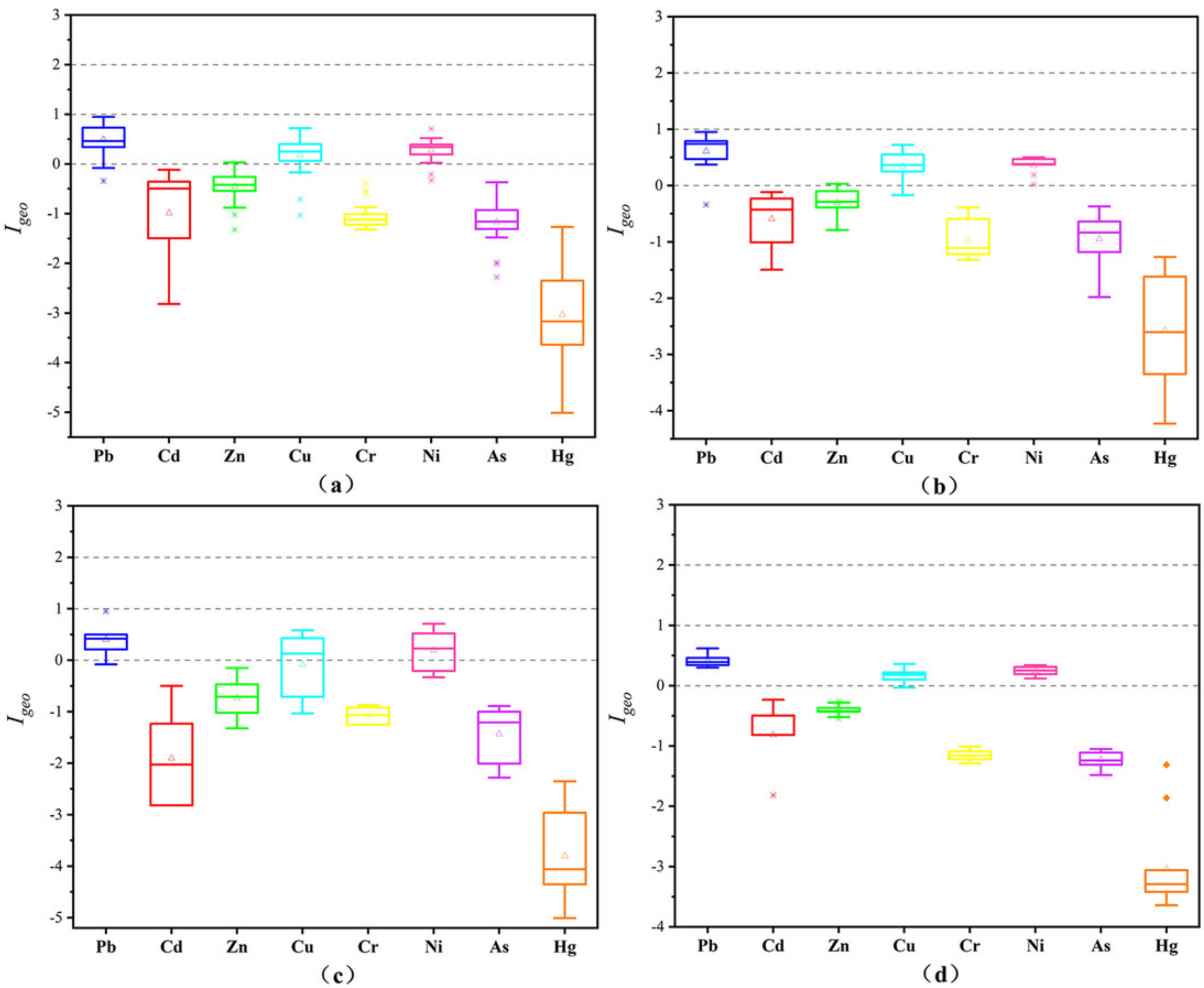

Figure 5

PLI for samples of ZZDL

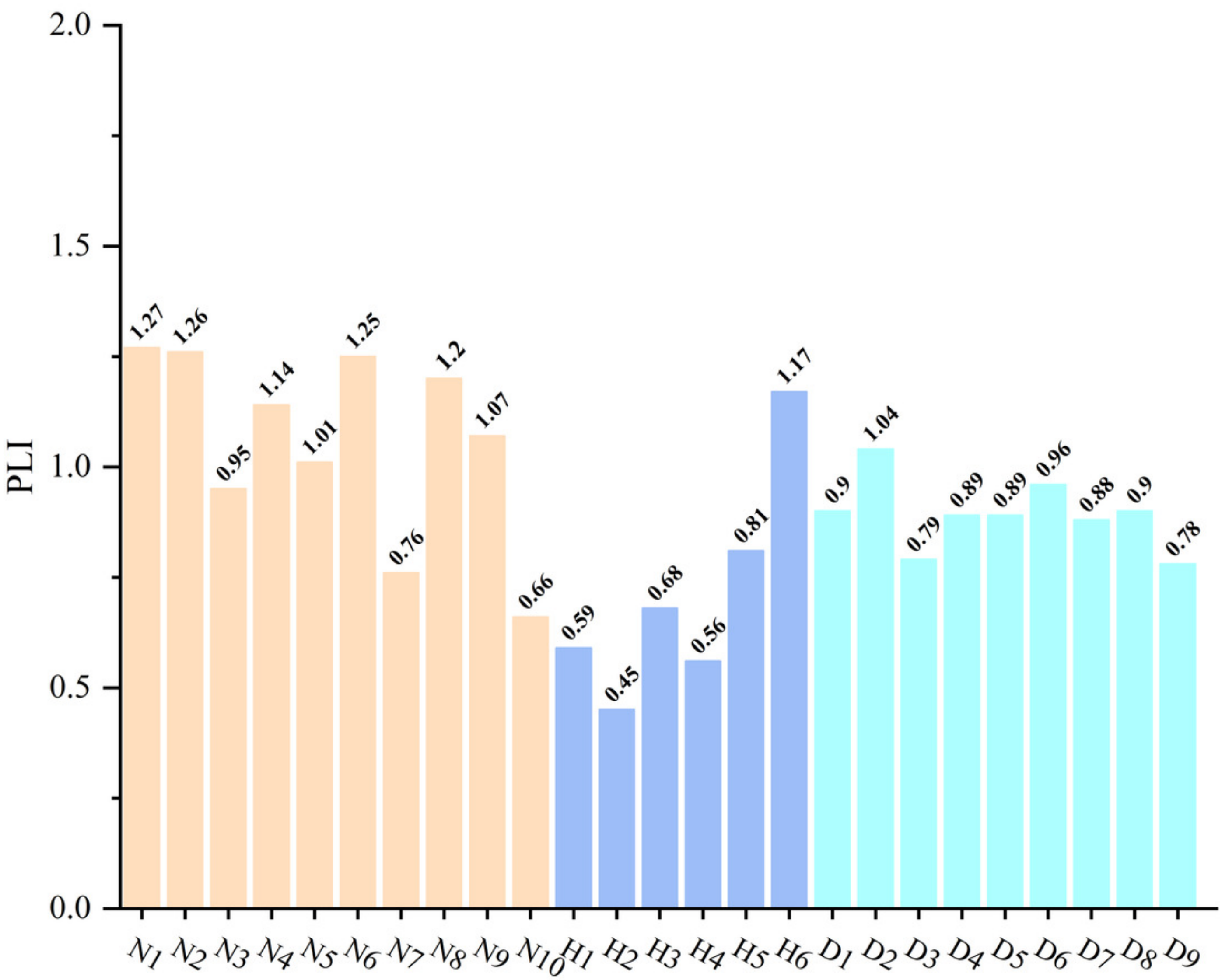

Figure 6

The E_r^i and RI of different regions (a~d represent the RI and E_r^i value of the whole study area, farmland area, ditch area, and lake area, respectively)

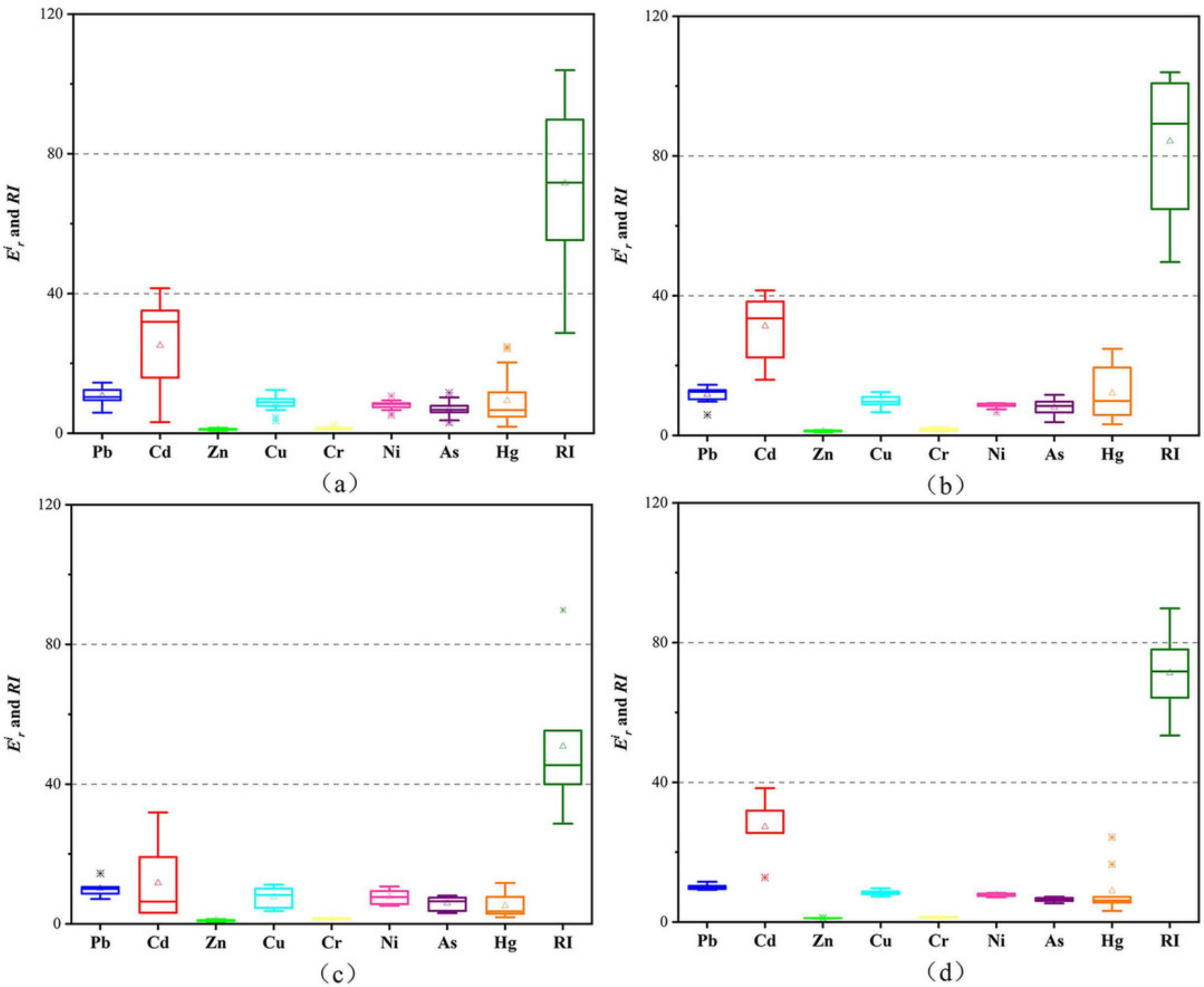

Figure 7

Correlation coefficients for heavy metals, As, and nutrients

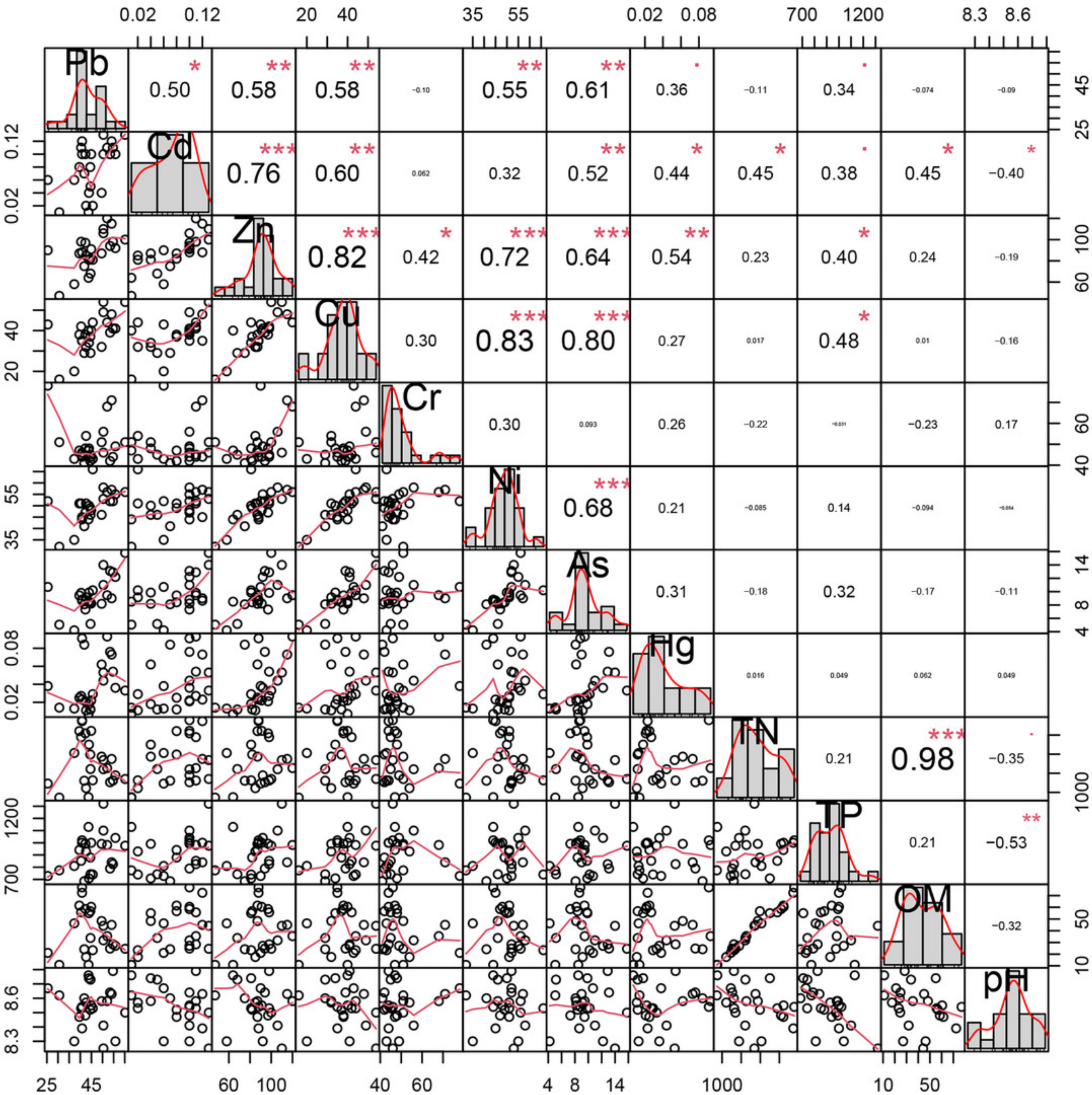

Figure 8

Heatmap of clustering heavy metals, As, and nutrients

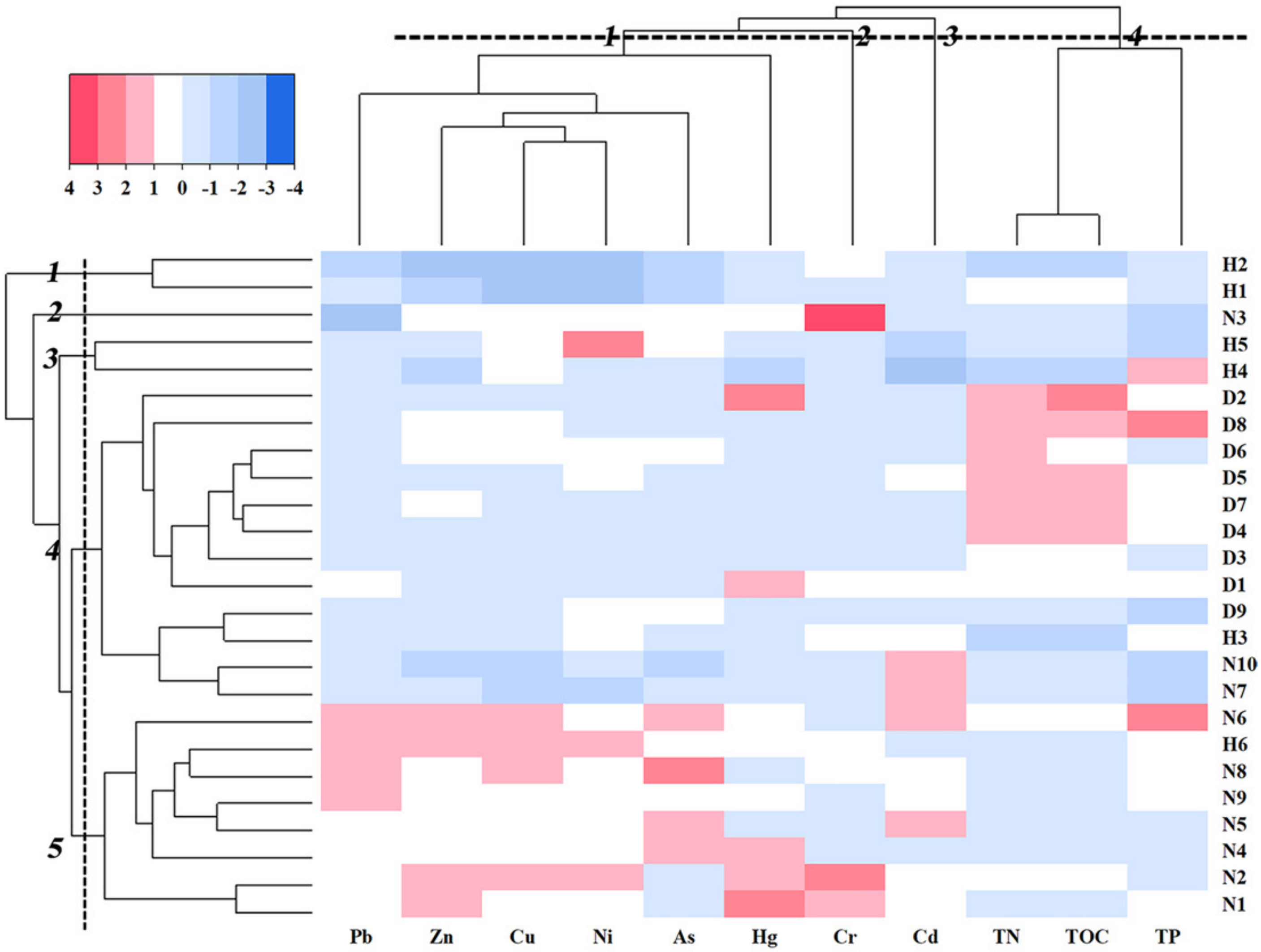

Figure 9

The principal component analysis loading plot

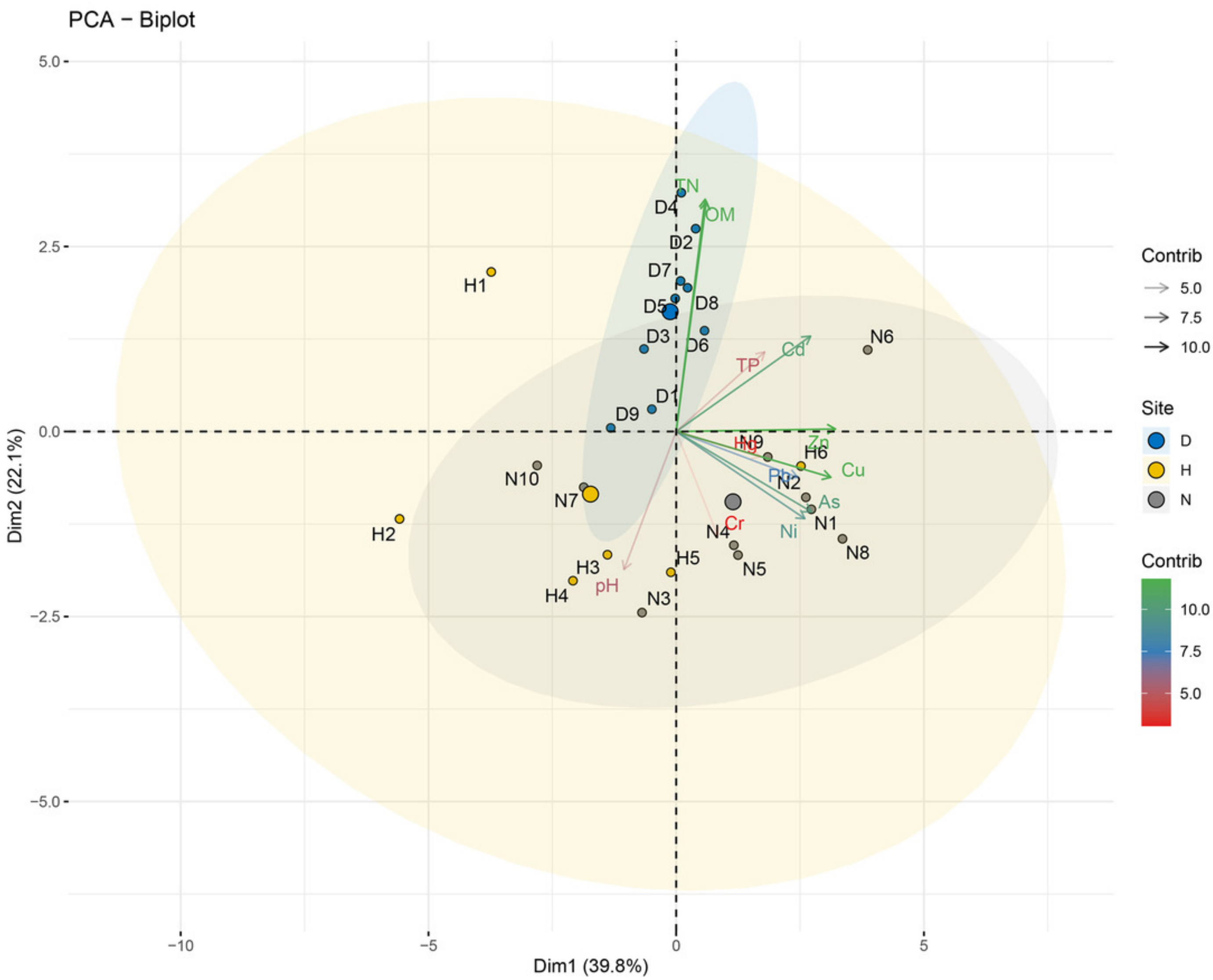

Table 1 (on next page)

Analysis on the difference of pollutants in different land types

Note: ** means p<0.01; * means p<0.05.

Table 1 Analysis on the difference of pollutants in different land types

	Background values	Land type (mean ± standard deviation)				
		Farmland (n=10)	Ditch (n=6)	Lake area (n=9)	*F*	*p*
Pb (mg/kg)	21.5	49.25±9.94	41.70±6.90	42.98±3.14	2.573	0.099
Cd (mg/kg)	0.094	0.10±0.03	0.04±0.04	0.09±0.03	7.908	0.003**
Zn (mg/kg)	78.4	96.20±16.44	73.50±21.34	88.78±5.83	4.281	0.027*
Cu (mg/kg)	21.8	42.20±8.68	33.50±13.29	36.78±3.11	2.086	0.148
Cr (mg/kg)	68.3	53.50±13.54	49.00±5.55	46.11±2.71	1.554	0.234
Ni (mg/kg)	30.8	52.00±4.99	47.67±13.22	47.89±2.57	1.014	0.379
As (mg/kg)	13.6	11.04±3.17	8.02±2.78	8.74±0.84	3.445	0.050*
Hg (mg/kg)	0.15	0.06±0.03	0.02±0.01	0.03±0.03	4.577	0.022*
TN (g/kg)	—	1.97±0.48	1.60±0.91	3.92±0.69	28.242	0.000**
TP (g/kg)	—	0.89±0.19	0.91±0.17	0.93±0.10	0.141	0.608
OM (g/kg)	—	30.86±8.45	23.48±13.72	60.61±8.91	31.2	0.000**

Note: ** means $p<0.01$; * means $p<0.05$.