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## Research Article


# Evaluation of Lead in Arable Soils, China

Lead (Pb) contamination in arable soils is one of the most serious ecological problems due to its high toxicity on human health. Thus, we need to understand the concentration level, contaminated area, and spatial distribution of Pb in arable soils on regional or national scale. This paper reviewed the studies on Pb concentrations throughout Chinese arable soils, based on relevant 537 studies from 2002 to 2014. The results showed that the average Pb concentration was 34.41 mg/kg, higher than its background of 23.50 mg/kg, indicating that Pb has been introduced into soil from exterior sources. Mining and smelting activities, irrigation by wastewater, and urban development greatly contributed to Pb accumulation in arable soils. North China had lower Pb concentrations than the south, and many hotspots existed on the Pb concentration map due to mining and smelting activities. On the provincial scale, arable soils in Yunnan, Guangxi, and Shaanxi Provinces were moderately polluted by Pb, Gansu and Shaanxi Provinces were slightly affected by Pb, while the other provinces showed relative safe levels.

**Keywords:** Contamination; Farmland; Heavy metals; Pollution

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## 1 Introduction

Lead (Pb) is a highly toxic element to humans when ingested or inhaled, particularly detrimental to the neurological development of children. Since the phase-out of leaded gasoline and the subsequent reduction of airborne lead, food and water are still the primary sources of Pb exposure to people [1, 2]. In 2002, the quality monitoring center for rice and rice products at China's Ministry of Agriculture reported that the most widespread problem was Pb contamination, with 28.40% of samples breaching limits ([www.chinadialogue.net/article/show/single/en/4197-China-stained-rice-trail](http://www.chinadialogue.net/article/show/single/en/4197-China-stained-rice-trail)). Another study in 2007 showed that about 12.09% of rice samples exceeded the limit of Pb content in grains [3].

Pb concentrations in arable soils have drawn wide attention [4, 5]. Most studies described the ranges of soil Pb concentrations at some specific locations and those affected by human activities [6–10], and concluded that regional Pb variations in arable soils greatly depended on initial concentrations in parent materials and exterior inputs, including mining and smelting, waste disposal, urban effluent, vehicle exhausts, sewage sludge, pesticides, and fertilizer application [11, 12].

On the national scale, it is essential to understand the size of the affected area by Pb, the level of soil Pb concentration and of its spatial distribution [6, 13]. Wei and Yang in reviewing 12 studies, found that Pb concentration was 37.55 mg/kg in Chinese agricultural soil in urban environment [5]; Niu et al. calculated an average Pb

of 33 mg/kg in farmland soil from 131 sampling points [14]; Song et al. reported that the average Pb was 34.86 mg/kg from reviewing 131 regions [15]. Although there have been several studies on Pb concentrations in arable soils on the national scale in China, it is not enough to understand the whole situation of China due to the limited number of sampling points [5, 14, 15].

China's Ministry of Environmental Protection and Ministry of Land recently reported that 1.50% of arable soil samples collected during 2005–2013 have been polluted by Pb ([www.sdpc.gov.cn/fzggz/mcjj/zhd/201404/t20140418\\_607888.html](http://www.sdpc.gov.cn/fzggz/mcjj/zhd/201404/t20140418_607888.html)). However, little is still known about general Pb concentrations and the spatial distribution in arable soils throughout China.

This study reviewed relevant studies on Pb concentrations in Chinese arable soils, aiming to acquire the spatial distribution of Pb concentrations, to identify the contribution of mining and smelting activities, wastewater irrigation, and urban development on Pb accumulations in arable soils, and to assess Pb pollution in arable soils in China.

## 2 Materials and methods

### 2.1 Data collection

The arithmetic Pb concentrations in surface arable soils (0–20 or 0–15 cm) were collected from studies of published papers. The papers were located by searching through (1) the ISI Web of Knowledge website using the keywords “soil heavy metal or Pb concentration”, and “arable land”, and “China”; (2) the CNKI website using the

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Chinese key words for “soil heavy metal or Pb concentration”, and “arable land”, and “China”; and (3) the relevant references in the papers obtained through the above mentioned methods. The following conditions were used to select the studies in these papers: (1) the paper was published during 2002–2014; (2) the paper included the information on the number of soil samples and the average of Pb concentrations. Finally, 537 peer reviewed articles consisting of 1194 data records on Pb concentrations were collected. Basic information, such as sample locations, Pb concentrations, land use, number of soil samples, and time of investigation, were collected from each study (Supporting Information Tab. S1).

Since 1995, China enacted a series of standards on sampling and monitoring heavy metal in soils, crops, and vegetables, such as the Environmental Quality Standards for Soils (GB 15618–1995) in 1995, and the Environmental Quality Risk Assessment Criteria for Soil at Manufacturing Facilities (HJ/T25-1999) issued by the State Environmental Protection Administration of China in 1999, etc. [16]. The sampling methods, soil pretreatment, and analyses followed these standards.

To map spatial distribution of Pb concentration in Chinese arable soils, the data at point and county scales (the map with the minimum administrative unit that we were able to obtain) were separately recorded in Supporting Information Tab. S1. If several data records were collected in one county or at one point, the sample-number-weighted mean was calculated as the Pb concentration in this county or at this point. The numbers of records at point and county scales were 291 and 387, respectively (Fig. 1).

To get general information of Pb concentrations, the data records at the prefectural level or provincial level, and the data at point and county scales were used. In total, 702 data records were used.

Among the 702 data records, the land category information including the areas around (i) mining and smelting plants, (ii) irrigation area, (iii) urban area, and (iv) remote area (the area without obvious point pollution sources) was collected from the original studies. Pb concentrations in these areas were separately recorded to study the contribution of potential sources for Pb

accumulation in arable soil. The number of the Pb concentration records was 133, 60, 103, and 406, respectively, in those areas.

## 2.2 Statistics methods

The statistical analysis of minimum, maximum, and standard deviation is conducted to understand Pb concentrations in arable soils. Since the sampling number in each study varied considerably, the arithmetic mean might not reflect the general situation of Pb concentrations. The sample numbers are taken as the weights to calculate the sample-number-weighted mean for Pb concentrations:

$$C = \frac{C_i N_i}{\sum_{i=1}^n N_i} \quad (1)$$

where  $N_i$  is the sampling number in the data record  $i$ ,  $C_i$  is the Pb concentration in data record  $i$ , and  $n$  is the number of the data records.

## 2.3 Spatial distribution

### 2.3.1 Trend analysis

Trend interpolation fits a smooth surface defined by a mathematical function to the input sample points, based on a global polynomial interpolation method. The trend surface changes gradually and captures coarse-scale patterns in the data. In this study, the trend analysis was used to model the general variation of Pb throughout Chinese arable soils.

### 2.3.2 Kriging interpolation method

Kriging, a geostatistical method, is used to estimate and map soil Pb concentrations in China. The main application of geostatistics

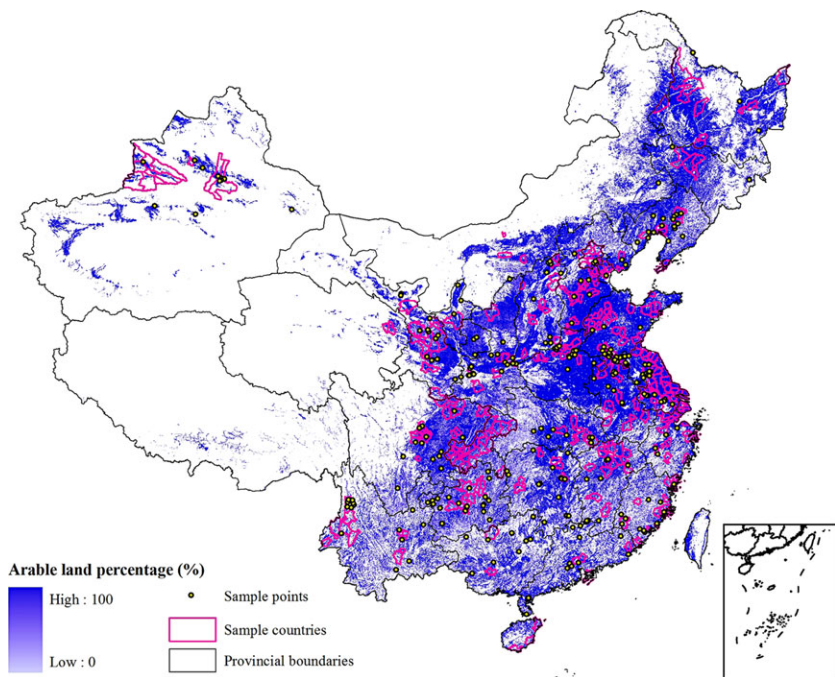


Figure 1. Spatial distribution of soil samples in arable soil in China.

to soil science is to estimate and map soil attributes in unsampled areas [17]. This method uses the semi-variogram to quantify the spatial autocorrelation and to model the spatial variation of a regionalized variable. Based on fitted semi-variogram models, the method of ordinary Kriging was used to map Pb distribution in China.

## 2.4 Assessment of Pb accumulation in arable soils

### 2.4.1 Pb accumulation assessment

The sample-number-weighted mean of Pb concentration on the provincial scale was calculated based on Eq. (1). Then, the Pb accumulations were calculated as:

$$A_m = C_m - B_m \quad (2)$$

where  $A_m$  is the Pb accumulation in province  $m$ ,  $C_m$  and  $B_m$  are the Pb concentration and background value in province  $m$ .

### 2.4.2 Geoaccumulation index

The index of geoaccumulation is used to assess the contamination by comparing current and background concentrations. It was computed using the following equation:

$$I_{geo} = \log_2 \frac{C_i}{1.5B_m} \quad (3)$$

where  $C_i$  is the measured Pb concentration, and  $B_m$  is the geochemical background value in province  $m$ . This index is distinguished into six grades. The detailed information has been described in the study [18].

## 3 Results and discussion

### 3.1 Pb concentrations in arable soils

The total investigated area was about 789 900 km<sup>2</sup>, accounting for 58.34% of the total arable area in China (1 353 850 km<sup>2</sup>, [http://news.xinhuanet.com/english/china/2013-12/30/c\\_133007338.htm](http://news.xinhuanet.com/english/china/2013-12/30/c_133007338.htm)); the number of soil samples in the 702 data records was 134 755. The

investigated area was distributed in 30 provinces, municipalities or districts, covering most of the mainland area of China.

The Pb concentrations widely ranged from 1.21 to 5579.66 mg/kg with the standard deviation (SD) of 340.60 mg/kg. The wide range of Pb concentrations and the high SD value denoted that Pb concentrations in separate studies were spread out over a large range of values. The sample-number-weighted mean of Pb concentration was 34.41 mg/kg. This value was higher than its background of 23.50 mg/kg [19], indicating that Pb has been introduced into arable soils from human activities, but it was much lower than the maximum permissible concentrations of potential toxic elements (250 mg/kg when pH < 6.5, 300 mg/kg when 6.5 < pH < 7.5, 350 mg/kg when pH > 7.5; China soil environment quality standard, GB15618-1995).

The average Pb concentration in this study was close to 34.86 mg/kg [15] and 33.00 mg/kg [14], but a little lower than 37.55 mg/kg [5]. Compared to Pb concentrations in other countries, Chinese arable soils had a much higher Pb concentration than 26 European countries, Malaysian, Thailand, and the USA, but close to the world average (Tab. 1).

The frequency of data records with Pb concentration is shown in Fig. 2. About 28.92% of data records had lower Pb concentrations than its background, while the others were higher than the background. In total, about 54.13% of the data records had the Pb concentration lower than the grade I reference (35 mg/kg), indicating more than half of investigated soil samples were not greatly affected by exterior sources. Ninety-two point thirty-one or 93.87% of the data records had lower Pb concentrations than the grade II value of 250 mg/kg (pH < 6.5) or 350 mg/kg (pH > 7.5), indicating that such arable soils are safe to plant crops or vegetables. The percentage of data records with Pb concentrations higher than grade II (350 mg/kg) was 6.13%, which is much higher than 0.72% provided by Song et al. [15].

### 3.2 Potential sources of Pb accumulations in arable soils

To evaluate the contribution of possible human activities on Pb accumulations in soil, the minimum, maximum, SD, and sample-number-weighted mean of Pb concentrations in the areas around mining and smelting activities, irrigation by wastewater, urban and suburban areas, and remote areas (where no obvious point Pb sources exist) were calculated separately (Tab. 2).

**Table 1.** Comparison of Pb concentration in Chinese arable soil with previously published surface soil Pb concentrations in China and other countries or regions

Country	Land use	Number of samples	Mean (mg/kg)	Minimum (mg/kg)	Maximum (mg/kg)	Reference
China	Arable soil	134 755	34.41	1.21	5579.66	This study
China	Arable soil	138 regions	34.85	3.79	341	[62]
China	Arable soil	131	33	10.1	184.2	[63]
China	Agricultural soil	12 regions	37.55	17.11	77.27	[5]
USA		4841	25.8	<0.5	12 400	b)
Malaysian	Crop soil	241	26.4	0.85	90	[64]
Thailand	Crop soil	318	17.5	0.1	550	[65]
Europe	Agricultural soil	2211	16 <sup>a)</sup>	1.6	1309	[66]
World			33.7	0.1	1520	[67]

<sup>a)</sup> Median.

<sup>b)</sup> <http://pubs.usgs.gov/ds/801/>

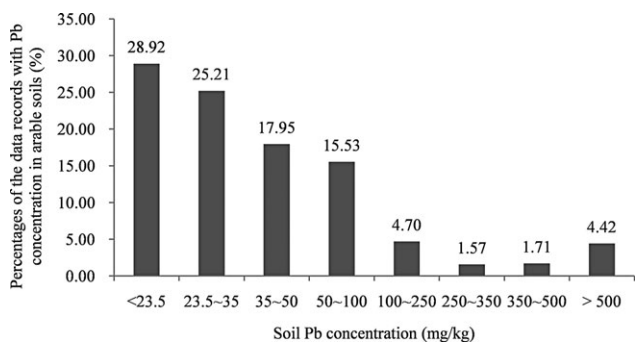


Figure 2. Frequency distribution of Pb concentrations in arable soil in China.

### 3.2.1 Pb concentrations in arable soils around mining and smelting activities

Arable soils around mining activities had an obvious higher Pb concentration than the other areas. The soil-sample-number weighted mean of Pb concentration was 9.70, 6.53, and 4.50 times of that in remote areas, urban environments, and irrigation areas. This is mainly because the mining and related activities, such as metal processing and smelting discharge large amounts of wastewater, waste gas, and solid waste to the environment, which often contained Pb [20]. Thus, Pb could enter into soil through atmospheric diffusion or surface runoff flushing or weathering [21, 22].

The range of Pb concentration in arable soils around mining and smelting activities was 5.95–5579.66 mg/kg, with a high SD value of 719.17 mg/kg. The highest value was 16 times of the grade II reference of 350 mg/kg, indicating some areas had been heavily polluted with Pb. The sample-number-weighted mean (266.49 mg/kg) was found to be much lower than the average of Pb concentration listed in the previous review paper of 72 examined mining areas in China (641.30 mg/kg) [18]. The big gap might be because this study only collected Pb concentration in arable soils, while the study by Li et al. collected soil samples in all kinds of land use soils [18]. Arable lands are often located apart from mining areas or smelting plants, and these areas had relative low Pb concentrations in soil [23, 24].

From the frequency distribution of the studies around mining activities (Fig. 3), about 29.32% of the data records are higher than the grade II reference of 350 mg/kg, and the others were in the range of 5.95–350.00 mg/kg. This also indicated that mining activities introduced a great amount of Pb into arable soils, leading to heavily polluted soil. Among the 18 data records with Pb values less than the

background of 23.50 mg/kg, most of them are distributed around the areas of coal mining (Tab. 3).

Table 3 shows the Pb concentrations in different kinds of mining and smelting areas. It is obvious that non-metallic minerals (including coal and the other non-metallic minerals) contained less Pb than non-ferrous metal ores. Among the non-ferrous mining and smelting areas, copper and gold activities introduced relative less Pb into arable soils, while the lead/zinc and antimony introduced a large amount of Pb into soil. In the area of manganese, tungsten, mercury mining and smelting, the average Pb concentrations were about 150 mg/kg, and in the iron and tin areas, the Pb was accumulated to about 250 and 350 mg/kg, respectively. Similar results were obtained by Li et al. [18].

### 3.2.2 Pb concentrations in arable soils in irrigation areas

Wastewater irrigation creates both opportunities and problems in the agricultural sector [25, 26], since it provides convenient disposal of waste products and has beneficial aspects of adding valuable plant nutrients and organic matter to soil [27, 28]. However, excessive Pb accumulation in soil through wastewater irrigation may not only result in soil contamination, but also affect food quality and safety [29]. In this study, the sample-number-weighted mean was 59.26 mg/kg, much higher than the background value of 23.50 mg/kg in China.

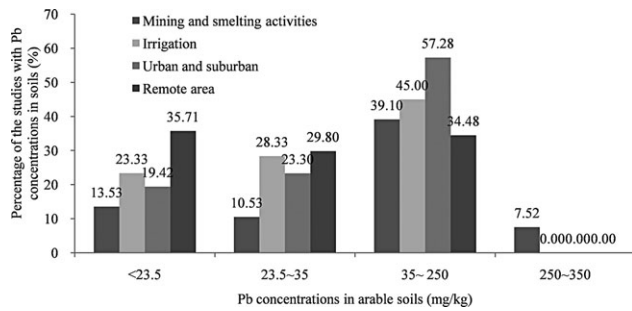
Pb concentrations in irrigated areas ranged from 9.63 to 573.61 mg/kg, with a high SD value (93.05 mg/kg). This demonstrated that Pb concentrations varied greatly in irrigated areas, mainly due to the quality of the irrigation water. If low Pb content was in the irrigation water, such as the Miqan River of Xinjiang [30], Hongsiyu River of Ningxia [31], Taiyuan River of Shanxi [32], Jinghuiqu River of Shaanxi [33], irrigated soil Pb concentration was also low. If a high Pb content could be detected in irrigation water, such as water affected by mining and smelting activities [34], or industry effluents [35], soil would be seriously polluted by Pb. Besides the water quality of the reclaimed water, Pb concentrations in arable soils are also determined by irrigation rate, soil properties, crop usage, and the irrigation period [36]. Therefore, to promote the safety of the reclaimed water for irrigation, the impacts of these influencing factors on Pb accumulation in soil should be studied [37].

The Pb concentrations <23.50 mg/kg accounted for 23.33%, and about 73.33% was within the range of 23.50–350.00 mg/kg (Fig. 3). This denoted that at least one quarter of water was good enough to be used to irrigate crops, where the others introduced Pb into arable soil. The data records with Pb exceeding 350 mg/kg in irrigation areas accounted for about 3.33%, which was lower than the ratio in the soil around mining activities. However, due to the large area of

Table 2. Pb concentrations in arable soil in those areas of mining and smelting activities, urban and suburban, irrigation, and remote areas

	Mining/smelting activity	Irrigation area	Urban/suburban area	Remote area
Number of study	133	60	103	406
Sample-number-weighted/mean	266.49	59.26	40.78	27.47
Minimum	5.95	9.63	8.71	1.21
Maximum	5579.66	573.61	193.42	209.56
SD	719.17	93.05	29.63	20.81

SD, standard deviation.



**Figure 3.** Frequency distribution of Pb concentrations in arable soil around areas of mining and smelting activities, irrigation, urban and suburban, and remote areas in China.

wastewater irrigation in China (about 14 000 km<sup>2</sup> in 2012), Pb concentrations in wastewater is one of the main causes of soil contamination in China [27, 38].

### 3.2.3 Pb concentrations in arable soils in urban areas

Soils around urban and suburban areas differ greatly from natural soils, since urban areas are the most densely populated regions of the world because of their strong industrial and economic activities [9, 39]. There are several reasons for a higher Pb concentration in arable soils in urban and suburban areas than in remote areas. Firstly, the large urban population generates a large amount of waste, some of which is applied to surrounding arable soils [40]. Secondly, vehicle emissions are considered to be the principal Pb source of urban soil, although Pb addition in gasoline have been banned [9, 41].

The sample-number-weighted mean of Pb was 40.78 mg/kg, which was lower than that in urban soils of 31 metropolises in China (45.30 mg/kg in topsoil) [9] and 47.34 mg/kg based on 3269 sampling points in 43 cities [8], 61.30 mg/kg calculated from 21 cities [42]. This big difference might be due to the fact that arable soil samples in urban areas were collected in this study, while the other studies collected soil samples in various kinds of land uses in urban environment. But the sample-number-weighted mean of Pb was a little higher than 37.55 mg/kg in agricultural soil of 12 cities [5].

Pb concentrations in urban environment ranged from 8.71 to 193.42 mg/kg, with an SD of 29.63 mg/kg. The percentage of data records with Pb concentrations <23.50 mg/kg was 19.42%, and the others were within a range of 23.50–250.00 mg/kg (Fig. 3). This

demonstrated that although arable soils had accumulated Pb in urban environment, it was still in the range of providing safe food.

### 3.2.4 Pb concentrations in arable soils in remote areas

In remote areas, the sample-number-weighted mean of Pb concentration was 27.47 mg/kg, which was much lower than that in the studies by Song et al. [15], Niu et al. [14], and Wei and Yang [5]. This indicated that the accumulation of Pb in remote areas was not that serious as the former studies described. It should be noted that the average of Pb was higher than its background value (23.50 mg/kg), indicating that Pb has been introduced into soil by anthropogenic activities in remote areas.

The data records with Pb concentrations lower than the background accounted for 35.71%, while the others were distributed in the range of 23.50–209.56 mg/kg. The exterior sources mainly referred to normal agricultural practices such as application of liquid and soil manure or inorganic fertilizers, or pesticide sprays [43, 44]. Phosphorus fertilizers and organic manure contained high Pb [45], and soil Pb concentrations were positively correlated with phosphorus concentrations [46]. Moreover, the application of plastic film in agricultural activities can introduce some amount of Pb into soil, because the film production process adds heat stabilizers containing Cd and Pb [15]. Most of the plastic films are not biodegradable, and the residual of plastic films will bring hazards to the arable soil.

## 3.3 Spatial distribution of soil Pb concentrations in arable soils in China

Data of Pb at points and countries were used to obtain spatial variation of Pb concentration in arable soils. Surface data at the country level were first converted into points at spatial resolution of 2 × 2 km<sup>2</sup>, and then the data were merged to a point data file, which contained 2003 Pb concentrations.

### 3.3.1 Trend distribution of Pb concentrations in arable soils

Polynomials with 1, 2, and 3 orders were performed to gain the spatial surface of Pb trends. Among the three surfaces, the third order model gained the lowest root mean square value (225.55). The result is described in Fig. 4a. The map showed that arable soils in southern China had higher Pb concentrations than the north, and the northwest and the northeast had lower Pb concentrations than

**Table 3.** Statistic of Pb concentrations in different kinds of mining and smelting areas

	Number of data records	Minimum	Maximum	Average
Coal	26	7.80	287.02	38.18
Other non-metallic minerals	12	21.06	98.76	58.31
Cu	12	19.33	116.38	56.73
Au	3	14.32	216.93	85.06
Mn	7	41.63	238.73	143.64
W	9	5.95	301.98	146.20
Hg	1	150.35	150.35	150.35
Fe	2	157.45	348.67	253.06
Sn	3	33.22	970.00	348.46
Pb/Zn	55	19.29	16702.50	1330.11
Sb	3	281.22	5579.66	3414.29

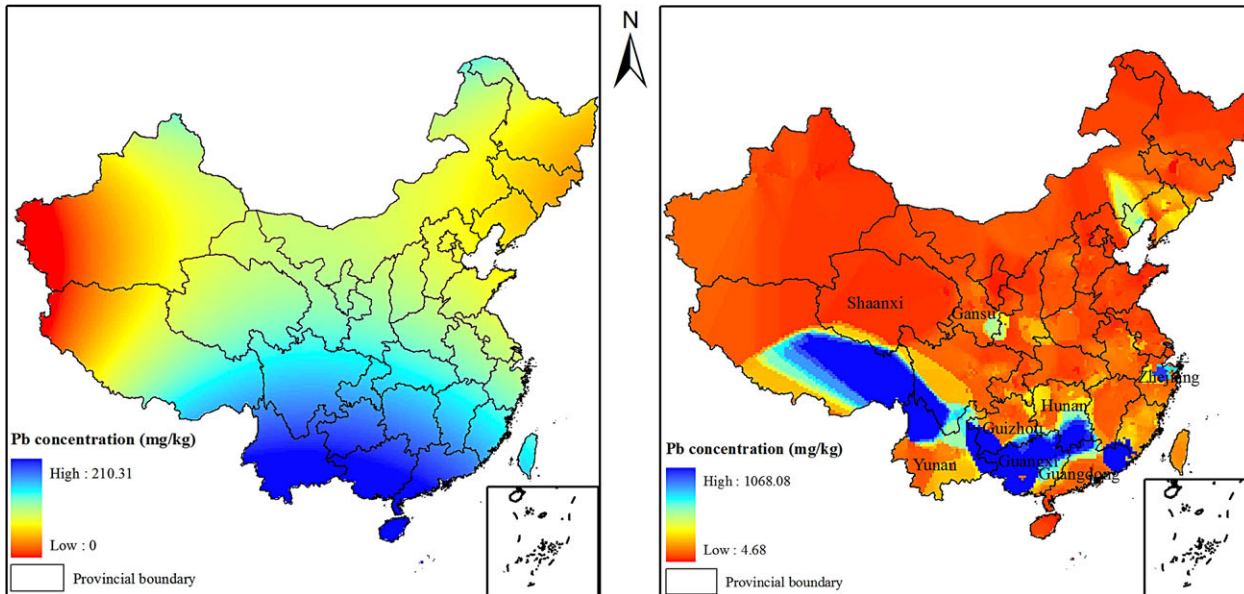


Figure 4. Spatial distribution of soil Pb concentration in China (a) by trend analysis, and (b) by Kriging method.

the central area. Niu et al. [14] and Song et al. [15] obtained similar spatial trends of Pb in China. However, although the trend surface showed obvious spatial variation of Pb concentrations on the national scale, it could not reflect the spatial variations on the local scale.

3.3.2 Spatial distribution of Pb concentrations by Kriging interpolation method

To obtain spatial distribution of soil Pb concentration, 2003 data pairs on Pb concentrations in arable soils were logarithmically transformed to conform to normal distribution. The summary of the four semivariogram models is given in Tab. 4. All four modes showed high accuracy of simulation, but the experimental semivariogram suggested that the theoretical exponential model was in reasonable agreement with the data for soil Pb concentrations, since it achieved the maximum R<sup>2</sup> value and the minimum root sum square value.

The ratio of nugget to sill (C<sub>0</sub>/(C<sub>0</sub> + C)) indicates the proportion of random components to system spatial heterogeneity. The ratio <0.25 for the exponential model denotes that the proportion of the spatial structure of soil Pb concentration had strong spatial autocorrelation [14]. To some extent, this indicator reflected that

an intrinsic factor, such as the initial concentrations of Pb in parent materials, is the predominant factor impacting the spatial variability of Pb in Chinese arable soils [47].

To understand the spatial patterns of soil Pb in arable soils, ordinary Kriging interpolation was used to obtain the filled contours map (Fig. 4b). Similar to the trend surface, this map also shows that northern China has relative lower Pb concentration in arable soils than the south, and the central has obviously higher Pb concentrations than the west and east.

Many hotspots exist on the Pb map throughout China, and most of them were located in southern China, for example, Lanping in Yunnan Province [48, 49], Qinglong and Liupanshui in Guizhou Province [50, 51], Hechi and Siding in Guangxi Zhuang Autonomous Region [52, 53], Lechang and Meixian in Guangdong Province [54, 55], Xiangxi, Chengzhou, Zhuzhou, and Hengyang in Hunan Province [56, 57]. Some hotspots were located in the central and north of China, such as Shangyu in Zhejiang Province [58], Qinling in Shaanxi Province [24], Huludao in Liaoning Province [59]. Most of the hotspots were mainly due to lead/zinc mining and smelting activities, while some of them were induced by antimony smelting. Such high Pb concentrations in soils around lead/zinc or antimony smelting activities were also found in Poland and England [60, 61].

Table 4. Semivariogram models of soil Pb concentrations and the parameters

Model	Nugget variance (C <sub>0</sub> )	Sill (C <sub>0</sub> + C)	C <sub>0</sub> /(C <sub>0</sub> + C)	Range (A, km)	Prediction error	
					R <sup>2</sup>	RSS
Exponential	0.20	0.94	0.21	1246	0.97	0.02
Linear	0.29	0.86	0.34	2044	0.93	0.03
Spherical	0.24	0.80	0.30	2147	0.96	0.02
Gaussian	0.31	0.79	0.39	1016	0.93	0.03

RSS, root sum square

### 3.4 Assessment of the accumulation of Pb in arable soils in China

Pb concentrations in arable soils grouped by Chinese provincial regions are shown in Supporting Information Tab. S2. The number of data records on the provincial scale ranged from 2 to 62, and the number of investigated samples ranged from 32 to 31 972. In some provinces, such as Qinghai Province and Ningxia Hui Autonomous Region, the general Pb concentrations and accumulations might not reflect the real situation of Pb in arable soils due to the limited sample numbers.

Three provinces, Yunnan, Guangxi, and Hunan, had Pb concentrations  $>100$  mg/kg, mainly due to large amounts of mining and smelting activities. Fujian, Guangdong, Guizhou, Shaanxi, and Sichuan had Pb concentrations ranging from 50 to 100 mg/kg. The high Pb concentrations in Fujian, Guangdong, and Guizhou are mainly due to high backgrounds of Pb concentrations and some mining and smelting activities in these provinces. Moreover, the coastal region of Fujian and Guangdong provinces are economically developed areas, with high population density and many factories. For the high Pb concentrations in Sichuan Province and Shaanxi Province, the combining effect of mining activities and wastewater irrigation might be the main reason.

The provinces of Gansu, Hubei, Jiangxi, and Zhejiang had Pb concentrations ranging from 35 to 50 mg/kg. Mining and smelting activities might be the main reason for the accumulation of Pb from exterior sources in Hubei and Jiangxi Provinces; irrigation by wastewater and mining activities might be the sources of Pb accumulation in Gansu Province, and the high level of economic development might have introduced Pb into soil in Zhejiang Province.

The other 19 provinces had Pb concentrations  $<35$  mg/kg in arable soils. Fertilizer applications might be the main source for the Pb accumulation in these soils. Among them, five provinces, Beijing, Shanghai, Heilongjiang, Jilin, and Qinghai, had lower Pb concentrations than their corresponding background values, indicating that arable soils in these provinces are safe enough for planting crops.

Considering the index of geoaccumulation for Pb concentrations, three provinces, Yunnan, Guangxi, and Hunan had values  $>1$ , indicating that arable soils in these three provinces had been moderately polluted by Pb. Shaanxi Province and Gansu Province had the accumulation index within the range of 0.50–1.00, indicating that large amounts of Pb had been introduced into arable soils since the background values of Pb concentrations were relatively low in the two provinces. The index in nine provinces, Fujian, Guangdong, Guizhou, Henan, Hubei, Shanxi, Sichuan, Tianjin, Zhejiang ranged from 0.00 to 0.50, denoting that the arable soils had been affected by Pb, but the pollution was not too serious or in the level of alarm. The remaining 16 provinces showed the index values  $<0$ , indicating that the soil in these provinces was not contaminated by Pb.

### 3.5 Limitations and uncertainties

Firstly, the total arable land in China is about 1 353 850 km<sup>2</sup>, while the investigated arable soils are about 789 900 km<sup>2</sup> in this study. Therefore, the investigated samples may not represent China's overall situation of Pb concentration in arable soils. Secondly, discrepancies in the sampling methods in the collected data from

literature may impact the consistency of the evaluation on Pb concentrations and the pollution assessments. Thirdly, the limited number of data records in some provinces, such as Qinghai Province and Ningxia Hui Autonomous Region, may not reflect the Pb accumulation status. Fourthly, many other factors, such as parent material, organic matter, and soil microbes can play important roles in influencing the concentration and distribution of Pb in soils. However, in this study, we did not consider the influence of these factors on Pb occurrence in soils due to the lack of such data.

## 4 Concluding remarks

This study presented Pb concentrations in arable soils in China based on reviewing relevant studies over a decade. The average Pb concentration was 34.41 mg/kg, higher than the background value in China. This indicated that Pb has been introduced into soil by human activities.

Mining and smelting activities, wastewater irrigation, and urban development had greatly contributed to the Pb accumulation in Chinese arable soils. Soils around mining and smelting activities, irrigation area, and urban area were more than nine, two, and one and a half time of that in remote areas, respectively.

Northern China had obvious lower Pb concentrations than the south, and the northwest and northeast had much lower Pb concentrations than the center. Moreover, many hotspots existed on the Pb map due to mining and smelting activities.

The Yunnan, Guangxi, and Hunan provinces had obviously high Pb concentrations and accumulations, indicating arable soils were moderate polluted by Pb; Shaanxi Province and Gansu Province had arable soils slightly polluted by Pb; nine provinces showed arable soils affected by Pb within alarm level; and the remaining 16 provinces had not been affected by Pb.

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## References

- [1] E. Emory, R. Pattillo, E. Archibold, M. Bayorh, F. Sung, Neuro-behavioral Effects of Low-Level Lead Exposure in Human Neonates, *Am. J. Obstetrics Gynecol.* **1999**, 181(1), S2–S11.
- [2] J. Liu, J. Wang, J. Qi, X. Li, Y. Chen, C. Wang, Y. Wu, Heavy Metal Contamination in Arable Soils and Vegetables Around a Sulfuric Acid Factory, China, *Clean–Soil Air Water* **2012**, 40(7), 766–772.
- [3] G. Zhu, C. Zhang, J. Wang, X. Wang, D. Chen, Investigation of Heavy Metal Pollution in Soil and Wheat Grains in Sewage-Irrigated Area in Sizhuangding, Xinxiang City, *J. Agro-Environ. Sci.* **2009**, 28(2), 263–268.
- [4] H. Chen, C. Zheng, C. Tu, Y. Zhu, Heavy Metal Pollution in Soils in China: Status and Countermeasures, *Ambio* **1999**, 28(2), 130–134.
- [5] B. G. Wei, L. S. Yang, A Review of Heavy Metal Contaminations in Urban Soils, Urban Road Dusts and Agricultural Soils From China, *Microchem. J.* **2010**, 94(2), 99–107.
- [6] X. Y. Zhang, F. F. Lin, M. T. Wong, X. L. Feng, K. Wang, Identification of Soil Heavy Metal Sources From Anthropogenic Activities and Pollution Assessment of Fuyang County, China, *Environ. Monit. Assess.* **2009**, 154(1–4), 439–449.



- [7] National Bureau of Statistics of China, *China Statistical Yearbook*, China Statistics Press, Beijing 2013.
- [8] Y. Wang, Y. C. Chen, Z. P. Li, Contamination Pattern of Heavy Metals in Chinese Urban Soils, *Environ. Chem.* 2012, 31(6), 8.
- [9] H. Cheng, M. Li, C. Zhao, K. Li, M. Peng, A. Qin, X. Cheng, Overview of Trace Metals in the Urban Soil of 31 Metropolises in China, *J. Geochem. Explor.* 2014, 139, 31–52.
- [10] Y. Fang, X. Sun, W. Yang, N. Ma, Z. Xin, J. Fu, X. Liu, et al., Concentrations and Health Risks of Lead, Cadmium, Arsenic, and Mercury in Rice and Edible Mushrooms in China, *Food Chem.* 2014, 147, 147–151.
- [11] P. Kosolsaksakul, J. G. Farmer, I. W. Oliver, M. C. Graham, Geochemical Associations and Availability of Cadmium (Cd) in a Paddy Field System, Northwestern Thailand, *Environ. Pollut.* 2014, 187, 153–161.
- [12] G. G. S. Holmgren, M. W. Meyer, R. L. Chaney, R. B. Daniels, Cadmium, Lead, Zinc, Copper, and Nickel in Agricultural Soils of the United States of America, *J. Environ. Qual.* 1993, 22(2), 335–348.
- [13] A. Ordóñez, R. Alvarez, S. Charlesworth, E. De Miguel, J. Loredó, Risk Assessment of Soils Contaminated by Mercury Mining, Northern Spain, *J. Environ. Monit.* 2011, 13(1), 128–136.
- [14] L. Niu, F. Yang, C. Xu, H. Yang, W. Liu, Status of Metal Accumulation in Farmland Soils Across China: From Distribution to Risk Assessment, *Environ. Pollut.* 2013, 176, 55–62.
- [15] W. Song, B. M. Chen, L. Liu, Soil Heavy Metal Pollution of Cultivated Land in China, *Res. Soil Water Conserv.* 2013, 20(2), 293–298.
- [16] Y. G. Teng, J. Wu, S. J. Lu, Y. Y. Wang, X. D. Jiao, L. T. Song, Soil and Soil Environmental Quality Monitoring in China: A Review, *Environ. Int.* 2014, 69, 177–199.
- [17] G. Pierre, Geostatistics in Soil Science: State-of-the-Art and Perspectives, *Geoderma* 1999, 89(1–2), 1–45.
- [18] Z. Y. Li, Z. W. Ma, T. J. van der Kuip, Z. W. Yuan, L. Huang, A Review of Soil Heavy Metal Pollution From Mines in China: Pollution and Health Risk Assessment, *Sci. Total Environ.* 2014, 468, 843–853.
- [19] China National Environmental Monitoring Center (CNEMC), *Chinese Soil Element Background Concentration*, Chinese Environment Science Press, Beijing 1990.
- [20] C. Zhang, Z. Y. Li, W. Yang, L. Pan, M. Gu, D. Lee, Assessment of Metals Pollution on Agricultural Soil Surrounding a Lead-Zinc Mining Area in the Karst Region of Guangxi, China, *Bull. Environ. Contam. Toxicol.* 2013, 90(6), 736–741.
- [21] V. Ettler, O. Sebek, T. Grygar, M. Klementova, P. Bezdicka, H. Slavikova, Controls on Metal Leaching From Secondary Pb Smelter Air-Pollution-Control Residues, *Environ. Sci. Technol.* 2008, 42(21), 7878–7884.
- [22] Y. G. Yang, C. Q. Liu, W. Pan, G. P. Zhang, W. H. Zhu, Heavy Metal Accumulation From Zinc Smelters in a Carbonate Rock Region in Hezhang County, Guizhou Province, China, *Water Air Soil Pollut.* 2006, 174(1–4), 321–339.
- [23] B. E. Davies, N. J. Houghton, Distance-Decline Patterns in Heavy Metal Contamination of Soil and Plants in Birmingham, England, *Urban Ecol.* 1984, 8, 285–294.
- [24] L. J. Wang, E. P. Zhu, The Study of Heavy Metals Distribution Character in Soil About Lead-Zinc Smelt Mill in Qinling, *J. Baoji Univ. Arts Sci. Nat. Sci.* 2008, 28(2), 150–152.
- [25] R. K. Yadav, B. Goyal, R. K. Sharma, S. K. Dubey, P. S. Minhas, Post-Irrigation Impact of Domestic Sewage Effluent on Composition of Soils, Crops and Ground Water—A Case Study, *Environ. Int.* 2002, 28(6), 481–486.
- [26] H. Yao, J. Lu, X. Yuan, J. Wu, J. Zhao, X. Yu, Y. Zhou, Concentrations, Bioavailability, and Spatial Distribution of Soil Heavy Metals in a Long-Term Wastewater Irrigation Area in North China, *Clean—Soil Air Water* 2014, 42(3), 331–338.
- [27] W. Liu, J. Zhao, Z. Ouyang, L. Söderlund, G. Liu, Impacts of Sewage Irrigation on Heavy Metal Distribution and Contamination in Beijing, China, *Environ. Int.* 2005, 31, 805–812.
- [28] J. Horswell, T. W. Speir, A. P. van Schaik, Bio-Indicators to Assess Impacts of Heavy Metals in Land-Applied Sewage Sludge, *Soil Biol. Biochem.* 2003, 35(11), 1501–1505.
- [29] A. Muchuweti, J. W. Birkett, E. Chinyanga, R. Zvauya, M. D. Scrimshaw, J. N. Lester, Heavy Metal Content of Vegetables Irrigated With Mixtures of Wastewater and Sewage Sludge in Zimbabwe: Implications for Human Health, *Agric. Ecosyst. Environ.* 2006, 112(1), 41–48.
- [30] Y. Y. Liu, H. F. Liu, Heavy Metal Pollution and Prohibition in the Irrigated Area by Wastewater in Miqian, Xinjiang, *J. Changji Coll.* 2007, 24(3), 45–48.
- [31] H. Y. Li, L. M. Mao, X. D. Ren, The Determination and Evaluation of Heavy Metals Content in Crop and Soil at Hongsiqu Region, *Ningxia Eng. Technol.* 2008, 7(1), 9–11.
- [32] W. Y. Xie, G. S. Fan, H. P. Zhou, J. F. Xie, C. L. Guan, Access of Heavy Metals Pollution of the Sewage Irrigation Region in Taiyuan, China, *J. Agro-Environ. Sci.* 2011, 30(8), 1553–1560.
- [33] X. Yi, X. J. Gu, Y. Q. Hou, X. H. Liu, J. Xie, Evaluation on Potential Ecological Risk of Heavy Metals in Soils of Jinghuiqu Irrigation District of Shaanxi, *Agric. Res. Arid Areas* 2010, 28(6), 217–221.
- [34] F. Y. Li, Z. P. Fan, P. F. Xiao, K. Oh, X. P. Ma, W. Hou, Contamination, Chemical Speciation and Vertical Distribution of Heavy Metals in Soils of an Old and Large Industrial Zone in Northeast China, *Environ. Geol.* 2009, 57(8), 1815–1823.
- [35] G. Zhu, X. Su, X. Wang, J. Fan, Investigation of the Pollution and Correlation of Heavy Metals in Sewage-Irrigated Area in Wangcun, Xinxing City, *J. Henan Normal Univ.* 2010, 38(6), 4.
- [36] B. F. Faria Pereira, Z. He, P. J. Stoffella, C. R. Montes, A. J. Melfi, V. C. Baligar, Nutrients and Nonessential Elements in Soil After 11 Years of Wastewater Irrigation, *J. Environ. Qual.* 2012, 41(3), 920–927.
- [37] W. Chen, S. Lu, C. Peng, W. Jiao, M. Wang, Accumulation of Cd in Agricultural Soil Under Long-Term Reclaimed Water Irrigation, *Environ. Pollut.* 2013, 178(0), 294–299.
- [38] P. Li, X. Wang, G. Allinson, X. Li, Z. Xiong, Risk Assessment of Heavy Metals in Soil Previously Irrigated With Industrial Wastewater in Shenyang, China, *J. Hazard. Mater.* 2009, 161(1), 516–521.
- [39] P. Bullock, P. J. Gregory, Soils: A neglected resource in urban areas, in *Soils in the Urban Environment* (Eds.: P. Bullock, P. J. Gregory), Blackwell Publishing, Oxford 2009, pp. 1–4.
- [40] H. Taghipour, M. Mosaferi, F. Armanfar, S. J. Gaemmagami, Heavy Metals Pollution in the Soils of Suburban Areas in Big Cities: A Case Study, *Int. J. Environ. Sci. Technol.* 2013, 10(2), 243–250.
- [41] A. Christoforidis, N. Stamatis, Heavy Metal Contamination in Street Dust and Roadside Soil Along the Major National Road in Kavala's Region, Greece, *Geoderma* 2009, 151(3–4), 257–263.
- [42] X. S. Luo, S. Yu, Y. G. Zhu, X. D. Li, Trace Metal Contamination in Urban Soils of China, *Sci. Total Environ.* 2012, 421, 17–30.
- [43] A. A. Abdelhafez, H. H. Abbas, R. S. Abd-El-Aal, N. F. Kandil, J. Li, W. Mahmoud, Environmental and Health Impacts of Successive Mineral Fertilization in Egypt, *Clean—Soil Air Water* 2012, 40(4), 356–363.
- [44] L. Wu, X. Pan, C. Chen, J. Huang, Y. Teng, Y. Luo, P. Christie, Occurrence and Distribution of Heavy Metals and Tetracyclines in Agricultural Soils After Typical Land Use Change in East China, *Environ. Sci. Pollut. Res.* 2013, 20, 8342–8354.
- [45] L. H. Chen, W. Z. Ni, X. L. Li, J. B. Sun, Investigation of Heavy Metal Concentrations in Commercial Fertilizers Commonly Used, *J. Zhejiang Sci. Tech. Univ.* 2009, 26(2), 223–227.
- [46] G. Nziguheba, E. Smolders, Inputs of Trace Elements in Agricultural Soils via Phosphate Fertilizers in European Countries, *Sci. Total Environ.* 2008, 390(1), 53–57.
- [47] C. Wu, J. Wu, Y. Luo, H. Zhang, Y. Teng, Statistical and Geostatistical Characterization of Heavy Metal Concentrations in a Contaminated Area Taking Into Account Soil Map Units, *Geoderma* 2008, 144(1–2), 171–179.
- [48] M. Peng, *PhD Thesis*, Dali College, Dali 2009.
- [49] H. Xie, X. H. Liu, T. B. Chen, X. Y. Liao, X. L. Yan, L. X. Wang, Concentration and Health Risk of Heavy Metals in Vegetables and

- Soils in Region Affected by an Ancient Tin Ore, *Environ. Sci.* **2008**, *29* (12), 3503–3507.
- [50] X. Y. Shan, S. Q. Xu, The Analysis and Assessment on the Pollution Condition of Heavy Metals in the Soil Around the Qinglong Dachang Antimony Mining area in Guizhou Province, *J. Guizhou Univ. Nat. Sci.* **2011**, *28*(1), 132–135.
- [51] Y. H. Dong, Q. H. Dai, Y. H. Deng, H. Q. Zhang, Distribution Characteristics and Pollution Evaluation of Heavy Metals in Different Lead-Zinc Waste Lands, *Guizhou Agric. Sci.* **2013**, *41*(5), 109–112.
- [52] M. Xiang, G. Zhang, L. Li, X. Wei, H. Li, The Characteristics of Heavy Metals in Soil Around the Hechi Antimony-Lead Smelter, Guangxi, China, *Earth Environ.* **2010**, *38*(4), 6.
- [53] R. Z. Yin, Y. P. Luo, J. C. Li, W. L. Luo, Y. N. Zhu, Evaluation of the Potential Ecological Risk of Heavy Metal Pollution in Soil and Bioaccumulation Characteristics of Dominant Plants in Siding Pb-Zn Mine, *J. Agro-Environ. Sci.* **2008**, *27*(6), 2158–2165.
- [54] Q. W. Yang, W. S. Shu, Z. Lin, L. Lin, H. L. Zou, C. J. Lan, Compound Pollution and Ecological Evaluation of Heavy Metals From Mining Waste Water to Soil-Rice Plant System, *J. Agro-Environ. Sci.* **2003**, *22*(4), 385–390.
- [55] H. N. Liu, Q. H. Yang, H. S. Yang, J. Q. Li, D. L. Liu, Characteristics of Absorption and Accumulation of Heavy Metals for Three Dominant Plants in Pb-Zn Mine Tailings Eastern Guangdong, *Guihaia* **2012**, *32* (6), 743–749.
- [56] J. W. Zhu, *PhD Thesis*, Hunan Agricultural University, Changsha **2012**.
- [57] H. Zhou, M. Zeng, X. Zhou, B. H. Liao, J. Liu, M. Lei, Q. Y. Zhong, et al., Assessment of Heavy Metal Contamination and Bioaccumulation in Soybean Plants From Mining and Smelting Areas of Southern Hunan Province, China, *Environ. Toxicol. Chem.* **2013**, *32*(12), 2719–2727.
- [58] Y. Wang, Q. L. Zhao, Y. Hu, X. Du, W. Ge, W. J. Liu, Survey and Contamination Assessment of Heavy Metals in Soil and Plants Around the Mine in Shangyu, Zhejiang Province, *Environ. Chem.* **2011**, *30*(7), 1354–1360.
- [59] C. A. Lu, J. F. Zhang, H. M. Jiang, J. C. Yang, J. T. Zhang, J. Z. Wang, H. X. Shan, Assessment of Soil Contamination With Cd, Pb and Zn and Source Identification in the Area Around the Huludao Zinc Plant, *J. Hazard. Mater.* **2010**, *182*(1–3), 743–748.
- [60] X. D. Li, I. Thornton, Chemical Partitioning of Trace and Major Elements in Soils Contaminated by Mining and Smelting Activities, *Appl. Geochem.* **2001**, *16*(15), 1693–1706.
- [61] J. O. Nriagu, J. M. Pacyna, Quantitative Assessment of Worldwide Contamination of Air, Water and Soils by Trace-Metals, *Nature* **1988**, *333*(6169), 134–139.
- [62] M. Arora, B. Kiran, S. Rani, A. Rani, B. Kaur, N. Mittal, Heavy Metal Accumulation in Vegetables Irrigated With Water From Different Sources, *Food Chem.* **2008**, *111*(4), 811–815.
- [63] L. Niu, F. Yang, C. Xu, H. Yang, W. Liu, Status of Metal Accumulation in Farmland Soils Across China: From Distribution to Risk Assessment, *Environ. Pollut.* **2013**, *176*, 55–62.
- [64] D. B. Smith, W. F. Cannon, L. G. Woodruff, F. Solano, J. E. Kilburn, D. L. Fey, *Geochemical and Mineralogical data for soils of the conterminous United States*, U. S. Geological Survey Data Series 801, US Geological Survey, Reston, VA **2013**, p. 19.
- [65] B. A. Zarcinas, C. F. Ishak, M. J. McLaughlin, G. Cozens, Heavy Metals in Soils and Crops in Southeast Asia, 1. Peninsular Malaysia, *Environ. Geochem. Health* **2004**, *26*(4), 343–357.
- [66] B. A. Zarcinas, P. Pongsakul, M. J. McLaughlin, G. Cozens, Heavy Metals in Soils and Crops in Southeast Asia, 2. Thailand, *Environ. Geochem. Health* **2004**, *26*(4), 359–371.
- [67] C. Reimann, B. Flem, K. Fabian, M. Birke, A. Ladenberger, P. Négrel, A. Demetriades, et al., Lead and Lead Isotopes in Agricultural Soils of Europe—The Continental Perspective, *Appl. Geochem.* **2012**, *27*(3), 532–542.