

Heavy metal contamination in soils of greenhouse vegetable production systems in a cold region of China

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Abstract: Heavy metal (HM) contamination in soils of greenhouse vegetable production (GVP) systems has drawn increasing attention in terms of food safety. In the present study, 64 surface soils were sampled, and the concentrations of select HMs were determined using atomic absorption spectroscopy. The results showed that the concentrations of cadmium (Cd), lead (Pb), zinc (Zn), copper (Cu), nickel (Ni) and chromium (Cr) in the soils were (0.2±0.2) mg/kg, (26.5±8.4) mg/kg, (101.4±43.2) mg/kg, (29.1±8.6) mg/kg, (24.5±3.3) mg/kg, and (56.5±6.3) mg/kg, and the corresponding accumulation index (AI) values were 2.30, 1.10, 1.43, 1.45, 1.07, and 0.97, respectively. The spatial distribution of the HMs suggested that Cd pollution displays a fractionation effect, which may be related to the source of Cd and its mobility. The concentration of Zn was significantly correlated with that of other HMs, implying that a comprehensive interactive effect might occur between Zn and other HMs. Furthermore, the values of the potential ecological risk index (RI) ranged from 41.23 to 185.91, meaning that attention should be paid to HM contamination of GVP soils to ensure food quality and safety.

Keywords: heavy metal contamination, cold region, greenhouse vegetable production (GVP), distribution, ecological risk, food quality, food safety

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1 Introduction

Greenhouse vegetable production (GVP) has rapidly expanded worldwide, especially in developing countries^[1-4]. This method has become a major contributor to vegetable production in China due increased living standards, increased demand for vegetable consumption and economic benefits for vegetable producers^[4,5]. In China, GVP areas were first established in 1983 but have expanded to 4.1 million hm² as of 2014^[6]. The total yield of greenhouse vegetables was 0.25 billion tonnes, which accounted for 30.5% of the total vegetable production in China. Although vegetable yields have increased in recent decades by means of intense agricultural practices, some ecoenvironmental issues have also been created because of large amounts of chemical inputs, high productivity and continuous cropping^[7,8].

Heavy metals (HMs) are among the most widely distributed and troublesome pollutants in the environment due to their persistent toxicity and bioaccumulation in the food chain, which can severely threaten human health and environmental safety^[9-12]. It is hard to avoid increasing HM accumulations in soils due to extraneous inputs from agricultural production and anthropogenic activities such as extensive applications of fertilizers, manure and

pesticides; wastewater irrigation; and atmospheric deposition^[6,13,14]. Specifically, manure, which can be used as an organic fertilizer to increase crop nutrient availability, soil organic matter (SOM), cation exchange capacity and water holding capacity, has the potential to be recycled in agricultural lands^[15]. However, manure applications have become an important source of certain HM inputs in soils^[16].

Since greenhouse vegetable consumption is important for the human diet, food security associated with GVP and consumption has received much attention^[17]. However, owing to its cold climate and long winters, the northeastern region of China experiences a contrasting supply and demand of vegetables; thus, intensive GVP can effectively alleviate this situation. Therefore, a systematic investigation and assessment of the occurrence and ecological risk of HMs in soils from GVP are necessary. Thus, the objectives of this study were (1) to investigate the levels of nutrients, pH, and SOM and the occurrence of HMs in GVP soils in the northeastern cold region of China; (2) to explore the spatial distribution characteristics of HMs; (3) to identify the variation characteristics and possible sources of HMs; and (4) to assess the potential ecological risk of HMs in the soils of the study area.

2 Materials and methods

2.1 Description of the study areas and soil sampling

Based on the different vegetable species (cucumber, potato, bean, eggplant and broccoli) planted and human activities, the sampling sites were divided into two groups: one group constituted an urban area, which included the Xiangfang, Daowai, Daoli, Nangang and Songbei districts, and the other group constituted a suburban country area, which included Acheng, Shuangcheng, Binxian, Bayan, Wuchang and Shangzhi. A total of 64 surface soil samples were collected in June 2014 (Figure 1); the samples were collected using a stainless steel auger. At each site, a composite sample was obtained from 5 random subsamples within an area of

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approximately 100 m². The soil samples were subsequently sieved through a 2 mm polyethylene sieve, and portions of the samples were sieved through a 0.15 mm mesh.

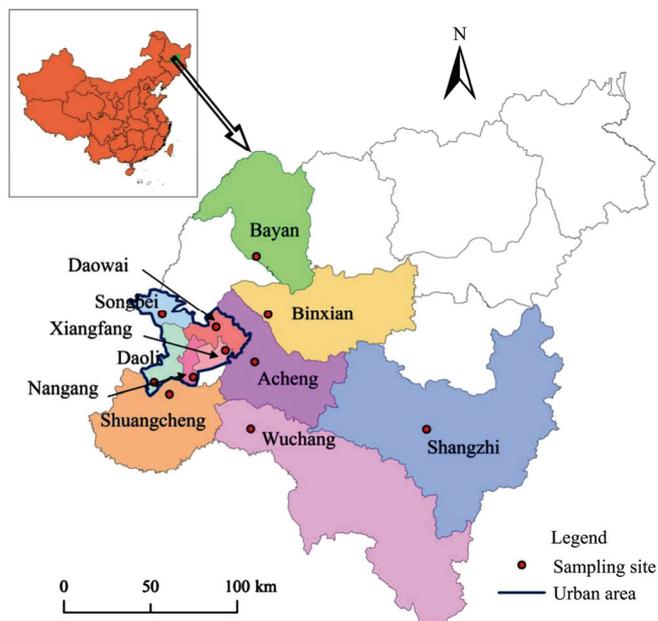


Figure 1 Study area and sampling sites

2.2 Chemical analysis

The soil properties, including the SOM, available nitrogen (AN), available phosphorus (AP), available potassium (AK), pH, and electrical conductivity, were measured using corresponding analytical methods of soil and agricultural chemistry^[18]. After they were digested with HClO₄, HNO₃ and HF according to the corresponding Chinese National Standards, HMs, including copper (Cu), cadmium (Cd), chromium (Cr), nickel (Ni), lead (Pb) and zinc (Zn), were detected by using flame spectrophotometry and a graphite furnace. Quality assurance and quality control throughout the whole HM analytical process were checked using soil standard references, procedure blanks and duplicate samples.

2.3 Potential ecological risk

A potential ecological risk index (RI) was developed by Hakanson^[19] to assess the degree of HMs and polychlorinated biphenyls (PCBs) for water pollution control purposes; this index has been widely applied to assess HM and PCB contamination levels in sediments and soils^[20-26]. This method is based on the assumption that the sensitivity of an aquatic system depends on its productivity. According to the toxicity of HMs and the response of the environment.

$$RI = \sum E_r^i \tag{1}$$

$$E_r^i = T_r^i C_f^i \tag{2}$$

$$C_f^i = C_0^i / C_n^i \tag{3}$$

where, RI is calculated as the sum of all risk factors for eight parameters (PCB and seven HMs); E_r^i describes the potential ecological risk factor for individual metals and includes five grades; T_r^i is the toxic response factor for metals and is based on the standardized HM toxicity factor developed by Hakanson^[19]; C_f^i is the contamination factor; C_0^i is the concentration of HMs in the soil, and C_n^i is a reference value for HMs (the local background value of the HMs was used).

In this study, only 6 types of HMs were detected, leading to the need to adjust the classification. In accordance with the results of Chen et al.^[22], the adjusted evaluation criteria based on the toxicity coefficient for these elements are presented in Table 1.

Table 1 Classification of the potential ecological risk of heavy metal pollution

E_r^i	Proposed by Hakanson RI	Improved RI	Ecological risk
$E_r^i < 40$	$RI < 150$	$RI < 55$	Low risk
$40 \leq E_r^i < 80$	$150 \leq RI < 300$	$55 \leq RI < 110$	Moderate risk
$80 \leq E_r^i < 160$	$300 \leq RI < 600$	$110 \leq RI < 220$	High risk
$160 \leq E_r^i < 320$	$RI \geq 600$	$RI \geq 220$	Very high risk
$E_r^i \geq 320$			Serious risk

3 Results and discussion

3.1 Soil properties and HM levels

A summary of the select soil physico-chemical properties, nutritive elements and concentrations of six HMs is presented in Table 2. The soil pH ranged from 6.1 to 7.3, with a mean value of 6.7, which equated to weakly acidic. The greenhouse SOM content in the study area was between 19.3 g/kg and 89.8 g/kg, with an average of 37.0 g/kg; this percentage was relatively greater than that recorded in Spain (2.0-42.0 g/kg)^[7] and in Beijing (6.0-71.0 g/kg)^[27] as well as in greenhouse soils along the Yellow Sea of China (7.6-51.6 g/kg)^[5]. The higher organic matter (OM) may be attributed to both the properties of the black soil in the northeastern region and over fertilization, especially with organic fertilizers^[5,28]. The OM and nitrogen (N) were significantly correlated ($R=0.604, p=0.000$), indicating that N fertilizer applications can enhance the content of OM in the soil. In addition, the SOM content increased with increasing greenhouse age ($R=0.721, p<0.05$). The concentrations of AN, AP, and AK were between 39.4 mg/kg and 144.9 mg/kg, 21.2 mg/kg and 299.4 mg/kg, 117.9 mg/kg and 968.7 mg/kg, respectively, with averages of 80.8 mg/kg, 169.5 mg/kg and 349.7 mg/kg, respectively. The soil conductivity ranged from 120.2 to 2320.0 μ S/cm, with a mean value of 808.2 μ S/cm, and widely varied in the study area.

Table 2 Soil properties and nutrient and heavy metal concentrations in greenhouse production systems in cold regions

	AN /mg·kg ⁻¹	AP /mg·kg ⁻¹	AK /mg·kg ⁻¹	pH	Conductivity / μ S·cm ⁻¹	OM /g·kg ⁻¹	Cd /mg·kg ⁻¹	Pb /mg·kg ⁻¹	Zn /mg·kg ⁻¹	Cu /mg·kg ⁻¹	Ni /mg·kg ⁻¹	Cr /mg·kg ⁻¹
mean	80.8	169.5	349.7	6.7	808.2	37.0	0.2	26.5	101.4	29.1	24.5	56.5
max	144.9	299.4	968.7	7.3	2320.0	89.8	1.1	56.3	226.0	54.7	33.1	75.4
min	39.4	21.2	117.9	6.1	120.2	19.3	0.0	13.6	43.3	16.1	15.5	40.0
standard	20.4	77.7	236.4	0.3	589.3	12.2	0.2	8.4	43.2	8.6	3.3	6.3
background values							0.086	24.2	70.7	20	22.8	58.6

The concentrations of HMs in all greenhouse soil samples are listed in Table 2. The mean concentrations of Cd and Cr in the greenhouse soils in Harbin were (0.2±0.2) mg/kg and (56.5±6.3)

mg/kg, respectively, which were lower than those from the GVP systems in Beijing, but the Pb concentration was 26.5 mg/kg, which was greater than that in Beijing^[27]. The results of the statistical

summary indicated that the HM concentrations in the greenhouse soils in the study area were in the following decreasing order: Zn>Cr>Cu>Pb>Ni>Cd. The HM content in all the studied soils was within the maximum limit levels of environmental standards for GVP in China^[29]. The surface soils of these 11 GVP sites in cold regions were slightly contaminated, and significant differences between the maximum and minimum HM concentrations may exist in both the urban and suburban areas. However, the values of the accumulation index (AI), which was calculated by dividing the average concentration by the background value of Cd, Pb, Zn, Cu and Ni, were 2.30, 1.10, 1.43, 1.45 and 1.07, respectively, and the value for Cr (0.97) almost reached the background value. These results indicated that long-term vegetable production can lead to significant HM cumulative effects in soils, especially for Cd, Zn and Cu.

3.2 Spatial distribution

Spatial heterogeneity is the most essential characteristic of HMs and other organic pollutants in soils and is influenced by many factors, such as the applications of fertilizers and pesticides, agricultural planting structure, wastewater irrigation, soil properties, different climatic conditions and atmospheric deposition^[30-32]. Overall, all these factors could be attributed to comprehensive

effects that result from human activities, the development of industry and agriculture and increasing material demands associated with the pursuit of better life.

The maximum and minimum concentrations of HMs in the GVP soils of 11 sampled sites are presented in Figure 2. The results showed that the maximum concentrations of the studied HMs were present in urban areas (at Xiangfang, except for Pb at Daoli) and in Shangzhi in the suburban country areas, with the exceptions of Ni and Cr. These findings also indicated that the spatial distribution of HMs in the greenhouse soils in the cold regions varied widely, depending on their location. Notably, the average concentration of Cd decreased from urban to suburban areas with increasing distance, except at Shangzhi. Thus, the occurrence of Cd presents a fractionation effect, which may be related to the distance from emission sources; i.e., the farther away from the source, the lower the concentration is. Local sources or atmospheric transport could lead to this distribution pattern, which contrasts with the results of a previous study by Sakata and Asakura^[32]. The study by those authors indicated that the relatively low dry deposition velocities for Cd could not lead to effective dry deposition via long-range atmospheric transport^[32]. However, Cong et al.^[31] reported that wet precipitation resulted in elevated

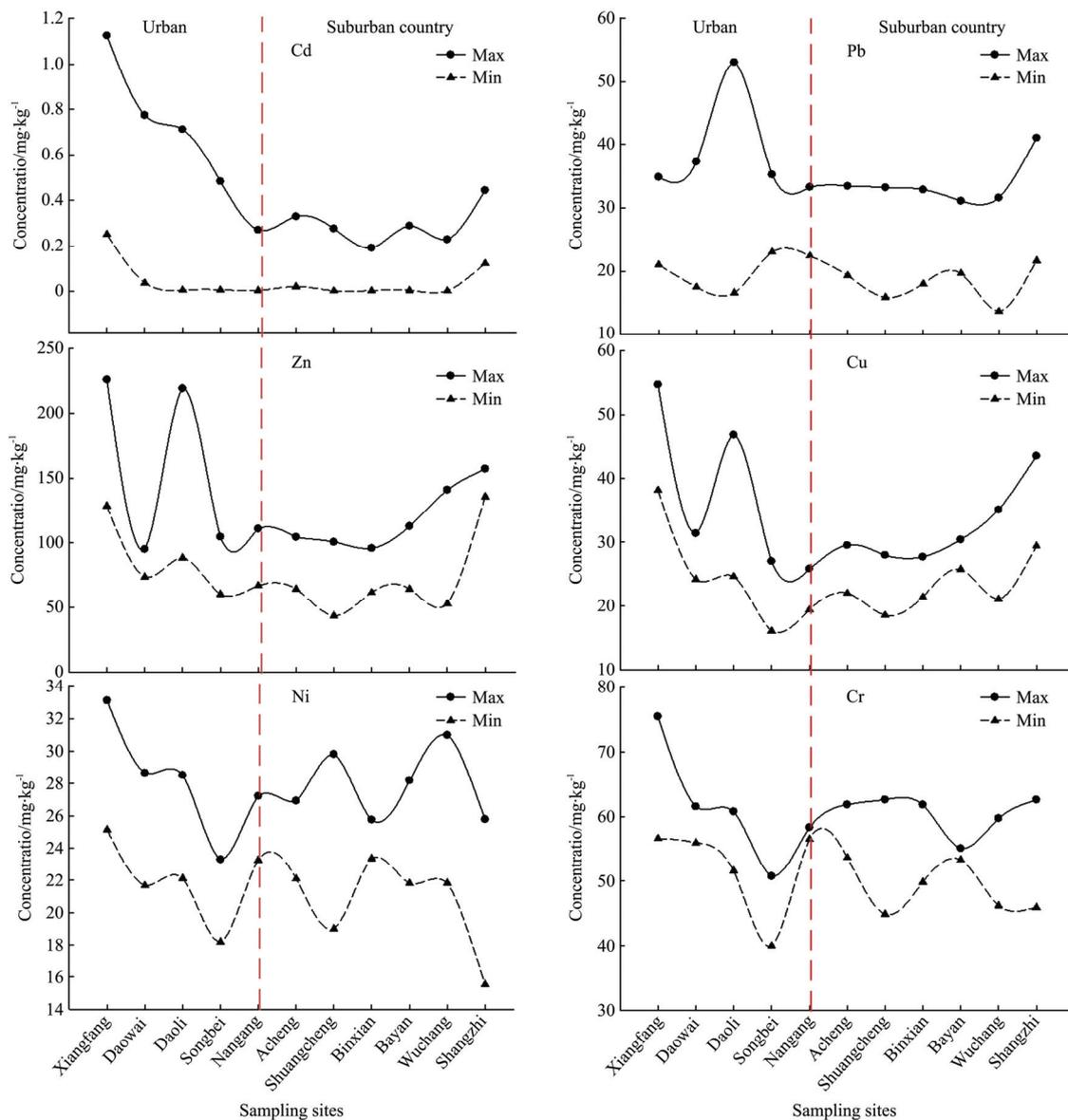


Figure 2 Distribution of heavy metals in the soils of greenhouse vegetable systems in cold regions

concentrations of Cd, Pb, Zn, Cu, Ni and Cr. Hence, the distribution pattern of Cd from urban to suburban areas in the present study may result from a combination of local sources, as well as dry and wet deposition. In addition, no significant spatial distribution characteristics were observed for the other HMs in the study area, which may be the result of many factors. In fact, point source emissions from industrial production, emissions from transportation sources and over application of fertilizers and pesticides could influence the high levels of Pb, Zn and Cu at Daoli and Shangzhi in the urban and suburban country areas, respectively.

3.3 Possible sources

In general, the coefficient of variation (C_v) is a useful statistic for comparing the degree of variation of one data series with that of Cui et al.^[26] The C_v values of HMs in the soils in the present study are presented in Figure 3, which shows that Cd, Zn, Cu, Pb and Ni and Cr correspond to moderate variability and weak variability, respectively. Cd, Zn, Cu and Pb present a relatively large degree of dispersion, which may result from different fertilizer and pesticide applications or atmospheric deposition. In contrast, there is a lower degree of dispersion for Ni and Cr, which could indicate similar sources. To explore and elucidate the possible sources of HMs in the greenhouse soils, the relationships between the different HMs and the age of the greenhouse were investigated by calculating the Pearson correlation coefficients for the concentrations of the different HMs and the age of the greenhouse among the sampling sites.

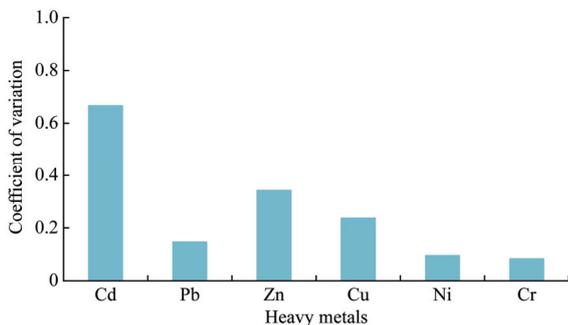


Figure 3 Coefficient of variation for heavy metals in the soils of greenhouse vegetable production systems in cold regions

The concentrations of all HMs are strongly correlated with greenhouse age (Table 3), indicating that the continuous accumulation of HMs in soils increases with increasing greenhouse longevity. Rotating vegetable species generally results in the absorption of soluble and ionic HMs, leading to a reduction in HM contents; however, a large amount of applied fertilizer or extraneous input of HMs can disrupt this status. Thus, the accumulation of HMs in greenhouses that have a long service life and the possible ecological risks should be causes of concern to the government and public. In the present study, nutrient contents and HMs were significantly positively correlated; the coefficients between N and P and Zn were 0.394 ($p < 0.01$) and 0.483 ($p < 0.01$), respectively; between N and Cu, 0.458 ($p < 0.01$); and between P and Cu, 0.410 ($p < 0.01$). No correlations between nutrient contents and the other HMs were found. Hence, the results of the present study showed that fertilizer applications can lead to Zn and Cu accumulations in GVP soils to some extent. In addition, the presence of Cu and Zn in the GVP soils in the study area may be attributed to organic fertilizer applications. Indeed, applications of animal manure as an organic fertilizer can lead to high concentrations of HM (Cu, Zn, Pb, Cd and Cr) contamination and can affect soil quality^[16]. Atafar et al.^[33]

also reported that Cd and Pb concentrations increased in cultivated soils with increasing fertilizer applications. Therefore, the application of different fertilizers might be among the major reasons for the differences in the spatial distribution of HMs. Notably, significant correlations between Zn and other HMs were found, suggesting that there are comprehensive interactions between Zn and other HMs, which are intimately or symbiotically related.

Table 3 Pearson correlation coefficients between heavy metals and greenhouse age

	Cd	Pb	Zn	Cu	Ni	Cr	greenhouse age
Cd	1						
Pb	0.273*	1					
Zn	0.359**	0.442**	1				
Cu	0.401**	0.245	0.798**	1			
Ni	0.238	0.308*	0.448**	0.527**	1		
Cr	0.250*	0.007	0.357**	0.527**	0.599**	1	
Greenhouse age	0.322**	0.313*	0.715**	0.548**	0.268*	0.309*	1

Note: *Correlation is significant at the 0.05 level (2-tailed); **Correlation is significant at the 0.01 level (2-tailed).

3.4 Potential ecological RI

According to the adjusted classification criteria, the spatial distribution of E_r^i and the RI in the present study is shown in Table 4. The individual RI s of the HMs were evaluated, and the potential ecological risk increased in the order of Zn < Cr < Ni < Pb < Cu < Cd. The average ecological risk of Cd in the GVP soils was 71.36, indicating that Cd could cause a moderate potential ecological risk to local vegetable production systems. The values of RI ranged from 41.23 to 185.91, indicating a large variation in the potential ecological risk (from low to high risk) in the GVP soils in this study. On the basis of calculated RI values, the risk posed by the accumulation of HMs at different sampling sites increased in the order of Wuchang < Bayan < Nangang < Binxian < Shuangcheng < Acheng < Daoli < Songbei < Shangzhi < Daowai < Xiangfang. In summary, a low ecological risk of HMs occurred at the Wuchang sampling site; a moderate ecological risk occurred at Bayan, Nangang, and Binxian as well as Shuangcheng, Acheng, Daoli and Songbei; and a high ecological risk occurred at Shangzhi, Daowai and Xiangfang, with the potential ecological risk caused primarily by Cd being a large concern. GVP in cold regions is an important agricultural activity, method for supplying vegetables and way for increasing farmers' income; thus, protecting soil quality and improving crop quality are very necessary.

Table 4 Evaluation of the potential ecological risk of heavy metals in soils

Sampling sites	E_r^i						RI
	Cu	Cr	Zn	Pb	Ni	Cd	
Xiangfang	11.34	2.22	2.47	6.29	6.15	157.44	185.91
Daowai	6.94	2.01	1.22	6.33	5.36	156.10	177.97
Daoli	8.20	1.95	1.80	6.00	5.66	61.63	85.23
Songbei	5.26	1.54	1.18	6.43	4.43	68.43	87.27
Nangang	5.95	1.95	1.35	6.07	5.63	38.37	59.33
Acheng	6.43	1.96	1.16	4.79	5.28	50.93	70.55
Shuangcheng	6.07	1.86	0.97	4.38	5.12	45.81	64.21
Binxian	6.42	1.99	1.02	5.07	5.39	41.32	61.20
Bayan	7.05	1.85	1.16	4.99	5.28	36.56	56.88
Wuchang	6.38	1.92	1.15	4.34	5.54	21.90	41.23
Shangzhi	8.65	1.89	2.07	6.05	4.44	106.51	129.62

4 Conclusions

The results showed that greenhouse age and N fertilizer applications affected SOM contents and that long-term vegetable production can lead to significant cumulative effects of HMs in soils, especially for Cd, Zn and Cu. In particular, the maximum concentrations of the studied HMs occurred mainly in urban areas. Different fertilizer applications or extraneous inputs could lead to the continuous accumulation of HMs in soils and comprehensive interactions between Zn and other HMs, which are intimately or symbiotically related. The ecological risk assessment results indicated that Cd could cause a moderate potential ecological risk to local vegetable production systems. The values of *RI* ranged from 41.23 to 185.91, indicating a large variation in ecological risk (from low to high risk) in GVP soils in this study, and this phenomenon should receive attention in terms of soil environmental quality and health risks. In fact, the motivation of this study was a preliminary investigation of the concentrations, sources and risks of HMs, and the objective was to enrich the basic database of HMs occurring in greenhouses in cold regions. Despite this objective, there is a strong need to improve and carry out additional detailed studies with better methodologies and investigate bioaccumulation effects in subsequent research.

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