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Evaluation of ecological risk and primary empirical research on heavy metals in polluted soil over Xiaoqinling gold mining region, Shaanxi, China

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Abstract: Soil, crop and residents’ hair over Xiaoqinling gold mining region, China, which was selected as a case study, were sampled and analyzed for Hg, Cd, Pb, Cu, Cr, As and Zn concentrations. The concentrations of heavy metals in soil or crop and hair samples were used to assess their potential ecological risks, or to find the responses to these metals as evidences to prove the potential risk was coming down to observed harm, respectively. The results showed that, these metals in soil were ranked by severity of ecological risk as Hg＞Cd＞Pb＞Cu＞Cr＞As＞Zn, based on their single-element indexes. In the view of the potential ecological risk indexes, of all soil samples, about half had significantly high or high potential ecological risk, which covered more than 74% of the studied region. Most of the risks were 97.41% from Hg, Pb and Cd, especially, 84.37% from Hg. Both the single-element and potential ecological risk indexes indicated that, the ecological risk grades had a special spatial characteristic, and increased from northwest to southeast generally. This was agreed with the spatial distribution of the strength in gold mining activities over the studied region. The concentrations of Hg and Pb were higher than their relative backgrounds in the corps, and were even 9.48 and 25.09 times higher than their relative backgrounds in residents’ hair, respectively. All these showed that the heavy metals in the soil had a high potential ecological risk, especially, had been affecting these crops’ growing and yield, and even the residents’ health through food strains. Obviously, these metals’ potential ecological harm had been coming down to observed harm to the ecology.
Key words: gold mining activity; soil pollution; heavy metal; potential ecological risk

1 Introduction

It is well known that, the soil, over a gold mining region, is always polluted by heavy metals, which are released from gold mining activities. Because these metals are toxic to living organisms, and even are several pollutants where ingestion of contaminated food has led to human death[1], such researches on ecological risk assessment of heavy metals in the polluted soil had gotten more and more attention[2−7]. It was found that the results of the ecological risk assessment can reveal the possibility for soil to be polluted, and even for the ecology to be harmed by concerned heavy metals[8], so these results were usually used to serve as a guide for all US Environmental Protection Agency (EPA) programs and regional offices to supplement or update the policies, practices and guidance. Therefore, such researches have got much attention from researchers over the world, and a great progress has been made[2−9]. Meanwhile，more and more studies and practices showed that, such results from the risk assessment have little capability to reveal the real degree of their potential toxic effects completely, especially, no capability to show whether the potential toxic effect is coming into an observed harm of ecology and its degree. So, based on the results, relative strategies and methods were taken to control the risk and even their practices usually were not operated in time. Therefore, a great harm of ecology would be happened in the end. Obviously, in order to obviate an ecology system to be harmful, it is important to do assessment of ecological risk; meanwhile, it is necessary to do primary empirical researches too.

Xiaoqinling gold mining region, which is the second larger Au-producing area in China, is located in eastern Shaanxi Province (N 34°23′−34°40′ and E 110°09′−110°25′), northwestern China (Fig.1). The Au production in the area from 1980 to 2003 reached 56.25 t

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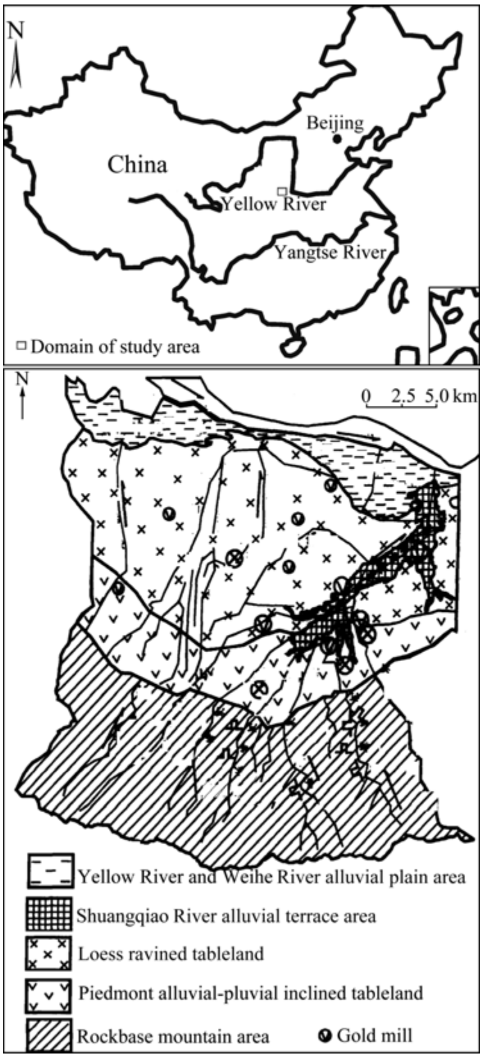

Fig.1 Location of studied region and its landscape distribution

[10]. During the 1980s and 1990s, artisanal Au mining activities were popular in the region and more than 15 000 people were involved in Au mining activities. Presently, there are 29 large Au mining companies involving 6 000 workers. Most of gold is extracted from original Au ores by amalgamation, meanwhile, waste water during these processes is directly discharged and resulted in both soil and water heavy metals pollution. Although several researches had been conducted on the soil heavy metals pollution and the source and transport paths of several metals were determined[11], there were few researches conducted on ecological risk assessment of these heavy metals, and no such studies were conducted to determine whether the potential toxic effect is coming into the observed harm of ecology and its degree. So, the goals of this study were to do the potential ecological risk assessment of heavy metals in the soil, and to find testimonies to confirm that the potential risk was coming into an observed ecological harm.

2 Materials and methods

Xiaoqinling gold mining region is situated in the Loess Plateau, and its average elevation is 1 500 m above sea level. It has a typical temperate zone terrestrial monsoon arid climate. The average annual rainfall is 625.5 mm, and the average annual temperature is 13 ℃.

Based on its geochemical landscape characteristics, the region is divided into five areas, the rock base mountain area, loess ravined tableland, piedmont alluvial-pluvial inclined tableland，Shuangqiao River alluvial terrace area, and Yellow River and Weihe River alluvial plain area. In the rock base mountain area, there are lots of gold ores and several gold mines (Fig.1). However, there is few soil, so it is suitable for agricultural activity and not included in the studied area, and the other four areas were selected as studied region. Among these four areas, Yellow River and Weihe River alluvial plain area is the only one which is occupied mainly by agricultural activities, and the others are cooperated in mixtures of agricultural and gold-mining activities. Although agricultural activities are conducted all over the other three, most of them are condensed in the area close to Tongguan county.

According to its local records, Au mining was first initiated in the region about 900 years ago[10]. Large-scale Au mining activities began in 1975[12]. About 72 abandoned ancient Au mines were known in the Tongyu, Haochayu, Taiyu, and Dongtongyu areas (Fig.1). Presently, there are many gold mills, and almost are mainly in Shuangqiao River alluvial terrace area (Fig.1), and the annual Au production reaches 2.5−3.0 t in the regiona[10].

The Au processing mills are distributed along several streams that are tributaries of the Yellow River (Fig.1) and wastewater from these mills is directly discharged into these streams. During the 1980s and 1990s, artisanal Au mining activities were popular in the region and more than 15 000 people were involved in Au mining activities, in which elemental Hg was used widely for Au extraction. Artisanal Au mining activities mainly occurred along the streams in Tongguan county. Artisanal Au mining in Tongguan county has declined significantly since the Chinese ban of such activities in 1996, but there are presently a few artisanal Au mines still operating. Generally, elemental Hg is used usually by these companies for extraction of Au. To date, at least eight million tons of tailings have been produced from mining in this area, and the average Hg content in the tailings is approximately 630 mg/kg, which is significantly elevated compared with the average Hg content (0.069 mg/kg) in original Au ores. So, the soil had been polluted by the heavy metals released from these gold mining activities[12].

Soil sampling was carried out in July 2006. The studied region was divided into a grid of 192×192 cells using a systematic grid sampling method with regularly spaced intervals of about 250−250 m. Within each grid cell five samples were randomly collected to a depth of 0−15 cm and then mixed thoroughly to give a composite sample. The sampling protocol was, therefore, a combination of systematic grid, simple, random and composite sampling methods.

After air-drying, mixing and milling, soil sub-samples were digested with aqua-regia (4:1 HCl-to-HNO_3 by volume) according to the Refs.[13−15] to determine the concentrations of Cu, Zn, Pb and Cd. The total concentrations in the solution of Cu, Zn and Pb were determined by flame atomic absorption spectrophotometry (Varian SpectrAA 220 FS) and of Cd with a Varian SpectrAA 220Z spectrophotometer using a graphite furnace.

Soil sub-samples were digested with HNO_3 and $HClO_4$ for determination of the metalloid As, and with HNO_3 and H_2O_2 for the metal Hg. The concentrations of these three elements in the solutions were determined by hydride generation-atomic fluorescence spectrometry (Titan AFS 930). Quality control was based on the combined use of the GSS series of Chinese standard soil reference materials[16] with internal control samples and duplicate analyses of each sample. Selected soil physicochemical properties, namely pH and mechanical composition were also measured. The data and heavy metal concentrations have been discussed elsewhere[17].

Soil heavy metal concentrations were analyzed by principal component analysis (PCA) and correlation analysis using Version 11.5 of the SPSS software package (SPSS Inc., Chicago, IL). In the PCA, varimax that was proposed by KAISER (1958) [18] was used as the rotation method in the analysis following standardization of the data.

According to the local climate, prevailing wind direction is from northwest to southeast generally. Under respecting to this wind and its effect on air transformation, Mengyuan, which was in the northwest of the studied region, was selected as the background site. Several soil samples were collected there and analyzed for metal concentrations. And Hg, Pb, Cu or As concentrations determined were 0.13, 32.57, 20.71 and 15.13 mg/kg as relative background, respectively.

34 crop samples which included wheat, B. Chinensis var. Communis, Lactuca sativa.L.var.romana, green vegetables and radish, and 20 hair samples from the residents over the regional were gotten and analyzed for the heavy metal concentrations.

Statistical analyses were conducted using Microsoft Excel and SPSS 10.01 software. Linear regression analysis and factor analysis of the concentration were performed using STATISTICA for Windows, Release 5.0, Copyright StatSoft, Inc. 1984−1995. The method suggested by HĀKANSON[19] was used to do the ecological risk assessment.

3 Results and discussion

The metal concentrations in 133 soil samples were done through statistical analyses with Microsoft Excel and SPSS 10.01 software, and descriptive parameters and probability distributed are obtained and listed in Table 1.

The Pb concentrations (about 3 450 mg/kg) (between the minimum and maximum values) was the largest, and its variation (Standard deviation=487.83), mean (216.93 mg/kg) and mode (32.00 mg/kg) were also the highest of all the studied samples. This suggests that there are several locations having great Pb concentrations, and the soils have been contaminated more severely by this metal. The ratios of mean to background value for most metals, except Cr and As, are larger than 1.0, and are ranked in following order: Hg＞Pb＞Cd＞Cu＞Zn. This means that the human activities have significant effects on the concentrations for almost metals in soils over this studied region, and the soil is polluted by these metals.

Correlation analysis was conducted to determine the extent of the relationships among metals in soils over the studied region. The correlation matrix in Table 2 shows that As is negatively correlated with any metal in soils

Table 1 Summary statistics of heavy metals in soils over studied region

Heavy metal	Minimum concentration/($mg\cdot kg^{-1}$)	Maximum concentration/($mg\cdot kg^{-1}$)	Mean concentration/($mg\cdot kg^{-1}$)	Standard deviation	Mean/ background
Hg	0.04	61.20	2.75	7.17	21.15
Pb	22.00	3470.00	216.93	487.83	6.66
Cd	0	45.20	0.55	3.91	3.24
Cr	31.50	69.80	44.72	6.64	0.98
As	3.15	35.70	14.04	4.30	0.90
Cu	31.00	600.00	54.13	71.18	2.61
Zn	46.00	781.00	118.06	85.26	1.85

Table 2 Correlations between heavy metal contents in studied soils over studied region

Heavy metal	Hg	Pb	Cd	Cr	As	Cu	Zn
Hg	1						
Pb	0.854 6	1					
Cd	0.018 2	0.029 6	1				
Cr	0.208 3	0.249 2	0.189 5	1			
As	−0.275 1	−0.271 1	0.108 9	−0.243 6	1		
Cu	0.773 2	0.965 4	0.087 8	0.262 0	−0.183 9	1	
Zn	0.182 4	0.234 7	0.102 2	0.058 8	−0.054 2	0.329 4	1

over the studied region, suggesting perhaps different soil sources from other metals. Hg, Pb, Cd, Cu, Cr and Zn are closely related to each other ($r>0.92$, $P<0.01$). This mentions that Hg, Pb, Cd, Cu, Cr and Zn may have another originate. Obviously, the soil pollution sounded complex in several kinds of heavy metals and their sources. This is agreed with XU et al[12], and mentions that it is difficult to protect the soil from the heavy metals pollution.

The assessment of soil contamination was conducted using the contamination factor and degree. In the version suggested by HĀKANSON[19], an assessment of soil contamination was conducted through reference of the concentrations in the surface layer of bottom sediments to pre-industrial concentrations:

$$C_{\mathrm{r}}^{i}=\frac{C_i}{C_{\mathrm{n}}^{i}} \tag{1}$$

where C_i is the mean concentration of an individual metal examined and C_{n}^{i} is the pre-industrial concentration of the individual metal. In our work, the background concentration of a metal in the regional soil (Table 3) was applied as the pre-industrial concentration of each individual metal. C_{r}^{i} is the single-element index. The sum of contamination factors for all metals examined represents the contamination degree (C_{d}) of the environment:

$$C_{\mathrm{d}}=\sum_{i=1}^{n} C_{\mathrm{r}}^{i} \tag{2}$$

E_{r}^{i} is the potential ecological risk index of an individual metal. It can be calculated by

$$E_{\mathrm{r}}^{i}=C_{\mathrm{f}}^{i}\times T_{\mathrm{r}}^{i} \tag{3}$$

where T_{r}^{i} is the toxic response factor provided by HĀKANSON[19] (T_{r}^{i} for Hg, Pb, Cd, Cr, As, Cu or Zn is 40, 5, 30, 2, 10, 5 or 1, respectively). R^i is the potential ecological risk index, which is the sum of E_{r}^{i}:

$$R^{i}=\sum_{i=1}^{n} E_{\mathrm{r}}^{i} \tag{4}$$

HĀKANSON[19] defined four categories of C_{r}^{i}, four categories of C_{d}, five categories of E_{r}^{i}, and four categories of R^i, as shown in Table 4.

Table 3 Evaluating standards of heavy metal contamination on farmland soils (mg·kg^{-1})

Heavy metal	Background in studied scale	Background in Guanzhong area	Limit for Grade 2*
Hg	0.13	0.086	1.0
Pb	32.57	16.300	350.0
Cd	0.17	0.118	0.6
Cr	45.83	65.700	250.0
As	15.53	12.700	25.0
Cu	20.70	23.500	100.0
Zn	63.97	65.800	300.0

* Environmental quality standard for soils (GB15618—1995), Ministry of Environmental Protection of the People's Republic of China[20].

Based on the single-element index (C_{r}^{i}) and its grades (Table 4), the soil around Tongguan county and over Yellow River and Weihe River alluvial plain area, was classified as considerably contaminated with As. The soil of piedmont alluvial-pluvial inclined tableland, Shuangqiao River alluvial terrace area, was contaminated with Hg, Pb, Cu, Cd and Zn. It was interesting that, the soil polluted seriously by As or Hg, Pb, Cu, Cd and Zn, was distributed over agricultural or gold mining area, respectively. This may be used to prove the guess mentioned above on metal sources and their complex to be right.

Among these metals, the range and means of the potential ecorisk index of Hg were the largest. The second was Pb, then followed by Cu, Zn, As and Cr (Table 5). This agreed with the results obtained from Table 1.

Of all soil samples, the ratios of the soil samples with low potential ecological risk, moderate potential ecological risk, high potential ecological risk and significantly high potential ecological risk, were determined, respectively. The results showed that, for all sampling sites, there were 21.05% of the sites had high risk, and 28.57% had significantly high potential ecological risk, and both were about 49.62%. And the significantly high potential ecological risk was mainly resulted from Hg, Pb, and Cd, especially, 84.37% from Hg. This means that, the heavy metals (Hg, Pb, Cd and others) from the gold-mining activities had not only polluted the soil seriously, but also caused the soil with significantly high and high potential ecological risk. The polluted soils with significantly high and high potential ecological risk covered about 74.54% of all the studied regions (Fig.2).

The potential ecological risk of each heavy metal and its main statistical parameters in the studied soil samples are shown in Table 6. E_r^i for Cr, As or Zn in all the studied soil samples was less than 40 (Table 6), and they also were in low potential ecological risk. Differently, E_r^i for Hg, Pb, and Cd in all the studied soil samples varied over a larger range in potential ecological

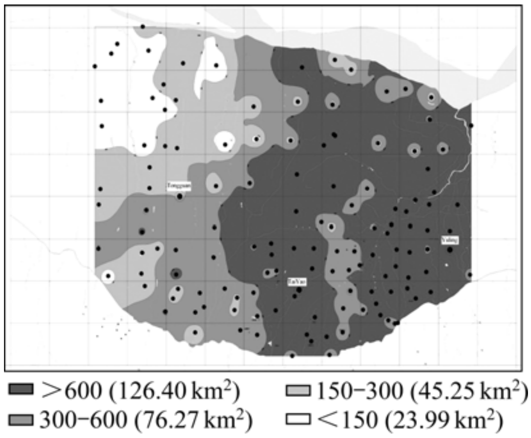

Fig.2 Distribution of grades on potential ecological risk of heavy metals in soil

Table 4 Risk grades indexes and grades of potential ecological risk of heavy metal pollution

E_r^i	Risk grade	R^i	Risk grade
<40	Low potential ecological risk	<150	Low potential ecological risk
40−80	Moderate potential ecological risk	150−300	Moderate potential ecological risk
80−160	Considerable potential ecological risk	300−600	High potential ecological risk
160−320	High potential ecological risk	≥600	Significantly high potential ecological risk
≥320	Significantly high potential ecological risk		

Table 5 Potential ecological risk of each heavy metal in soil and their main mean values in soil

Heavy metal	Potential ecorisk index range	Mean value	Standard deviation
Hg	12.31−18 830.77	845.16	2 205.77
Pb	3.38−532.70	33.30	74.89
Cd	0−7 976.47	97.36	689.76
Cr	1.37−3.05	1.95	0.29
As	2.03−22.99	9.04	2.76
Cu	3.14−144.93	13.07	17.19
Zn	0.72−12.21	1.85	1.33
R^i	37.91−19 457.27	1 001.74	2 397.86

Table 6 Grade and mean of potential ecological risk (E_r^i) for each heavy metal

Heavy metal	Low potential ecological risk		Moderate potential ecological risk		Considerable potential ecological risk		High potential ecological risk		Significantly high potential ecological risk	
	Ratio*/%	Mean of E_r^i	Ratio/%	Mean of E_r^i	Ratio/%	Mean of E_r^i	Ratio/%	Mean of E_r^i	Ratio/%	Mean of E_r^i
Hg	6.02	24.62	5.26	61.10	26.32	128.70	20.30	223.70	42.11	1807.80
Pb	82.71	12.50	11.28	59.12	2.26	98.25	1.50	239.64	2.26	464.54
Cd	78.95	22.92	11.28	57.77	7.52	111.35	0.75	213.53	1.50	4174.41
Cr	100.00	1.95								
As	100.00	9.04								
Cu	96.24	10.19	2.26	60.15	1.50	127.06				
Zn	100.00	1.85								

* Ratio means samples with relative risk in all studied samples.

risk index. Most of these potential ecological risk indexes, especially for Hg, were much larger than 80, and even 18 830.77, so that the risk grades were in considerable potential ecological risk, high potential ecological risk and significantly high potential ecological risk, and these samples were 88.73% of all the studied samples. On contrary, out of all the studied soil samples, samples with significantly high and high potential ecological risks for Pb, Cd and Cu were 6.02%，9.77% and 1.50%, respectively. Obviously, the studied heavy metals can be ranked by severity of ecological risk as follows: Hg＞Cd＞Pb＞Cu＞As＞Cr＞Zn.

The potential ecological risk indexes (R^i) for all the studied soil samples were distributed with a site-specific characteristic (Fig.2). The soil samples with significantly high potential ecological risk were distributed over the east part from Taiyao village, and the soil samples with high potential ecological risk were distributed over the area between Taiyao village and Tongguan county. Over the last part of the studied region, the west part from Tongguan county, there were the soil samples with low potential ecological risk. Generally, the grade of potential ecological risk was increased from northwest to southeast. This characteristic was agreed well with the distribution of the gold mining activities (Fig.1).

From all above, it was concluded that these heavy metals in the studied regional soil were resulted mainly from the gold mining activities, and had large potential to affect the ecology. The results of the study conducted by CAO et al[2] showed that, the heavy metals in the polluted soil had the potential to affect crops, and the effect could be observed. Actually, the metals accumulated to shield concentrations and then caused toxic to the crops, so the effect could be observed. Otherwise, the effect was not easy to be observed, although the stress of the metals existed. Under this condition, the heavy metals concentration is a good index to show the stress of the metals to the crops.

The common crops (wheat, B. Chinensis var. Communis, Lactuca sativa.L.var.romana, green vegetables and radish) were sampled in the studied region and the background, and were analyzed for Hg, Pb, Cd, Cu and Zn concentrations (the data were not shown). The concentrations in the crop samples from the studied region were higher than relative ones from background. For examples, the mean concentrations for Hg, Pb and Cd in the wheat from the studied region, were 5.11, 5.69 and 5.12 times higher than their relative concentrations from background, and also higher than their relative tolerance limit of iron in foods in national standards of China. Therefore, the yield of the crops and its quality were in a great risk to be affected by the heavy metals from the gold mining activities. This also mentioned that these heavy metals in the crops would pass into human body through food chain, thus the resident health was hardly escaped from the stress of these heavy metals.

In order to reveal the effect of the gold mining activities on the resident’s health, 20 hair samples from the residents over the studied region, and 4 hair samples from background were obtained and analyzed for Hg, Pb, Cd, Cu and Zn concentrations (the data was not given). The concentrations showed that, these heavy metals, which existed in both the soil and the crops, were also found in hair samples from the residents, no matter whether how old, and male or female they were. And the concentrations were not only higher than their relative backgrounds[18], but also higher than relative limits in GBW-07601[21]. Especially, the means of Pb and Hg concentrations, were 9.48 and 5.09 times higher than their relative backgrounds, and were 8.64 and 7.64 times higher than their relative limits in GBW-07601, respectively. It was worth mentioning that, 15 of the surveyed 20 residents did not feel well and had something wrong with their healthy, such as weak chest, dizzy and high blood pressure. On the contrary, these phenomena were not commonly found among the residents in the background. All these strongly revealed that these heavy metals in the soils and crops due to the gold mining activities, had being affected the residents’ health.

4 Conclusions

1) Hg, Pb, Cu, As and Cr heavy metals in the polluted soil, were sourced from gold mining activities, and these metals with higher concentrations were mainly distributed around the gold mills over the studied region.

2) The potential ecological risk indexes of these metals in most soil samples were more considerable, and in about 49.62% soil samples were high or significantly high potential ecological risk, which covered 74.54% of all the studied area. All these mean not only that the soil had been polluted seriously by heavy metals, but also these heavy metals were ready to be toxic to the environment.

3) These heavy metals concentrations in crops were higher than their relative backgrounds and even than relative limits in the national food standards (GBW-07601), respectively. Unfortunately, these metals were also found in hair from the residents and their concentrations were higher than relative backgrounds. These proved that the risk had come into observed harm to environment and even to human beings.

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