Distributions, Source and Pollution Status of Heavy Metals of Urban Soil in Xining, China

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Abstract. In recent years, the topsoil has suffered from remarkable heavy metal contamination with a rapid development of urbanization and industrialization in Xining. However, there are less detailed studies on the heavy metals contamination in Xining topsoil. We collected 40 soils samples from the upper 5 cm of the soil layer and detected the concentrations of Pb, Cu, Zn, Cd, As, Sh, Cr, Ni in the soil by inductively coupled plasma-mass spectrometry (ICP-MS) and that of Hg was tested by mercury detector. Compared with Chinese other part of soil, the concentrations of heavy metals in urban soils of Xining were significantly higher, and the average values of Zn, Cd, Sh, Pb and Hg were 9.13, 72.96, 5.30, 6.13, and 8.23 times as high as their corresponding background values, respectively. On the basis of the results of the correlation coefficient analysis, principal component analysis (PCA), cluster analysis (CA), the spatial distribution pattern analysis and heavy metal ratio analysis, we discovered that Hg, As, Cu, Sh, Pb, Cd and Zn had similar patterns, and their hotspots were primarily associated with three industrial districts in Xining, especially the Ganhe Industrial District in the southwest of Xining, while Cr and Ni mainly came from the natural source (loess around Xining). Heavy metal enrichment factor (EF) further confirms the source identification and degree of contamination. Moreover, the wind direction analysis shows the southwest and northwest winds transport lots of pollutions from the industry districts to the urban zones. The results of this paper clearly highlight that all people in xining are in sore need of pulling together to control industrial emissions/site, and meanwhile remedy seriously contaminated urban soils.

1. Introduction

Heavy metal pollution in urban soils has been an issue of great concern because of their extensive sources, high toxicity, non-biodegradable characteristics and accumulative properties. It is generally believed that Urban soils have become a sink of heavy metals and other contaminations from varieties of sources including industrial activities, coal and fuel combustion, vehicle emissions, urban waste disposal and natural origin [1-4]. The excessive accumulations of soil ecosystem have threatened human health human and safety, and at the same time brought along a series of other environmental issues. Hence, quantifying the concentrations and spatial variability of heavy metals in the soils were as important as determining their sources.

At present, soil contaminated by heavy metal has been considered a one of the major environmental issues worldwide [5]. Especially in many developing countries, heavy metal pollution of topsoil has reached a quite serious level [4, 6-7]. China, the world’s largest developing country, has achieved
rapid economic development and its GDP has grown at an average annual rate of 10% in the past two decades. However, the cost of this economic success was at the sacrifice of the environment [8]. With the rapid development of industrialization and urbanization in China, heavy metal pollution has become rather serious, especially in urban road dust, urban soils and agricultural soils. A good deal of researches on heavy metals pollutants in urban soils have been reported in China [2, 4, 9-19]. Metal concentrations were high in old industrial cities (for example, Shenyang) where metals deposited from power plants and industrial processes [16]. According to heavy metal investigation, the big metropolitan area (for example, Beijing, Shanghai and Guangzhou) suffers from the pollutions of the emissions of traffic, power plants, and industrial processes [9-11, 14-15]. The concentrations of metals in Jinchang and Changsha were particularly high because of the wholesale mining and smelting operations of metals [20] in the neighborhood. With the exception of geographical differences in background levels, the content of trace metals in urban soils also vary largely from city to city. At the same time, the differences in socioeconomic development and enforcement of environmental regulations are also part of reasons, and the differences are largely related to particular industrial activities and the degree of air pollution in urban areas [2]. However, there is few reports about heavy metal contamination of soils in Xining in the northeast of Tibetan Plateau.

This research was carried out in Xining. The Xining, a provincial capital, is situated at the northeast part of the Tibetan Plateau, with an area of 7649 km² and a total population of 2.2 million people. This city is located in Huangshui valley and surrounded by Quaternary loess (figure 1), with an average annual mean air temperature of 7.4-7.8 °C, and the annual precipitation of about 400 mm. With the rapid development of urbanization and industrialization in Xining in recent years, three industrial zones have been built surrounding Xining (figure 1): (1) Beichuan Industrial District (BCID), an industrial area for non-ferrous metal smelting, chemical raw materials, new materials, and building materials deep processing, includes chemical plant, aluminium plant and power plants; (2) Xining steel plant (XNSP), has had three main industries-iron and steel smelting, iron mining and dressing, and coal cooking; (3) Ganhe Industrial District (GHID), forming a main industry chain for lead, zinc and aluminium processing, fertilizer and cement production. In the past ten years, these industries produce a large of heavy metal and led to serious environmental pollution, however, up to now, there was less report about soil pollution in Xining.

Figure 1. Topographic map showing the locations of major industries and soil sampling sites of Xining city in the northeast China.
2. Sample Collection and Analysis Methods

2.1. Sampling and Analysis
Forty soil samples were collected in the Xining city in September 2015, including three industrial districts and Xining downtown area (figure 1). The geographical information of all sample sites were obtained by taking advantage of a portable global positioning system (GPS). Because of Xining locating at the Huangshui river valley (figure 1), we mainly sample along major streets and river. A stainless steel hand auger was used to collect soil samples. Every topsoil samples (0-5 cm) were composed of 3-5 sub-samples in different directions, which were homogenizing or analysed separately. Moreover, we collected two loess samples and two atmospheric dust from GHIB to represent two possible sources.

First of all, all samples were placed in an oven at 40 °C for 2 days, then poured the dried samples in a 1-mm plastic sieve to remove impurities, such as large plant roots, gravel-sized stone, etc. Finally, the sieved samples were ground and mixed uniformly with a polypropylene mortar following sieved through a diameter 66 um of 200 meshes sieve. The treated samples were digested with a 5:2:3 mixture of HNO\textsubscript{3}-HClO\textsubscript{3}-HF. The digested solutions would be used to analysed the contaminations of Pb, Cu, Cd, Zn, Ni, As, Sb, Hg, Cr in the samples by use of ICP-MS and mercury detector in the State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography & Limnology, Chinese Academic Sciences. Quality control mainly includes the following aspects: 15 random samples and 6 national standard samples were analysed, and the samples were selected randomly in order to ensure that the average deviation was less than 3%.

2.2. Descriptive Analysis and Correlation Coefficient Analysis
Descriptive data analysis was often used to conduct and analyse the data information, which mainly contained mean, standard deviation (SD), minimum and maximum concentrations, skewness, variation coefficient etc. Combining SD and mean, variation coefficient (VC) was expressed as SD/mean, and was utilized to estimate the degree of discrete distribution of the different metal element concentrations. In addition to variation coefficient, Skewness was also used to indicate degree of heavy metal distribution in the samples. Furthermore, correlation coefficients between different metals were computed to determine their relationships.

2.3. Multivariate Analysis
In order to determine the relationships between heavy metals in urban soils and their possible sources, a commercial statistics software package SPSS version 17.0 for Windows (SPSS Inc, USA) was applied to carry out Pearson’s correlation coefficient analysis, principal component analysis (PCA) and cluster analysis (CA). The strength of the mutual relation between the two heavy metals was often measured by use of correlation coefficient. The principal component analysis (PCA) method main used orthogonal transformation to convert a set of variables that may be related to a set of linearly uncorrelated variables (principal components, PCs), which was applied to analyse relationships between heavy metals. On the basis of similarities in chemical characters, the heavy metal elements coming from different sources would be further classified by using of Cluster analysis (CA).

2.4. Enrichment factor
The enrichment factor (EF) of elements was an important indicator to quantitatively evaluate the degree and the source of pollution. It selected an element meeting certain conditions as reference elements (or standardized elements), which usually had an obvious characteristic of low occurrence variability, for example, the most frequently-used elements were: Al, Fe, Ti, Si, Sr, K, Mn, etc. [1, 21]. The enrichment factor was defined the ratio of the concentration of the contaminating element in the sample and background area to the that of the corresponding reference element. The EF calculation formula was expressed as follows:
EF = \left[ \frac{C_x}{C_{\text{ref}}} \right]_{\text{Sample}} - \left[ \frac{C_x}{C_{\text{ref}}} \right]_{\text{Background}}

where $C_x$ refers to the concentration of the element to be studied in soil samples and $C_{\text{ref}}$ refers to the concentration of reference element to be normalized. When EF values is less than 5.0, it was considered to be irrelevant, because such a small amount of the enrichment may be caused by differences between the composition of local soils and the reference soil applied in above formula [21]. However, for the enrichment ratio and/or factor method, there was no recognized contamination grade system or classification of degree of contamination so far. According to the value of enrichment factor, the following are currently accepted five pollution categories: EF<2, refers to the minimal enrichment, EF=2-5 indicates moderate enrichment, EF=5-20 means significant enrichment, EF=20-40 indicates quite high enrichment and EF>40 states means extremely high enrichment [1, 22].

2.5. Geostatistical Analysis

The Exploratory Spatial Data Analysis (ESDA) tools of ArcGIS software are applied to analyse geostatistical data. Every ESDA tool can provide a different view for data, and at the same time displayed views in a separate window. These views can be operated and explored, which are related to each other through connection and the data displayed in ArcMap. In order to analyze distribution of the metals in urban soils, the data of heavy metals concentration were inputted for a grid based contouring maps. Moreover, ArcGIS provides different spatial interpolating methods in the software, including kriging and inverse distance weighted (IDW).

3. Discussions

3.1. Analysis of Content of Heavy Metals in the Soil

Descriptive statistics of the contents of studied heavy metals in Xining city and their corresponding background values [23] were shown in table 1. The data in the table show the concentrations of heavy metals in the sample change significantly. The contents of Cr, Ni, Cu, Zn, As, Cd, Sb, Pb and Hg varied between 48.55 and 1021.03, 20.32 and 173.31, 19.21 and 228.76, 54.99 and 9308.2, 8.79 and 92.95, 0.19 and 130.37, 0.38 and 1.15, 16.25 and 1573.3, 0.08 and 1.45 mg/kg, with mean contents of 142.93, 40.89, 46.34, 545.75, 16.89, 6.57, 3.02, 0.5, 124.4 and 0.24 mg/kg, respectively. The average concentrations of heavy metals in Xining soils were much higher by comparison with the background values of Chinese soils. The average content of Cd element was nearly 80 times higher than its reference value. The average values of urban dusts divided by their corresponding reference value, showing a decreasing trend: Cd>Zn>Hg>Pb>Sb>Cr>Cu>As>Ni. However, the mean concentration of Ni was similar to its reference value, indicating that Ni perhaps principally derive from the natural source.

3.2. Correlation Analysis between Heavy Metals

Correlation analysis can provide an effective way to reveal the relationships among different multiple variables, which is conducive to understand the influencing factors and identify the sources of chemical composition, so it has been extensively used in environmental research. The relationships between heavy metals in the soil are usually complex. If the correlation between heavy metals in soil was high, it perhaps suggested that these heavy metals come from similar pollution sources. The analysis results of the Pearson’s correlation coefficients and their significance levels (table 2) indicated that the concentration of Pb was significantly correlated with Hg (0.849), Sb (0.717), Cd (0.959), As (0.928), Zn (0.888) and Cu (0.577). However, the concentrations of Cr and Ni were weakly correlated with Zn, As, Cd, Sb, Pb and Hg, indicating that Cr and Ni were from the same sources, while Cd, As and Hg came from other sources.
Table 1. The heavy metals concentrations and background values (mg/kg) of urban topsoils in Xining.

|   | Minimum | Maximum | Mean  | Median | SD    | CV%  | Skewness | Kurtosis | Background value |
|---|---------|---------|-------|--------|-------|------|----------|----------|-----------------|
| Cr | 48.55   | 1021.03 | 142.93| 87.88  | 181.45| 126.95| 3.79     | 15.72    | 54.20           |
| Ni | 20.32   | 173.31  | 40.89 | 30.61  | 31.34 | 76.65| 3.47     | 12.54    | 33.80           |
| Cu | 19.21   | 228.76  | 46.34 | 34.09  | 38.00 | 81.99| 3.42     | 14.16    | 18.20           |
| Zn | 54.99   | 9308.20 | 545.75| 107.86 | 1577.56| 289.06| 4.94     | 26.44    | 59.80           |
| As | 8.79    | 92.95   | 16.89 | 12.55  | 15.67 | 92.76| 3.96     | 16.36    | 8.20            |
| Cd | 0.19    | 130.37  | 6.57  | 0.77   | 21.86 | 332.96| 5.16     | 28.65    | 0.09            |
| Sb | 0.93    | 19.13   | 3.02  | 1.65   | 4.12  | 136.32| 3.23     | 9.94     | 0.57            |
| Pb | 16.25   | 1573.30 | 124.45| 33.25  | 282.89| 227.32| 4.21     | 19.35    | 20.30           |
| Hg | 0.09    | 1.46    | 0.25  | 0.14   | 0.26  | 107.07| 3.17     | 11.61    | 0.03            |

Table 2. Correlations between heavy metal concentrations.

|   | Cr   | Ni   | Cu   | Zn   | As   | Cd   | Sb   | Pb   | Hg   |
|---|------|------|------|------|------|------|------|------|------|
| Cr | 1    |      |      |      |      |      |      |      |      |
| Ni | 0.861*| 1    |      |      |      |      |      |      |      |
| Cu | 0.536*| 0.709*| 1    |      |      |      |      |      |      |
| Zn | 0.163 | 0.341**| 0.591**| 1    |      |      |      |      |      |
| As | 0.277 | 0.465* | 0.767**| 0.898**| 1    |      |      |      |      |
| Cd | 0.154 | 0.311 | 0.609**| 0.953**| 0.956**| 1    |      |      |      |
| Sb | 0.36  | 0.546* | 0.797**| 0.743**| 0.852**| 0.769**| 1    |      |      |
| Pb | 0.212 | 0.297 | 0.577**| 0.888**| 0.928**| 0.959**| 0.717**| 1    |      |
| Hg | 0.186 | 0.404**| 0.623**| 0.801**| 0.878**| 0.874**| 0.738**| 0.849**| 1    |

Note: **P<0.01; *P<0.05.

3.3. Principal Component Analysis (PCA)

Principal Component Analysis (PCA) are usually applied to analyze the sources of heavy metals pollution (Facchinelli et al., 2001). The results of the PCA of metal concentrations in Xining soils were computed in table 3. The two principal components considered were on the basis of the results of the original eigenvalues, accounting for more than 80% of the total variance. When only two factors were Considered, the communalities shown by the variables, rang from 80.1% for Cu to 97.8% for As. Therefore, all the two elements can represent the two main components very well. The initial composition matrix (table 4) showed that Pb, Cu, Zn, Cd, As, Sb and Hg were strongly linked to the first principal component (PC1), explaining 62% of the total variance, while Cr and Ni were chiefly associated with the second principal component (PC2), explaining 18% of the total variance. However, different heavy metals may be distributed on different component, for instance, Cr and Ni were mainly related to PC2, while a part is associated with PC1. The metals in the PC1 showed anthropogenic sources were main pollution source. Cr and Ni in PC2 had a strong correlation and were clearly separated from other heavy metals in the analysis of PCA and correlation coefficient indicating that Cr and Ni mainly come from natural sources.

3.4. Cluster Analysis

In order to analyze the standardized bulk concentration data, the cluster analysis (CA) were applied by use of Ward’s method, with the Euclidean distance as the criterion for forming element clusters. This form of CA was generally considered effective, though it tends to create small clusters. Figure 2 shows three clusters: (1) Zn-Cd-Pb; (2) As-Hg-Cu-Sb; (3) Cr-Ni. However, it can be observed that clusters 1
and 2 joining together at rather higher level may imply the same source, while Cr and Ni at a long distance in cluster 3 possibly indicate that it is different from the other one.

![Hierarchical Dendogram](image)

**Figure 2.** A hierarchical dendogram for 9 elements obtained by Ward’s hierarchical clustering method (the distances indicate the degree of correlation between different elements).

**Table 3.** Total variance of explained and component matrixes.

| Component | Initial eigenvalues | Extraction sums of squared loadings | Rotation sums of squared loadings |
|-----------|---------------------|-------------------------------------|----------------------------------|
|           | Total % of variance | Cumulative % | Total % of variance | Cumulative % | Total % of variance | Cumulative % |
| 1         | 6.224 62.241        | 62.241      | 6.224 62.241        | 62.241      | 5.763 57.632        | 57.632       |
| 2         | 1.829 18.287        | 80.528      | 1.829 18.287        | 80.528      | 2.290 22.897        | 80.528       |
| 3         | 0.996 9.956         | 90.485      |                      |             |                    |              |
| 4         | 0.390 3.901         | 94.386      |                      |             |                    |              |
| 5         | 0.192 1.915         | 96.301      |                      |             |                    |              |
| 6         | 0.164 1.643         | 97.944      |                      |             |                    |              |
| 7         | 0.132 1.321         | 99.264      |                      |             |                    |              |
| 8         | 0.044 0.438         | 99.702      |                      |             |                    |              |
| 9         | 0.020 0.196         | 99.898      |                      |             |                    |              |

**Table 4.** Matrix of the principal component analysis loading of heavy metals.

|          | Cr     | Ni     | Cu     | Zn     | As     | Cd     | Sb     | Pb     | Hg     |
|----------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| PC1      | 0.128  | 0.333  | 0.658  | 0.939  | 0.969  | 0.983  | 0.81   | 0.952  | 0.916  |
| PC2      | 0.921  | 0.861  | 0.606  | 0.031  | 0.18   | 0.002  | 0.381  | 0.013  | 0.067  |

3.5. Spatial Distribution of Heavy Metals and Source Identification

The spatial distribution of metal concentrations is helpful to assess the possible sources and determine hotspots of high concentrations of heavy metal. The estimation maps of Pb, Cu, Cr, Ni, As, Cd, Zn, Hg, Sb were shown in figure 3, and at the same time several hotspots with high concentration of heavy metal were determined through the geochemical maps. A lot of isolated locations with an industrial history indicating quite high values in urban areas were called hotspots. In the southwest of the Xining, there were distinct hotspots of Hg, As, Cu, Sb, Pb, Cd and Zn. The highest concentration of Hg, As, Cu, Sb, Pb, Cd and Zn all appeared at this location, up to 1.45, 92.95, 228.75, 19.13, 1573.3, 130.37, and 9308.2 mg/kg, respectively. These concentrations mainly came from GHID. Although GHID was built in 2002, it had built 63 factories, including fertilizer plant, hydropower plant, aluminum plant, chemical plant, mineral and metallurgical plant, etc, and was the largest industrial district in the
Qinghai province. To a certain extent, it indicated that Hg, As, Cu, Sb, Pb, Cd and Zn may be from the identical source and industrial activities had played a large role in the urban soil pollution. Moreover, the pollution in industry district had seriously destroyed local air and crops, and resulted in blood lead level increasing in local children. There were another two pollution hotspots at the northern and western of the study area. It can be seen that Hg and Pb pollution were most serious, followed by As and Cd in the northern area (BCID). There were a lot of industrial enterprises in this region, including metal casting, non-ferrous metals and chemical industry. Main pollution of the western area (XNSP) was Zn, Sb, Pb, Cu and Cr. The main cause of this pollution was due to smelting iron and steel in the study area. The results of PCA and CA analyses corresponded with above interpretations. A strong correlation between Hg, As, Cu, Sb, Pb, Cd and Zn in both analyses can be considered as an indicator of the industrial origin.

Figure 3. The concentration maps for Ni, Sb, Cu, Cr, Zn, Mn, Cd, As and Hg (mg/kg) estimated by Kriging.

According to the result of the skewness and CA analyses, it gave the impression that the source of Ni and Cr may be different from other elements, because the concentrations of Cr and Ni were close to their corresponding background values, which indicates a natural source. Because Xining suffers from the dust storms in the spring of each year, the wind brings loess around Xining to the urban. Above analysis shows the Cd seriously comes from the industrial origin, while the Ni and Cr almost originate
from the natural source. So we use the relationship between the Cr/Cd and Ni/Cd ratios of the topsoils and the major sources to identify the potential pathways of the heavy metal in the Xining topsoils (figure 4). As shown in figure 4, a good linear relationship between the topsoil, atmospheric dust in the GHID and loess around Xining shows the industrial process and loess around Xining are the two main sources for the heavy metal of topsoil in Xining.

It can be seen from the concentration patterns of eight kinds of heavy metals studied in the Xining city that anthropogenic factors have a significant influence on the concentrations of heavy metal in topsoil, but natural factors would not be considered, because the compositions of topsoils almost approach to atmospheric dust in GHID (figure 4).

### 3.6. Enrichment Factor Analysis

Relative to the background value of elements in Chinese soil, the enrichment factors of heavy metals in each soil samples were calculated [23], and Ni was selected as the reference element. The change ranges of EF values of Cr, Cu, Zn, As, Cd, Sb, Pb and Hg are 0.72-21.46, 0.88-15.95, 0.78-133.68, 0.73-9.73, 1.91-1244.1, 1.49-42.6, 0.65-66.56 and 2.87-41.74, with an average of 2.66, 2.48, 8.44, 1.94, 66.16, 5.31, 5.54 and 7.6, respectively (figure 5). The average EF values of Zn, Cd, Sb, Pb and Hg is all higher than 5, meaning there is obvious pollution; while the mean EF of Cr and Cu between 2 and 5, were considered as moderate contamination. The maximum EF can reflect the degree of local pollution affecting every metal. The maximum EF values of Zn, Cd and Pb are greater than 50, suggesting that the soils from GHID have been seriously polluted. The maximum EF of Hg and Sb (40-50), which means very high contamination, comes from the GHID topsoils. This EF analysis, similar with hotspots map, shows the GHID is the most contaminated zone in the Xining city.

**Figure 4.** The Cr/Cd vs Ni/Cd plot obtained from topsoil, loess in Xining and atmospheric dust in GHID.

**Figure 5.** The average monthly maximum wind direction in Xining City during 1988-2010.

### 4. Conclusion

This study investigated the concentrations of heavy metal in the soil in Xining, east of the Tibetan Plateau. The results showed the EF values of Hg, As, Cu, Sb, Pb, Cr, Cd, Zn were high, which revealed the influence of industrial activities on the accumulation of heavy metals in the soil in the studied region. Lots of hotspots polluted with Hg, As, Cu, Sb, Pb, Cd, Zn were in the GHID, XNSP, BCID, while Cr and Ni mainly come from loess. Moreover, the southwest and northwest wind transport lots of pollutions from the industry districts to the urban zone. This study clearly shows there is an urgent need to work together to get command of industrial emissions/site while remedying heavily contaminated urban soils.
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