Heavy metal pollution comprehensive evaluation of contaminated soil in lead-zinc mining area based on the fuzzy mathematics

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Abstract. A new comprehensive evaluation method of heavy metal pollution based on the fuzzy mathematics was constructed. The results showed that mean concentrations of the heavy metals in the contaminated soils decreased in the order of Zn > Pb > Cd > Cu > As > Ni >Cr. Leaching experiment results showed that concentration of heavy metal in leaching agent of soil is not proportional to the content of heavy metal in soil. Through fuzzy comprehensive evaluation, the solid soil environmental quality grade was heavy pollution and leaching liquid water quality standards was grade V.

1. Introduction

Heavy metal, as a kind of persistent potentially toxic pollutants, once be released into the environment can not be biodegradable and exist in environment for a long time, which is one of the most serious environmental problems (Zhao et al. 2014). More attention are paid to metal pollution, because of heavy metals’ persistence and hazardous effects on the environment and human health (Bhargava et al. 2012). Therefore, the objective and practical comprehensive evaluation of the pollution is of great significance.

The fuzzy evaluation method based on fuzzy mathematics, quantifies some obscure boundary and not easy to quantitative factors. It can reflect objective existence fuzziness and uncertainty factors, which is widely used in the assessment of water quality (Soenmez et al. 2013) and water classification (Wang et al. 2013). The fuzzy comprehensive evaluation was used in many other respects. A fuzzy synthetic evaluation model with four air pollutants was designed to evaluate air quality (Gorai et al. 2014); Jiang introduced fuzzy mathematics into urban ecological security evaluation and established an evaluation model (Jiang et al. 2011). Zhao applied the membership function of fuzzy synthetic evaluation to assess the multi-metals comprehensive accumulation ability of the trees (Zhao et al. 2014). Through these researches it can be discover that fuzzy evaluation research mainly aimed at making the evaluation of the single way of pollution, however, the fuzzy evaluation of composite contamination is rare relatively. In this research, a new pollution comprehensive evaluation method of heavy metal based on the fuzzy mathematics was constructed, and the mixture of slag and contaminated soil as pollution source was evaluated more objectively and practically which can provide more reasonable basis for pollution protection and management.
2. Materials and methods

2.1. Research materials
The study was conducted in Maxipu in Yuanling County, Huaihua City in Hunan province (Fig. 1). There is a Pb/Zn mine, named jingshi mining. Initially, the land around the mine was relatively flat and was cultivated with rice every year. However, some lands were left uncultivated because of the low productivity caused by pollution. Sampling was conducted randomly in an area around the mine, with 8 composite soil samples marked as S1-S8. Four sub-samples were collected in each sample site and the sampling depth was between 0-20 cm.

![Fig. 1 Location of investigation area and sampling sites](image)

Soil samples collected were stored in polyethylene bags for transport and storage, and were air-dried, then sieved through a 2.0-mm mesh sieve to remove stones, coarse materials and other debris. After that, the samples were placed in sealed polyethylene container, labeled and wait to use.

2.2. methods
Accurately weighed sample powder 0.2 g, take samples and placed in Pte digestion pot, add 2-3 drops of deionized water to wetting, add 6 ml nitric acid, 6 ml hydrofluoric acid and 2 ml hydrochloric acid. After microwave digestion, cooling add to 50 ml with 1% nitric acid, save at 4°C.

The mixed sample was quartered. Then each part was divided into two parts, one for microwave digestion and one for leaching experiments. Leaching experiments of the sample was conducted referring to HJ557-2009 《Solid waste-Extraction procedure for leaching toxicity-Horizontal vibration method》. Add mixed acid (m(H₂SO₄):m(HNO₃)=2:1) to deionized water (1 L water about 2 drops of mixture), ensure pH=3.20±0.05. Accurately weighed soil powder 0.2 g and placed in extraction flask, add 100 mL extracting agent, overturn and vibration for 18±2h (at 23±2 °C), rotate speed is 30±2 r/min, after which collect the supernatant, then membranes and collect the leach liquor, saved at 4 °C and wait to use. Set up three groups of parallel experiments and two groups of blank tests.

2.3. XRD analysis
The voltage was 40 KV and the current was 40 mA. The 2θ step of 0.02°/min with scanning range of 10.00°~80.00° and a longer dwell time than normal were used to reduce interferences from background noise and to increase the counting statistics. Also peak/background ratios can be increased by using longer dwell times.

2.4. Construction of fuzzy comprehensive evaluation model

2.4.1. Establishment of sets. The factors set \( U = \{ u_1, u_2, \ldots, u_m \} \) is established as a collection of factors that affect evaluation object. The evaluation set \( V = \{ v_1, v_2, \ldots, v_n \} \) is a collection of environmental quality levels. The weight set \( A = \{ a_1, a_2, \ldots, a_m \} \) is a collection of factor weight,
which is determined by the value of the pollution degree. Where, \( \Sigma a_i = 1, a_i \geq 0 \), m is the number of selected environment factors, n is the number of environment quality standard grades, and \( a_i \) is weight which can be calculated by formula (1).

\[
a_i = \frac{c_i/s_i}{\sum_{k} c_k/s_k}
\]  

(1)

where, \( c_i \) is measured concentration of factor i, \( s_i \) is average of grading standards of contamination factor i.

2.4.2. Determining fuzzy relationship matrix. The membership grades are calculated by membership function based on the measured concentration values, which constitutes the corresponding relationship matrix shown as the formulation (2). If there are m evaluation factors attaching to n membership grades, it can constitutes matrix R (order of the matrix is m\( \times \)n).

\[
R = \begin{bmatrix}
r_{11} & r_{12} & \cdots & r_{1n} \\
r_{21} & r_{22} & \cdots & r_{2n} \\
\vdots & \vdots & \ddots & \vdots \\
r_{m1} & r_{m2} & \cdots & r_{mn}
\end{bmatrix}
\]

(2)

The membership grade can be calculated by classification standards and environmental pollution values. (Li et al. 2008). Assuming the actual contaminated concentration of the factor i is \( x \), then membership grade of the contaminated factor in each environmental quality level can be calculated as formula (3).

\[
r_{i(x)} = \begin{cases}
\frac{1}{e(2) - e(1)} & x \leq e(1) \\
0 & e(1) < x < e(2) \\
\frac{e(1) - e(2)}{e(3) - e(2)} & x \geq e(2)
\end{cases}
\]

(3)

Where, \( r_{i(x)} \) is the membership grade of environmental quality \( x \) for quality standard i; \( e(i) \) is classification standard value.

2.4.3. Comprehensive evaluation computing. Composite operations are done between fuzzy weight vector A and fuzzy relation matrix R, namely fuzzy comprehensive evaluation vector B, where, \( b_j \) is assessment index which is the degree of membership of the evaluation object for the element j in the evaluation set when considering the impact of all factors. Fuzzy comprehensive evaluation vector B is calculated as formula (4),

\[
B = A \cdot R = A = \{a_1, a_2, \ldots, a_m\} \cdot \begin{bmatrix}
r_{11} & r_{12} & \cdots & r_{1n} \\
r_{21} & r_{22} & \cdots & r_{2n} \\
\vdots & \vdots & \ddots & \vdots \\
r_{m1} & r_{m2} & \cdots & r_{mn}
\end{bmatrix} = \{b_1, b_2, \ldots, b_m\}
\]

(4)

\( b_j \) is the degree of membership of level j of environmental quality standard for comprehensive environmental classification index. Calculation model B [27] is expressed as:
B is normalized according to the principle of maximum membership degree of fuzzy math, and select the greatest level as rating of environmental quality. When the operation result is in the emergence of two identical (or nearly equal) maximum value, level is determined in accordance with the principles of the second largest level.

3. Results and analysis

3.1. Concentration and correlation of heavy metals in the soil

The maximum value, minimum value, average value and standard values of deviation are listed in Table 1. As the results shown, the average concentrations of Cr, Ni, Cu, Zn, As, Cd and Pb were 9.25, 37.96, 191.29, 3606.98, 120.64, 342.44, 1260.38 mg·kg\(^{-1}\). The highest concentration of heavy metals in the soil is Zn, and the lowest is Cr. Mean concentrations of the heavy metals in the contaminated soils decreased in the order of Zn > Pb > Cd > Cu > As > Ni > Cr.

![Table 1](image)

**Table 1.** Concentrations of heavy metal in the lead-zinc mining area

|       | Cr    | Ni    | Cu    | Zn    | As    | Cd    | Pb    |
|-------|-------|-------|-------|-------|-------|-------|-------|
| Max   | 17.52 | 52.81 | 360.69| 6311.57| 323.18| 831.33| 3265.33|
| Min   | 0.98  | 23.11 | 21.88 | 902.39| 32.85 | 185.45| 403.26|
| Avg   | 9.25  | 37.96 | 191.29| 3606.98| 120.64| 342.44| 1260.38|
| SD    | 5.90  | 10.54 | 112.55| 1758.30| 93.30 | 200.47| 921.92|
| CV    | 63.78%| 27.77%| 58.84%| 48.75% | 77.34%| 58.54%| 73.15%

Max: Maximum; Min: Minimum; Avg: Average; SD: Standard deviation; CV: Coefficients of variation (SD/average · 100%).

The results from correlative analysis showed that combined heavy metal pollution exists in the soil. The highest correlation coefficient between different heavy metals was Cd and Pb (The correlation coefficient = 0.952). Significant positive correlations existed between the seven elements (Table 2), which indicated that the heavy metal pollution was led to by the same pollution source. Cu and Cd were main associated components of Pb/Zn mine, furthermore, the different kinds of heavy metals had greater significant positive correlation, therefore, heavy metal contamination of the soil around the lead-zinc mining area mainly originated in Pb/Zn smelting production.

![Table 2](image)

**Table 2.** Pearson correlations between difference heavy metal elements in the soils

|       | Cr    | Ni    | Cu    | Zn    | As    | Cd    | Pb    |
|-------|-------|-------|-------|-------|-------|-------|-------|
| Cr    | 1     |       |       |       |       |       |       |
| Ni    | .800* | 1     |       |       |       |       |       |
| Cu    | .916**| .912**| 1     |       |       |       |       |
| Zn    | .889**| .823* | .872**| 1     |       |       |       |
| As    | .827* | .755* | .846**| .794* | 1     |       |       |
| Cd    | .945**| .759* | .881**| .859**| .936**| 1     |       |
| Pb    | .928**| .752* | .916**| .895**| .937**| .952**| 1     |

Correlation is significant at the ** 0.01 or * 0.05 level (two-tailed). (n = 8)

3.2. Results of leaching experiment

Heavy metals are determined after microwave digesting of residue which is in the lead-zinc slag pile area and the test results are shown in Table 3.
Table 3. Concentration of heavy metal in the soils (mg/kg)

| Number | 52(Cr) | 60(Ni) | 65(Cu) | 66(Zn) | 75(As) | 111(Cd) | 206(Pb) |
|--------|--------|--------|--------|--------|--------|---------|---------|
| N1     | 8.63   | 30.31  | 120.32 | 3311.41| 103.92 | 428.51  | 1365.05 |
| N2     | 5.35   | 33.65  | 108.33 | 2985.86| 102.35 | 383.05  | 1230.63 |
| N3     | 4.08   | 27.92  | 119.38 | 2919.49| 98.33  | 400.53  | 1209.26 |
| average| 6.02   | 30.63  | 116.01 | 3072.25| 101.53 | 404.03  | 1268.31 |

Leaching experiments were operated referring to standard HJ557-2009 "level oscillation for leaching toxicity of solid waste". The supernatant was collected, extract was obtained after filtration membrane, contents of heavy metals were determined by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) and Atomic Fluorescence Spectrometry (AFS). The results are shown in Table 4.

Table 4. Concentration of heavy metal in leaching agents (mg/L)

| Number | 52(Cr) | 60(Ni) | 65(Cu) | 66(Zn) | 75(As) | 111(Cd) | 206(Pb) |
|--------|--------|--------|--------|--------|--------|---------|---------|
| N1     | 8.64E-04| 4.80E-03| 1.01E-02| 1.88E-01| 5.30E-03| 2.63E-01| 1.09E-03|
| N2     | 7.82E-04| 4.75E-03| 1.00E-02| 2.38E-01| 5.02E-03| 2.77E-01| 1.40E-03|
| N3     | 7.54E-04| 4.93E-03| 1.02E-02| 1.63E-01| 5.35E-03| 2.91E-01| 1.34E-03|
| average| 8.00E-04| 4.83E-03| 1.01E-02| 1.96E-01| 5.22E-03| 2.77E-01| 1.28E-03|

The leaching concentration of different heavy metals in the leaching agent and the heavy metal concentration of contaminated soil were compared, shown in Fig.3. The heavy metal with highest content in contaminated soil is zinc, up to 3072.25 mg/kg. Its leaching concentration in leach agent is 1.96E-01 mg/L, and the concentration is not the corresponding maximum value in concentrations of heavy metals. The heavy metal with highest content in leaching agent is cadmium, and its concentration is 2.77E-01 mg/L. The concentration of cadmium in contaminated soil is 404.03 mg/kg, and its total amount in soil is not the highest, indicating that concentration of heavy metal in leaching agent of soil is not proportional to the content of heavy metal in soil. The dissolubility of all metals was correlated to many factors (Milinovic.2014; Guo.et al.2013; Wang et al.2012).

![Fig. 2 Comparison of heavy metals concentrations in residue and leaching agent](image-url)
3.3. XRD analysis
The analysis showed that main minerals of the sample included α-quartz, β-feldspar potassian, χ-kaolinite, δ-montmorillonite, etc. Fig. 4 shows the X-ray diffraction of the soil around lead-zinc mine. The most abundant inorganic mineral is quartz. It is the most stable silicate which is namely silicon dioxide (SiO₂). Its crystal is oxide mineral with trigonal system. Its adsorption capacity on metal ion is weak. Feldspar group mineral is silicate with frame structure which has obvious mineral characteristics of pore structure. Kaolinite is clay mineral, which is mainly the product of natural alteration of feldspar and other silicate minerals. Montmorillonite crystal is silicate mineral with hydrous layered structure which is belong to monoclinic.

Fig. 3 The X-ray diffraction patterns of soils

3.4. Results of solid contamination level evaluation
The soil contamination level was assessed using fuzzy evaluation method. Table 5 "Level standard of soil heavy metal pollution" was used as a single factor score sheet when evaluating. The soil contamination level was assessed using fuzzy evaluation method. Table 5 "Level standard of soil heavy metal pollution" was used as a single factor score sheet when evaluating.

Table 5. Level standard of heavy metal pollution

| Pollution factor | Clean | Still clean | Light pollution | Moderately pollution | Severe pollution | Average of grading standards |
|------------------|-------|-------------|-----------------|----------------------|-----------------|------------------------------|
| 52(Cr)           | 74.88 | 99.54       | 150             | 350                  | 500             | 234.88                       |
| 60(Ni)           | 77.51 | 82.56       | 94.21           | 102.56               | 154.25          | 102.22                       |
| 65(Cu)           | 28.37 | 40.63       | 120             | 280                  | 400             | 173.80                       |
| 66(Zn)           | 83.68 | 116.75      | 240             | 560                  | 800             | 360.09                       |
| 75(As)           | 82.56 | 95.43       | 124.26          | 164.28               | 201.38          | 133.58                       |
| 111(Cd)          | 0.12  | 0.25        | 0.60            | 1.40                 | 2.00            | 1.03                         |
| 206(Pb)          | 23.35 | 36.09       | 150             | 350                  | 500             | 211.89                       |

3.4.1. Standard allowable values and weights. The weight of single factor index in water quality assessment sub-index is not directly related to grading standard of a certain factor, so intermediate standard or average of five level standards can be defined as the demarcation point between "clean" and "pollution". The average of standard levels was taken as the allowable value of standard si in this study, and the calculation results are shown in Table 6.
Table 6. Results of weight calculation

| Item | 52(Cr) | 60(Ni) | 65(Cu) | 66(Zn) | 75(As) | 111(Cd) | 206(Pb) |
|------|--------|--------|--------|--------|--------|---------|---------|
| S(mg/kg) | 234.88 | 102.22 | 173.8 | 360.09 | 133.58 | 1.03 | 211.89 |
| Ci/Si | 0.03 | 0.30 | 0.67 | 8.53 | 0.76 | 392.26 | 5.99 |
| Weight $a_i$ | 6.27E-05 | 7.33E-04 | 1.634E-03 | 0.02 | 1.86E-03 | 0.96 | 0.01 |

Thus available, weight set $A = \{a_1, a_2, \ldots, a_m\} = \{6.27E-05, 7.33E-04, 1.634E-03, 0.02, 1.86E-03, 0.96, 0.01\}$.  

### 3.4.2. Fuzzy relationship matrix
According to the weight from the table above, membership of each environmental factor to each level of environmental standard can be obtained combining with the formula 3 given in 2.5.2, and fuzzy relation matrix $R$ can also be obtained.

$$R = \begin{bmatrix}
1 & 0 & 0 & 0 & 0 \\
1 & 0 & 0 & 0 & 0 \\
0 & 0.05 & 0.95 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 \\
0 & 0.79 & 0.21 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 \\
0 & 0 & 0 & 0 & 1 \\
\end{bmatrix}$$

### 3.4.3. Comprehensive evaluation computing
According to the formula of fuzzy comprehensive evaluation vector $B$ given in 2.5.3, fuzzy comprehensive evaluation vector $B$ is got by composite operating combining with the fuzzy weight vector $A$ calculated in 2.5.1 and fuzzy relationship matrix $R$ calculated in 2.5.2, and $B = A \cdot R = \{7.96E-4, 1.55E-03, 1.94E-03, 0.00, 0.99\}$, after normalization, $B = A \cdot R = \{8.04E-4, 1.57E-03, 1.96E-04, 0.00, 1.00\}$. From the results, membership of clean is 8.04E-04; membership of still clean is 1.57E-03; membership of light pollution is 0.00; the highest membership of comprehensive evaluation vector is 1.00 which is heavy pollution. Therefore, its environment quality rating is heavy pollution.

### 3.5. Results of leaching agent quality standards evaluation
Impact of leaching agent of residue in Lead-zinc slag pile area on water quality was assessed using fuzzy evaluation method. "Quality standard of surface water environment" was used as a single factor score sheet when evaluating. Quality standard of surface water environment of associated heavy metals in leaching agent are shown in Table 7.

Table 7. Quality standard limits of surface water environment (mg/L)

| Pollution factor | Clean | Still clean | Light pollution | Moderately pollution | Severe pollution | Average of grading standards |
|------------------|-------|-------------|-----------------|----------------------|-----------------|-----------------------------|
| 52(Cr)           | 0.01  | 0.05        | 0.05            | 0.05                 | 0.1             | 0.052                       |
| 60(Ni)           | 0.02  | 0.02        | 0.02            | 0.02                 | 0.02            | 0.02                        |
| 65(Cu)           | 0.01  | 1           | 1               | 2                    | 2               | 1.202                       |
| 66(Zn)           | 0.05  | 1           | 1               | 2                    | 2               | 1.21                        |
| 75(As)           | 0.05  | 0.05        | 0.05            | 0.1                  | 0.1             | 0.07                        |
| 111(Cd)          | 0.001 | 0.005       | 0.005           | 0.005                | 0.01            | 0.0052                      |
| 206(Pb)          | 0.01  | 0.05        | 0.05            | 0.05                 | 0.1             | 0.052                       |

Note: No grading standard data of nickel, referring to the standard limits of nickel in surface water.
3.5.1. **Standard allowable values and weights.** As described in 3.4.1, intermediate standard or average is defined as the demarcation point. The average of standard levels was taken as the allowable value of standard $si$ in this study, and the weight of each element is shown in Table 8.

| Item       | 52(Cr)   | 60(Ni)   | 65(Cu)   | 66(Zn)   | 75(As)   | 111(Cd)  | 206(Pb)  |
|------------|----------|----------|----------|----------|----------|----------|----------|
| S(mg/kg)   | 5.20E-02 | 2.00E-02 | 1.20E+00 | 1.21E+00 | 7.00E-02 | 5.20E-03 | 5.20E-02 |
| Cu/Si      | 1.54E-02 | 2.41E-01 | 8.42E-03 | 1.62E-01 | 7.46E-02 | 5.32E+01 | 2.45E-02 |
| Weight ai  | 2.86E-04 | 4.49E-03 | 1.57E-04 | 3.02E-03 | 1.39E-03 | 9.90E-01 | 4.56E-04 |

Table 8. Results of weight calculation

Thus available, weight set $A=\{a_1, a_2, \ldots, a_m\} = \{2.86E-04, 4.49E-03, 1.57E-04, 3.02E-03, 1.39E-03, 9.90E-01, 4.56E-04\}$.

3.5.2. **Fuzzy relationship matrix.** According to the weight from the table above, membership of each environmental factor to each level of environmental standard can be obtained combining with the formula 4 given in 2.5.3, using fuzzy relation matrix $R$ to express.

$$
R = \begin{bmatrix}
1 & 0 & 0 & 0 & 0 \\
1 & 0 & 0 & 0 & 0 \\
0.9999 & 0.001 & 0 & 0 & 0 \\
0.31 & 0.19 & 0 & 0 & 1 \\
1 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 \\
1 & 0 & 0 & 0 & 0
\end{bmatrix}
$$

3.5.3. **Comprehensive evaluation computing.** According to the formula of fuzzy comprehensive evaluation vector $B$ given in 2.5.4, fuzzy comprehensive evaluation vector $B$ is got by composite operating combining with the fuzzy weight vector $A$ calculated in 2.5.1 and fuzzy relationship matrix $R$ calculated in 2.5.2, and $B = A \cdot R = \{9.22E - 03, 5.74E - 04, 0.00E - 0, 0.00E - 0, 9.93E - 01\}$. After normalization, $B = A \cdot R = \{9.28E - 03, 5.78E - 04, 0.00, 0.00, 1.00\}$. Membership of level I in $B$ is 9.28E-03; membership of level II is 5.78E-04; the highest membership of comprehensive evaluation vector is 1.00 which is belong to V water body. Therefore, its water quality standard is level 5.

4. **Conclusion**

(1) The results showed that mean concentrations of the heavy metals in the contaminated soils decreased in the order of $Zn > Pb > Cd > Cu > As > Ni > Cr$. The significant positive correlations existed between the seven elements, which indicated that the heavy metal pollution was leaded to by the same pollution source.

(2) The mixture of slag and heavy metal contaminated soil in the lead-zinc mining area was analyzed by xrd and the results showed that component is complicated including quartz, feldspar, kaolinite and montmorillonite, etc, the maximum inorganic mineral composition is SiO$_2$.

(3) After the fuzzy comprehensive evaluation of the heavy metal contaminated soil, soil pollution levels and leaching liquid environment quality standards were evaluated synthetically. The results shows that the fuzzy comprehensive evaluation vectors of soil pollution levels and leaching liquid environment quality standards are $B = A \cdot R = \{9.22E - 03, 5.74E - 04, 0.00, 0.00, 9.93E - 01\}$, $B = A \cdot R = \{8.04E - 04, 1.57E - 03, 1.96E - 04, 0.00, 1.00\}$ respectively. Through analyzing, the soil environmental quality grade is heavy pollution and leaching liquid water quality standards is grade 5, belonging to V class water.

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