Spatial Variation and Assessment of Heavy Metal and Radioactive Risk in Farmland around a Retired Uranium Mine

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Abstract. In recent years, heavy metal contamination in the environment has been attracted worldwide attention due to their toxicity, persistence, extensive sources and non-biodegradable properties. We herein investigate variation trend and risk of heavy metal and radiation distribution in the former mine stope, former mineral ore stockyard, and mine road with surface soils of a retired uranium mine in the mid-south of China. The mean concentrations (mg/kg) of Pb, Cd, Cu, Zn, As, Hg, Cr, Mn, Ni, U, and $^{232}$Th were analyzed according to the corresponding background values in Hunan, China. The Geo-accumulation index ($I_{geo}$) were used for the assessment of pollution level of heavy metals and the radioactive elements of U and $^{232}$Th. Then, Pollution load index (PLI) and GIS technique were integrated to assess spatial distribution of heavy metal contamination and radioactive contamination. Results confirmed that three areas in the retired uranium mine was a primary source of pollution, which showed anthropogenic origin mainly from agricultural runoff, hydrometallurgy from chemical industries, radioactive tailings, and electroplating industries finally drained into Zishui River and Xiangjiang River. Based on the actual situation, some suggestions were put forward for the treatment of the retired uranium mine in conclusion.

1. Introduction
In recent years, heavy metal and radioactive elements contamination in the environment has been attracted worldwide attention due to their toxicity, persistence, extensive sources and non-biodegradable properties [1]. The agricultural soil around the mining area, and the quantities are more than the carrying capacity of heavy metals. The food chain can serious impact one the health of the human body, and ultimately cause health risks to the human body[4]. The result of the principal
components (PCs) and the factor analyze (FA) showed that the anthropogenic activities are primary sources of soil contamination with heavy metals and radioactive elements. Combined with multivariate statistical analysis and geographic information system (GIS) technology[7], we investigated the three regions around the retired uranium mine of heavy metal and radioactive pollution sources in the mid-south China. Specifically, the heavy metal and radioactive element was sampled and analyzed in the former mine stope, former mineral ore stockyard and mine road. Therefore, the following three aims are considered in this study.(1) analyzing pollution status of nine heavy metals (Pb,Cd,Cu,Zn,As,Hg,Cr,Mn, and Ni) and two natural radioactivity (U and $^{232}$Th), (2) using potential geo-accumulation index($I_{geo}$), and combined pollution load index (PLI) and GIS to assess spatial variation of heavy metal contamination and (3) using the Pearson's coefficient to analysis the correlation with factor analysis (FA) between the heavy metal elements and radioactive elements.

2. Materials and Methods

2.1. Study Area

The uranium mine, retired in 1996, is located in Hunan Province, middle and southern part of China. The type of ore deposit belongs to the hard rock type uranium deposit in the little hilly area with the elevation of 200-300m. The average annual temperature is 18 - 20°C, and the mean annual precipitation is 1286-1372mm. The polluted surface soil contains heavy metal elements, such as Pb, Zn, Cd, and so on, and radioactive elements of U and $^{232}$Th. The corroded soil form farmland flows into the river, lies in the south of the study area.

![Figure 1](image_url)

**Figure 1.** Study area and sampling sites of surface soils in farmland along a retired uranium mine.

2.2. Sampling

In Spring season of 2016, a total of ten sites were collected in the farmland around the retired (Figure 1). In the former mine stope, former mineral ore stockyard and mine road collected nearly 60 soil samples. In each samples sites, the former mine stope, former mineral ore stockyard and mine road collected respectively 24, 16 and 18 soil samples. S1, S2, and S3 were located in mine stope. S4, S5, S6 and S7 were located in mineral ore stockyard. S8, S9, and S10 were located in mine road. The polyethylene bags were packed and sent back to the laboratory for cold storage.

2.3. Analytical methods and quality control
The sample was dried up in an oven at 100°C for 24 h, until constant weight sieved with 40 mesh sieve for homogenization, and homogenized and separated into two parts. A sample of 0.5 g was weighed and transferred to airtight Teflon vessels and added by HNO₃, HF, HClO₄ which were digested with a mixture. The other sample of 328.0 g was weighed and sealed in airtight polyethylene cylindrical container, and then placed for 30 days to reach secular equilibrium before further gamma-ray measurement. Inductively coupled plasma-mass spectrometry (ICP-MAS), followed by the Chinese DZ/T0223-2001, was used to analyze Pb, Cd, Cu, Zn, Cr, Ni. Atomic fluorescence spectrometry (AFS) was used to analyze As, Hg, Mn. Gamma spectrometry (GS) was used to analyze ²³²Th and U, follow by the Chinese GB/T 11743-2013.

In the analysis process, the quality control of the soil composition analysis standard materials (GBW07401, Institute of Geophysical and Geochemical Exploration) was added to meet the quality control requirements. The analytical precision was conducted with the repetitive rate of 10%.[8] The results met the quality control requirement of The Technical Specification for Soil Environmental Monitoring (HJ/T 166-2004) and Determination of radionuclides in soil by gamma spectrometry (GB/T 11743-2013).

2.4. Geo-accumulation index \( I_{geo} \)

\( I_{geo} \) index was introduced by Muller(1969).[9] It is a geochemical criterion to evaluate heavy metal pollution and radioactive pollution in uranium mining area. The equation is also considered the background value change which caused by the natural diagenesis. The \( I_{geo} \) was calculated by the equation [10]:

\[
I_{geo} = \log_2 \left( \frac{C_n}{1.5B_n} \right)
\]

Where \( C_n \) is the measured concentration of element (n), mg/kg and \( B_n \) is the geochemical background value of the element (n), mg/kg. In the equation, average values were used, and 1.5 is the factor used for lithologic variations of trace metals. According to the soil background values in Hunan(1988), background values of heavy metal and radioactive contents in a uranium mine were used as \( B_n \) for calculation. The level classification of \( I_{geo} \) is defined as unpolluted (<0), unpolluted to moderately polluted (0-1), moderately polluted (1-2), moderately to strongly polluted (2-3), strongly polluted (3-4), strongly to super strongly polluted (4-5) and super-strongly polluted (>5).

Table 1. Summary statistics of heavy metal and radioactive concentrations in the retired uranium mine, compared with guide values of China(mg/kg, dry weight).

|             | Pb   | Cd   | Cu    | Zn   | As   | Hg   | Cr   | Mn   | Ni   | ²³²Th | U   |
|-------------|------|------|-------|------|------|------|------|------|------|-------|-----|
| Former mine stope |      |      |       |      |      |      |      |      |      |       |     |
| Min         | 173.8| 1.78 | 38.33 | 280.5| 18.1 | 165  | 29.6 | 300.5| 3.8  | 18.82 | 16.2|
|             | 4    | 2    | 3     | 4    |      |      |      |      |      | 3     |     |
| Max         | 2319 | 22.5 | 228.8 | 4173 | 226. | 1881 | 60.6 | 572.2| 18.2 | 84.62 | 217.|
|             | 0    | 4    | 9     | 2    | .76  | 6    | 1    | 6    | 2    | 22    |     |
| Mean        | 3568 | 10.8 | 119.6 | 2150.| 57.8 | 839. | 47.4 | 435.1| 12.3 | 27.89 | 101.|

\( \log (1/1.5) \)
|          | 57   | 3    | 6    | 49   | 6    | 69   | 5    | 4    | 5    | 46   |
|----------|------|------|------|------|------|------|------|------|------|------|
| S.D.     |      |      |      |      |      |      |      |      |      |      |
|          | 5854. | 6.06 | 57.97| 1250. | 45.3 | 554. | 8.05 | 74.31| 3.4  | 16.79| 53.3 |
|          | 71    | 2    | 32   | 7    | 34   |      |      |      |      |      |      |
| CV (%)   | 1.64  | 0.56 | 0.48 | 0.58 | 0.78 | 0.66 | 0.17 | 0.17 | 0.28 | 0.6  | 0.53 |

Former mineral ore stockyard

|          |      |      |      |      |      |      |      |      |      |      |
|----------|------|------|------|------|------|------|------|------|------|------|
| Min      | 145.7 | 0.63 | 21.8 | 184.8 | 15.8 | 113. | 31.2 | 333.4 | 4.17 | 22.29| 14.1 |
| Max      | 1737  | 10.2 | 157.2| 2548. | 92.1 | 509. | 64.7 | 504.6 | 17.0 | 60.01| 527. |
| Mean     | 2479. | 2.69 | 45.57| 655.0 | 37.9 | 225. | 47.1 | 429.7 | 11.9 | 30.47| 91.2 |
|          | 48    |      | 3    | 4    |      |      |      |      |      |      |      |
| S.D.     | 4948. | 2.73 | 41.5 | 720.8 | 19.9 | 107. | 8.96 | 45.12 | 3.98 | 10.18| 126. |
|          | 66    |      | 9    | 76   |      |      |      |      |      |      |      |
| CV (%)   | 2.0   | 1.01 | 0.91 | 1.1  | 0.53 | 0.48 | 0.19 | 0.11 | 0.33 | 0.33 | 1.39 |

Mine Road

|          |      |      |      |      |      |      |      |      |      |      |
|----------|------|------|------|------|------|------|------|------|------|------|
| Min      | 50.7  | 0.11 | 12.84| 63.98| 15.2 | 62.1 | 29.0 | 310.3 | 8.43 | 20.81| 6.88 |
| Max      | 1323  | 12.4 | 58.58| 1976 | 62.8 | 1082 | 56.3 | 494.4 | 17.1 | 28.12| 43.7 |
| Mean     | 370.7 | 2.79 | 30.37| 515.2| 43.3 | 300. | 43.8 | 398.8 | 11.6 | 23.63| 22.2 |
|          | 8     | 7    | 57   | 1    | 8    | 3    |      |      |      |      |      |
| S.D.     | 383.8 | 3.4  | 13.42| 567.4| 13.8 | 297. | 6.69 | 40.76 | 2.24 | 1.99 | 10.9 |
|          | 5     |      | 6    | 4    | 78   |      |      |      |      |      |      |
| CV (%)   | 1.04  | 1.22 | 0.44 | 1.1  | 0.32 | 0.99 | 0.15 | 0.1  | 0.19 | 0.08 | 0.49 |
| Mean of 3| 2275. | 6.09 | 71.51| 1230. | 47.8 | 502. | 46.2 | 422.3 | 12.0 | 27.28| 74.0 |
| fields   | 69    |      | 47   | 7    | 81   | 2    | 9    | 1    |      |      |      |
| Backgroun| 27    | 0.08 | 26   | 94   | 14   | 0.09 | 68   | 441   | 32   | 17   | 4.2  |
|          | 5     |      | 6    |      |      |      |      |      |      |      |      |

\(^a\)S.D.: standard deviation

\(^b\)CV: coefficient of variation

\(^c\)Background: According to the Soil background values in Hunan (1988)

\[\text{2.5. Pollution load index (PLI)}\]

Pollution load index (PLI) is a kind of evaluation method proposed by TOMLINSON[12] in the classification of heavy metal pollution levels[13]. Contamination factor(CF) is the ratio of measured
concentration by background value. For the entire sampling site, PLI has been determined as the nth root of the product of the nCF [14]:

\[ CF = C_n / B_n \]  

\[ PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n)^{1/n} \]  

The evaluation steps include the calculation of the highest pollution coefficient and the calculation of the pollution load index, and to provide a straightforward and comparative method for evaluating the level of heavy metal and radioactive contamination. The level classification of PLI is defined as unpolluted (<1), moderately polluted (1-2), strongly polluted (2-3) and super-strongly polluted (>3).

2.6. Multivariate statistical and GIS-based analyses

In this study, we used the Pearson correlation coefficient to reflect the linear relationship between variables. All the statistical analyses were performed using SPSS 18.0 for Windows. ArcGIS 10.0 software was used to analyze the spatial variations of heavy metal and radioactive elements [16].

3. Results and discussion

3.1. Soil contamination characteristics

The mean contents of Pb, Cd, Cu, Zn, As, Hg, Cr, Mn, Ni, 232 Th, and U in three areas were presented in Table 1, along with background values in Hunan, China. In surface soils of a uranium mine, heavy metal elements and radioactive elements were higher than the background values, such as Pb, Cd, Cu, Zn, As, Hg, and U. Pb, Cd, Cu, Zn, Hg, and U in the former mine stope and the former mineral ore stockyard were approximately twice to forth as high as that of mine road. The pollution level of Pb, Cd, Cu, Zn, Hg, and U has increased the degree of pollution during the three areas studied in Figures 2 and Table 2. Hg was more than two times in the super-strongly pollution level. The same level of Cd pollution is not optimistic, and the serious is only less than Hg. Zn and U were increased quickly with all elements. Pb was in the moderately level. According to Igeo, average pollution level decreased following the order Hg > Cd > Pb > U > Zn > As > Cu > 232 Th > Mn > Cr > Ni.

![Figure 2. Geo-accumulation index (Igeo) of heavy metals and radiances in a retired uranium mine for three areas.](image-url)
Table 2. The level classification of Geo-accumulation index ($I_{geo}$) of heavy metals and radiations in a retired uranium mine for three areas.

|                | Former mine stope                  | Former mineral ore stockyard | Mine road                          | Mean                                |
|----------------|------------------------------------|------------------------------|------------------------------------|-------------------------------------|
| Pb             | Super-strongly polluted            | Strongly polluted            | Moderately to strongly polluted    | Strongly to super-strongly polluted |
| Cd             | Super-strongly polluted            | Strongly polluted            | Strongly polluted                  | Strongly to super-strongly polluted |
| Cu             | Moderately polluted                | Unpolluted                   | Unpolluted                         | Unpolluted to moderately polluted   |
| Zn             | Strongly polluted                  | Moderately polluted          | Moderately polluted                | Moderately to strongly polluted     |
| As             | Moderately polluted                | Unpollotted to moderately polluted | Unpollotted to moderately polluted | Unpollotted to moderately polluted  |
| Hg             | Super-strongly polluted            | Super-strongly polluted      | Super-strongly polluted            | Super-strongly polluted             |
| Cr             | Unpolluted                         | Unpolluted                   | Unpolluted                         | Unpolluted                          |
| Mn             | Unpolluted                         | Unpolluted                   | Unpolluted                         | Unpolluted                          |
| Ni             | Unpolluted                         | Unpolluted                   | Unpolluted                         | Unpolluted                          |
| $^{232}$Th     | Unpolluted                         | Unpollotted to moderately polluted | Unpollotted to moderately polluted | Unpollotted to moderately polluted  |
| U              | Strongly polluted                  | Strongly polluted            | Moderately polluted                | Moderately to strongly polluted     |

3.2. Spatial risk assessment of heavy metal and radioactive elements

Spatial risk map of heavy metal in a retired uranium mine was shown in Figures 3. To facilitate the analysis, with different colors to represent the various points in the extreme heavy pollution conditions over the multiple samples. In the former mine stope, it might be affected by the residue soil and hydrometallurgical mill which left the radioactive contamination and chemical pollution. In the former mineral ore stockyard, S4, S5, S6, were also the more severe contamination area, which was nearly the uranium deposit cave and hydrometallurgical mill. There are many ditches, and finally, import to Zishui River. Infected by rain and slag mucks, the heavy metal and radioactive were polluted around the farmlands. S7 is lower than S4, S5, and S6, due to it was far from the ditches. In mine road, S8, S9, and S10, the area were located nearly the battery factory and chemical plant. The heavy metal pollution was more serious than radioactive contamination. Similarly affected by the ditches, the pollution levels were only below the mine yard.
3.3. Correlation and factor analysis

Correlation analysis of heavy metal and radioactive in soils were shown in Table 3. In the former mine stope, a strong positive correlation between Pb, Cd, Cu, As, Cr, Mn, Ni, $^{232}$Th and U ($P \leq 0.01$) was presented. In the former mineral ore stockyard, a strong positive correlation between Cd, As, Hg, Cr, Mn, Ni, $^{232}$Th, and U is shown. In mine road, both all elements were found a definite positive. The $^{232}$Th and U were also found a strong positive correlation in three fields. They were randomly distributed in the surface soil and considered in association with other heavy metal elements.

![Spatial distribution maps using pollution load index (PLI) for heavy metal and radioactive risk in a uranium mine.](image)

**Figure 3.** Spatial distribution maps using pollution load index (PLI) for heavy metal and radioactive risk in a uranium mine.

**Table 3.** Pearson correlation matrix for heavy metal and radioactive concentration in a retired uranium mine.

|          | Pb   | Cd   | Cu   | Zn   | As   | Hg   | Cr   | Mn   | Ni   | $^{232}$Th | U   |
|----------|------|------|------|------|------|------|------|------|------|-----------|-----|
| Former mine stope |      |      |      |      |      |      |      |      |      |           |     |
| Pb       | 1    |      |      |      |      |      |      |      |      |           |     |
| Cd       | 0.274$^a$ | 1    |      |      |      |      |      |      |      |           |     |
| Cu       | 0.412$^a$ | 0.6$^a$ | 1    |      |      |      |      |      |      |           |     |
| Zn       | 0.419 | 0.96 | 0.64 | 1    |      |      |      |      |      |           |     |
| As       | 0.199$^a$ | 0.50 | 0.11 | 0.502 | 1    |      |      |      |      |           |     |
| Hg       | 0.083$^b$ | 0.54 | 0.44 | 0.493 | 0.399 | 1    |      |      |      |           |     |
| Cr       | -0.579 | -0.4 | -0.24 | -0.51 | -0.40 | -0.54 | 1    |      |      |           |     |
| Mn       | -0.113 | -0.38 | -0.15 | 0.267 | -0.17 | 0.256 | 1    |      |      |           |     |
| Ni       | -0.79$^a$ | -0.2 | -0.29 | -0.28 | -0.24 | -0.11 | 0.573 | 0.23 | 1    |           |     |
| $^{232}$Th | -0.232 | 0.97 | 0.09 | 0.299 | 0.26$^a$ | 0.10 | -0.06$^a$ | -0.29 | 0.332 | 1         |     |
| Element | Former mineral ore stockyard |   |   |   |   |   |   |   |   |   |
|---------|-------------------------------|---|---|---|---|---|---|---|---|---|
| U       | 0.456a                        | 0.75 | 0.86 | 0.781 | 0.295 | 0.40 | -0.30 | -0.11 | -0.29 | 0.33 | 1   |
|         | 4a                            | 2   | a   | 7a   | 4a   | 2a   | 5a   | 2a   |   |   |

**Mine road**

| Element |   |   |   |   |   |   |   |   |   |   |
|---------|---|---|---|---|---|---|---|---|---|---|
| Pb      | 1 |   |   |   |   |   |   |   |   |   |
| Cd      | 0.918 | 1 |   |   |   |   |   |   |   |   |
| Cu      | 0.988 | 0.96 | 1 |   |   |   |   |   |   |   |
|         | 1a |   |   |   |   |   |   |   |   |   |
| Zn      | 0.951 | 0.99 | 0.97 | 1 |   |   |   |   |   |   |
|         | 4a | 9a |   |   |   |   |   |   |   |   |
| As      | 0.089 | 0.19 | 0.11 | 0.176 | 1 |   |   |   |   |   |
|         | 1a | a |   |   |   |   |   |   |   |   |
| Hg      | 0.784 | 0.94a | 0.84 | 0.918 | 0.124 | 1 |   |   |   |   |
|         | 8a | b | a |   |   |   |   |   |   |   |
| Cr      | -0.494 | -0.4 | -0.49 | -0.47 | -0.23b | -0.34 | 1 |   |   |   |
|         | 75a | 6   | 1a | 6a |   |   |   |   |   |   |
| Mn      | -0.504 | -0.5 | -0.52 | -0.5b | -0.15 | -0.29 | 0.763 | 1 |   |   |
|         | 15b | 1a | 5a | 1a | a |   |   |   |   |   |
| Ni      | -0.693 | -0.6 | -0.68 | -0.63 | -0.29 | -0.43 | 0.92a | 0.81 | 1 |   |
|         | 27a | 4a | 5a | 5a | 1a | 4a |   |   |   |   |
| $^{232}$Th | 0.915 | 0.79 | 0.88 | 0.843 | -0.06 | 0.66 | -0.4a | -0.35 | -0.54 | 1 |
|         | 7a | 6   | a | 6a | 5a | 2a |   |   |   |   |
| U       | 0.958 | 0.8a | 0.91 | 0.85a | 0.107 | 0.62 | -0.49 | -0.43 | -0.68 | 0.93 | 1 |
|         | 3   | 9a | 9a | 1b |   |   |   |   |   |   |
Correlation is significant at the 0.01 level (2-tailed)
Correlation is significant at the 0.05 level (2-tailed)

Loading of the principal components (PCs) for three areas were shown in Figure 4a-4c. In the formelmire stope, PC1 (53.1% of the total variance) was dominated mainly by Zn, Cd, and Hg, and the origin could be from chemical industries. PC2 (35.6% of the total variance) was dominated primarily by Cu, As, and Pb, the origin could be from agricultural runoff. PC3 (17.2% of the total variance) was dominated mainly by U and $^{232}$Th. The origin could be from radioactive tails. In the former mineral ore stockyard, PC1 explained 41.6% of the total variance and dominated by Zn, and Hg, it might be chemical industries. PC2 explained 39.2% of the total variance and dominated by Pb; it might be electroplated industries. PC3 explained 16.4% of the total variance and dominated by U and $^{232}$Th; it might be radioactive tailings. In mine road, PC1 (43.5% of the total variance) was dominated mainly by Hg and As; the origin could be from agricultural runoff. PC2 explained 41.7% of the total variance and dominated by Pb, Cd, and Zn; it might be chemical industries. PC3 accounted for 12.9% with the strong loading of U and $^{232}$Th; it was similarly to mineral ore stockyard and mine stope.

Figure 4. Loadings of the PCs for heavy metals and radiations: (a) in the former mine stope, (b) in the former mineral ore stockyard and (c) in mine road.
4. Conclusion
In this article, three-fields longitudinal assessment and analyses were conducted in the present study in a uranium mine during dry seasons. After the investigation, we carried out nine heavy metals and two radioactive elements and revealed their pollution condition. Pb, Cd, Hg, Zn, and U were in potentially high risk with considerable loadings and increased contents. Especially Hg and Cd were in super-strongly pollution status. Hg was more than two times. Due to the growth of agricultural runoff, chemical industries from hydrometallurgy, radioactive tailings, and electroplate industries, they were the factors that lead to heavy metal and radioactive risk. Spatial distribution analyses indicated the former mine stope and the former mineral ore stockyard were the biggest sources of pollution, and the a key to retire governance. A strong positive correlation between Pb, Cd, As, Cr, Mn, Ni, $^{232}$Th and U ($P \leq 0.01$) is found. Finally, we suggested taking measures to reduce the risk in a retired uranium mine.

Due to the soil-plant system is a source of bio-energy, the plants can clean up the potential risk of the heavy metal pollution [17]. Restore soil fertility is a way of accelerate ecological succession, such as different grass species planting, legumes and native species rotating [18]. Natural and artificially constructed wetlands get helpful to control the elements of heavy metal in acid mine water, and avoid spreading the heavy metal pollution in surface and subsurface water [19]. The vegetables with radionuclides can not to feed the livestock in the uranium mine, and they are growing on the sludge and the stream wetland [20]. Avail the susceptibility of the long half-life of radionuclides, such as post-reduction minerals to reoxidation and remobilisation via microbial metabolism or abiotic mechanisms, and it reduces the radioactivity in the environmental remediation [21]. The chemical treatment get helpful to control the radionuclides in tailings, such as CaO/CaCO$_3$ and BaCl$_2$, and cover with clay materials and other soils [22].

This study indicated that the environmental pollution problem of the retired uranium ore can not be ignored and got the first line information of the risk assessment of the domestic retired uranium mine. The obtained data can be not only used as a reference data for monitoring possible pollution in future but also can be applied to radioactive site remediation technology, phytoremediation technology, and cover experiment.

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