Assessment of Potential Heavy Metal Contamination in the Agricultural Soils based on various improved evaluation methods in Beijing, China

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Abstract

The evaluation of the soil contaminated by heavy metals can help to judge whether the soil meets the standard and whether the pollution will threaten human health and the ecological environment. In this study, the farmland soil from eight districts in Beijing was used as the research object, and the concentration of heavy metal elements, Pb, As and Cd in the soils and agricultural products were analyzed. The analysis results showed that: (1) The evaluation based on the improved Hakanson method suggested that the crops exhibit a significantly higher ability to absorb Cd than to absorb Pb and As. Pb, As and Cd are all at normal level of ecological risk; among them, Cd is mainly in a moderate ecological risk, without strong ecological risk. (2) Based on the Improved analytic hierarchy process(AHP) of evaluation, 0.2317 is the average value of the integrated index of heavy metal pollution of soil in the study area, which is a mild level of pollution. (3) Through the calculation of
various parameters in the Influence index of comprehensive quality (IICQ) of soil and agricultural products, it was found that $0 < IICQS < 1$, suggesting that the environmental quality of soil is at a clean level. In summary, the pollution of heavy metals Pb, As and Cd in the farmland soils and crops in the eight districts of Beijing, including Fangshan, Daxing, Shunyi, and Shijingshan is at a low level, and no significant impact has been brought to the surrounding environment.

Keywords: Heavy metals, Risk evaluation, Pollution assessment, Soil, Crops, Source identification

1. Introduction

In recent years, the rapid economic and industrial growth in China has triggered some prominent environmental problems, especially the serious industrial heavy metal pollution to a bulk amount of arable land. Heavy metal pollution of arable land in China has seriously harmed people’s health and agricultural economic development (Fu et al. 2020). Most soils contaminated by heavy metals contain various toxic metal elements, which will not only reduce crop yields, but also threaten food safety after penetrating the crops (Niu et al. 2013). Surveys showed that more than 70% of the arable soil in China has been contaminated by heavy metals, accounting for 1/6 of China’s total area of cultivated soil. Therefore, it is not hard to see that the agricultural land in China is suffering severe heavy metal pollution (Zhang et al. 2015).

Meanwhile, One of the main sources of heavy metal contamination in crops is heavy metals in the soil. When crops grow on arable land contaminated by heavy metals, their physiological, biochemical and developmental processes will be subject to adverse effect (Teng et al. 2010). For example, high content of cadmium in the soil will directly affect the normal growth of crops, resulting in crop failure or even death. Cadmium may also damage the chlorophyll structure of crop leaves, thereby weakening the ability of crops to absorb nutrients, sunlight and water; Plumbum will reduce the photosynthesis ability of crops and hinder the
normal absorption of water by crops; high concentration of arsenic will inhibit crop growth and undermine the chlorophyll structure of crops, leading to biomass reduction, slow growth, and even death of the crops (Zhai et al. 2008; Zhang et al. 2013; Zhang et al. 2015). Moreover, edible parts of the crops contaminated by heavy metals pose direct threats to human health and safety through food chain spread.

In summary, it is imperative to evaluate the risk of soil. At present, the main method of soil heavy metal risk assessment at home and abroad is the index evaluation method, while some people also use the fuzzy theory evaluation method and the spatial characteristics method of heavy metal pollution (Merry et al. 1981). The relationship between heavy metal content and soil background values can be measured directly by the index evaluation method, thus objectively evaluating the risk level of heavy metals in soil (He et al. 2013), and is a common evaluation method based on the total amount of heavy metals. According to different evaluation standards, the index evaluation method can be divided into single pollution index method (Merry et al. 1981), Nemerow index method (Barona and Romero 1997), geoaccumulation index method (Saaty 1990), and potential ecological risk index method (Hakanson 1980), etc. Karak, Tanmoy and other scholars have conducted in-depth study on the micro-characteristics of heavy metals (Karak 2010; Karak et al. 2013), they found that the transportation and transformation of heavy metals in the soil-plant system and their ecological toxicity are closely related to their content and morphology. Heavy metals of different forms have different bioavailability (Li and Jia 2018; Niu et al. 2013; Qin et al. 2012). On this basis, morphology-based evaluation methods have been developed, such as risk evaluation coding method (Singh et al. 2005), secondary phase and primary phase ratio method (Dudka and Miller 1999), and TCLP method (Guo et al. 2006). The risk of heavy metals in the soil can be reflected by total and morphology-based evaluation methods. However, each method also has its limitations (Wei and Yang 2010a), for example, the relationship between the heavy metal concentration of soil and background values, although it can be reflected by the single pollution index method, cannot provide a
comprehensive evaluation of heavy metal pollution. The degree of heavy metal contamination in the soil can be expressed by the Nemero index method, but incorporating differences in heavy metal biotoxicity (Wei and Yang 2010b). The content and biotoxicity of heavy metals are represented in the potential ecological risk index method, but the biological effectiveness of heavy metals is unclear; the biological effective state of heavy metals is addressed in the risk assessment coding method, but the application of heavy metal enrichment characteristics and biotoxicity is lacking (Lu et al. 2012; Niu et al. 2013; Wang et al. 2019). Therefore, the evaluation of heavy metal risk in soils requires different evaluation methods and the comprehensive consideration of the environmental effects and behavior characteristics of heavy metals, so that the pollution risks of heavy metals in soils can be reflected in an objective and comprehensive way.

2 Test method

2.1 Introduction to the research area

In this study, eight typical agricultural soils in Beijing were selected as the study area, and the ecological risk evaluation of soil heavy metals was carried out by analyzing the pollution characteristics of soil heavy metals and selecting multiple evaluation methods. By analyzing and comparing the evaluation conclusions of different methods, we aim to accumulate experience and methods for the ecological risk evaluation of soil heavy metals in this region, and also provide scientific basis for local land integration and utilization, and guide the development of green agriculture.

2.2 Data collection

The main heavy metals studied in this paper are Pb, As, and Cd. The data were collected from 2013 to 2020,
and screened from academic journals in the China National Knowledge Infrastructure (CNKI) and Web of Science (WOS) databases. Among the 20 selected papers, 64 soil samples and 80 samples of agricultural products containing Pb, As and Cd from 8 different locations were documented. After summarizing, the average concentration of Pb, As and Cd in the farmland soils and agricultural products from 8 districts of Beijing was calculated. All the original papers used for the calculation of the data on heavy metals in farmland soils from 8 typical districts in Beijing.

### 2.3 Method of Ecological risk assessment

#### 2.3.1 Improved Hakanson method

In 1980, Hakanson, L proposed a risk assessment method that comprehensively considers the types of pollutants in sediments, environmental abundance, sedimentation effects, toxicity sensitivity and other factors, called the Hakanson method (Hakanson 1980). On this basis, Liu and others proposed an improved Hakanson method that comprehensively considers the soil-crop system, which can effectively reflect the bioavailability of soil heavy metals and the quality and safety of agricultural products (Liu et al. 2019).

#### 2.3.2 Improved analytic hierarchy process and weighted average method

The Analytic Hierarchy Process (AHP) was proposed by the famous American operations researcher Satty T. L in the late 1970s (Saaty 1990). It is an unstructured multi-criteria decision-making method that combines qualitative analysis and quantitative analysis to bring people’s thinking. The process is hierarchical and quantified, which is especially suitable for situations where the target structure is complex. The improved analytic hierarchy process comprehensively considers the nature of the arable land, and adds the limit value of
heavy metals in crops ("Limits of Contaminants in Food" GB2762-2017). At the same time, the toxicity response coefficient of heavy metals (Hakanson, 1980, Liu et al. 2019) and the influence of heavy metals on crops planted on cultivated soil are used as the basis for the application of the analytic hierarchy process.

The first layer of the improved analytic hierarchy process structure is the target layer, that is, the heavy metal pollution evaluation of the soil in a certain area, defined as layer A; the second layer is the standard layer, that is, the selected three heavy metals of Pb, As, and Cd are in the crops Limit values (respectively 0.2 mg·kg$^{-1}$, 0.2 mg·kg$^{-1}$ and 0.5 mg·kg$^{-1}$) and heavy metal toxicity response coefficient (respectively 1, 20, 10) two standards, defined as B level, of which the two standards are defined as B1, B2; The third layer is the specific evaluation factors, namely three heavy metal elements of lead, cadmium, and arsenic, which are defined as layer C; this layer analysis is a three-layer structure type (Chen et al. 2019; Yang et al. 2020).

### 2.3.3 Soil and Agricultural Products Influence index of comprehensive quality

IICQ in the compound influence of heavy metals in farmland (or cultivated land) soil is composed of the comprehensive soil quality influence index (IICQ$_S$) and the comprehensive quality influence index of agricultural products (IICQ$_{AP}$), and the background of soil elements is also considered Factors such as soil element standards and valence effects, the content of target elements in agricultural products and pollutant limit standards (Romero et al. 1987, Wang et al. 2016).

### 3 Results and analysis

#### 3.1 Comprehensive evaluation based on the improved Hakanson method

In this paper, the “absorption effect” of crops was used to replace the “sedimentation effect” and “sensitivity
effect” in the Håkanson method. The toxicity response coefficient was calculated by supplementary information formula (3), and standardized to ensure its numerical range matches the range of pollution factor $C_f^i$. Finally, the product of the crop’s absorption effect coefficient $\delta$ and the relative abundance is the modified toxicity response coefficient $V_{Ti}^r$ of each pollutant (Table 1).

Through the pollutant category-absorption coefficient correction method, the index of the Hakanson method was improved, and the limits of grading standard $E_i^r$ and $RI$ are shown in Table 2.

### 3.1.1 The content of heavy metals in crops and root soils

The content of heavy metals in crops is shown in Table 4. The range of $w$(Pb), $w$(As) and $w$(Cd) is $(0.01-0.099), (0.002-0.071), (0.002-0.046) \text{ mg/kg}$ respectively. According to the “Maximum Levels of Contaminants in Foods” GB2762-2017, the content of $w$(Pb), $w$(As) and $w$(Cd) in crops are within the standard limits.

The range of the content of $w$(Pb), $w$(Cd) and $w$(As) in the soils of crop roots is $(10.4-29.12), (0.116-0.27)$ and $(2.722-10.792) \text{ mg/kg}$ respectively, lower than the screening values of soil contamination risk stipulated in the “Risk Control Standard for Soil Contamination of Agricultural Land” GB15618-2018, indicated no risk of contamination.

Through the statistics on the content of heavy metals in root soils and crops, box plots were drawn and compared (Fig. 2 and Fig. 3). It can be found that of the 80 Pb samples of agricultural products, only 4 have mild outliers; the content of Cd and As in crops is not abnormal; Pb samples of root soils also have the same trend, and only 3 samples have mild outliers; the content of As and Cd in root soils is not abnormal, with the outliers in the error range.

The statistical characteristics of crop’s heavy metal absorption coefficient $\delta$ are shown in the table. The
average value of δ is \( \delta_{Cd}(0.0371) > \delta_{As}(0.0026) > \delta_{Pb}(0.0015) \). The crops have significantly higher ability to absorb Cd than to absorb Pb and As. Guo et al. studied the accumulation characteristics of As, Cd and Pb in 16 kinds of wheat, and found that Cd is easier to accumulate to higher level than As and Pb in wheat; by investigating the accumulation characteristics of heavy metals in four types of rice, Liang et al. discovered that the accumulation level of Cd is the highest; An et al. determined the absorption status of five different plants (tomato, corn, green cabbage, cabbage and alfalfa), and reported that Cd and Pb accumulate more in the root system. The conclusions of this paper are consistent with the above results, indicating that Cd in the soil is more easily absorbed by crops.

3.1.2 Evaluation results

In the evaluation results obtained by the improved correction method, the average value of \( E^f_r \) is \( E^Cd_f \) (30.67) > \( E^As_f \) (10.59) > \( E^Pb_f \) (1.02). Pb, Cr and As are all at normal level of ecological risk; the proportion of low, moderate and strong ecological risk of Cd is (3.1%) and (96.9%) respectively, suggesting that Cd is mainly in the moderate ecological risk without strong ecological risk. The contribution of \( E^Cd_f \), \( E^As_f \) and \( E^Pb_f \) to RI is 72.54%, 25.06% and 2.4% respectively. It can be seen that \( E^Cd_f \) have the largest contribution.

The study showed that the overall background value of As in Beijing is small, and only point source pollution characteristics have been found, thus the pollution risk of As is at a low level. Although the surface pollution characteristics of Cd and Pb have been observed, Pb exhibits excellent performance in soils in Beijing, with a normal level of ecological pollution risk; the environmental quality of Cd element is generally good, and light pollution exists in some areas.
### 3.2 Comprehensive evaluation based on improved analytic hierarchy process and weighted average method

According to the comprehensive evaluation results, when the soil heavy metal pollution in the study area is at an average level, the comprehensive index is 0.2317, indicating a light pollution level. The pollution index varies from 0.1273 to 0.3079, suggesting that there is little difference in pollution; 64 soil samples and 80 samples of agricultural products have not been contaminated, and they are all clean samples with low level of pollution.

It can be found in Fig. 5 that the limit value of heavy metals in crops in the improved AHP accounts for 83% of the total analysis method, demonstrating that this method fully considers the relationship between heavy metals, soils and crops, and the evaluation results are more convincing.

### 3.3 Comprehensive evaluation based on IICQ

The calculation of the evaluation parameters showed that the soil exceeds the background $Y \geq 1$, and the soil exceeds the standard $X=0$, $0 < IICQS < 1$, indicating invasion and accumulation condition (contaminated, but not exceed the standard), and the environmental quality of soil is at a clean level (the value of IICQ indicates the relative degree of deviation from the background value).

It can be known from the Figure 7 and Figure 8 that the IICQ value in 8 typical districts of Beijing in a descending order is: Mentougou > Shijingshan > Daxing > Fangshan > Miyun > Shunyi > Changping > Huairou. Huairou District has the smallest IICQ value and Mentougou District has the highest value.

Mentougou District has the highest IICQ value, and its farmland soil pollution is more serious than other districts. The booming mining industry in Mentougou District not only destroys the ecology, but also causes water pollution, and the sewage disposal efficiency is poor; second, the mining in Mentougou District and
industrial production in the surrounding areas released a large amount of harmful pollutants, which polluted the air and also affected the growth of crops. These are the reasons why Mentougou District has a slightly higher IICQ value than other districts. In summary, the IICQ method incorporates the soil background values, the content of heavy metals in soil samples and agricultural products into the calculation process. In addition, the evaluation parameters of the IICQ method involve the actual land use and the quality indexes of edible parts of agricultural products, solving the problem of not being able to consider both soil and agricultural products in soil environmental quality evaluation.

3.4 Comparative analysis of improved Hakanson method, AHP method and IICQ method

The risk grading standard in the improved Hakanson method is closely related to the types of pollutants. The re-determination of the grading standard according to the types of pollutants is the basis for the application of standard in ecological risk evaluation of farmland soil. In addition, since there are significant differences between the farmland soil environment and the water body sedimentation environment, introducing the heavy metal absorption coefficient of crops into the heavy metal toxicity response coefficient of soil can help to evaluate the farmland soil and crops as a system. Based on the goals of ensuring rice quality and safety, and conservative principle, the type of pollutants-absorption coefficient-improved Hakanson method can more accurately evaluate the ecological risks of Pb, Cd, and As in soil, and the quality and safety of Pb, As, and Cd in crops are also considered.

From the perspective of arable soil use, the improved Analytic Hierarchy Process determines the weights between heavy metal elements based upon the comprehensive consideration of the heavy metal toxicity response coefficient and food safety, and constructs an evaluation model of heavy metal contamination in cropland soil.
based on AHP and weighted average. The results of comprehensive evaluation suggested light pollution, but the sample concentration is at a clean level. The results showed that the mean value of the composite index was 0.2317, which is close to 0.2, indicating light clean pollution. In addition, the evaluation method incorporates the current status and spatial distribution of heavy metal pollution in agricultural soils, and is applicable to the evaluation of heavy metal contamination of farmland in mining areas with multiple pollution sources. In addition, the toxicity response coefficients of heavy metals when the method was applied in this study were the results obtained by the improved Hakanson method, allowing a closer integration of the soil-crop system. This method has higher sensitivity than other methods when applied to study the case where pollution is serious or complex and multiple pollution sources exist (e.g., pollution in mining area).

The quality of agricultural products is indispensable in the evaluation of environmental quality of farmland soil. The current evaluation parameters constructed by the IICQ of soil and agricultural products take the actual situation of land use into account, and add the quality index of the edible parts of agricultural products affected by heavy metals. Meanwhile, soil and agricultural products are considered simultaneously. By calculating the background value, standard and valence state effect of soil elements, the content of target elements in agricultural products and the pollutant limit, it was found that the farmland soils in Beijing’s eight districts are within the clean range. In general, the IICQ of soil and agricultural products is applicable to the evaluation of the combined and individual influence of heavy metals in soil, and this method fully considers the valence effect of heavy metals, and has made a new breakthrough in the analysis of the effective state of heavy metals.

4 Analysis of sources of heavy metals in farmland soils

Both soil parent material and human activities can influence the sources of heavy metals in soil. The main sources of soil pollution in Beijing include wastewater, waste gas and waste residue discharged from industrial
and mining enterprises. In addition, a large amount of pesticides and chemical fertilizers are used in the agricultural production process. In life, a large amount of domestic waste, enterprise relocation and automobile exhaust also cause heavy metal pollution. The correlation of heavy metal content in the soils of the study area can be analyzed to infer whether the regional sources of heavy metal pollution are the same. If there is a significant correlation between the content of several heavy metals, it indicates high homology, otherwise, the sources may be complicated and interfered by various factors.

4.1 Analysis of correlation between heavy metals in soils in Beijing

Spearman correlation analysis was conducted on three kinds of heavy metals in farmland soils in Beijing, and the results are shown in Table 10. It can be seen that Pb, As, and Cd in farmland soils in Beijing are significantly correlated with each other, with the level of P<0.05. Considering that there may be differences in the accumulation of heavy metal elements in the soils in different areas, the correlation analysis was further performed on the heavy metals in the soils in different districts.

Table 8 shows that the significant correlation between the three kinds of heavy metals is roughly the same in different districts of Beijing, but the correlation intensity is different. There is a significant correlation between Pb and Cd in all districts, a very strong significant correlation has been found in Fangshan, Daxing, Mentougou, and Miyun, and a significant correlation is presented in Shunyi, Shijingshan, and Changping; significant correlation between Pb and As is shown in some districts, and extremely strong significant correlation has been observed in Mentougou, Changping, and Miyun; Cd and As are weakly correlated or not correlated in most districts.

In general, the correlation between Cr and As is relatively not strong in 7 districts of Beijing, but the strong
correlation has been found in Mentougou. There is a correlation between Pb and Cd, and between Pb and As. The correlation between heavy metals is relatively strong in the soils in Daxing, Shijingshan, and Changping, while relatively weak correlation exists in the soils in Fangshan, Shunyi, and Huairou.

4.2 Principal component analysis of heavy metals in Beijing

In order to better understand the relationship between the three kinds of heavy metal elements in the soils in Beijing and explore the correlation and homology of the data, SPSS statistics 26.0 software was used to perform principal component analysis on the content of seven heavy metals in 64 soil samples. It can be seen from 4.1 that there is a linear correlation between the heavy metals. KMO and Bartlett tests were conducted on the data. The coefficient of KMO test is 0.604>0.600, and the Bartlett test result is P=0.000<0.001. Therefore, the data structure can be extracted for principal components. The varimax method was employed to rotate the data, so that the component factors can be better explained. The results of principal component analysis are shown in Table 12. In this paper, the components with eigenvalues greater than 1 are selected as principal components, and two principal components are extracted, which can explain 86.2% of the total variance.

Figure 9 and Table 9 show that component 1 mainly reflects the composition information of Pb and Cd, and these two heavy metals also have a strong correlation. Cd and Pb almost all exceed the background value at the point location, thus it is speculated that the sources of these two elements in the research area are mainly the human activities such as agricultural fertilizers, sewage irrigation, and industrial production.

Component 2 mainly reflects the information of As. As has a weak correlation with other metal elements. As is a diagenetic element, and its accumulation in soil comes from both natural and artificial sources. The artificial sources involve the use of agrochemical products, mining, and industry.
Eight districts including Mentougou, Shijingshan, Daxing, Fangshan, Miyun, Shunyi, Changping, and Huairou are the main planting bases of crops in Beijing, which play a vital role in the daily life and health of residents. Therefore, it is of great significance to evaluate the heavy metal risk in these 8 districts. The evaluation results from the improved Hakanson method showed that the average concentration of Pb, As and Cd is 23.84 mg·kg\(^{-1}\), 0.199 mg·kg\(^{-1}\), and 8.7 mg·kg\(^{-1}\) respectively, lower than the screening value of soil contamination risk. The ecological risk index of Pb, Cd, and As is 1.02, 30.67, and 10.59 respectively, indicating no contamination risk. The evaluation by the improved Hakanson method can directly reflect the ability of crops to absorb pollutants, so that certain heavy metal can be handled in a targeted manner; the evaluation results from the improved analytic hierarchy process demonstrated that the average value of comprehensive pollution index is 0.2317, indicating light pollution. However, the AHP method is more suitable for studying the farmlands in mining area where multiple and mixed pollution sources exist. Its evaluation of the soil with a single pollution source is slightly biased; the method of IICQ of soil and agricultural products calculated the evaluation parameters, and the results showed that 0< IICQS <1 in 8 districts of Beijing, suggesting the environmental quality of soil is at a clean level. The IICQ method comprehensively considers the interaction between heavy metals in soils and agricultural products and the environmental quality of farmland soils. It is more applicable to the evaluation of the combined and individual influence of heavy metals in soil, and this method takes the valence state effect of heavy metals into full consideration, so that it can effectively analyze the effective state of heavy metals. In addition, through source analysis of farmland soils in Beijing’s eight districts, it was found that the pollution mainly comes from artificial sources such as agricultural fertilizers and irrigation. Therefore, it is necessary to strengthen the management of agricultural fertilizers and sewage irrigation.

In summary, the quality of urban agricultural soil is vital to human health. In order to effectively reduce the
risk of heavy metal pollution in urban agricultural area and further develop reliable protection measures, risk
evaluation is required. In the evaluation, different evaluation methods should be adopted according to different
soil pollutants, different valence states of heavy metals and different pollution sources, so that the potential
environmental risk of farmland soil in different areas can be accurately, comprehensively and objectively
evaluated and predicted.

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