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Leaching of Heavy Metals from Farmland Soil in China: The Status and Ecological Risk Assessment

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Abstract

In this paper, using the leaching models, we quantified the leaching content of Cd, Pb, Cu, and Zn, and estimated the ecological risk changes in farmland soil caused by leaching and the ecological risk in leachate in China. Jiangxi, Guangxi, Guizhou, Hainan, Hunan, Zhejiang, Guangdong, and Chongqing are hotspot areas. The leaching of Cd in these regions exceed reported mean values in Europe (2.56 g ha^{-1} $year^{-1}$). Although the total ecological risk of heavy metals in the soil of various provinces (ranged from 20 to 130) was generally low, Cd was the most important contributor to ecological risks, while 9 provinces exhibited considerable ecological risk from Cd. The calculated Cd, Pb, and Zn in leachate exceed drinking water standards (GB 5749-2022) in five provinces. Overall, the leaching of heavy metals in Chinese agricultural soils, particularly in the southern regions, is a critical issue that warrants attention. Soil pH is the most prominent factor influencing heavy metal leaching. A 5% increase in pH reduces leaching by 31.2% for Cd, 25.42% for Pb, 22.07% for Cu, and 38.37% for Zn. Adjusting the pH to 6 can effectively solve the problem of excessive heavy metal content in leachate in most areas. The study recommends prioritizing groundwater monitoring in critical provinces such as Jiangxi and adjusting the soil pH of farmland in key regions.

Keywords: farmland soils; heavy metals; leaching; ecological risk

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1. Introduction

The pollution of agricultural soils, a vital resource, emerged as a significant global challenge [1]. Among various pollutants, heavy metal pollution presents a particularly serious threat and attracts widespread attention [2,3]. Excessive amounts of heavy metals not only reduce crop yield and quality through accumulation, but also threaten the health of animals and humans and endanger ecosystem safety [4,5]. According to the Report of the National General Survey of Soil Contamination (Ministry of Environmental Protection, PRC, 2014) [6], the exceedance rate of soils pollution in China reached 16.1%, and for agricultural soils, the percentage of exceedance is even greater at 19.4% (equivalent to approximately 26 million ha assuming that that the area is proportional to the number of survey samples).

Among the polluted sites, 82.8% were contaminated by eight heavy metals/metalloids, including cadmium (Cd), mercury (Hg), arsenic (As), lead (Pb), chromium (Cr), copper (Cu), nickel (Ni), and zinc (Zn). The geographical distribution pattern shows that soil pollution in southern China is more serious than in northern China. Notably, the southwest and south-central regions exhibit the most widespread exceedances of heavy metal standards. This may be attributed to the high industrial density [7], frequent mining activities [8], and intensive agricultural practices in southern regions [9], leading to severe heavy metal contamination in soils. In particular, the southwestern and south-central regions, due to their abundant mineral resources and long-term intensive agricultural activities [10], exhibit especially prominent heavy metal pollution. Farmland soils are not only major sinks, but also sources of heavy metals [11], as heavy metals in these polluted farmland soils could migrate to surface water and groundwater. Thus, the leaching of heavy metals in farmland soils deserves further attention. We selected Cd, Pb, Cu, and Zn for this study due to their widespread distribution in farmland soil and groundwater, high toxicity, and significant ecological and human health risks [12,13].

Studies confirmed that leaching to groundwater constitutes a significant pathway for Cd output in China, Europe, and Japan [14]. Notably, Xia et al. reported that nearly 100% of Cd, Pb, Cu, and Zn output in southern Heilongjiang resulted from leaching [15]. Up to now, the research on heavy metal leaching from farmland in China has been conducted at the regional scale, such as in the Yangtze River Delta and Heilongjiang [15,16], rather than the national scale. China's immense territory exhibits pronounced heterogeneity in soil type, climatic regimes, levels of economic development, industrial activities, and lifestyles. These regional disparities result in substantial variability in heavy metal leaching dynamics [17]. A comprehensive, nationwide study is therefore essential to characterize leaching across diverse regions, identify hotspot regions, key factors, and inform the prioritization of monitoring and management interventions. However, the comparison of leaching among different regions of China is difficult due to the different methods used for leaching amount; for example, field surveys [11,16], runoff experiments [18,19], and literature retrieval and contrast analysis [20]. Although several models for heavy metal leaching in agricultural soils have been developed (such as HYDRUS-2 D), existing studies predominantly focus on the migration mechanisms of heavy metals, with relevant numerical simulations concentrating on exogenous heavy metal movement in soil [21]. Furthermore, these models are frequently constrained by limited soil type diversity and small sample sizes, hindering their applicability across broader regional scales. Crucially, the model combining the heavy metal soil distribution coefficient (Kd) with the soil water balance model has been applied to large-scale regional studies in America and Europe [22,23].

Leaching has been demonstrated to drive heavy metal migration to deep soil layers and further contaminate surface water and groundwater [23,24]. Many methods have been used to assess the contamination rate and ecological risk of soils, including geo-accumulation index (Igeo), contamination factor (CF), contamination degree (CD), pollution load index (PLI), and ecological risk index (ERI) [25]. Among them, ERI, has been widely employed to assess the potential risks posed by heavy metals in soil, water, and sediment [26–28]. Although China established the preliminary environmental monitoring network for soil and groundwater (22,427 soil sites and 1912 groundwater wells) [29,30], a significant gap remains: there is currently no coordinated monitoring mechanism specifically for agricultural land to track both soil and groundwater. Consequently, the impacts of soil leaching from farmland on subsoil and groundwater lack sufficient data, and the potential ecological risks remain largely unknown.

To fill this knowledge gap, we used the leaching models based on the soil water balance to quantify heavy metal leaching from agriculture in China. The objectives of this study are as follows: (1) to evaluate the leaching of heavy metals, including Cd, Pb, Cu, and Zn, in farmland soils at a national scale; (2) to estimate the ecological risk changes of heavy metals in farmland soil caused by leaching and in leachate; and (3) to identify key factors controlling heavy metal leaching. This work can enhance scientific understanding of heavy metal leaching from farmland soil and provide scientific and comprehensive knowledge for soil management and pollution control.

2. Materials and Methods

2.1. Leaching Models

The leaching was estimated according to the method used by Six and Smolders [22], i.e., multiplying the amount of water leaving the topsoil (W_{lea}, mm year^{-1}) by the Cd concentration in the soil solution ($[Me]_w$, mg L^{-1}):

$$Q_{lea} = 10w_{lea}[Me]_w \quad (1)$$

Based on empirical coefficients (1/6 and 1/12) obtained through actual measurements, the formula was revised. For further details, see Text S1. The updated formula is provided below:

$$Q_{lea} = 10w_{lea}[Me]_w/12 \quad (2)$$

$$Q_{lea} = 10w_{lea}[Me]_w/6 \quad (3)$$

2.2. Estimation of the Infiltration Water

The soil water balance model includes precipitation, irrigation, evapotranspiration, surface runoff, soil storage of water, and infiltration water [31]. Baes and Sharp [23] assumed that post-1930 farming techniques made surface runoff and soil storage of water negligible, and they ultimately adopted a simplified formula to calculate infiltration water from farmland soils in the United States: rainfall plus irrigation minus evaporation. The formula used by European researchers is more simplified, the infiltration water was calculated by subtracting the annual evaporation from the annual precipitation [22,32]. Considering the irrigation practices in farmland planting activities in China, combining irrigation and irrigation water use efficiency, we devised the following equation for estimating the infiltration water:

$$W_{lea} = P + I \times (1 - \eta) - E \quad (4)$$

where P (mm year^{-1}) is the total annual precipitation, I (mm year^{-1}) is the total annual irrigation, η is the irrigative water use efficiency, which is the actual effective utilization of water to the total inflow of the channel head that does not include deep seepage and field loss, and E is the total annual evapotranspiration (mm year^{-1}). In this study, average annual precipitation was provided by the dataset published by Xie and Jiang [33]. The data of I and η for each city and district were collected from the Water Resources Bulletin issued by each province from 2019 to 2021. For provinces without a water resource bulletin, data were obtained from the national Water Resource Bulletin from 2019 to 2021 (Ministry of Water Resources, PRC, 2019–2021) [34–36]. Average annual precipitation adopted data released by National Tibetan Plateau Data Center (http://data.tpdc.ac.cn) [37]. The data processing and analysis were performed using ArcGIS 9.3 (ESRI Inc., Redlands, CA, USA).

2.3. Soil Heavy Metal Content and Other Parameters

K_D value is widely used to represent the distribution of heavy metals between the solid phase $[Me]_s$ (mg kg^{-1}) and solution phase or pore water (water held in pore space between soil particles), $[Me]_w$ (mg L^{-1}) [38].

$$K_D = \frac{[Me]_s}{[Me]_w} \tag{5}$$

The model adopted by Six and Smolders [22] was used to predict the K_D value of Cd (Equation (6)). The prediction formulas collected by Degryse et al. [39] were used to predict the K_D value of Pb (Equation (7)), Cu (Equation (8)), and Zn (Equation (9)).

$$K_D = \frac{[Me]_s}{[Me]_w} \tag{6}$$

$$\log(K_{D2}) = 1.32 + 0.40pH_{CaCl_2} + 0.50\log(OC) \tag{7}$$

$$\log(K_{D3}) = 0.45 + 0.34pH_{CaCl_2} + 0.65\log(OC) \tag{8}$$

$$\log(K_{D4}) = -1.77 + 0.66pH_{CaCl_2} + 0.79\log(OC) \tag{9}$$

where $pHcacl_2$ is the soil pH measured in 0.01mol L^{-1} $CaCl_2$ solution, the OC is the soil organic carbon content (% mass). The heavy metal content in farmland soil in China was based on the research results of Chen et al. [40] (see Table S1 for details). The soil pH measured in water (pH_{water}) and soil organic matter (SOM) of each province were obtained from published literature (see Table S2 for details). The commonly applied fixed OC to SOM transfer factor (CF_{SOC} = 1.724) was used to convert SOM to OC [41]. The soil pH measured in water (pH_{water}) was transformed into values measured in 0.01 mol L^{-1} $CaCl_2$ solution (pH_{CaCl2}), according to the relationship used by Sterckeman et al. [32]:

$$pH_{CaCl_2} = pH_{water} - 0.54 \left(R^2 = 0.88,\ n = 86\right). \tag{10}$$

2.4. Ecological Risks

The ecological risk index (ERI) model [27,42] was employed to evaluate the potential ecological risks posed by heavy metals in soil and leachate. The ERI was calculated using Equation (11).

$$ERI = \sum_{i=1}^{n} EI \tag{11}$$

$$EI = T_i \times \frac{C_i}{B_I} \tag{12}$$

where ERI corresponds to the sum of all the risk factors for heavy metals. EI is the individual potential ecological risk factor; n represents the number of target heavy metals. T_i is the biological toxicity factor of a target heavy metal. When estimating the risk of heavy metals in soil, C_i (mg kg^{-1}) is the concentration of metal in the soil, and B_i (mg kg^{-1}) is the background value for metals in soil. When estimating the risk of heavy metals in leachate, Ci (mg L^{-1}) is the concentration of metal in the leachate, Bi (mg L^{-1}) value of target heavy metal was set as the Class III concentration threshold of "Standard for groundwater quality (GB/T 14848-2017)" [43]. The values of T_i and B_i were illustrated in Table S3. The ERI and EI are divided into 5 grades [42], see Table S4 for details.

3. Results and Discussion

3.1. Average Annual Infiltration Water in China

The geographical distribution of the average annual precipitation, irrigation, and evapotranspiration in China is depicted in Figure S1, Figure S2 and Figure S3, respectively. Soil water infiltration is an important process of the water cycle in farmland soil, which influences the water budget of crops, potential topsoil loss by erosion, runoff, and groundwater recharge [44]. The infiltration data were processed by applying the zonal analysis tool in ArcGIS to determine the average infiltration values for each administrative region, as summarized in Table S5 and shown in Figure S4. The average annual infiltration water of farmland soil varied widely among different regions. The amount of infiltration water in southern China was significantly higher than that in northern China. The infiltration water of Jiangxi, Guangdong, Guangxi, and Hainan provinces was over 1200 mm. Meanwhile, the infiltration water of Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia, and Shandong provinces was below 200 mm.

3.2. Average Annual Heavy Metal Leaching in China

Traditionally, the southern and northern parts of China are marked by the Qinling Mountain and Huai River. There are significant differences between South and North China in terms of climate, landscape, economy activities, and agricultural practices. After calculating the average annual infiltration water for each administrative region, Equations (1)–(3) were used to simulate the leaching amounts of heavy metals and labeled the outcomes as L, L_{12}, and L_6, respectively. These results are presented in Table S5.

3.2.1. Comparison of Model Prediction Results with Previous Literatures

Our previous study on the fluxes of Cd in farmland soil demonstrated notable differences between the north and south regions of China [14]. Therefore, we divided China into north and south regions for further discussion, see Table S5 for details. The calculation results of Equations (1)–(3) were compared with reported measurements in the literature.

As Figure 1 shows, the leaching amount of heavy metals calculated by Equation (1) was the largest. The average leaching amounts of Cd, Pb, Cu, and Zn in North China were approximately 2 g ha^{-1} $year^{-1}$, 6 g ha^{-1} $year^{-1}$, 100 g ha^{-1} $year^{-1}$, and 500 g ha^{-1} $year^{-1}$, respectively, while in the South China they were about 40 g ha^{-1} $year^{-1}$, 120 g ha^{-1} $year^{-1}$, 1300 g ha^{-1} $year^{-1}$, and 16,800 g ha^{-1} $year^{-1}$, respectively. The average leaching of Cd in Hebei province was 0.032 g ha^{-1} $year^{-1}$ [19], within the range of Cd leaching calculated by Equation (2) (0.02 g ha^{-1} $year^{-1}$) and Equation (3) (0.041 g ha^{-1} $year^{-1}$). Additionally, it was closest to the average Cd leaching of the 15 northern provinces calculated by Equation (2) (0.139 g ha^{-1} $year^{-1}$). Previous studies reported the leaching amounts of four heavy metals (i.e., Cd, Pb, Cu, and Zn) in Jiangsu, Shanghai, Zhejiang, and Hunan provinces [11,16,18,31,45]. The average leaching amounts of Cd (3.425 g ha^{-1} $year^{-1}$), Cu (109.002 g ha^{-1} $year^{-1}$), and Zn (1399.045 g ha^{-1} $year^{-1}$) in the southern provinces calculated by Equation (2) were closest to those reported in the literature. As for the leaching of Pb in various southern regions, there were significant differences with average values of 43.207 g ha^{-1} $year^{-1}$, 131.240 g ha^{-1} $year^{-1}$, 16.340 g ha^{-1} $year^{-1}$, and 5.115 g ha^{-1} $year^{-1}$ in Jiangsu, Shanghai, Zhejiang, and Hunan, respectively. The Pb leaching values reported in Jiangsu and Zhejiang were closest to the average of southern provinces calculated by Equation (3), while Shanghai was closest to average calculated by Equation (1), and Hunan was closest to the average calculated by Equation (2). Above all, the prediction results by Equation (2) are closest to the measured values of Cd, Cu, and Zn leaching in previous literature, while also avoiding overestimating of the leaching of Pb. Additionally, intergroup significant difference analysis regarding Cd, Pb, Cu, and Zn was performed between

our estimated values for southern provinces and literature-reported values. The results indicate that the Cd, Pb, Cu, and Zn leaching amounts calculated using Equation (2) had the least significant differences compared to the literature-reported values (Table S6). Thus, we chose the values calculated from Equation (2) of Cd, Cu, Zn, and Pb for further analysis.

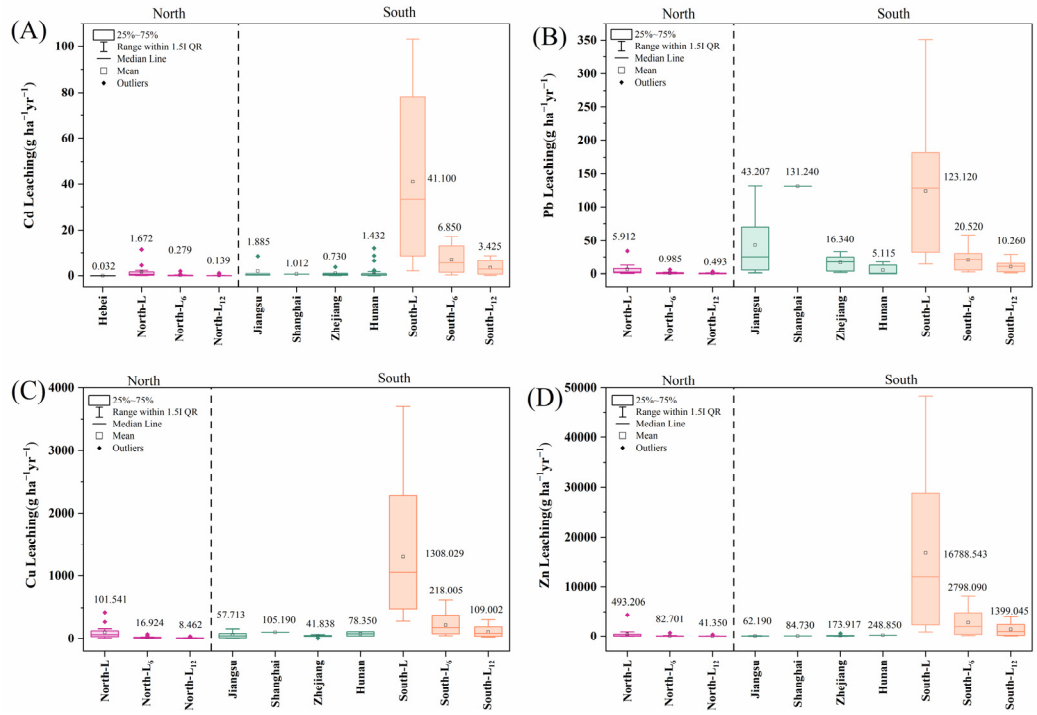


Figure 1. The comparison between calculated value and previously reported values of the leaching of heavy metals. North-L, North-L_6, and North-L_{12} represent the leaching in the 15 provinces of North China, calculated by Equation (1), Equation (3) and Equation (2), respectively. S-15-L, S-15-L_6, and S-15-L_{12} denote the leaching in the 15 provinces of South China, calculated by Equation (1), Equation (3) and Equation (2), respectively. (**A**–**D**) depict the leaching of Cd, Pb, Cu, and Zn, respectively.

3.2.2. Average Annual Heavy Metal Leaching in Different Administrative Regions

The leaching of Cd, Pb, Cu, and Zn in different provinces is illustrated in Figure 2. In most of North China, the leaching amount of Cd ranged from 0.02 g ha^{-1} year^{-1} to 0.2 g ha^{-1} year^{-1}, whereas in most of South China, it ranged from 2 g ha^{-1} year^{-1} to 9 g ha^{-1} year^{-1} (Figure 2A). Guangxi province had the highest Cd leaching (8.605 g ha^{-1} year^{-1}), which was 1000 times higher than the lowest (Beijing). Figure 2B shows that the leaching amount of Pb in most of North China was below 0.2 g ha^{-1} year^{-1}, while in South China, it ranged from 1.2 g ha^{-1} year^{-1} to 30 g ha^{-1} year^{-1}. Jiangxi province had the highest Pb leaching (29.265 g ha^{-1} year^{-1}), while Beijing had the lowest (0.041 g ha^{-1} year^{-1}). The leaching of Cu and Zn in South China was substantially higher than in North China. In most of North China, Cu and Zn leaching were below 10 g ha^{-1} year^{-1} and 100 g ha^{-1} year^{-1}, respectively. In most of South China, Cu and Zn leaching were over 40 g ha^{-1} year^{-1} and 200 g ha^{-1} year^{-1}, respectively. Consistent with the results of Pb, Jiangxi province had the highest leaching amount of Cu and Zn, while Beijing had the least. The differences in heavy metal leaching flux may be attributed to the higher levels of heavy metal contamination in southern Chinese soils compared to northern soils [6], coupled with the acidic nature of southern soils [46], which promotes the presence of heavy metals in free ionic forms. Additionally, precipitation in Southeast China is significantly higher than in the northwest [47]. However, the current division of northern and southern regions is based on the Qinling Mountain and Huai River, and there are still significant differences in climate, soil, hydrology, and other aspects within

the region. Therefore, focusing on specific provinces is more conducive to identifying key areas and implementing effective management measures.

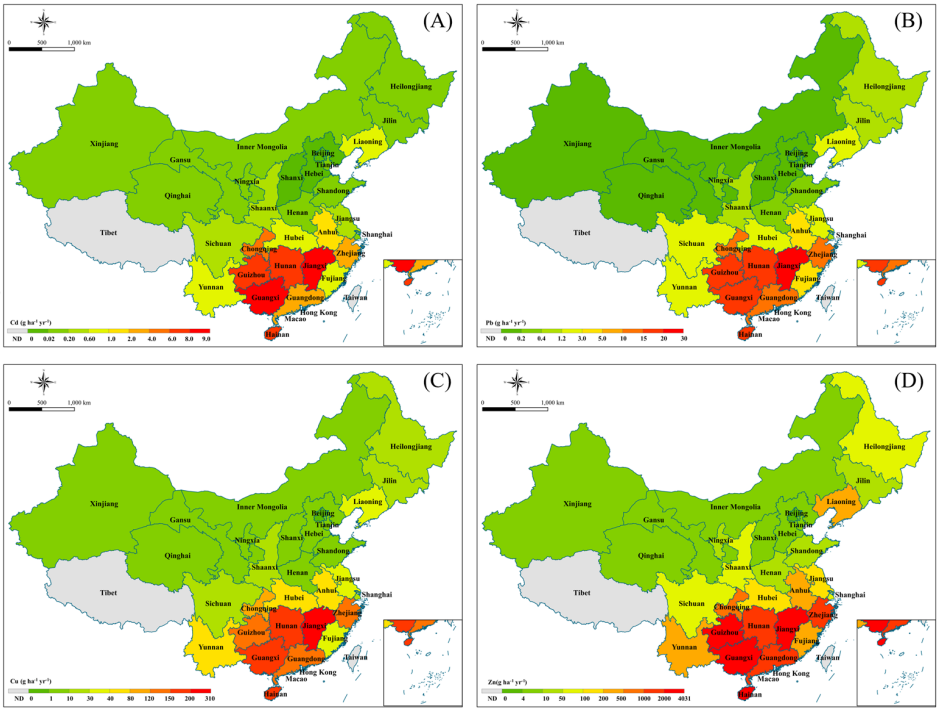


Figure 2. Geographical distribution of Cd (**A**), Pb (**B**), Cu (**C**), and Zn (**D**) leaching (L_{12}) in China.

Previous studies on heavy metal leaching mainly focused on Cd. To further evaluate the leaching degree of heavy metals, the calculated results were compared with research results from other regions around the world. Figure 3 shows that the average of Cd leaching in China was 1.78 g ha^{-1} year^{-1}, which was lower than the average of Japan (3.86 g ha^{-1} year^{-1}) [48] and Europe (2.56 g ha^{-1} year^{-1}) [49]. However, Cd leaching in eight southern provinces exceeded European averages, while in six provinces, it surpassed reported values in Japan. Jiangxi and Guangxi provinces were even over double of that in Japan.

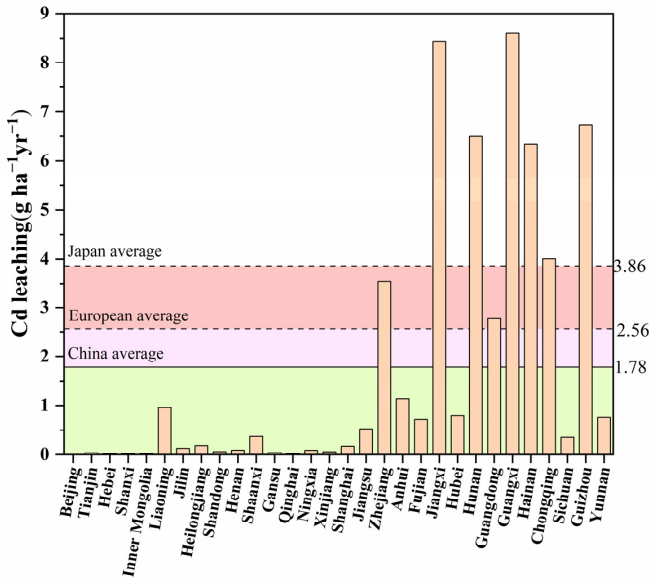


Figure 3. The comparison between calculated Cd leaching (L_{12}) in China and the reported average values in Europe and Japan.

Figure 4 shows the results of normalization processing for Cd, Pb, Cu, and Zn leaching amounts using ArcGIS. Areas with high Cd leaching also had higher leaching for Pb, Cu, and Zn. Jiangxi province exhibited the highest heavy metal leaching. Furthermore, Zhejiang, Hunan, Chongqing, Guizhou, Guangxi, Guangdong, and Hainan provinces are crucial areas needing attention. The pollution pressure brought by heavy metal leaching into deep soil and water cannot be ignored in the above provinces.

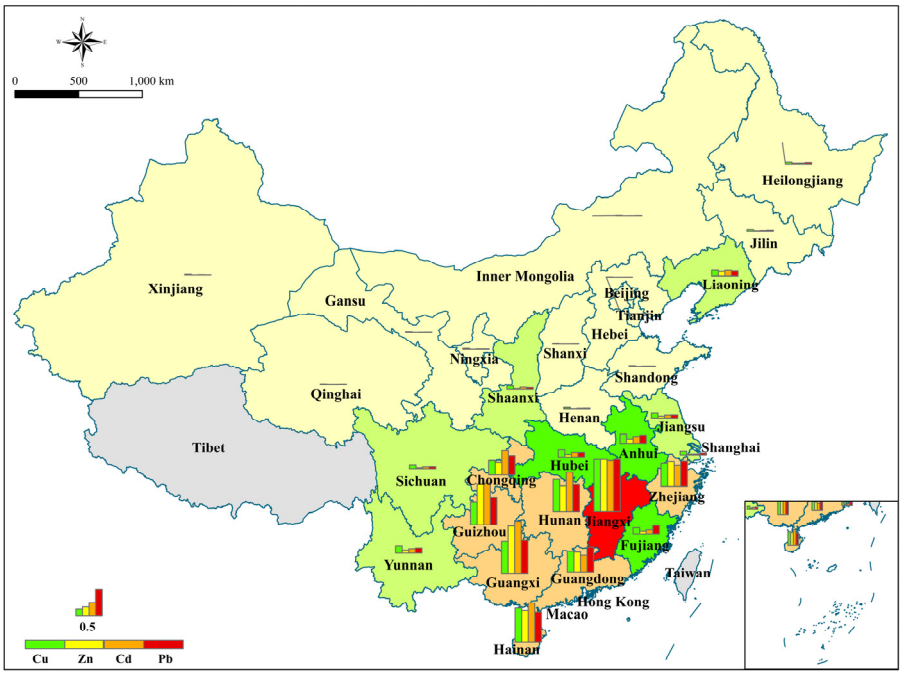


Figure 4. Geographical distribution of Cd, Pb, Cu, and Zn leaching in China after normalization treatment.

3.3. The Impact of Leaching on the Ecological Risk of Heavy Metals in Soil and Leachate

At present, the ERI values of heavy metals in the farmland soil range from 20 to 130 around China (Table 1), indicating low potential ecological risk. The EI values of Cd are between 40 and 80 in most regions, indicating that the potential ecological risks of Cd in these areas were moderate (Table S7). The EI values of Cd in Tianjin, Inner Mongolia, Zhejiang, Fujian, Henan, Hunan, Guangdong, Sichuan, and Shaanxi provinces exceed 80, indicating considerable ecological risks associated with Cd. It should also be noted that Cd average content in agricultural soils in Hunan, Guangxi, Chongqing, Guizhou, and Shaanxi exhibit levels exceeding the risk screening values stipulated in GB 15618-2018 (with pH $\leq$ 7.5 and concentrations > 0.5 mg/kg). In the case of simulations with the leaching rate (Equation (2)), without considering other input and output pathways of heavy metals, as depicted in Table 1, leaching could lead to a steady decrease in the ecological risk of heavy metals in the soil. The ERI values of Zhejiang, Jiangxi, Hunan, Guangdong, and Hainan provinces especially will significantly decrease (Variation% = −46.61%, −70.13%, −47.1%, −42.5%, −55.03%) after 100 years, mainly due to the reduction in the EI values of Cd. A century later, the ecological risk of Cd in Zhejiang, Hunan, and Guangdong provinces will shift from considerable ecological risk to moderate ecological risk, while the ecological risk of Cd in Jiangxi and Hainan provinces will decrease from moderate ecological risk to low ecological risk. However, research by Yang et al. [50] demonstrated that Cd, Pb, Cu, and Zn can migrate downward through various soil layers—including sandy layers, silty clay layers, and silty layers—reaching depths of up to 30 m and attaining the water table in some profiles. Therefore, while leaching effectively reduces heavy metal concentrations and associated ecological risks in surface soil, this benefit is offset by the

transfer of contamination to deep soil profiles and groundwater compartments, posing significant secondary pollution threats.

Table 1. Evolution of the potential ecological risks of heavy metals in soil under leaching models.

Province	ERI of Heavy Metals in Soil				Variation, %		
	Initial	10 years	20 years	100 years	10 years	20 years	100 years
Beijing	72.55	72.54	72.53	72.44	−0.01	−0.03	−0.15
Tianjin	129.92	129.87	129.82	129.41	−0.04	−0.08	−0.39
Hebei	71.00	70.97	70.94	70.68	−0.04	−0.08	−0.45
Shanxi	57.69	57.67	57.65	57.48	−0.03	−0.07	−0.36
Inner Mongolia	96.67	96.61	96.54	96.01	−0.06	−0.13	−0.68
Liaoning	74.76	73.55	72.35	63.60	−1.62	−3.22	−14.93
Jilin	70.72	70.55	70.36	68.93	−0.24	−0.51	−2.53
Heilongjiang	59.69	59.39	59.10	56.80	−0.50	−0.99	−4.84
Shanghai	41.85	41.66	41.47	40.04	−0.45	−0.91	−4.32
Jiangsu	71.77	71.19	70.61	66.19	−0.81	−1.62	−7.77
Zhejiang	105.04	98.34	92.14	56.08	−6.38	−12.28	−46.61
Anhui	90.32	88.71	87.15	75.65	−1.78	−3.51	−16.24
Fujian	110.27	108.95	107.68	97.93	−1.20	−2.35	−11.19
Jiangxi	76.51	66.48	57.98	22.85	−13.11	−24.22	−70.13
Shandong	81.91	81.82	81.72	80.94	−0.11	−0.23	−1.18
Henan	119.58	119.42	119.25	117.93	−0.13	−0.28	−1.38
Hubei	56.93	56.29	55.66	50.89	−1.12	−2.23	−10.61
Hunan	105.51	98.77	92.49	55.82	−6.39	−12.34	−47.10
Guangdong	119.40	112.82	106.64	68.65	−5.51	−10.69	−42.50
Guangxi	48.87	44.51	40.63	21.55	−8.92	−16.86	−55.90
Hainan	73.51	67.56	62.15	33.06	−8.09	−15.45	−55.03
Chongqing	58.33	56.37	54.47	41.94	−3.36	−6.62	−28.10
Sichuan	113.20	112.60	111.99	107.33	−0.53	−1.07	−5.19
Guizhou	27.72	26.21	24.81	16.85	−5.45	−10.50	−39.21
Yunnan	43.23	42.73	42.26	38.64	−1.16	−2.24	−10.62
Tibet	/	/	/	/	/	/	/
Shaanxi	116.94	116.4	115.87	111.68	−0.46	−0.91	−4.5
Gansu	69.24	69.18	69.14	68.77	−0.09	−0.14	−0.68
Qinghai	50.35	50.32	50.3	50.11	−0.06	−0.1	−0.48
Ningxia	68.45	68.34	68.23	67.36	−0.16	−0.32	−1.59
Xinjiang	80.47	80.4	80.33	79.8	−0.09	−0.17	−0.83

As shown in Figure 5, the ERI values of leachate in various regions ranged from 1.22 to 82.04, which suggested that heavy metals exerted low total ecological risks in leachate. The EI values of Cd in Jiangxi, Hunan, Guangxi, and Guizhou provinces were between 40 and 80, indicating moderate ecological risk of Cd of leachate in these regions. It is worth noting that in some areas, the concentration of heavy metals in the leachate exceeded the standard limits for Cd, Pb, Cu, and Zn in drinking water (GB 5749-2022) [51] and Class III of groundwater (GB/T 14848-2017) (Figure S5, Table S8). The Zn concentration in the leachate of Liaoning, Zhejiang, Jiangxi, Hunan, Guangdong, Guangxi, Hainan, Chongqing, and Guizhou provinces all surpassed the standard limit (1 mg L^{-1}), with Guizhou province (4.893 mg L^{-1}) exceeding it by over four times. The Pb concentration in leachate from Zhejiang, Jiangxi, Hunan, Guangdong, Guangxi, Hainan, Chongqing, and Guizhou provinces also exceeded the standard requirement (0.01 mg L^{-1}), with Jiangxi being the most severe at 2.6 times higher than the standard. The Cd concentration in the leachate of Jiangxi, Hunan, Guangxi, Chongqing, and Guizhou provinces exceeded the standard limit (0.005 mg L^{-1}), while Guizhou province (0.107 mg L^{-1}) was more

than twice that. Research indicates that consuming groundwater with elevated heavy metal concentrations increases the risk of chronic diseases and cancers, particularly in children [52,53]. Reports show that approximately 220 Chinese cities utilize groundwater as a drinking water resource, a pattern particularly prominent in Northwest China [54]. Hence, heavy metal leaching from farmland constitutes a public health concern that warrants critical attention.

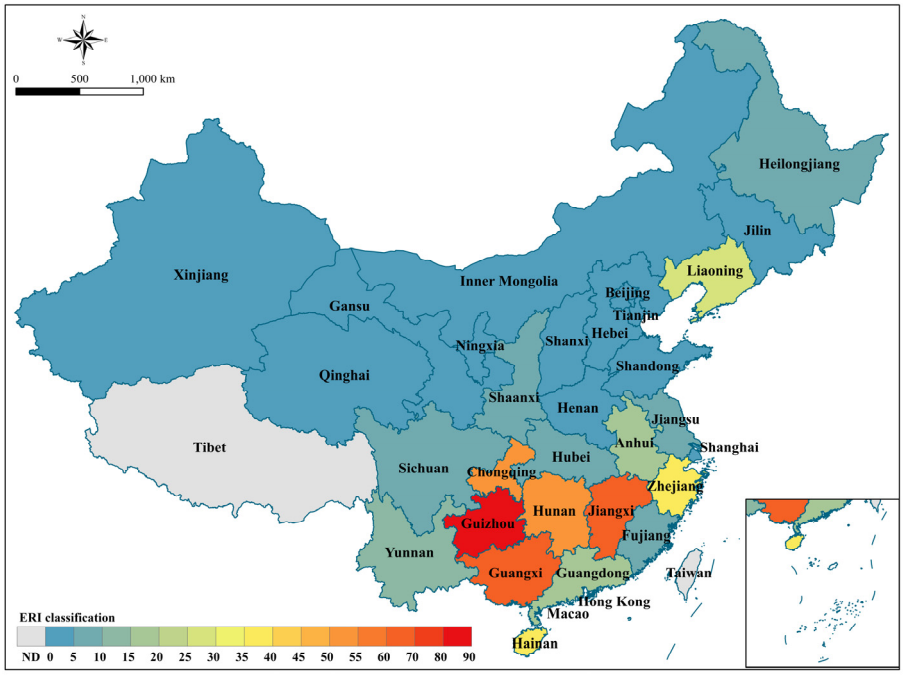


Figure 5. The potential ecological risks of heavy metals in leachate.

3.4. Factors Influencing Heavy Metals Leaching

Referring to the study of Feng et al. [18], we conducted parameter sensitivity analysis for the leaching model by systematically increasing each parameter by 5%. The sensitivity analysis shows that soil pH had the greatest impact on the leaching of Cd, Pb, Cu, and Zn, followed by annual precipitation (P) > heavy metal content in soil ([Me]s) > annual evapotranspiration (E) > soil organic matter content (SOM) > the irrigative water use efficiency (η) > annual irrigation (I) (Figure 6). An increase in pH by 5% leads to a reduction of 31.2% in Cd, 25.42% in Pb, 22.07% in Cu, and 38.37% in Zn leaching. This may be greatly due to the significant influence of pH on the adsorption and speciation behaviors of heavy metals in soil [38]. A decrease in soil pH will lead to an increase in the mobility of metals [55]; with a pH below 6, the exchangeable fraction Cd increases regularly along the field profile in neutral loamy [56]. Whereas an increase in soil pH is conducive to the adsorption of metals on the soil surface, there was 100% Cd sorption on six series soil at pH > 6 [57]. Furthermore, the formula reveals significant interaction effects between variables. Taking the influence of pH and rainfall on Cd leaching flux as an exemplar: (1) when precipitation increases to 1300 mm, Cd leaching flux rises by 16%; (2) when pH declines to 6, Cd leaching flux increases by 54%; and (3) when precipitation rises to 1300 mm concurrently with pH reduction to 6, leaching flux increases by 108%, yielding a synergistic amplification factor of 1.54. Environmental management implications include the following: (1) Low-pH conditions coupled with high precipitation constitute a high-risk combination. Emergency monitoring protocols for leaching should be activated during extreme rainfall events. (2) In low-pH soil zones, implement infiltration control measures such as impermeable covers and engineered drainage systems. (3) In high-precipitation regions, initiate soil pH

amendment projects, as every 0.5 unit pH increase reduces synergistic amplification effects by approximately 40%.

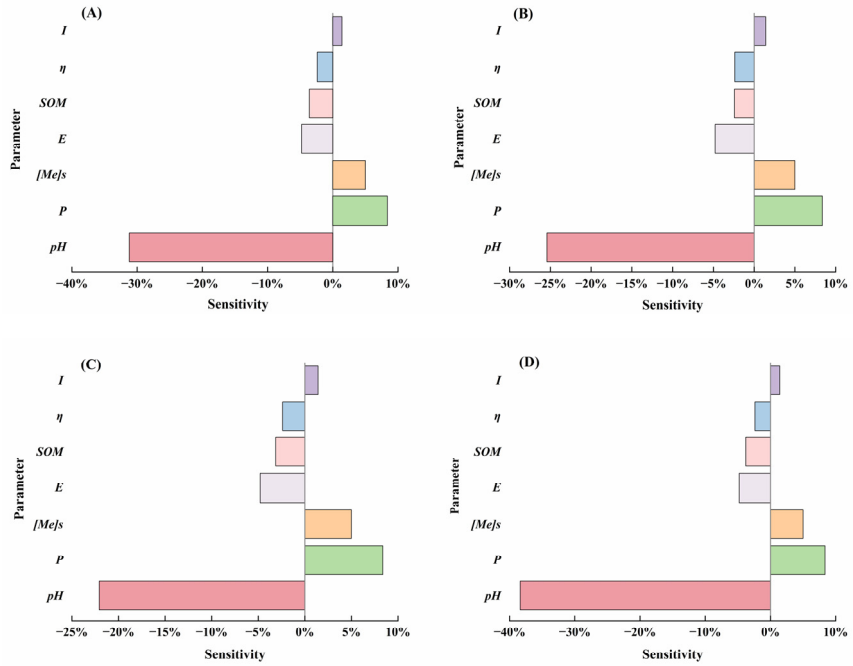


Figure 6. The influences of changes in input parameters on Cd (**A**), Pb (**B**), Cu (**C**), and Zn (**D**) leaching (each parameter was increased 5%). *pH*: the soil pH measured in $CaCl_2$; *P*: the total annual precipitation; $[Me]_s$: heavy metal content in soil; *E*: the total annual evapotranspiration; *SOM*: soil organic matter; η: the irrigative water use efficiency; and *I*: the total annual irrigation.

With the emission of large amounts of acidic substances caused by industrialization, the area of acidic soil in China expanded [58]. Although soil acidification improved in recent years, the pH of paddy fields in the middle and lower reaches of the Yangtze River and dryland soils in Northeast China is still showing a downward trend [59]. The amount of heavy metal leaching increases with higher heavy metal concentrations in the soil and decreases with increasing organic matter content in the soil. Luo et al. [60] revealed that the content of Cd, Zn, Cu, and Pb in Chinese farmland soils increased at rates of 0.004 mg $kg^{-1}year^{-1}$, 0.45 mg $kg^{-1}year^{-1}$, 0.21 mg $kg^{-1}year^{-1}$, and 0.1 mg $kg^{-1}year^{-1}$, respectively. Precipitation, evaporation, irrigation, and the irrigative water use efficiency impact metal leaching by influencing infiltration water. Precipitation was the second most influential factor after soil pH and positively correlates with heavy metal leaching. Cd, Zn, Cu, and Pb leaching increases 8.37% with a 5% increase in precipitation. Wu et al. [61] indicated that average precipitation is projected to rise by 8% or even 12% by the end of the century. On the contrary, heavy metal leaching decreases by 4.79% for each 5% rise in evaporation. According to public reports by the Ministry of Water Resources of the People's Republic of China, the irrigation in the farmland of China decreased from 2019 to 2021, and the irrigative water use efficiency increased. However, we estimate that this resulted in only a 0.91% annual reduction in the average infiltration water volume. Therefore, the impact of η and *I* in the future is limited. In summary, there is an increasing risk of heavy metal leaching in farmland soil in China in the future, especially in the middle and lower Yangtze River and Northeast China. Human-controllable factors influencing leaching include pH, [Me]s, SOM, η, and I, with pH exerting the most significant impact. Continuous monitoring of changes in various influencing factors in the key areas is essential, especially soil pH.

3.5. Management Implications

The above calculations quantified leaching in various provinces and assessed associated ecological risks. They identified pH as the most significant factor influencing leaching. These findings hold significant management implications: (1) the importance of prioritizing groundwater quality monitoring in critical regions (Guizhou, Jiangxi, Guangxi, Hunan, and Chongqing) and (2) implementing pH adjustment for agricultural soils in areas where groundwater exceeds the standard. As demonstrated in Table S9, adjusting farmland soil pH to 6 effectively reduces heavy metal concentrations in leachates below drinking water and groundwater (Class III) standards across all provinces except Hunan and Chongqing. For Hunan and Chongqing, where both soil and leachate heavy metals exceed thresholds, soil restoration materials (e.g., lime, phosphate amendment and biochar amendment) can be applied. These approaches simultaneously suppress metal leaching by increasing soil pH and reducing crop metal uptake [62,63]. Overall, this study provides the scientific basis and insights for optimizing farmland soil management and prioritizing groundwater monitoring.

4. Conclusions and Implication

Based on the calculated leaching model results, this study described the heavy metal (i.e., Cd, Pb, Cu, and Zn) leaching status of farmland land in China, analyzed the impact of leaching on the ecological risk of heavy metals in soil and leachate, and evaluated the impact of various factors on leaching. The average leaching amount of heavy metals in South China was over 10 times higher than in North China. Jiangxi, Guangxi, Guizhou, Hainan, Hunan, Zhejiang, Guangdong, and Chongqing provinces were critical regions for heavy metal leaching. The leaching of Cd in these regions exceeded the reported average in Europe. Furthermore, the Pb, Cu, and Zn leaching in these regions were also among the top ones in China. At present, the ecological risks of soils in various regions of China all exhibited low total potential risk (ERI < 150), while Cd was the dominant contributor to the risks. There were 16 provinces with moderate ecological risk of Cd and 9 provinces with considerable ecological risk of Cd. Although, without considering other input and output pathways, leaching reduces surface soil heavy metal concentrations and ecological risks, and this comes at the cost of pollutant transfer to deep soil layers and groundwater, ultimately posing a severe secondary contamination threat. The heavy metals in leachate exerted low total ecological risk; however, the concentration in some areas exceeded drinking water and groundwater (Class III) standards, posing a risk to human health. The ranking of the effects of various factors on heavy metal leaching was soil pH > precipitation > heavy metal content in soil > evapotranspiration > soil organic matter content > the irrigative water use efficiency > irrigation. Regulating pH is an effective measure to control heavy metal leaching from farmland. Adjusting the pH to 6 can effectively reduce heavy metal concentrations in leachates below drinking water and groundwater (Class III) standards across all provinces except Hunan and Chongqing. In the future, there will be a trend of further increases in heavy metal leaching in China. Therefore, it is crucial to prioritize groundwater monitoring in Guizhou, Jiangxi, Guangxi, Hunan, and Chongqing, and to regulate the pH of farmland in key regions. However, the lack of field monitoring validation represents a potential limitation. Future research should focus on field monitoring to assess the model's applicability and subsequently refine it based on empirical measurements. This study did not explore temporal variability, such as the impact of seasonal rainfall changes on leaching flux. Future research should include field monitoring of the temporal distribution of heavy metal leaching to better address extreme rainfall events.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/agronomy15092126/s1, Text S1: Empirical coefficients (1/12 and 1/6) [49,64–66]; Figure S1: Geographical distribution of average annual precipitation in mm in China; Figure S2: Geographical distribution of average annual irrigation in mm in China; Figure S3: Geographical distribution of average annual evapotranspiration in mm in China; Figure S4: Geographical distribution of average annual infiltration water in mm in China; Figure S5: The calculated heavy metals content in the leachate exceeds the drinking water quality (GB 5749-2022). The red area exceeds the standard, and the blue area does not exceed the standard; Table S1: Mean soil characteristics used in Leaching models [40]; Table S2: Soil pH and SOM values in various provinces in China [67–77]; Table S3: Biological toxicity factor, background value in soil, and standard concentration in groundwater of the target heavy metal [27,43,77–81]; Table S4: Evaluation criteria for potential ecological risk index method; Table S5: The annual leaching amounts of Cd, Pb, Cu, and Zn under different leaching models calculated using Equations (1)–(3); Table S6: Analysis of significant differences among multiple groups for Cd, Pb, Cu, and Zn; Table S7: EI and ERI values of the heavy metals in soil; Table S8. The concentration, EI and ERI value for heavy metals (Cd, Pb, Cu, and Zn) in leachate [51]; Table S9. Heavy metal content in leachate in critical provinces before and after pH control.

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Abbreviations

The following abbreviations are used in this manuscript:

OC	The soil organic carbon content
SOM	Soil organic matter
ERI	The ecological risk index

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