Land use assessment using indices of heavy metal contamination in soils from intensive agricultural areas

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Abstract. Soil contamination by heavy metals is often caused by industrial and agricultural activities, which directly affects environment quality and food safety, especially for farmlands. To develop indicators for making proper decisions regarding land use, the Soil Heavy Metal Contamination Evaluation System including the Single Pollution Index (Pi), the Nemerow Pollution Index (PINemerow), the Potential Ecological Risk Index (RI), and the Contamination Severity Index (CSI) was developed using data from 499 samples collected from an area of 2017 km². A risk assessment zoning solution based on PPRC (the Soil Heavy Metal Contamination Evaluation System including Pi, PINemerow, RI, and CSI) was proposed, and the study area was mapped for four risk assessment zones as follows: no risk zone, very low risk zone, low risk zone, and intermediate risk zone. Four land use strategies (priority protection, protection, comprehensive monitoring, and warning and control) were proposed. Generally, the risk assessment zoning solution based on PPRC-defined minimizes the potential ecological risk to the land for the scientific evaluation of heavy metal pollution in areas of intensive agricultural activity.

1. Introduction
Soil contamination by heavy metals is becoming increasingly serious, and a comprehensive method for quantifying soil quality is very important [1]. The assessment of heavy metal contamination in soils can be performed using appropriate indices [2-5]. Common indices of pollution used for the assessment of soil quality are the Geoaccumulation Index (Igeo), the Enrichment Factor (EF), the Nemerow Pollution Index (PINemerow), the Potential Ecological Risk (RI), and the Contamination Severity Index (CSI) [6-10]. These indices enable determination of ecological risk and help to protect ecosystems [11]. So far, the connotation of evaluation methods is transitioning from single to comprehensive [10,12-13]. However, a scientific framework for the evaluation, risk assessment zoning, and control according to classifications of regional land has been lacking, which results in hidden dangers of land usage [14].

In this study, PPRC (the Soil Heavy Metal Contamination Evaluation System including Pi, PINemerow, RI, and CSI) was proposed for the comprehensive evaluation of heavy metals, and a risk assessment zoning solution based on PPRC was provided to realize control according to the classification of...
2. Materials and methods

2.1 Study area
The study was performed in Qixia County, which is a typical agricultural area in the east of China with an area of 2017 km². This region has mountainous topography and a subhumid warm temperate monsoon climate with four distinct seasons. Annual mean values for air temperature, rainfall, frost-free days, and sunshine are 11.3 °C, 650 mm, 207 days, and 2690 hours, respectively. The majority of this land is used for intensive crop production, of which over 4333 ha are planted for apple trees.

2.2 Sample collection and preparation
A total of 499 sampling sites with an area of approximately 4 km² for each site were established in the study area using the Mesh Points Method based on Geographical Information System data. One representative sample from the 0-20 cm surface layer was collected in each area of approximately 4 km². For each sampling site, a composite soil sample (~1 kg) was collected from 0-20 cm of depth at five sampling points (four corners and the center) of each of five subplots (2500 m² each). The samples were dried at room temperature, ground, and passed through a 2 mm mesh sieve. After acid digestion, soil samples were analyzed for total As, Cd, Cr, Cu, Hg, Ni, Pb and Zn content using ICP – MS.

2.3 Data analysis
(1) Principal component analysis
Principal component analysis (PCA) was performed for data collected in this study. Kaiser–Meyer–Olkin (KMO) and Bartlett’s tests were performed to check the suitability of the data for PCA. KMO is a measure of sampling adequacy that demonstrates the proportion of variance in variables that might be caused by underlying factors. High values (close to 1.0) generally indicate that a PCA may be useful. Bartlett’s test of sphericity indicates whether the correlation matrix is an identity matrix, which would indicate that the variables are unrelated. Small values (less than 0.05) of the significance level indicate that there are significant relationships among variables and that PCA can be beneficial[15-17]. All mathematical and statistical calculations were conducted using SPSS.

(2) Inverse distance weighted interpolation
There are many interpolation methods, such as kriging interpolation[18], local polynomial interpolation and inverse distance weighted interpolation. Spatial distribution characteristics of heavy metals in soil are closely related to distance. The inverse distance weighted interpolation is based on the similarity principle and is of high precision for the case in which the entire interpolation area is covered uniformly with sample points. Inverse distance weighted interpolation was used in this study.

2.4 PPRC
Developing the Soil Heavy Metal Contamination Evaluation System is critical for a comprehensive assessment of soil pollution [2,4]. In this study, the Soil Heavy Metal Contamination Evaluation System (PPRC) consisted of the Single Pollution Index (Pi); the Nemerow Pollution Index (PINemerow); the Potential Ecological Risk Index (RI); and the Contamination Severity Index (CSI)[8] (The calculation formulas are given in the Table1).

| Table 1 Indices of pollution used in this study |
|-----------------|----------|-----------------|----------|
| **Index** | **Formula** | **Explanations** | **Limit values** |
| Pi | \( \frac{c_i}{s_i} \) | \( c_i \)-measured content of heavy metal \( i \) in analyzed sample of soil; \( s_i \)-evaluation limit of heavy metal \( i \) according Environmental Quality | Pi classes according to Technical Regulation for assessment of soil pollution in China (2008) |
| | | | I. \( \leq 1 \) no pollution; |
Standard for soils (GB 15618-2008)

- I. 1-2 slight pollution;
- II. 2-3 low pollution;
- III. 3-5 moderate pollution;
- IV. >5 heavy pollution.

**Pollution classes according to the Technical Specification for soil Environmental monitoring (HJ/T166-2004)**

- I. ≤0.7: clean;
- II. 0.7-1: warning limit;
- III. 1-2: slight pollution;
- IV. 2-3: moderate pollution;
- V. ≥3: heavy pollution.

**Classification of RI [6]**

- I. ≤150 low;
- II. 150-300 moderate;
- III. 300-600 strong;
- IV. ≥600 very strong;

**Classification of CSI**

- I. < 0.5 uncontaminated
- II. 0.5-1 very low severity
- III. 1-1.5 low severity
- IV. 1.5-2 low - moderate severity
- V. 2-2.5 Moderate severity
- VI. 2.5-3 Moderate-high severity
- VII. 3-4 High severity
- VIII. 4-5 Very high severity
- IX. ≥5 Ultra high severity

(1) Single Pollution Index (Pi)

The Single Pollution Index (Pi) is used to assess pollution from a single heavy metal in the soil. To calculate Pi, the evaluation limits of heavy metals are fixed according to Environmental Quality Standard for soils (GB 15618-2008). The evaluation limits for Cu, Zn, Cr, Ni, Pb, Cd, As and Hg are 50, 200, 150, 80, 80, 0.3, 40, and 0.35, respectively.

(2) Nemerow Pollution Index (PINemerow)

PINemerow is used to assess the degree of contamination in a soil environment [19]. The calculated values of Pi were used to calculate PINemerow.

(3) Potential Ecological Risk Index (RI)

The Potential Ecological Risk Index (RI), introduced by Hakanson [6], is used to investigate the ecological risk of heavy metals in the study area. To calculate RI, the toxic response coefficients were given by Hakanson [6]. The coefficients for Cu, Zn, Cr, Ni, Pb, Cd, As and Hg are 5, 1, 2, 5, 5, 30, 10,
and 40, respectively [6,20-21].

(4) Contamination Severity Index (CSI)

CSI, introduced by Pejman [8], relates to the intensity of concentration of heavy metals in the soil. It is also useful for determining the limit of toxicity and adverse impacts in the soil environment. Effects range low (ERL) and effects range median (ERM) values were given by Long [22] to calculate the CSI. The ERL values for Cu, Zn, Cr, Ni, Pb, Cd, As and Hg are 34, 150, 81, 20.9, 46.7, 1.2, 8.2, and 0.15, respectively. The ERM values for Cu, Zn, Cr, Ni, Pb, Cd, As and Hg are 270, 410, 370, 51.6, 218, 9.6, 70, and 0.71, respectively. The weighted values \( W_i \) for each metal were computed using the following formulas for calculating the CSI:

\[
\begin{align*}
\text{f}_i &= \frac{\text{PC}_j \cdot \text{loadvalue}_i}{\sqrt{\text{Eigenvalue}_j}} \\
\text{F}_i &= \frac{\sum_{j=1}^{n} \text{f}_j \times TV_j}{\sum_{j=1}^{n} TV_j} \\
\text{w}_i &= \frac{\text{F}_i}{\sum_{i=1}^{m} \text{F}_i}
\end{align*}
\]

where \( f_j \) is the coefficient of index \( i \) (heavy metal \( i \)) in the linear combination of principal component \( j \), \( \text{PC}_j \cdot \text{loadvalue}_i \) is the principal component \( j \) loadvalue of index \( i \) (heavy metal \( i \)), \( \text{Eigenvalue}_j \) is the eigenvalue of the principal component \( j \), \( F_i \) is the coefficient of index \( i \) (heavy metal \( i \)) in the constructed model, \( n \) is the number of the principal components with eigenvalues >1, \( TV_j \) is the percentage total variance that principal component \( j \) explains, \( W_i \) is the weighted values of the heavy metal \( i \), and \( m \) is the number of indices (heavy metals). These indices were based on concentrations of heavy metals in the 499 samples collected and were calculated by the formulas given in Table 1.

2.5 Risk assessment zoning solution

Dimensionless classifications of environmental quality were provided by 600 (5×5×4×6=600) permutations and combinations based on the grades of the four pollution indices. Based on a summary of these combinations, detailed descriptions of risk grades and risk assessment zones are presented in the Table 2. Land use strategies are given below:

(1) Risk grade I: No risk zone. Four pollution indices showed zero or low levels of contaminants. The quality of soils is good, and a strategy of preventing contamination is required.

(2) Risk grade II: Very low risk zone. The levels of contaminants are slightly higher than those of background levels in ambient soils. There is a slight risk when using these soils, and a strategy of preventing contamination is required.

(3) Risk grade III: Low risk zone. Contaminants have accumulated in these soils and the four pollution indices slightly exceed the limits. Risk is still low but exists for soil use, and a strategy of comprehensive monitoring is required.

(4) Risk grade IV: Intermediate risk zone. The levels of contaminants are high than those in ambient soils. The four pollution indices definitely exceed established limits. Using these soils poses a significant threat to the growth of crops and to human health. Therefore, a warning and control strategy is required.

(5) Risk grade V: High risk zone. The levels of contaminants are much higher than those in ambient soils. The four pollution indices seriously exceed established limits. The quality of these soils poses a serious threat to the growth of crops and to human health. A restricted utilization strategy is required.

| Grades      | Type combination |
|-------------|------------------|
| Risk grade I| \((D_{R1}=1)\) and \((D_{Pi}merow}=1)\) and \((D_{R1}=1)\) and \((D_{CSI}<2)\) |

No risk zone

Table 2 Descriptions of risk grades and risk assessment zones.
3. Results

3.1 Principal component analysis
The suitability of data for PCA, KMO and Bartlett’s Sphericity tests were evaluated. The calculated value of KMO and the significance level of Bartlett’s Sphericity indicated compatibility of data for PCA. PCA was applied to the normalized dataset to identify the factors affecting each variable in this study. The values of the eigenvalues specify the significance of the PCs, and eigenvalues greater than 1 are considered significant [16,23]. The result of PCA demonstrated that two PCs with eigenvalues >1 were extracted, which explain 63.1% of the total variance in the respective dataset. PC1 accounts for 37.885% of total variance and PC2 explains 25.205% of total variance. The values of PCA including load values, eigenvalues and %total variance were obtained.

3.2 Pollution indices

3.2.1 Single Pollution Index (Pi)
The results of single pollution indices in this study area are presented in Table 3. Point spatial distributions of soil heavy metals are shown in Fig 1, and spatial distributions of soil heavy metals are shown in Fig 2.

| Index grade | I       | II      | III     | IV      | V       |
|-------------|---------|---------|---------|---------|---------|
| As          | Points  | 100.00  |         |         |         |
|             | Area    | 100.00  |         |         |         |
| Cd          | Points  | 94.99   | 5.01    |         |         |
|             | Area    | 97.11   | 2.89    |         |         |
| Cr          | Points  | 97.80   | 2.20    |         |         |
|             | Area    | 99.16   | 0.84    |         |         |
| Cu          | Points  | 64.33   | 32.87   | 2.81    |         |
|             | Area    | 63.22   | 36.52   | 0.26    |         |
| Hg          | Points  | 97.19   | 2.81    |         |         |
|             | Area    | 99.02   | 0.98    |         |         |
| Ni          | Points  | 98.60   | 1.40    |         |         |
|             | Area    | 99.51   | 0.49    |         |         |
| Pb          | Points  | 98.20   | 1.80    |         |         |
|             | Area    | 99.60   | 0.40    |         |         |
a. The entire study area (a total of 499 study points with a total area of 2017 km²) shows no As pollution (see Table3, Fig1 (a), and Fig 2 (a)).
b. There are 25 points comprising 56 km² of land that exhibit slight Cd pollution and are mainly distributed in the north portion of the study area (see Table3, Fig1 (b), and Fig2 (b)).
c. There are only 11 points comprising 17 km² of land that exhibit slight Cr pollution (see Table3). The point spatial distribution of Cr in the soil is shown in Fig1(c). The spatial distribution Cr in the soil is shown in Fig2 (c).
d. There are 164 points showing slight Cu pollution, and 14 points showing low Cu pollution. The point spatial distribution of Cu in the soil is shown in Fig1(d). There is 736 km² of land exhibiting slight Cu pollution and 5 km² of land with low Cu pollution (Fig 2 (d)).
e. There are 14 points comprising 20 km² of land that show slight Hg pollution. These areas are mainly distributed in the midwestern portion of the study area (see Table3, Fig1(e), and Fig2(e)).
f. There are only 7 points comprising 10 km² of land that show slight Ni pollution (see Table3, Fig 1 (f), and Fig2(f)).
g. There are only 9 points that comprise 8 km² of land showing slight Pb pollution. These are mainly distributed in the northern part of study area (see Table3, Fig1(g), and Fig2(g)).
h. The entire study area shows no Zn pollution (see Table3, Fig1 (h), and Fig 2(h)).

3.2.2 Potential Ecological Risk Index (RI)
The results for the Potential Ecological Risk Index (RI) in the study area show that a total of 499 points and 2017 km² of land area are at low risk (see Table3, Fig1(i), and Fig2(i)).
Fig 1 Point spatial distribution of soil heavy metals (a-As; b-Cd; c-Cr; d-Cu; e-Hg; f-Ni; g-Pb; h-Zn; i-RI; j-PInemerow; k-CSI)

3.2.3 Nemerow Pollution Index (PINemerow)
The results for the Nemerow Pollution Index (PINemerow) for the study area are presented in Table 3 (according to the PINemerow grades shown in Table 1). There are a total of 150 points comprising 802 km² of land area at or above the Nemerow warning limit. A total of 89 points representing 196 km² of land area exhibit slight pollution according to this index (see Table 3, Fig1(j), and Fig2 (j)).

3.2.4 Contamination Severity Index (CSI)
Using formulas (1), (2), and (3) and the obtained values of PCA, the weighted values for heavy metals were calculated. The weighted values for Cu, Zn, Cr, Ni, Pb, Cd, As and Hg are 0.146, 0.180, 0.208, 0.212, 0.033, 0.111, 0.053, and 0.057, respectively.

The results of the Contamination Severity Index (CSI) for the study area are shown in Table 3 (based on the CSI grades shown in Table 1). There are 272 points comprising 997.29 km² of land with
low contamination severity, 213 points comprising 992.87 km$^2$ of land with low contamination severity, 10 points comprising 24.57 km$^2$ of land show low to moderate contamination severity, and only 4 points comprising 2.27 km$^2$ of land exhibit moderate contamination severity (see Fig 1 (k)). The spatial distribution of soil heavy metals CSI is shown in Fig 2 (k).

3.3 Risk assessment zoning
According to the risk assessment zoning solution (Table 2), the study area was divided into four grades: no risk zone, very low risk zone, low risk zone, and intermediate risk zone. The spatial distributions of four grades are shown in Fig3.

(1) Grade I: The area of the no risk zone is 758.31 km$^2$ or 37.59% of the total study area.
(2) Grade II: The area of the very low risk zone has the largest land coverage, covering 1037.5 km$^2$ or 51.44% of the total study area.
(3) Grade III: The area of the low risk zone is 218.91 km$^2$, encompassing 10.85% of the total study area.
(4) Grade IV: The area of the intermediate risk zone covers only 2.28 km$^2$, which is 0.12% of the total study area.

Areas in the no risk and very low risk zones total 1795.81 km$^2$, which is 89.03% of the study area. The low risk zones are distributed as noncontiguous patches. Lands in the intermediate risk zone are scattered in the middle part of Qixia.

Fig3 Zones of land safe utilization based on PPRC

3.4 Land use strategy
Results of the assessment show that the areas of with high PI Nemerow values are distributed in the north and south-central regions where the content of Cu and Cd is high. Qixia is a main apple producing area where large amounts of the traditional Bordeaux mixture (a fungicide) have been applied by farmers. Due to this, a high concentration of Cu has accumulated in the soils. Additionally, the use of phosphate fertilizers, plastic or reflective films and the influence of chelation and adsorption of soil organic matter result in a slightly higher Cd concentration. A copper-based nutrition protective agent to reduce the accumulation of copper is now being manufactured. Using this agent rather than the traditional Bordeaux mixture not only can meet the copper requirements of the plants during the entire growth period but can also reduce the accumulation of copper in soil.

The spatial distribution of no risk zones in Qixia is shown in Fig3. No risk zones and very low risk zones take up most of the study area; the low risk zones are distributed in the form of noncontiguous patches; lands in intermediate risk zones are scattered in the middle part of the study area. Generally, Qixia is an area of typical nonpoint source pollution. Different zones have different characteristics and require different methods and protection measures.

(1) Grade I: A priority protection strategy is adopted in no risk zones. Strict precautions are taken against pollution and to maintain a safe soil environment.
(2) Grade II: A protection strategy is adopted in very low risk zones. A thorough investigation of pollution sources is performed. Pollution sources are controlled, and dynamics are monitored.

(3) Grade III: A comprehensive monitoring strategy is used in low risk zones. Constant monitoring of soil, water, and atmospheric pollution are employed. A thorough investigation of pollution sources is conducted along with strict control of these pollution sources. These pollution source include pesticides containing Cu, phosphate fertilizers, plastic films and reflective films. Repairing soil environmental quality is advised.

(4) Grade IV: A warning and control strategy is adopted in intermediate risk zones. Soils, water, and atmospheric pollution are constantly monitored. Warnings regarding pollution levels are issued. Thorough investigations of pollution sources are conducted along with strict control of pollution sources. Repair of the soil environmental quality must be enforced.

4. Conclusion
In this study, using an innovative risk assessment zoning solution based on the soil heavy metal standards evaluation system (PPRC) composed of the Single Pollution Index (Pi), the Nemerow Pollution Index (PI\textsubscript{Nemerow}), the Potential Ecological Risk (RI), and the Contamination Severity Index (CSI), Qixia County was classified into four grades as follows: no risk zones, very low risk zones, low risk zones, and intermediate risk zones. We determined that, risk assessment zoning solutions based on PPRC provide reasonable risk assessment zones, which are beneficial to conduct differential control strategies which are based on different zone classifications. This paper provides an important method for ensuring the rational utilization and protection of land resources by conducting regional land safety use evaluations and zoning studies. The results in this paper show only statistical analyses from the results of the Pi, PI\textsubscript{Nemerow}, RI, and CSI pollution indices. A classification scheme for safety utilization was then proposed after qualitative analysis. Studies based on quantitative classification schemes of soil safety utilization based on larger amounts of data should be continued in the future.

Acknowledgements
This work was supported by the National Natural Science Foundation of China (No.41671346).

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