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Spatial Distribution of Mercury (Hg) Concentration in Agricultural Soil and Its Risk Assessment on Food Safety in China

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Abstract: Soil mercury (Hg) pollution in some areas of China is a serious problem and has aroused a lot of attention on a local scale. However, there are few studies on Hg pollution on a national scale. This study collected 444 published papers during 2005–2015 on Hg concentrations in agricultural soil throughout China, under seven land uses, namely: dry land, paddy field, vegetable field, tea garden, orchard, traditional Chinese medicine field and tobacco field, to assess the spatial distribution of Hg concentration and evaluate its influence on food safety. The averaged Hg concentration (0.108 mg/kg) was higher than its background (0.065 mg/kg), but much lower than the guidelines (GB15618-1995 II) for crop production. The spatial distribution of Hg throughout China showed great variability, with some hotspots due to Hg related mining and smelting activities. According to the Environment Quality Standard for soil in China (GB15618-1995 II), 4.2% of agricultural soil should be abandoned due to Hg pollution, and 2.0% faced a high risk of Hg pollution.

Keywords: Hg concentrations; food safety; agricultural soil; land uses; China

1. Introduction

Environmental pollution and food safety are two of the most important issues [1]. Among the pollutants reported, soil heavy metals are considered as one of the greatest risks to food safety in China (MEP & MLR 2014). Mercury (Hg) is extremely toxic to human and animal's health through various absorption pathways such as ingestion, dermal contact, and diet through soil-plant system [2–4]. Recent studies in Guizhou and Zhejiang provinces of China showed that the consumption of grains was the primary Hg exposure pathway for local residents in inland Hg polluted areas [5–8]. Soil is one of the important sources of Hg in crops and vegetables since their roots can take up Hg from soil, and transfer it to seeds and edible parts [9]. Thus, public concerns over food safety have grown due to the potential accumulation of Hg in agricultural soil [10].

Mercury contamination is a serious problem in some areas of China [11], where there has been rapid progress in economic and industrial developments related to Hg emissions and accumulations [7,12,13]. Natural Hg concentration in soils depends primary on the geochemistry of the parent material [14]. This can explain the spatial variability of Hg over heterogeneous lithologies.

Soil pH, organic matter content, atmospheric deposition, sewage irrigation, fertilizer and pesticides applications and other human activities can also influence Hg concentration in soil [15–17]. Particularly, China has abundant Hg reserves, which mining and smelting are considered as hotspots of Hg pollution [18]. Some industry activities, such as electric producing, coal combustion, agriculture practices, can emit large amounts of Hg into environment [19–21]. Agricultural soils around Hg-related factories and mines might have been contaminated by Hg.

To prevent further soil Hg pollution and to carry out remediation, it is essential to understand the level of Hg concentrations and pollutions, and their influence on food safety on a national scale. Understanding the Hg concentration in agricultural soil is of critical importance to assess human impact on soil Hg and have great significance in terms of agricultural production. The Chinese government investigated 6300,000 km^2 soils from 2005 to 2013 across China, and reported that 1.6% of the investigated samples exceeded Hg reference, but it did not give the Hg contaminated locations and areas (MEP & MLR 2014). Song et al. reviewed 121 regions and concluded that the averaged Hg in Chinese farmland was 0.160 mg/kg with a pollution rate of 3.3% [22]. However, the spatial distribution of Hg concentration in agricultural soil on a national scale is still unknown. It is therefore difficult to assess the threat to safe crop production posed by soil Hg contamination on a national scale.

This study aims to obtain the spatial distribution of Hg concentrations in agricultural soil, and evaluate the risk of soil Hg contamination on food safety across China, based on the meta-data analysis method. Firstly, the Hg concentrations in Chinese agricultural soil were collected from the published papers during 2005–2015; secondly, the soil-sample weighted averages of Hg concentration in soil under seven land uses are calculated; thirdly, the spatial distribution of Hg concentration is obtained based on a kriging method; and finally, the risk of Hg on food production is assessed based on the Environment Quality Standard for soil in China (GB15618-1995 II).

2. Data Collection and Methods

2.1. Data Collection

The data on Hg concentration in topsoil (0–20 or 0–15 cm) were collected under seven land uses: dry land, paddy field, vegetable field, tea garden, orchard, traditional Chinese medicine field and tobacco field, from the studies published during 2005–2015 in China. These studies were selected through the ISI (Institutes for Scientific Information) Web of Knowledge and CNKI (China National Knowledge Infrastructure) web using key words "soil heavy metal", "mercury/Hg" or "land use" and "agricultural soil". The detail information about the selection process of the Hg-related studies and the description of Hg concentrations in agricultural soil could be found in the study by Zhang et al. [23]. In total, 444 peer reviewed articles consisting of 821 data records on Hg concentration were collected. Figure 1 illustrates the locations of the collected data records.

Soil pH and land use have strong impacts on Hg bioavailability (or toxicity) to the crops and vegetables [24], thus they are used to assess the influence of Hg on food production. The soil pH map in China came from "Atlas of Soil Environmental Background Value in the People's Republic of China" [25], which were classified into 6 grades with the soil pH < 5, 5–6, 6–6.5, 6.5–7.5, 7.5–8.5, >8.5. The land use map of dry land, paddy field and woodland was freely obtained from the Data Sharing Network of Earth System Science [26].

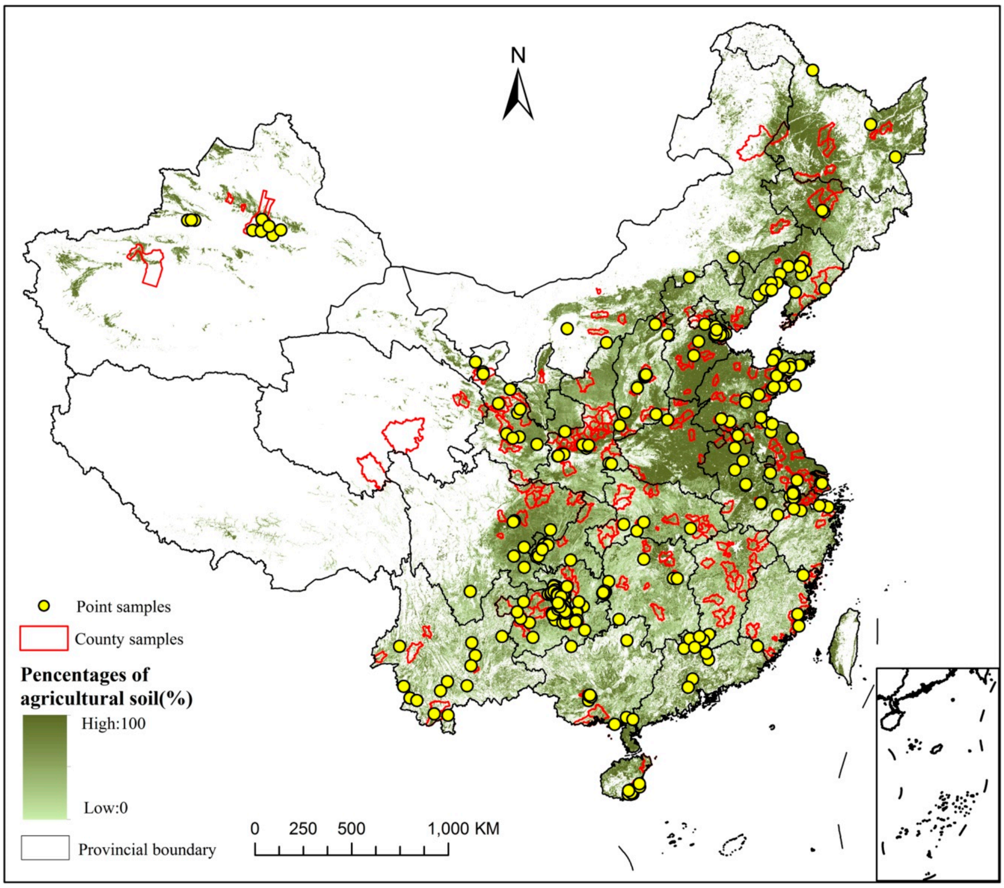


Figure 1. Spatial distribution of the collected soil Hg samples and agricultural land in the mainland of China.

2.2. Statistical Analyses

The sample-number-weighted mean is used to represent the average of Hg concentration on a national scale, since the arithmetic mean might not reflect the general situation of Hg concentrations due to the varied sample numbers and sampling methods in each study.

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

where C_A is the sample-number-weighted mean, C_i is the Hg concentration in the data record *i*, *n* is the number of the data records, N_i is the sampling number in the *i*th data record.

In addition to the role of parent material, the concentration of Hg in soil under different land uses may be distinct from one another due to human activities. The Formula (1) is used to calculate the sample-number-weighted mean of Hg concentrations under the seven kinds of land uses.

2.3. Spatial Distribution of Soil Hg Concentrations and Risk Assessment on Food Safety

The Ordinary Kriging method is used to estimate and map soil Hg concentrations on a regional scale [27], based on the collected point and county samples. The data from the counties and the points were used to obtain the spatial variation of Hg concentrations in agricultural soil. Surface data in counties were firstly converted into points at spatial resolution of 10 km × 10 km, and then these data would be merged to point data. Several models, including Linear, Spherical, Exponential and Gaussian are used to simulate the semi-variogram of Hg in soil, and then the model with the highest R^2 will be selected to map the Hg concentration in China.

The soil environmental quality standard (China Environmental Protection Bureau, 1995) was developed through numerous experiments and soil surveys. This Standard specifies the index value for the maximum allowable concentration of Hg in soil. According to the standard, the II reference values (0.300 mg/kg under soil pH < 6.5, 0.500 mg/kg under soil 6.5 < pH < 7.5, 1.000 mg/kg under soil pH > 7.5) guarantee agricultural production and protect human health, and the III references (1.500 mg/kg) guarantee agricultural production and plant growth in the areas that have high Hg concentration in parent rocks.

3. Results and Discussion

3.1. Hg Concentration in Chinese Agricultural Soil

The number of total investigated samples was 139,334. Hg concentrations in agricultural soil ranged from 0.003 to 150.000 mg/kg, with a standard deviation (SD) of 6.031 mg/kg. This wide range was close to the reported results that Hg concentrations vary widely from 0.010 mg/kg to 1.000 mg/kg in rural and remote areas, and from 0.100 mg/kg to >10.000 mg/kg in urban, industrial, and mineralized/mined lands [28,29].

The wide range of Hg concentrations and the high SD value denoted that Hg concentrations in separate studies were spread out over a large range of values in Chinese agricultural soil. The sample-number-weighted mean of Hg concentration was 0.108 mg/kg, higher than its background of 0.065 mg/kg [7], indicating that Hg had been introduced into agricultural soil from human activities in some areas. Also, this value was much lower than Hg concentration (0.160 mg/kg) in Chinese farmland obtained by Song et al. [22]. Compared to other countries or regions, Chinese agricultural soil had close Hg content with United Kingdom [30], higher value than Europe [31], Thailand [32], and United States of America [33], but lower than Malaysian [34] and Belgium [35] (Table 1). The different soil Hg occurrence in different regions might be due to the components of physical parents and the human activities related to Hg emissions.

Table 1. Comparison of Hg concentration in agricultural soil of China with previously published surface soil Hg concentrations in China and other countries or regions.

Country	Land Uses	Number of Samples	Mean (mg/kg)	Minimum (mg/kg)	Maximum (mg/kg)	Reference
China	Agricultural soil	139,334	0.108	0.003	150.000	This study
China	Farmland soil	121 regions	0.160	0.030	1.350	[22]
United Kingdom	Rural soil	898	0.095			[30]
Belgium	Agricultural soil	316	0.240	0.030	4.190	[35]
United states of America	All of land uses	4841	0.050	<0.010	56.400	[33]
Malaysian	Crop soil	241	0.147	0.002	0.860	[34]
Thailand	Crop soil	318	0.040	0.010	0.270	[32]
Europe	Agricultural soil	2108	0.030	<0.003	1.600	[31]

According to the frequency distribution of Hg concentrations in Figure 2, 46.0% of data records had lower Hg concentrations than Hg background in China. The remaining were higher than the background, indicating Hg had been introduced from exterior sources in some regions. About 96.5% or 99.2% of Hg concentrations were lower than grade II value of 0.300 mg/kg (pH < 6.5) or 1.000 mg/kg (pH > 7.5), and these agricultural soils were safe for planting crops or vegetables. The percentage of Hg concentrations higher than 1.000 mg/kg was 0.8%, which was lower than the finding of 3.3% in the study by Song et al. [22]. This indicated that although the general Hg concentration in Chinese soil is at a safe level, there are still some local areas facing serious Hg pollution. Thus it is essential to understand the size of the Hg affected area, the level of soil Hg concentration and its spatial distribution [13,36].

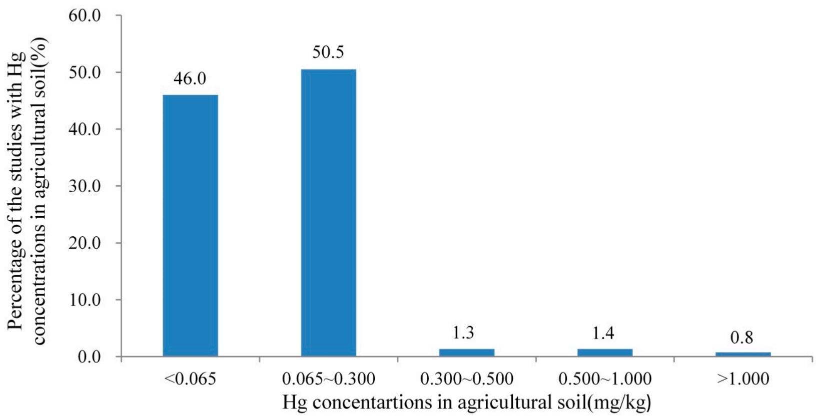


Figure 2. Frequency distribution of Hg concentrations in agricultural soil in China.

The sample-number-weighted mean of Hg concentrations under the seven land uses are illustrated in Table 2. The Hg concentrations in orchard, dry land, and tobacco field were at low levels, but all of the averaged Hg concentrations under the three land uses were higher than the background of 0.065 mg/kg [7]. The basic sources of Hg in soil are parent material, atmospheric deposition, industrial waste, sewage irrigation, fertilizer and pesticides applications, and other human activities can also influence Hg concentration in soil [15,16,37]. Generally, the orchard field had no tillage and received less fertilizer in comparison with arable land, leading to the lower Hg concentration in orchard than other land uses [38–40]. Dry land and tobacco field have allied means of land use, therefore they have close pollution level and source. The external pollution source of Hg in dry land and tobacco field are mainly fertilizer application and atmospheric deposition [15,41].

Table 2. The sample-number-weighted Hg concentration in the soil under different land uses (unit: mg/kg).

	Dry Land	Paddy Field	Vegetable Field	Tea Garden	Orchard	Medicine Field	Tobacco Field
Sample number	80735	45426	8171	1578	2137	625	662
Minimum	0.007	0.012	0.010	0.030	0.010	0.003	0.011
Maximum	66.490	150.000	1.089	0.180	0.617	0.850	0.140
Weighted means	0.097	0.116	0.172	0.161	0.074	0.145	0.092
Standard deviation	4.889	11.575	0.213	0.124	0.118	0.181	0.045

The pollution of Hg in paddy field was at moderate level. Considering the tillage methods of paddy field, sewage irrigation and the application of agricultural chemicals may result in the accumulation of Hg in paddy soil [42]. In addition, the paddy field was mainly located in south China, which has relatively high background value of soil Hg. This could make paddy fields have comparatively high Hg concentration.

Compared with those in the other land uses, the Hg concentrations in the vegetable field, tea garden and traditional Chinese medicine field were at a high level, particularly in the vegetable field. This might be due to the high application of agrochemicals and fertilizers to guarantee the high production [43]. Usually, different fertilizers have diverse kinds and amounts of heavy metals due to distinct origin ore and the process used for fertilizer production [44]. Long-term application of excessive fertilizers and organic manures for the vegetable field can lead to the accumulation of soil heavy metals and the content of heavy metals in soil will increase with increasing vegetable production history [45,46]. These factors may lead to Hg concentration in vegetable field being obviously higher than in other land uses. Most of the tea gardens were located in South China, where the concentration of heavy metals is higher than North China due to the high concentration of heavy metals in parent materials [47]. The application of large amount of organic fertilizer and sewage irrigation can also increase the content of Hg in the tea garden [48]. The high Hg concentration in traditional Chinese medicine field may due to the high content of heavy metals in the plantations where there may be a lot of organic matter which contains high Hg content and an accumulation of Hg due to the long-term utilization of pesticides and fertilizers [17,49,50].

3.2. Spatial Distribution of Hg in Agricultural Soil

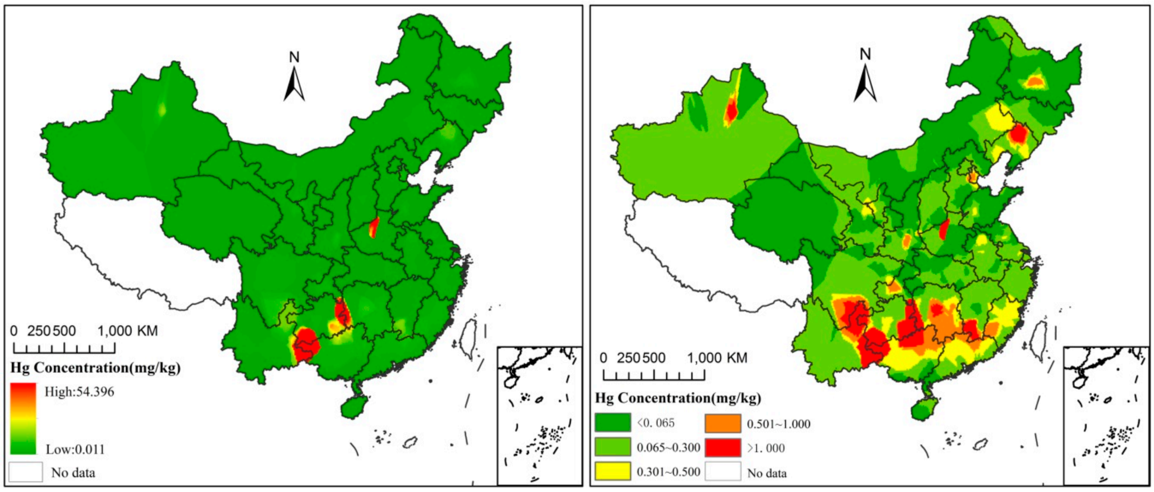
After merging the data of counties and points, 5764 Hg concentrations were used to map the spatial distribution of Hg concentrations over China. These data were logarithmically transformed to conform to normal distribution, and then four semi-variogram models of Linear, Spherical, Exponential and Gaussian were constructed to explore the degree of spatial continuity and the range of spatial dependence. The results in Table 3 showed that the theoretical Spherical model was in reasonable agreement with the data for soil Hg concentrations, since it achieved the maximum R^2 value (0.907).

Table 3. Semi-variogram model parameters of soil Hg.

Semi-Variogram Model	Nugget (C_0)	Sill (C_0 + C)	Range (A)	SS	R^2	C/(C_0 + C)
Linear	0.851	1.482	24.930	0.328	0.583	0.426
Spherical	0.382	1.309	9.920	0.073	0.907	0.708
Exponential	0.213	1.328	10.740	0.086	0.891	0.840
Gaussian	0.510	1.309	8.350	0.079	0.900	0.610

The relatively wide range of Hg suggested that large scale factors including soil parent material may have great influence on the spatial distribution of soil Hg [51]. The ratio of partial sill to sill (C/(C_0 + C)) of Spherical was 0.708 (Table 3), which was between 0.25 and 0.75, denoting that Hg in agricultural soil had moderate spatial dependence [52]. To some extent, this indicator reflected that intrinsic factors such as parent materials and topography are the predominant factors impacting the spatial variability of Hg in Chinese agricultural soil, but the anthropogenic factors changed its spatial correlation through industrial production, mining and smelting activities, fertilization and other soil management practices [13,53].

The spatial distribution of Hg concentrations in agricultural soil in China is shown in Figure 3. On the Hg map, several hotspots existed in Hunan, Guangxi, Guizhou, Yunnan, Sichuan province due to Hg, Mo, Sb, Pb/Zn mining and smelting activities [4,54–59], in Liaoning, Heilongjiang and Jiangxi due to industrial pollution and mining activities [60–65], in Henan due to coal mining [66], and in Xinjiang due to sewage irrigation [67]. Mercury mining areas are considered hot spots of Hg pollution, since the cinnabar ore roasting generally generates huge quantities of mine waste [5]. Soil Hg concentrations in Hg mining areas in Guizhou province could reach to 150.00 mg/kg [68], about 500 times the grade II reference in agricultural soil. The Sb, Pb/Zn, Mo and coal mining and related activities discharge also could lead to large amounts of Hg with the waste water, waste gas and solid waste being added to the environment.



(**a**) (**b**)

Figure 3. Spatial distribution of estimated Hg concentrations in agricultural in the mainland of China illustrated in (**a**) stretched map and (**b**) graded map.

Figure 3b shows that Hg concentrations in agricultural soil were generally higher in southern China than those in the north, which was consistent with the result of Zheng et al. (1994) [25]. The whole southern China had high concentration of Hg in soil. In particular, the pollution of Hg was more serious in the southwest China where there are lots of Hg mining smelting activities [69]. The regions with high risk to food safety (soil Hg concentrations higher than 1.000 mg/kg) were mainly located in the above mentioned hotspots. Other areas such as the provinces of Shaanxi due to Au mining [70], Anhui Province due to coal ores [71], Tianjin due to irrigation by sewages from industry and urban development also have high Hg concentration in soil (soil Hg concentrations in the range of 0.501~1.000 mg/kg) [72].

The regions with soil Hg concentrations lower than 0.065 mg/kg indicated these areas were seldom influenced by exterior factors. The remaining areas had Hg concentrations higher than 0.065 mg/kg, this showed that the soil in most of the areas had introduced Hg from anthropogenic activities, such as the agricultural practices of applying liquid and soil manure or inorganic fertilizers [73].

3.3. Risk Assessment of Soil Hg Concentrations on Food Safety

To investigate the influence of soil Hg on food production in China, agricultural soils were graded into four grades according to the soil environmental quality standard (China Environmental Protection Bureau, 1995) (Figure 4). The spatial distribution of dry land, paddy fields and agricultural woodland were used to evaluate the levels of Hg concentrations. The statistical information is listed in Table 4. It shows that 62.5% of agricultural soils were in Grade I, indicating these areas were not greatly influenced by Hg from exterior sources. Another 31.3% was in Grade II, indicating that, in total, 93.8% of agricultural soils were in the safe level for food production (within Grade I and Grade II). The agricultural soils within Grade III accounted for 2.0%, where they could be used for agricultural production but with high risks of Hg pollution. About 4.2% of agricultural soils were beyond the Grade III range, indicating these areas should not be used as farmlands or other agricultural land.

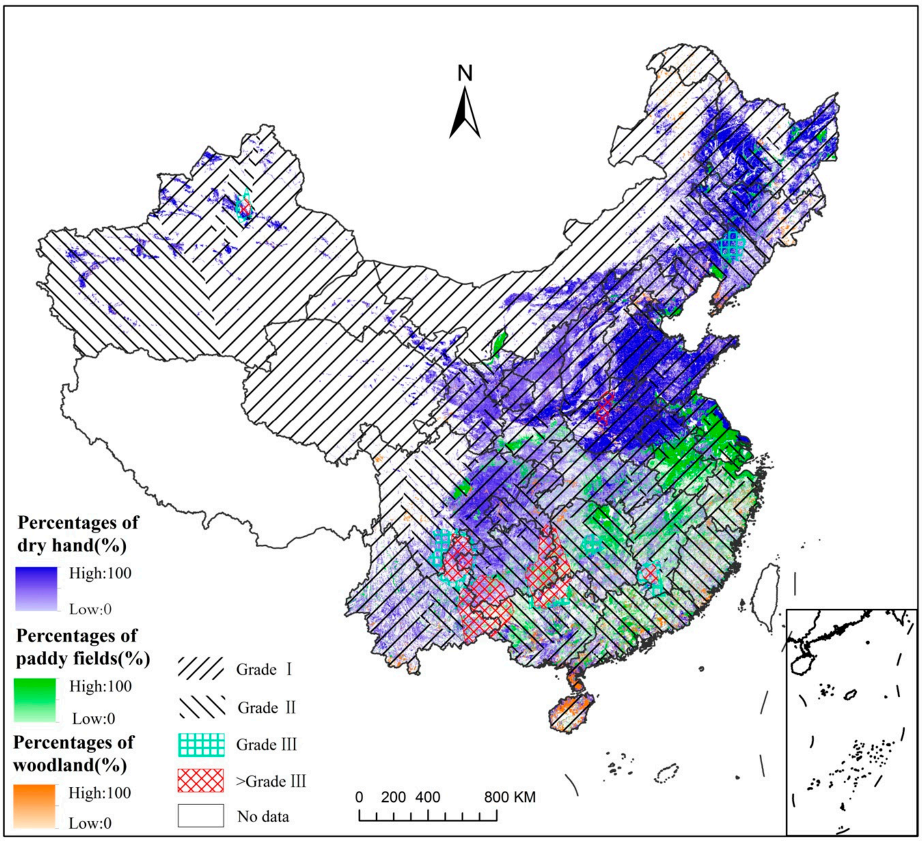


Figure 4. Assessment results of Hg pollution in agricultural soil in China. Note: The dry land in this figure includes dry land, vegetable field and tobacco field, woodland includes tea garden, orchard and traditional Chinese medicine field.

Table 4. Percentages of agricultural soils in Grades of Hg pollution.

Grades	Dry Land (%)	Paddy Field (%)	Woodland (%)	Dry Land, Paddy Field and Woodland (%)
Grade I	66.9	52.2	57.9	62.5
Grade II	27.2	40.4	39.1	31.3
Grade III	2.0	2.2	1.0	2.0
>Grade III	3.9	5.2	2.0	4.2

Note: The dry land in this table includes dry land, vegetable field and tobacco field, woodland includes tea garden, orchard and traditional Chinese medicine field.

Compared with paddy fields, dry land had lower Hg risk: about 66.9% of dry land was within the Grade I, while only 52.2% of paddy field was within this grade (Table 4), which indicates that paddy fields faced much higher risk of Hg pollutions than that of dry lands. This might be because the paddy fields were mainly distributed in South China, where there is a relatively higher Hg concentration than the north due to the large amount of Hg mining activities in the south [4,54–59,62]. Normally, woodland had no tillage and received less fertilizer, therefore it had low Hg risk [38–40], with 57.9% of woodland within Grade I. The other reasons for the difference might be that the fertilizer applications were not consistent under different land use conditions. The soils in Grade I and II for dry land accounted for 94.1%, while this value for paddy field and woodland were 92.6% and 97.0%. Moreover, the percentages of woodland beyond of Grade III was the lowest while that of paddy field was the highest, which indicated that 3.9% of crop, 5.2% of rice production and 2.0% of tea, fruit and other garden production will be decreased. Moreover, 2.0% of crops, 2.2% rice and 1.0% tea, fruit and other garden production were at high risk of Hg pollution.

4. Limitations and Uncertainties

Since this study was based on the soil Hg concentrations collected from the published papers, some limitations and uncertainties should be clarified. First, discrepancies in the sampling methods and the limited agricultural soils in the collected data may impact the consistency of the evaluation on Hg concentrations and the pollution assessments. The ununiformed distribution of the collected samples, such as the relatively small number of samples in some areas may affect the accuracy of Hg distribution.

Second, the interpolation method, Kriging, although having many advantages as mentioned in the method section, also has several disadvantages—including smoothing local acute hotspots of contamination, and expanding the high value in a large area [74]. This might introduce high estimation of Hg concentrations around mining and smelting areas, and directly introduce higher risk estimation than the actual situation.

Third, the soil pH maps were obtained in 1994 while the land use maps were obtained in 2000. At present, the soil pH and the spatial distribution of paddy field, dry land and woodland in some areas might change. However, we lack a current map of soil pH and land use, and the large areas of soil pH and agricultural land are hard to obtain. This might influence the assessment risk results.

5. Conclusions

Based on the Hg concentrations in agricultural soils throughout China, the averaged Hg concentration in Chinese agricultural soil was 0.108 mg/kg, higher than the background value in China. The spatial distribution of Hg concentrations showed high variations and there were some hotspots due to human activities. Overall, the Hg pollutions in the South China are more serious than in the north. Among the seven land uses, the vegetable field had the highest Hg concentration while the orchard had the lowest. The tea garden and traditional Chinese medicine field had relatively higher concentrations than those of dry land, paddy field, and tobacco field. In total, 4.2% of agricultural soil should be abandoned due to serious Hg pollution.

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