agronomy

MDPI

*Article*

# Environmental Assessment of Soils and Crops Based on Heavy Metal Risk Analysis in Southeastern China

**Xinyu Mao [1,*], Jingjing Sun [1], Hiba Shaghaleh [2], Xiaosan Jiang [3], Huaizhi Yu [1], Senmao Zhai [1] and Yousef Alhaj Hamoud [4,*]**

[1] College of Agricultural Science and Engineering, Hohai University, Nanjing 210098, China
[2] College of Environment, Hohai University, Nanjing 210098, China
[3] Taizhou Research Institute, Nanjing Agricultural University, Taizhou 225300, China
[4] College of Hydrology and Water Recourses, Hohai University, Nanjing 210098, China
* Correspondence: mxy880731@163.com (X.M.); yousef-hamoud11@hotmail.com (Y.A.H.)

**Abstract:** Heavy metal pollution in soil–crop systems has attracted great attention globally, caused by rapid urbanization and intensive industrialization. The research aims to investigate the environmental quality of the agricultural production area in Taizhou City, a typical economic region that is along the Yangtze River in the Southeast of China. A total of 370 sampling sites were chosen, with 370 soil, rice (*Oryza sativa* L.) and wheat (*Triticum aestivum* L.) samples collected, respectively, for measuring and analyzing the status, spatial distribution and pollution level of different heavy metals. The mean values of soil Cr, Pb, Cd, As and Hg were 66.78, 32.88, 0.23, 8.16 and 0.16 mg/kg, which were lower than the risk control standard values (RCV). However, the mean values of Pb, Cd and Hg were 1.25-, 1.77- and 2-fold larger than their soil background values (SBV) due to the intensive anthropogenic activities. The average content of Cd in rice exceeded its food safety limiting values (FCV) by 0.05 mg/kg, and the average contents of Pb in rice and wheat both exceeded the relevant FSV by 0.42 and 0.186 mg/kg, respectively. In addition, the maximum As and Cr contents in rice and wheat could be 0.13, 0.46 mg/kg and 0.63, 3.5 mg/kg larger than the relative FCVs in certain areas. Most of the high-value areas of soil and crop heavy metals were mainly located in Xinghua City, Taixing City and Jiangyan District, which had a similar distribution pattern with local industries or anthropogenic activities. The heavy metal pollution in soils and crops was found to be inconsistent, as 8.94% of the arable land possessed lightly metal pollution, while 3.18% of the area of rice and 4.0% of the area of wheat suffered severe pollution, with excessive accumulation of Cr, Pb and Cd. Based on the heavy metal pollution assessment of soil–crop systems, approximately 83% of the study area possessed medium or higher environmental quality, which was preferable for agricultural production. Our results implied that the spatial distribution and pollution level of the heavy metals in soil–crop systems were significantly influenced by industrial activities, followed by agricultural sources, transportation emissions and so on. Therefore, continuous monitoring and source control of heavy metals, especially for Cr, Pb and Cd, should be conducted to ensure the regional environmental quality and food security.

**Keywords:** heavy metal pollution; spatial distribution; pollution risk assessment; soil–crop system; environment partition

check for updates

**Citation:** Mao, X.; Sun, J.; Shaghaleh, H.; Jiang, X.; Yu, H.; Zhai, S.; Hamoud, Y.A. Environmental Assessment of Soils and Crops Based on Heavy Metal Risk Analysis in Southeastern China. *Agronomy* **2023**, *13*, 1107. https://doi.org/10.3390/agronomy13041107

Academic Editors: Ting Li, Ming Li and Kang Tian

Received: 22 February 2023
Revised: 4 April 2023
Accepted: 8 April 2023
Published: 12 April 2023

**Copyright:** © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/).

## 1. Introduction

Soil heavy metal pollution, which resulted from rapid urbanization and intensive industrial and agricultural production, has attracted both academic and public attention within the last few decades [1]. Heavy metal refers to metal (loid) with a density larger than 5 g/cm$^3$, such as lead (Pb), chromium (Cr), cadmium (Cd), mercury (Hg) and arsenic (As) [2]. Due to the characteristics of toxicity, non-degradability and persistence, the accumulation of heavy metals in soil might pose a serious threat to food safety and human

health [3]. In China, approximately 16% of the arable land is facing different levels of heavy metal contamination, and 2.6% of it is at a moderate or severe level, according to the National Communique of Soil Pollution Survey [4]. In such regions, soil heavy metals were mainly derived from anthropogenic sources, which could also threaten the surrounding aquatic life via surface runoff or leaking in underground water [5,6]. Therefore, it is necessary and important to assess the environmental quality based on the analysis of the overall heavy metal contamination status, thus providing scientific instructions for reducing pollution risk before caring out agricultural activities.

Prior to classifying the potential risk of heavy metals in soil and determining the appropriate solution for risk control, it is vital to identify the total amount, distribution pattern and contamination level. The total amount of soil heavy metals can provide information on the overall pollution status, which may be valuable for making police and updating relevant threshold values [7]. Many publications have addressed that the spatial distribution of soil heavy metals is complex and significantly influenced by soil parent properties and anthropogenic activities, especially in the industrial areas [8–11]. This is due to the different sensitivities of heavy metals to the environmental factors such as soil and plant types, point source pollution inputs and tillage methods [12]. Therefore, to quantify the correlation between the studied elements in the soil and environmental variables using Pearson correlation analysis is necessary, which is helpful for the source identification of pollutants. The contamination level of a pollutant is conventionally evaluated through potential ecological risk indexes [13] such as the geo-accumulation index [14], single-factor pollution index [15], Nemerow comprehensive pollution index [16], enrichment factor [17] and target hazard quotient index [18]. These approaches are widely accepted and primarily focus on a single or multiple heavy metals by comparing the content of target heavy metal to its background and standard values.

Numerous previous studies have been conducted to assess the environment quality by using the statistical analysis results of the above indexes, while heavy metal contents in both soil and crops have not been taken into account [19–21]. In fact, it has been proved that not all heavy metal pollution in crops comes from soil, which highlights the demand for detecting the quality of soil and crops together [22]. Recently, much more attention is needed regarding food security in light of the request from the intense population, especially in economically developed areas. Although a few studies have been implemented to explore the in-between relationship of quality and ecological risk in soil–crop systems, they are mainly focused on some specific heavy metals in certain crops, and regional-scale pollution data of multiple heavy metals are still lacking [23–25]. Lately, the environment partition method of the production area has been proposed and applied to achieve an integrated evaluation by considering the soil–crop system quality in relation to heavy metals [26]. This approach is more rational for combining the soil environmental quality and agricultural product security by using the geographic information systems (GIS) technique and clustering analysis [27,28]. It also indicates the possible relationship between the agricultural soil–water resources security and farm produce quality, which might act as a useful tool for guiding the development of local sustainable agriculture [29].

In this work, a total of 370 sampling sites were chosen, with 370 soil, rice (*Oryza sativa* L.) and wheat (*Triticum aestivum* L.) samples collected, respectively, in Taizhou City, a typical economic development area of Southeastern China. The main objectives of this study were to: (1) investigate the overall heavy metal pollution status of soil–rice and soil–wheat systems; (2) illustrate the spatial distribution of heavy metals and identify the relevant hotspot clusters in the study area; (3) estimate the heavy metal pollution levels of soil–crop systems and identify the correlation between different metals with anthropogenic activities; and (4) classify the agricultural environment quality based on a comprehensive analysis in combination with soil environment quality and agricultural product security. The results of this study might provide practical references for evaluating environmental quality in other similar areas with intensive agriculture and would also offer a reliable data basis for local governments to mitigate heavy metal pollution with high risks.

## 2. Materials and Methods

### *2.1. Description of the Study Area*

The study area is Taizhou City (Figure 1), which is a typical agricultural region located in the central of Jiangsu Province, China (32°01′57″–33°10′59″ N, 119°38′24″–120°32′20″ E). It is the industrial belt of the Yangtze River Delta region and the coastal economic development area in Southeastern China, which consists of Xinghua City, Taixing City, Jingjiang City, Hailing District, Gaogang District and Jiangyan District, with a total area of 5787 km$^2$. The area is within a subtropical humid monsoon climate zone, with an annual average temperature and precipitation of about 14.4–15.1 °C and 1037.7 mm, respectively [30].

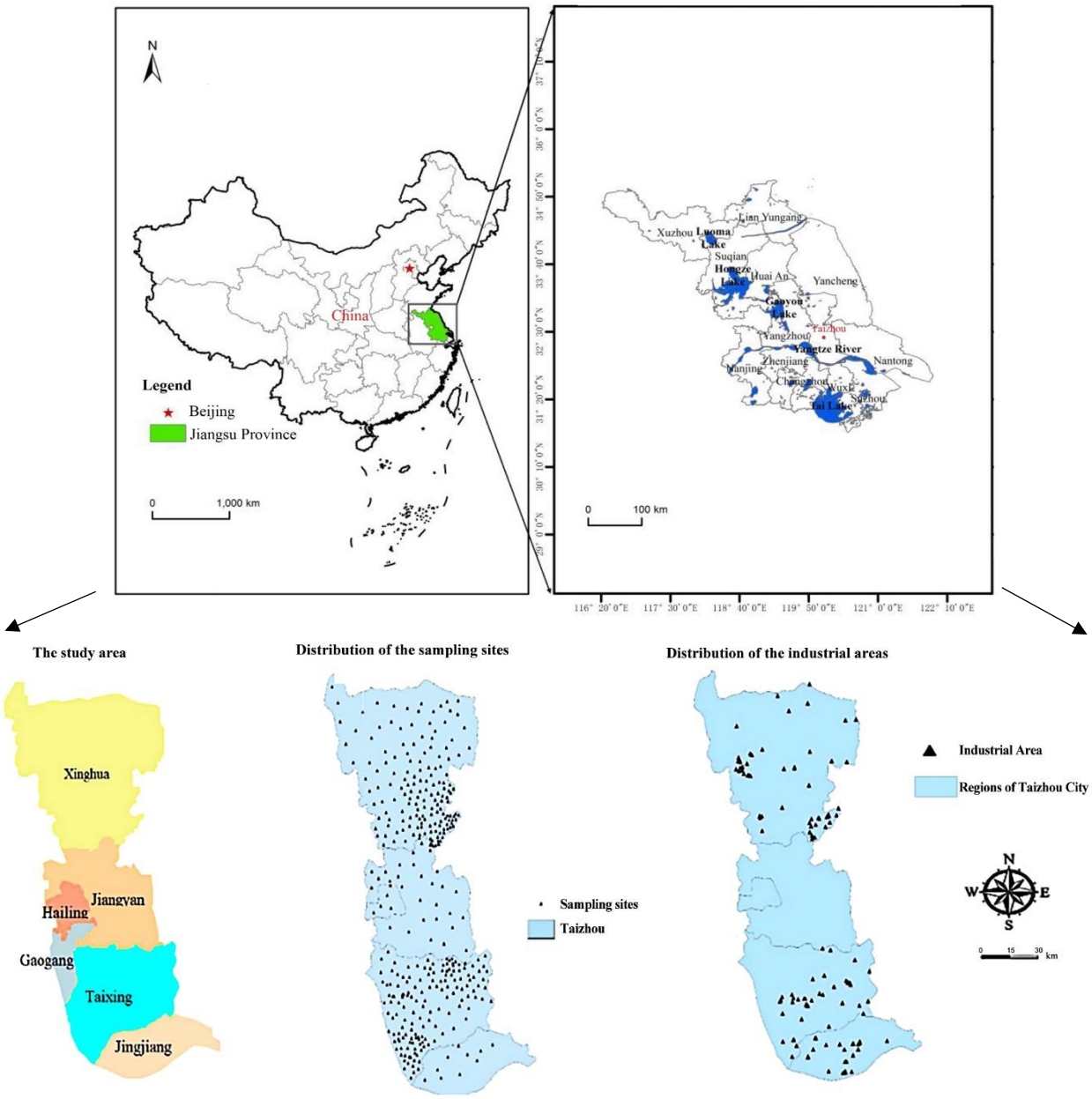


**Figure 1.** Distribution of sampling sites and some industrial areas in the studying area.

The main soil types in the study region are moisture soil and paddy soil, and the texture gradually becomes sandy from north to south. The average content of soil organic matter (OM), total nitrogen (TN), available phosphorus (AP) and available potassium (AK) is 21.4 g/kg, 1.36 g/kg, 25.2 mg/kg and 86.7 mg/kg, respectively [31,32]. By the end

of 2020, the total registered population of the city was 505.19 million. The overall grain production is 306.68 million tons, with an acre yield of 490.9 kg and wheat, rice, oilseed rape (*Brassica napus*) and vegetables being the major crops [33]. According to the statistics, the grain crop production in the study region was steadily increased at a rate of 2.3–3.5% within the last five years, ranking the first in Jiangsu Province [34]. The extensive input of chemical fertilizer, livestock and poultry manure and pesticides resulted in this satisfying agricultural production, while it raised the risk of heavy metal accumulation in both soils and crops. Industrial regions that include electronics, the chemical industry, medicine and smelting are mainly located in the north and south of the study area (Figure 1). Sewage and fumes emitted from these industrial factories also increased the heavy metals accumulation in soil–crop systems [35].

### *2.2. Sample Collection and Analysis*

#### 2.2.1. Layout of Sampling Points

According to the requirement for the layout of monitoring points, the selection of a sampling site should be typical, representative and homogeneous in order to accurately reflect the pollution status of heavy metals. The sampling sites were geo-located with a global positioning software (ArcGIS 10.3) after a four-step process that included fishnet creation (4 km × 4 km grid), central point establishment (center point to the interior of the grid), location calibration (calibrating the location of the sampling points) and coordinate system transition (deleting or moving the location of the sampling points) [36]. Then, a total of 370 sampling points were determined, and their spatial distribution is shown in Figure 1.

#### 2.2.2. Soil and Crop Sampling

For each sampling site, surface soils (0–20 cm) were collected with a stainless-steel drill from June to November 2020 through the quincunx resampling method, according to "Technical Specification for Monitoring Farmland Soil Environmental Quality" [37]. A total of 370 soil samples were collected. Each sample was approximately 2.5 kg and was a composite of five subsamples from a nearby 10 $m^2$ area from the center point of the sampling site. The soil samples were air-dried and passed through a 2 mm nylon sieve after grinding. Winter wheat and summer rice were chosen as the objective crops, and their grains were collected in June and November 2020, respectively, according to "Technical Specification for Contamination Monitoring of Agricultural Products" [38]. The sampling positions of grain were in accordance with those of soils, and about 250 g of each sample that consisted of five subsamples was collected. The sample number of rice and wheat was also 370 for each. The grain samples were initially cleaned by deionized water and dried to constant weight; then, they were passed through a 200 mm nylon sieve after grinding. All the samples were stored in hermetically sealed polyethylene bags before chemical analysis.

#### 2.2.3. Chemical Analysis

Soil pH was measured using potentiometry at a 1:2.5 (soil:water) ratio with a pH meter (pH 700, Thermo Scientific, Waltham, MA, USA). Soil OM was measured by the potassium dichromate volumetric method. Soil TN was determined using a Kjeldahl nitrogen analyzer (SKD-3000, Thermo Scientific, Waltham, MA, USA). Soil AK was determined by sodium bicarbonate extraction–molybdenum antimony resistance colorimetry using an ultraviolet visible spectrophotometer (Evolution 200, American Thermo Scientific). Soil AK was measured by the ammonium acetate extraction–atomic absorption spectrophotometry (200-seriesAA, Agilent Technologies, Santa Clara, CA, USA) method.

For heavy metal determination, the soil and grain samples were initially digested by HCl-HF-$HClO_4$-$HNO_3$: 0.5 g of the soil or grain sample was dried at 105~110 °C and was placed in a 30 mL polytetrafluoroethylene crucible. The samples were moistened by deionized water and then homogeneously mixed with a 10 mL HF and 5 mL $HCLO_4$-$HNO_3$ (*v*:*v* = 1:1) mixture; After 1 h and 4 h of digestion at low (100 °C) and high temperatures

(250 °C), respectively, another 5 mL $HCLO_4$-$HNO_3$ was added to the mixture to fully decompose the organic carbides. When the black organic carbides disappeared, 5 mL HCl (2 mol/L) was added to further digest the residue until the white smoke was exhausted. The residue was then transferred to a 25 mL volumetric flask and diluted with deionized water. After digestion, the contents of soil and grain Cd, Pb, As and Cr were measured using inductively coupled plasma mass spectrometry (ICP-MS, 7500 Cx, Agilent Technologies, Santa Clara, CA, USA) [39]. For the determination of soil Hg, 5 g of the soil sample was first added in a 150 mL conical flask and mixed with 40 mL deionized water, 10 mL $H_2SO_4$ and 20 mL $KMnO_4$ solution. The mixture was digested for 1 h under a temperature of 80 °C. After cooling, the mixture was added with $NH_2OH{\cdot}HCl$ solution and shaken until the purplish red and brown color faded away. Deionized water was applied to get the mixture to a constant volume, and 2 mL $SnCl_2{\cdot}H_2O$ was added to reduce the $Hg^+$ to Hg. Then, the Hg was measured by cold vapor atomic absorption spectroscopy under ultraviolet light with a wavelength of 253.7 nm [40]. For the determination of grain Hg, the microwave digestion method was applied for the pretreatment of the grain: a 0.3 g grain sample was mixed with 5 mL $HNO_3$ and 2 mL $H_2O_2$ and added into a digester tank. The digester tank was put in the microwave digestion system and initially digested under conditions of 30 min boost time, 5 min holding time, 343 kpa pressure and 50% power. The secondary digestion was conducted under conditions of 30 min boost time, 7 min holding time, 686 kpa pressure and 75% power. The tertiary digestion was implemented under conditions of 30 min boost time, 5 min holding time, 1096 kpa pressure and 90% power. Then, the grain sample was completely digested and mixed with $HNO_3$ at a volume ratio of 1:9, and the content of Hg was further detected by hydride generation–atomic fluorescence spectrometry [41]. The recovery rates of Cd, Pb, As, Cr and Hg in the samples with the addition of standard reference solutions were between 92.6 and 106.4%. Duplicates were simultaneously analyzed, with relative standard deviations less than 5.0% compared to the measurements of national standard reference materials (GBW07402, GBW10011 and GBW10011 were obtained from the National Standard Detection Research Center, Beijing, China).

### *2.3. Data and Statistical Analysis*

#### 2.3.1. Evaluation of Soil Heavy Metal Pollution Status

In this research, the single-factor pollution index was introduced to distinguish the key accumulated heavy metals and estimate their pollution status in the study area, which was calculated using the following equation:

$$P_i = \frac{C_i}{S_i}$$

where $P_i$ is the single-factor pollution index of soil heavy metal, $C_i$ (mg/kg) is the measured value of soil heavy metal in the sampling site and $S_i$ (mg/kg) is the pollution risk control threshold of soil heavy metal under different soil pH conditions (pH $\leq$ 5.5, 5.5 < pH $\leq$ 5.5, 6.5 < pH $\leq$ 7.5, pH > 7.5) [42].

The Nemero comprehensive pollution index is a kind of weighted multi-factor environmental quality index which gives specific consideration to the most serious pollution factor in order to avoid the reduction in the heavy metal weighted value caused by the influence of average calculation. This method is most widely used and is extended to assess the pollution level of soil heavy metal as follows:

$$P_{com} = \frac{\overline{P_i}^2 + P_{imax}^2}{2}$$

where $P_{com}$ denotes the Nemero comprehensive pollution index of soil heavy metal, $\overline{P_i}$ denotes the average value of the soil heavy metal single-factor pollution index and $P_{imax}$ denotes the maximum value of soil heavy metal single-factor pollution index.

2.3.2. Assessment of Agricultural Product Heavy Metal Accumulation

The accumulation of heavy metals in agricultural products was also estimated by both the single-factor pollution index and Nemero comprehensive pollution index. In accordance with the crop types and relevant national standard for food safety [43], the single-factor pollution index of agricultural product heavy metals ($P_n$) can be calculated as follows:

$$P_n = \frac{C_n}{S_n}$$

where $C_n$ and $S_n$ refer to the detected value and safety threshold of the heavy metals in grain samples, respectively.

The Nemero comprehensive pollution index of agricultural product heavy metals ($P_{ncom}$) can be computed by the following formula:

$$P_{ncom} = \frac{\overline{P_n}^2 + P_{nmax}^2}{2}$$

where $\overline{P_n}$ and $P_{nmax}$ represent the average value and the maximum value of the heavy metal single-factor pollution index regarding the sampling grains.

2.3.3. Environment Partition of the Production Area

In this research, the soil quality was applied as the main indicator to define the environmental quality of the production area. Comprehensively considering the spatial distribution of heavy metals in soil–crop systems and the yield of crop, the K-means clustering algorithm was used to quantitatively classify the environment partition. Five major influencing factors were selected: (1) the comprehensive pollution index of soil heavy metals ($P_{com}$), (2) the comprehensive pollution index of rice ($P_{n1com}$), (3) wheat heavy metals ($P_{n2com}$), (4) the yield of rice ($Y_1$) and (5) wheat ($Y_2$). The mathematical expression of K-means clustering is as follows:

$$E = \sum_{i=1}^{k} \sum_{x \in C_i}^{k} \| x - u_i \|_2^2$$

where the given samples in the study area were divided into $K$ clusters according to the distance between the samples. The setup of the $K$ value is important and should be based on the prior experience of the data through cross-validation. Assuming that the cluster is divided into $C_1, C_2, \ldots, C_K$ points, which should be set as close together as possible, the goal is to minimize the squared error $E$. In the equation, $u_i$ refers to the mean vector of the cluster $C_i$, also known as the centroid, which can be expressed as follows:

$$u_i = \frac{1}{C_i} \sum_{x \in C_i}^{k} x$$

The location of $K$ initialized centroids should not be too close due to their significant influence on the final clustering results and running time. Then, the minimum values of the above equation can be found to partition the cluster $C_i$.

*2.4. Data Analysis*

The spatial distribution of five heavy metals in soils and crops and relevant food safety risks were mapped via the inverse distance weight method in ArcGIS 10.3 (Esri Inc., Redlands, CA, USA). The distribution map of sampling sites and industrial areas was drawn according to the data offered by the Cultivated Land Quality Protection Station of Jiangsu Province and Shanghai MetroDataTech co. Ltd. using ArcGIS 10.3 (Esri Inc., Redlands, CA, USA). The statistical and correlation analysis of the heavy metal concentrations and environmental partition of the production areas were implemented using Excel 2016

(Microsoft Corp., Redlands, CA, USA) and PASW Statistics v18.0 (IBM Corp., Armonk, NY, USA).

## 3. Results and Discussion

### *3.1. Descriptive Statistical Analysis of Soil Physicochemical Properties and Heavy Metals Accumulation in Soils and Crops*

The physicochemical properties of the surface soils in the study area are descriptively listed in Table 1. The soil pH ranged from 3.40 to 8.28, with an average value of 6.67, indicating that the sampling soils are mainly slightly acidic to alkaline. The soil OM ranged from 4.87 to 77.5 g/kg, with the majority of soils possessing more than 2% of OM. The soil had low TN (ranged from 0.25 to 2.49 g/kg), while the AP and AK contents were relatively higher, with mean values of 25.2 and 86.7 mg/kg, respectively, compared to the results of the Second National Soil Survey of China [44]. In general, the farmland soils of Taizhou City are fertile, which is attributed to the over-application of organic and chemical fertilizers during the intensive agricultural production [45].

**Table 1.** Descriptive statistical analysis of soil physicochemical properties and heavy metals in the study area.

| Indexes | Cr | Pb | Cd | As | Hg | pH | OM | TN | AP | AK |
|---|---|---|---|---|---|---|---|---|---|---|
| | (mg/kg) | | | | | | (g/kg) | | (mg/kg) | |
| Maximum value | 807.0 | 123.0 | 1.19 | 24.70 | 0.70 | 8.28 | 77.5 | 2.49 | 49.7 | 475.8 |
| Minimum value | 19.0 | 10.50 | 0.06 | 0.01 | 0.02 | 3.40 | 4.87 | 0.25 | 12.3 | 15.1 |
| Mean value | 66.78 | 32.88 | 0.23 | 8.16 | 0.16 | 6.67 | 21.4 | 1.36 | 25.2 | 86.7 |
| Standard deviation | 49.28 | 10.53 | 0.11 | 4.49 | 0.87 | 0.87 | 8.75 | 0.49 | 42.4 | 51.2 |
| Coefficient of variation | 0.74 | 0.32 | 0.49 | 0.55 | 0.55 | 0.13 | 0.38 | 0.30 | 0.82 | 0.34 |
| SBV [a] | 77.8 | 26.2 | 0.13 | 10.0 | 0.08 | - | - | - | - | - |
| Over SBV rate (%) | 20.9 | 69.1 | 88.6 | 20.9 | 86.2 | - | - | - | - | - |
| RCV [b] | 200 | 250 | 0.30 | 30 | 0.30 | - | - | - | - | - |
| Over RCV rate (%) | 1.40 | 0 | 11.70 | 0 | 5.96 | - | - | - | - | - |

Note: [a] SBV: the soil background values of soil heavy metals in Jiangsu province that were obtained from the Chinese Environmental Monitoring Station (1990), [b] RCV: the risk control standard values for soil heavy metals that refer to II level standard of soil environment quality (GB15618-2018).

The average contents of Cr, Pb, Cd, As and Hg in the sampling soils were 66.78, 32.88, 0.23, 8.16 and 0.16 mg/kg, respectively (Table 1), which were lower than the risk control standard values (RCV) referred to II Level Standard of Soil Environment Quality [37]. In addition, the results were consistent with the data in the National Communique of Soil Pollution Survey [4] for the central region of Jiangsu Province, China, indicating that the majority of the studied soils might be safe for plantation and human health. The mean values of soil Cr and As were 66.78 and 8.16 mg/kg, respectively, which were both lower than their soil background values (SBV). However, the mean values of Pb, Cd and Hg were 1.25-, 1.77- and 2-fold larger than their SBVs, which maintained high over-SBV rates of 69.1%, 88.6% and 86.2%, respectively. Therefore, soil heavy metals, particularly for Cd and Hg, should receive attention, as 11.70% and 5.96% of the sampling sites exceeded the relevant RCVs. Accordingly, the mean value of soil Cd was similar to that in the Yangtze River Delta region but higher than that in Guangzhou City (0.21 mg/kg) and Shanghai City (0.196 mg/kg) [46–49]. The average content of soil Hg was much higher than that in Hebei Province (0.034 mg/kg), Chongqing City (0.08 mg/kg) and Tianjin City (0.043 mg/kg) [48–50]. The coefficients of variation (CV) of soil Cd and Hg contents were 0.49 and 0.55, respectively, suggesting a high spatial variability of soil Cd and Hg that was attributed to the influence of local anthropogenic activities [51]. Moreover, the contents of soil Cd were positively correlated with that of soil Pb (Table 2), which might imply their homology to accessing the soil environment such as atmospheric deposition, sewage irrigation and pesticide application [52].

**Table 2.** Pearson correlation analysis of the five heavy metals in soils.

| | Soil−Cr | Soil−Pb | Soil−Cd | Soil−As | Soil−Hg |
|---|---|---|---|---|---|
| Soil−Cr | 1.00 | – | – | – | – |
| Soil−Pb | 0.06 | 1.00 | – | – | – |
| Soil−Cd | 0.13 | 0.17 * | 1.00 | – | – |
| Soil−As | −0.09 | −0.24 * | −0.05 | 1.00 | – |
| Soil−Hg | −0.16 | −0.13 | −0.26 * | 0.15 | 1.00 |

Note: * Significantly correlated at the 0.05 level.

The accumulation of different heavy metals in rice and wheat is shown in Table 3. Except for Hg (mean value < 0.05 mg/kg), all other heavy metals could raise the food safety risk due to their detected contents in crop grain exceeding the food safety limiting values (FSV) referred to in the national standard [38] at different rates. The average contents of Cd in grains were 0.15 mg/kg for rice and 0.049 mg/kg for wheat, and the Cd content in rice was observed to be 0.05 mg/kg larger than its FSV. The average contents of As in grains were less than the relevant FCV, while in certain regions, the maximum As contents in rice and wheat reached 0.63 and 0.96 mg/kg, which were 0.13 and 0.46 mg/kg larger than the FCV. Like As, the average contents of Cr in grains did not exceed the FCV, but the maximum Cr concentration in rice and wheat achieved 1.63 and 4.50 mg/kg, which were much higher than the FCV. In addition, a higher average content of Pb was obtained, which was approximately threefold and twofold larger than the relative FSV in rice and wheat grains, respectively.

**Table 3.** Descriptive statistical analysis of the heavy metals content of agricultural products in the study area.

| Indexes | Cr | | Pb | | Cd (mg/kg) | | As | | Hg | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Rice | Wheat | Rice | Wheat | Rice | Wheat | Rice | Wheat | Rice | Wheat |
| Maximum value | 1.630 | 4.500 | 5.400 | 2.600 | 0.684 | 0.246 | 0.630 | 0.960 | 0.017 | 0.005 |
| Minimum value | 0.180 | 0.263 | 0.060 | 0.041 | 0.005 | 0.02 | 0.023 | 0.006 | 0.001 | 0.001 |
| Mean value | 0.480 | 0.388 | 0.620 | 0.386 | 0.150 | 0.049 | 0.099 | 0.056 | 0.005 | 0.002 |
| Standard deviation | 0.201 | 0.253 | 0.483 | 0.157 | 0.097 | 0.028 | 0.067 | 0.035 | 0.003 | 0.001 |
| FSV [a] | 1.0 | | 0.2 | | 0.1 | | 0.5 | | 0.02 | |
| Over FSV rate (%) | 1.27 | 7.28 | 3.18 | 17.22 | 12.74 | 1.32 | 0.64 | 0.66 | 0 | 0 |
| Coefficient of variation | 0.42 | 0.65 | 0.78 | 0.41 | 0.65 | 0.57 | 0.68 | 0.63 | 0.60 | 0.50 |

Note: [a] FSV: Food safety limiting values of heavy metals for cereal that refer to the national standard (GB2762-2017).

The comparatively higher contents of Cr and Pb in grains were probably due to their high contents in the study soil, which were in accordance with current reports [53]. The enrichment of Pb was comparably higher than that of Cr, which might be explained by the fact that the exchangeable Pb in soil is more easily absorbed by the crop and transported from the root to the grain [54]. Furthermore, as reported, soil Cr mainly exists in two valence forms of Cr (III) and Cr (VI). After entering the soil, Cr (III) tends to be adsorbed and fixed by the soil or forms $Cr(OH)_3$ precipitation in weak acidic or alkaline environments, and its content is generally higher than that of Cr (VI), making it difficult to migrate [55]. In regard to Cd, the over-FSV rate (12.74%) observed in rice was significantly higher than those of Cr (1.27%) and Pb (3.18%), with a much lower mean concentration of 0.15 mg/kg. In fact, Cd is known as the most bioavailable toxic element that is easily transported from soil to the food chain, even though its background concentration is low [56,57]. Several studies have pointed out that the enrichment and transport capacity of rice on Cd are much stronger than those of other cereal crops due to the effect of the transport protein OsNramp5 [58–60].

Actually, the average content of Cd in rice was found to be 0.33–0.69 mg/kg in the polluted area of Southeastern China, which was significantly higher than the safety threshold [61]. Hence, some preventive engineering measurements should be taken to mitigate the risks induced by the soil Cd contamination. Moreover, the accumulation of Cr in rice and wheat was positive corelated with As and Pb, respectively, while the accumulation of Cd in rice was negative corelated with As (Table 4), indicating the synergetic and competitive accumulation effects between these metals [62–64].

**Table 4.** Pearson correlation analysis of the five heavy metals in rice and wheat, respectively, in the study area.

| | | **Cr** | **Pb** | **Cd** | **As** | **Hg** |
|---|---|---|---|---|---|---|
| Rice | Cr | 1.00 | | | | |
| | Pb | 0.04 | 1.00 | | | |
| | Cd | 0.02 | 0.11 | 1.00 | | |
| | As | 0.24 * | 0.01 | −0.02 | 1.00 | |
| | Hg | 0.02 | 0.07 | 0.06 | 0.04 | 1.00 |
| | | Cr | Pb | Cd | As | Hg |
| Wheat | Cr | 1.00 | | | | |
| | Pb | 0.18 * | 1.00 | | | |
| | Cd | 0.01 | 0.01 | 1.00 | | |
| | As | 0.11 | −0.01 | −0.27 * | 1.00 | |
| | Hg | −0.09 | −0.05 | 0.10 | 0.01 | 1.00 |

Note: * Significantly correlated at the 0.05 level.

### *3.2. Spatial Distribution of Heavy Metals in Soils and Crops*

The spatial distribution of soil heavy metals is presented in Figure 2. Overall, the spatial distribution of soil heavy metals was apparently different from the south to north of the study area without a regular variation trend. The hotspots of high Cr distributed in the northern area of Jiangyan District and the low-value areas were relatively concentrated in the south of Taizhou City. The high-value areas of Pb were scattered and distributed in Xinghua City. Compared with that of Pb, the scattered distribution of Cd was more obvious, mainly situated in the south of Xinghua and Jingjiang City. Furthermore, higher contents of As and Hg were located in the southwest and southeast of Jingjiang City. The results showed that most of the high-value areas of soil heavy metals were located in Xinghua and Jingjiang City, which had a similar distribution pattern with local industries (Figure 1) to some extent. As reported, the spatial distribution of heavy metals in farmland soils is significantly affected by point source pollution, which mostly comes from the industrial emissions, which accounted for more than 50% of soil heavy metal accumulation [65]. Therefore, the densest human activities, particularly industrial activities such as metallurgy, nonferrous casting, electronic product processing and cotton spinning, of the study area resulted in the high contents of Cd, As, Cr, Hg and Pb in the surroundings [66]. Transportation, fertilization and swage irrigation are also important sources of soil heavy metal in the study area [67]. For instance, on the Ningtong Highway and Provincial Road, with a developed transportation network and high traffic steam, crossing the study area might lead to high levels of Pb, Cd and Hg in the surroundings through exhaust gas emissions and tire wear [68]. In addition, the southern study area adjoins the Yangtze River, and the high concentrations of soil Cd, Hg, As and Cr were likely due to the sewage irrigation and the excessive application of chemical fertilizer [69].

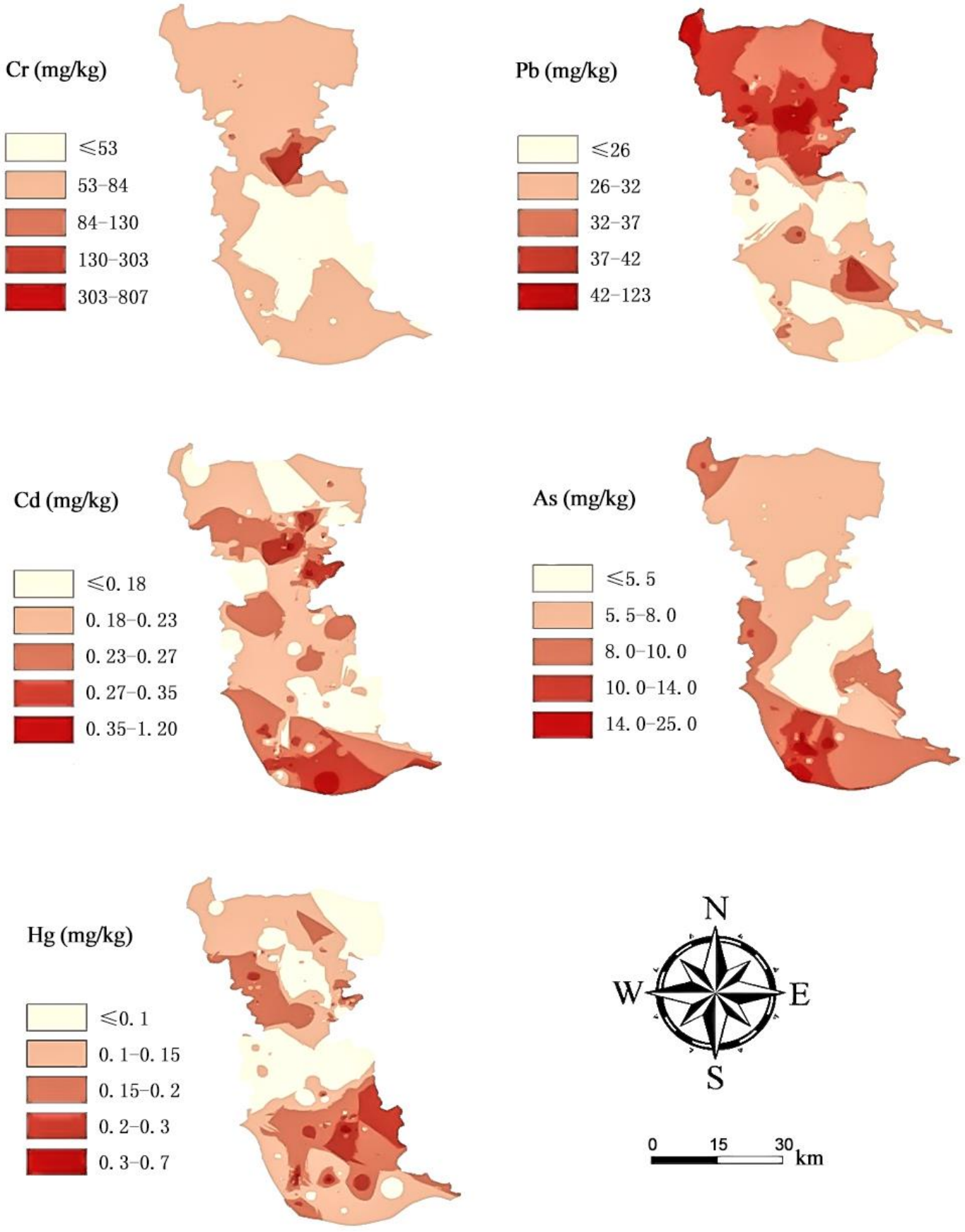


**Figure 2.** Spatial distribution of soil heavy metals in the study area.

The spatial distribution of heavy metals in crops is shown in Figure 3. Similarly, the high-value area of five heavy metals was scattered and distributed without a regular variation trend. Their concentrations in paddy grains showed a positive correlation with those of soil heavy metals, but not a linear relationship, which might be related to the inhibition mechanism of the root assimilation and plant conduction of heavy metals [70]. In terms of rice, higher contents of Cr, Pb and Cd were found in north-central regions, while higher contents of As and Hg were observed in south-central regions. For wheat, high-value Cr, Pb and As were distributed in south-central areas, and high-value Cd and Hg were distributed in north-central areas. In short, most of the hotspots with high-level heavy metals, especially for Cr, Cd and As in rice and for Pb and Hg in wheat, which exceeded the safety threshold, were mainly situated in the central sections of Taizhou City (including the Jiangyan, Gaogang and Hailing Districts). These results might be due to the local discrepancy that central Taizhou is downtown and possesses a larger population density, fewer industries and a higher contamination load compared to the other regions [71]. In fact, except for point source pollution, the bioaccumulation of heavy metals in food crops could be significantly influenced by urbanization, agrochemical application, plant species as well as environmental and climate conditions [72]. In general, the average contents of Cr, Pb, Cd, As and Hg in edible parts of the crops were 0.48, 0.62, 0.15, 0.099 and 0.005 mg/kg for rice and 0.388, 0.386, 0.049, 0.056 and 0.002 mg/kg for wheat, respectively. These results were detected to be similar or slightly lower than the current data gathered in Southeastern China from the published reports [73].

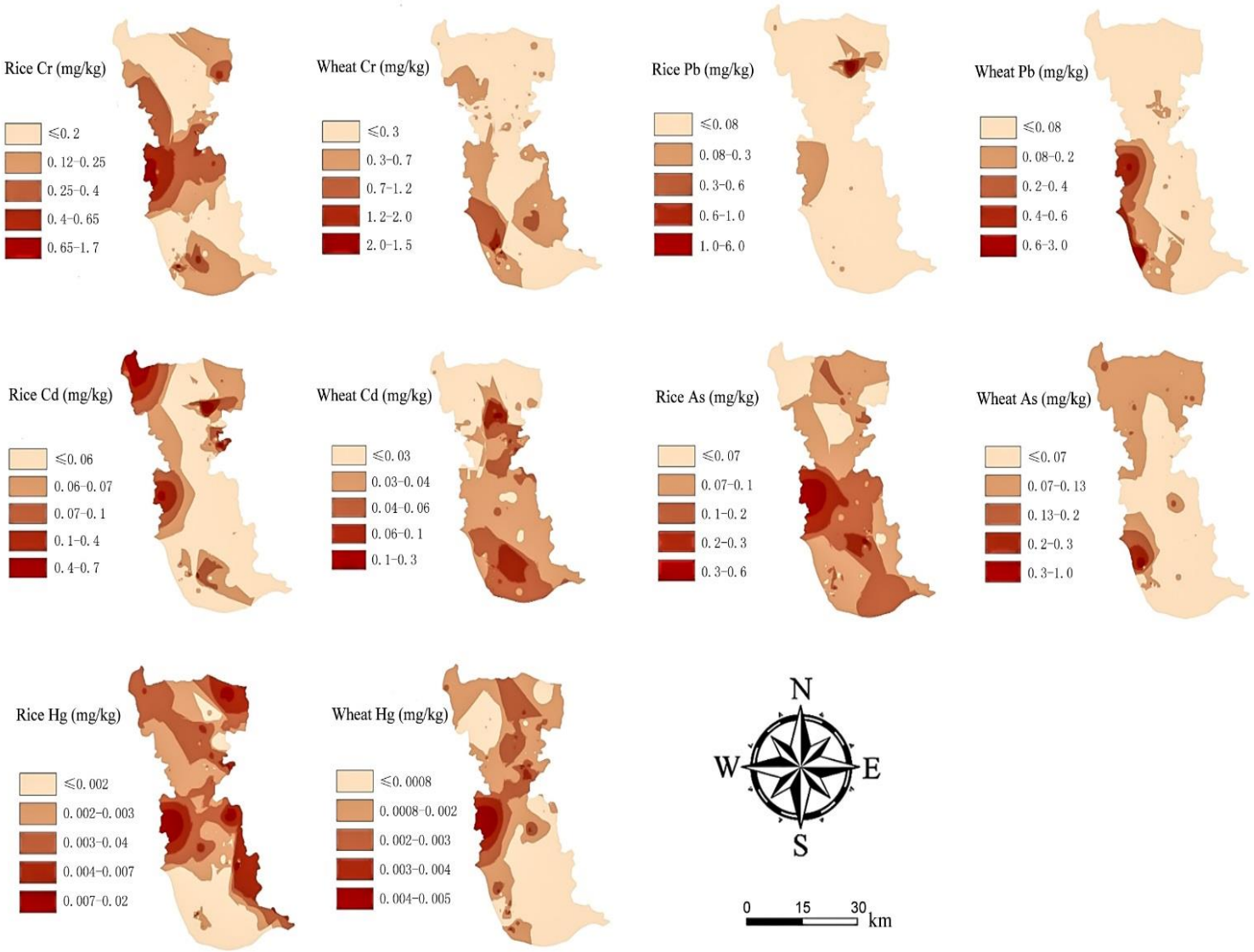


**Figure 3.** Spatial distribution of heavy metals in agricultural products in the study area.

### *3.3. Heavy Metal Pollution Assessment in the Study Area*

Table 5 presents the heavy metal contamination degrees of sampling soils and their potential environmental risks. According to the calculated single-factor pollution index ($P_i$) values, the soil samples were free of Hg and Pb pollution due to all their $P_i$ values being obtained to be less than 1. The pollution levels of Cr and As were light ($1.0 < P_i \leq 2.0$), with pollution rates of 0.75% and 0.27%, respectively. Soil Cd deserved more attention, since 2.98% of the samples reached a light or moderate pollution level. The results showed that more than 97% of the topsoil samples were not polluted with toxic metals, demonstrating a low potential environmental risk in the study area. The Nemero comprehensive pollution index ($P_{com}$) is able to assess the soil quality in a more complete way than the evaluation of only one single element. Nevertheless, it is worth mentioning that $P_{com}$ excessively emphasizes the impact of the maximum studied element, which might artificially exaggerate or neglect the influence of some other factors in the evaluation process. The $P_{com}$ values showed that most of the sampling sites were within the safety threshold, as 91.06% and 7.86% reached the security level ($P_{com} \leq 0.7$) and warning level ($0.7 < P_{com} \leq 1.0$), respectively. Apparently, most of the sampling soils were appropriate for food crop planting; only 1.08% of them reached a light pollution level, which was much lower than the average value (20%) of arable land in China published by the Chinese Ministry of Land and Resources [74]. In addition, the results based on $P_i$ and $P_{com}$ were similar, suggesting that the soil quality assessment in this work was reliable.

**Table 5.** Proportion of soil heavy metal pollution levels analyzed by the single-factor pollution index (A) and Nemero comprehensive pollution index (B) methods, respectively.

| | **Single-Evaluation Index** | **Class of Pollution** | **Proportion of Different Pollution Levels (%)** | | | | |
|---|---|---|---|---|---|---|---|
| | | | **Cr** | **Pb** | **Cd** | **As** | **Hg** |
| A | $P_i \leq 1.0$ | Non-pollution | 98.92 | 100 | 97.02 | 99.73 | 100 |
| | $1.0 < P_i \leq 2.0$ | Light pollution | 0.75 | 0 | 2.71 | 0.27 | 0 |
| | $2.0 < P_i \leq 3.0$ | Medium pollution | 0 | 0 | 0.27 | 0 | 0 |
| | $P_i > 3.0$ | Heavy pollution | 0 | 0 | 0 | 0 | 0 |
| | Comprehensive evaluation index | Class of pollution | Proportion of different pollution levels (%) | | | | |
| B | $P_{com} \leq 0.7$ | Security level | 91.06 | | | | |
| | $0.7 < P_{com} \leq 1.0$ | Warning level | 7.86 | | | | |
| | $1.0 < P_{com} \leq 2.0$ | Light pollution level | 1.08 | | | | |
| | $2.0 < P_{com} \leq 3.0$ | Medium pollution level | 0 | | | | |
| | $P_{com} > 3.0$ | Heavy pollution level | 0 | | | | |

Note: $P_i$ and $P_{com}$ are the single-factor pollution index and Nemero comprehensive pollution index of soil heavy metals, respectively.

The heavy metal contamination levels of the sampling food crops and their potential ecological risks are shown in Table 6. The results of the pollution index ($P_n$) suggested that both rice and wheat were not contaminated with Hg, as their obtained $P_n$ values were less than 1. The pollution rate of rice and wheat caused by four other heavy metals were ranked as Cd (12.1%) > Pb (3.18%) > Cr (1.27%) > As (0.64%) and Pb (17.33%) > Cr (7.33%) > Cd (1.34%) > As (0.67%) in rice and wheat, respectively. A much higher accumulation of Cd was observed in rice, with 3.82% of the samples reaching the heavy pollution level ($P_n > 3.0$). Inversely, the contamination of Cd in wheat was not obvious, while it had a serious-pollution rate of Cr (2.0%) and Pb (5.33%). These differences are significantly correlated with the bioavailability of soil heavy metals, which is usually ranked as Cd > Pb > As > Cr > Hg [75]. The tillage is another important influencing factor, as rice is grown under flooded conditions, which would improve the transformation of metals such as Cr (VI) into its more stable form (Cr (III)), thus decreasing the bioaccumulation of Cr [55].

Moreover, the translocation mechanism also has a great effect on metal bioaccumulation—for instance, the transport protein OsNramp5 of rice significantly improves the adsorption and translocation of Cd compared to other metals [58]. Generally, the pollution area of wheat was larger than that of rice, which was also reflected in the results of the Nemero comprehensive pollution index ($P_{ncom}$). The total polluted area of wheat was 21.33%, which was approximately twofold that of rice (11.46%). The contaminated area of rice and wheat ranked by different $P_{ncom}$ values was in same order: light pollution > heavy pollution > moderate pollution, and more ecological hazards were observed in wheat at each pollution level, which might be mainly caused by the integrative excessive accumulation of Cr, Pb and Cd.

**Table 6.** Proportion of crop heavy metal pollution degree, as analyzed by the single-factor pollution index (A) and Nemero comprehensive pollution index (B) methods, respectively.

| | **Single-Evaluation Index** | **Class of Pollution** | **Proportion of Different Pollution Levels (%)** | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | **Cr** | | **Pb** | | **Cd** | | **As** | | **Hg** | |
| | | | **Rice** | **Wheat** | **Rice** | **Wheat** | **Rice** | **Wheat** | **Rice** | **Wheat** | **Rice** | **Wheat** |
| A | $P_{n1} \leq 1.0$ | Non-pollution | 98.73 | 92.67 | 96.82 | 82.67 | 87.90 | 98.66 | 99.36 | 99.33 | 100 | 100 |
| | $1.0 < P_{n1} \leq 2.0$ | Light pollution | 1.27 | 4.0 | 1.91 | 8.67 | 7.01 | 0.67 | 0.64 | 0.67 | 0 | 0 |
| | $2.0 < P_{n1} \leq 3.0$ | Medium pollution | 0 | 1.33 | 1.27 | 3.33 | 1.27 | 0.67 | 0 | 0 | 0 | 0 |
| | $P_{n1} > 3.0$ | Heavy pollution | 0 | 2.0 | 0 | 5.33 | 3.82 | 0 | 0 | 0 | 0 | 0 |

| | Comprehensive evaluation index | Class of pollution | Proportion of different pollution levels (%) | |
|---|---|---|---|---|
| | | | Rice | Wheat |
| B | $P_{n1com} \leq 0.7$ | Security level | 84.08 | 64.67 |
| | $0.7 < P_{n1com} \leq 1.0$ | Warning level | 4.46 | 14.0 |
| | $1.0 < P_{n1com} \leq 2.0$ | Light pollution level | 6.37 | 14.0 |
| | $2.0 < P_{n1com} \leq 3.0$ | Medium pollution level | 1.91 | 3.33 |
| | $P_{n1com} > 3.0$ | Heavy pollution level | 3.18 | 4.0 |

Note: $P_{n1}$ and $P_{n2}$ is the single-factor pollution indexes of rice and wheat, respectively; $P_{n1com}$ and $P_{n2com}$ are the Nemero comprehensive pollution indexes of rice and wheat, respectively.

It could be seen that the soil pollution risk and crop pollution risk caused by heavy metals were inconsistent, which might be mainly attributed to the distribution of point source pollution. Figure 4 presents the spatial distribution of heavy metal pollution risk in crops based on the $P_{ncom}$ values. The most of heavy pollution areas of rice and wheat were located in the southeast of Xinghua City and the west of Taixing City, respectively. These regions were close to the relatively densest industries and the traffic arteries and streams that increased the local inputs and bioaccumulation of heavy metals via atmospheric deposition and irrigation [50,54]. Moreover, since the discrepancies of external influences such as climate, acid rain, tillage and fertilization would alter soil properties and the heavy metal morphology, they ultimately increased the uptake of heavy metals by crops [43,57,76]. Therefore, an objective environmental assessment of a production area should consider the integrated aspects of soil environmental quality and agricultural product quality.

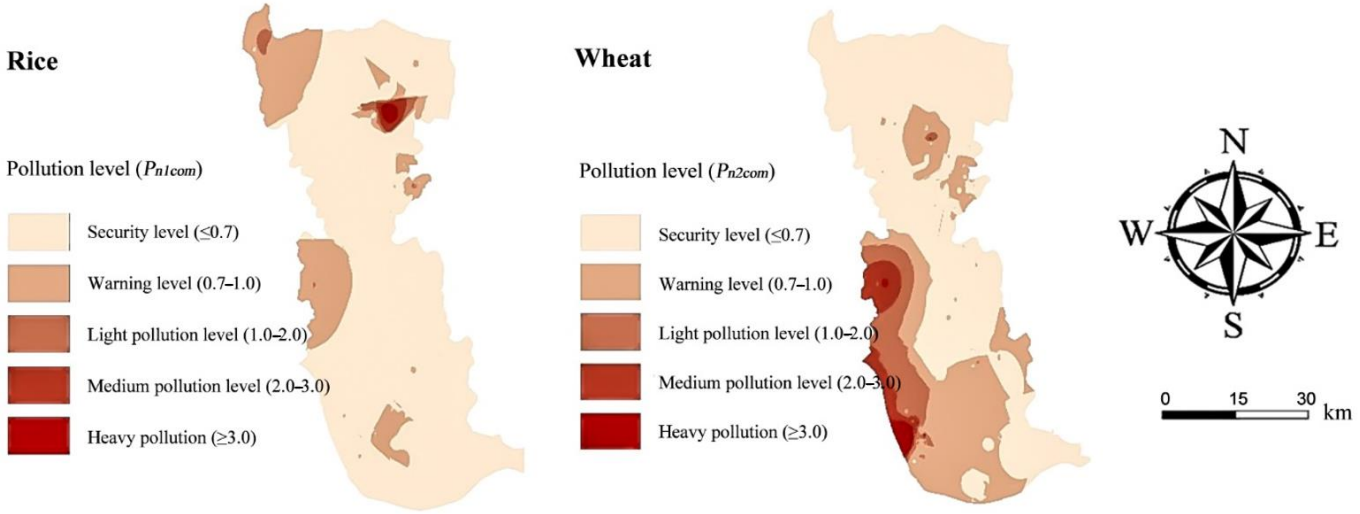


**Figure 4.** Comprehensive risk map of heavy metals in rice and wheat, respectively.

### *3.4. Environment Partition of the Production Area*

The environment partition of the production area in Taizhou city based on clustering analysis is shown in Table 7 and Figure 5. According to the determined clustering indexes, the environmental quality of the production area was classified into five grades: high quality, good, medium, common and poor quality, named as 1–5. The order for the proportion of the production area with different qualities was as follows: good (58.38%) > common (15.14%) > medium (12.16%) = high quality (12.16%) > poor quality (2.16%). The results indicated that 82.7% of the 370 sampling sites possessed medium or higher environmental quality, which was totally appropriate for the local agricultural production. The production area with poor quality was mainly located in Xinghua City, Taixing City and Jiangyan District, where were surrounded by intensive industries. Considering the pollutant inputs, continuous monitoring is suggested for the soil quality in these regions, along with the organization of the correlative enterprises working with other stakeholders to seek potential pathways for alleviating the risks from heavy metals pollution.

**Table 7.** Environment partition of the production area in Taizhou city based on clustering analysis.

| Clustering Index | Cluster | | | | |
|---|---|---|---|---|---|
| | **2** | **5** | **3** | **4** | **1** |
| $P_{com}$ | 0.48 | 0.37 | 0.47 | 0.42 | 0.45 |
| $P_{n1com}$ | 0.46 | 0.28 | 0.39 | 0.42 | 0.46 |
| $P_{n2com}$ | 0.51 | 0.52 | 0.85 | 0.80 | 0.44 |
| $Y_1$ | 619 | 353 | 613 | 550 | 666 |
| $Y_2$ | 427 | 400 | 421 | 410 | 436 |
| Case number | 216 | 8 | 45 | 56 | 45 |
| Area proportion (%) | 58.38 | 2.16 | 12.16 | 15.14 | 12.16 |

Note: $Y_1$ and $Y_2$ are the yield of rice and wheat, respectively; numbers 1–5 refer to the environment partition rank of the production area, named high quality, good, medium, common and poor quality, respectively.

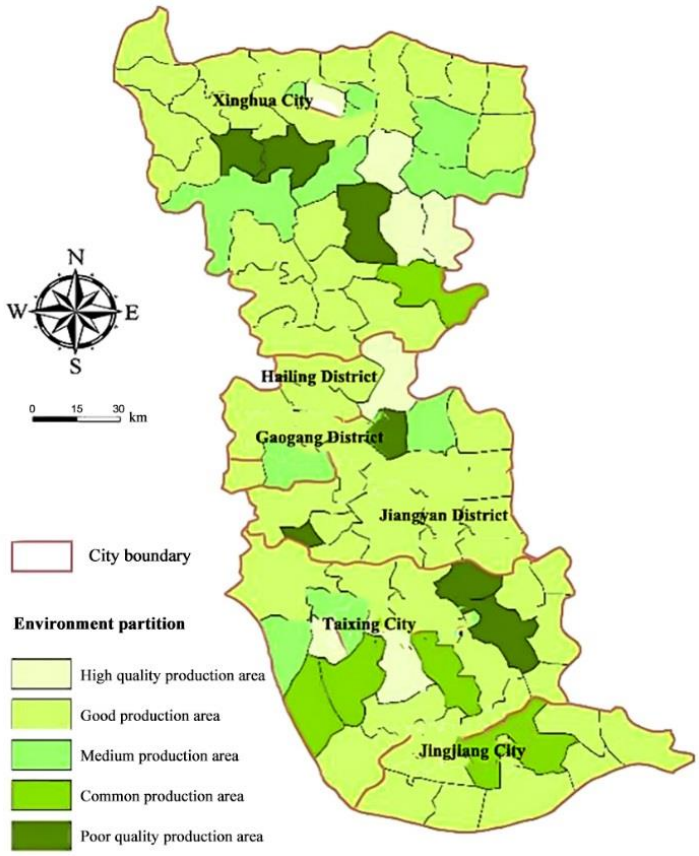


**Figure 5.** Environment partition of the production area in Taizhou City.

## 4. Conclusions

In this work, the environment quality of the production area in Taizhou City, a typical agricultural and industrial region of Jiangsu Province, was assessed by characterizing the contamination status, spatial distribution, correlation and risk of heavy metals in soils and crops simultaneously. The results indicated that the mean concentrations of soil Cr, Pb, Cd, As and Hg were 66.78, 32.88, 0.23, 8.16 and 0.16 mg/kg, respectively, which were lower than the RCV. Except for Hg, all other heavy metals could raise food safety risk issues. A higher content value of Cd was obtained in rice, which was 0.05 mg/kg larger than its FSV. The average content of Pb in rice and wheat both exceeded the relative FSV by threefold and twofold, respectively. In addition, in some certain areas, the maximum As and Cr contents in rice and wheat could reach 0.63 and 0.96 mg/kg and 1.63 and 4.50 mg/kg, which were much higher than their FCVs. The higher accumulation of heavy metals in crops was generally in accordance with the spatial distribution of soil heavy metals that were mainly distributed in Xinghua City, Taixing City and Jiangyan District, indicating the intensive industrial or anthropogenic activities in these regions. Influenced by the metal's species, bioavailability and enrichment mechanism and the transport capacity of the crop, Cr, Cd and As in rice and Pb and Hg in wheat exceeded the safety thresholds in some areas, even though the relevant metal's contents in soils were within the safety control standards. According to the statistical analysis, 8.94% of the arable land possessed light metal pollution, which resulted in 3.18% of the area of rice and 4.0% of the area of wheat suffering from severe pollution with the excessive accumulation of Cr, Pb and Cd. Furthermore, in consideration of both soil and crop qualities, the overall environmental quality of the production area in Taizhou was suggested to be preferable; only 2.16% of the area maintained poor quality. Thus, regular monitoring, source control and integrated environmental management that focuses on heavy metals should be continued to guarantee the sustainability of local agriculture.

**Author Contributions:** Project administration and funding acquisition, X.M.; formal analysis, data curation and writing, J.S., X.J., H.Y. and S.Z.; writing—review and editing, H.S.; supervision, conceptualization, methodology and validation, Y.A.H. All authors have read and agreed to the published version of the manuscript.

**Funding:** This work was financially supported by the "Youth Program of National Natural Science Foundation of China" (51809076) and the "Fundamental Research Funds for the Central Universities" (2019B08514).

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Informed consent was obtained from all subjects involved in this study.

**Data Availability Statement:** The data presented in this study are available on request from the corresponding author.

**Acknowledgments:** We are grateful to all the patients who participated in this study, to the technicians from Taizhou Research Institute of Nanjing Agricultural University for the assistance and to the management and staff of Taizhou Science and Technology Bureau for their cooperation and support.

**Conflicts of Interest:** The authors declare no conflict of interest. The funders had no role in the design of the study, in the collection, analyses or interpretation of the data, in the writing of the manuscript or in the decision to publish the results.

## References

1. Seleiman, M.F.; Santanen, A.; Mäkelä, P.S.A. Recycling sludge on cropland as fertilizer–Advantages and risks. *Resour. Conserv. Recy.* **2020**, *155*, 104647. [CrossRef]
2. Hubbard, A. Heavy metals in the environment: Origin, interaction and remediation. *J. Colloid Interface Sci.* **2005**, *291*, 307. [CrossRef]
3. Rehman, Z.U.; Junaid, M.F.; Ijaz, N.; Khalid, U.; Ijaz, Z. Remediation methods of heavy metal contaminated soils from environmental and geotechnical standpoints. *Sci. Total Environ.* **2023**, *867*, 161468. [CrossRef] [PubMed]
4. MEPC (Ministry of Environmental Protection of China). *National Communique of Soil Pollution Survey*; MEPC: Beijing, China, 2014. (In Chinese)
5. Wu, Q.M.; Hu, W.Y.; Wang, H.F.; Liu, P.; Wang, X.K.; Huang, B. Spatial distribution, ecological risk and sources of heavy metals in soils from a typical economic development area, Southeastern China. *Sci. Total Environ.* **2021**, *780*, 146557. [CrossRef] [PubMed]
6. Borris, M.; Österlund, H.; Marsalek, J.; Viklander, M. Contribution of coarse particles from road surfaces to dissolved and particle-bound heavy metal loads in runoff: A laboratory leaching study with synthetic stormwater. *Sci. Total Environ.* **2016**, *573*, 212–221. [CrossRef] [PubMed]
7. Xiang, M.T.; Li, Y.; Yang, J.Y.; Lei, K.G.; Li, Y.; Li, F.; Zheng, D.F.; Fang, X.Q.; Cao, Y. Heavy metal contamination risk assessment and correlation analysis of heavy metal contents in soil and crops. *Environ. Pollut.* **2021**, *278*, 116911. [CrossRef]
8. Marrugo-Negrete, J.; Pinedo-Hernández, J.; Díez, S. Assessment of heavy metal pollution, spatial distribution and origin in agricultural soils along the Sinú River Basin, Colombia. *Environ. Res.* **2017**, *154*, 380–388. [CrossRef]
9. Štrbac, S.; Ranđelović, D.; Gajica, G.; Hukić, E.; Stojadinović, S.; Veselinović, G.; Orlić, J.; Tognetti, R.; Kašanin-Grubin, M. Spatial distribution and source identification of heavy metals in European mountain beech forests soils. *Chemosphere* **2022**, *309 Pt 1*, 136662. [CrossRef]
10. Lazo, P.; Steinnes, E.; Qarri, F.; Allajbeu, S.; Kane, S.; Stafilov, T.; Frontasyeva, M.V.; Harmens, H. Origin and spatial distribution of metals in moss samples in Albania: A hotspot of heavy metal contamination in Europe. *Chemosphere* **2018**, *190*, 337–349. [CrossRef]
11. Gan, Y.D.; Huang, X.M.; Li, S.S.; Liu, N.; Li, Y.C.C.; Freidenreich, A.; Wang, W.X.; Wang, R.Q.; Dai, J.L. Source quantification and potential risk of mercury, cadmium, arsenic, lead, and chromium in farmland soils of Yellow River Delta. *J. Clean Prod.* **2019**, *221*, 98–107. [CrossRef]
12. Anaman, R.; Peng, C.; Jiang, Z.C.; Liu, X.; Zhou, Z.R.; Guo, Z.H.; Xiao, X.Y. Identifying sources and transport routes of heavy metals in soil with different land uses around a smelting site by GIS based PCA and PMF. *Sci. Total Environ.* **2022**, *823*, 153759. [CrossRef] [PubMed]
13. Hakanson, L. An ecological risk index for aquatic pollution control. A sedimentological approach. *Water Res.* **1980**, *14*, 975–1001. [CrossRef]
14. Ran, J.; Wang, D.; Wang, C.; Zhang, G.; Zhang, H. Heavy metal contents, distribution, and prediction in a regional soil-wheat system. *Sci. Total Environ.* **2016**, *544*, 422–431. [CrossRef] [PubMed]
15. Koner, S.; Chen, J.S.; Rathod, J.; Hussain, B.; Hsu, B.M. Unravelling the ultramafic rock-driven serpentine soil formation leading to the geo-accumulation of heavy metals: An impact on the resident microbiome, biogeochemical cycling and acclimatized eco-physiological profiles. *Sci. Total Environ.* **2023**, *216 Pt 4*, 114664. [CrossRef] [PubMed]

16. Liu, R.P.; Xu, Y.N.; Zhang, J.H.; Wang, W.K.; Elwardany, R.M. Effects of heavy metal pollution on farmland soils and crops: A case study of the Xiaoqinling Gold Belt, China. *China Geol.* **2020**, *3*, 402–410.
17. Napoletano, P.; Guezgouz, N.; Iorio, E.D.; Colombo, C.; Guerriero, G.; Marco, A.D. Anthropic impact on soil heavy metal contamination in riparian ecosystems of northern Algeria. *Chemosphere* **2023**, *313*, 137522. [CrossRef]
18. USEPA (United States Environmental Protection Agency). *Integrated Risk Information System*; USEPA: Washington, DC, USA, 2019.
19. Beinabaj, S.M.H.; Heydariyan, H.; Aleii, H.M.; Hosseinzadeh, A. Concentration of heavy metals in leachate, soil, and plants in Tehran's landfill: Investigation of the effect of landfill age on the intensity of pollution. *Heliyon* **2023**, *9*, e13017. [CrossRef]
20. Guo, G.H.; Li, K.; Lei, M. Accumulation, environmental risk characteristics and associated driving mechanisms of potential toxicity elements in roadside soils across China. *Sci. Total Environ.* **2022**, *835*, 155342. [CrossRef]
21. Lemessa, F.; Simane, B.; Seyoum, A.; Gebresenbet, G. Analysis of the concentration of heavy metals in soil, vegetables and water around the bole Lemi industry park, Ethiopia. *Heliyon* **2022**, *8*, e12429. [CrossRef]
22. Wan, M.X.; Hu, W.Y.; Wang, H.F.; Tian, K.; Huang, B. Comprehensive assessment of heavy metal risk in soil-crop systems along the Yangtze River in Nanjing, Southeast China. *Sci. Total Environ.* **2021**, *780*, 146567. [CrossRef]
23. Doabi, S.A.; Karami, M.; Afyuni, M.; Yeganeh, M. Pollution and health risk assessment of heavy metals in agricultural soil, atmospheric dust and major food crops in Kermanshah province, Iran. *Ecotoxicol. Environ. Saf.* **2018**, *163*, 153–164. [CrossRef] [PubMed]
24. Rai, P.K.; Sonne, C.; Kim, K.H. Heavy metals and arsenic stress in food crops: Elucidating antioxidative defense mechanisms in hyperaccumulators for food security, agricultural sustainability, and human health. *Sci. Total Environ.* **2023**, *874*, 162327. [CrossRef] [PubMed]
25. Wei, L.Y.; Li, Z.H.; Sun, J.T.; Zhu, L.Z. Pollution characteristics and health risk assessment of phthalate esters in agricultural soil and vegetables in the Yangtze River Delta of China. *Sci. Total Environ.* **2020**, *726*, 137978. [CrossRef]
26. Liu, M.Y.; Liu, D.Y.; Zhang, W.; Chen, X.X.; Zhao, Q.Y.; Chen, X.P.; Zou, C.Q. Health risk assessment of heavy metals (Zn, Cu, Cd, Pb, As and Cr) in wheat grain receiving repeated Zn fertilizers. *Environ. Pollut.* **2020**, *257*, 113581. [CrossRef]
27. Anderson, R.H.; Farrar, D.B.; Thoms, S.R. Application of discriminant analysis with clustered data to determine anthropogenic metals contamination. *Sci. Total Environ.* **2009**, *408*, 50–56. [CrossRef]
28. Puthusseri, R.M.; Nair, H.P.; Johny, T.K.; Bhat, S.G. Insights into the response of mangrove sediment microbiomes to heavy metal pollution: Ecological risk assessment and metagenomics perspectives. *J. Environ. Manag.* **2021**, *298*, 113492. [CrossRef] [PubMed]
29. Dong, B.; Zhang, R.Z.; Gan, Y.D.; Cai, L.Q.; Freidenreich, A.; Wang, K.P.; Guo, T.W.; Wang, H.B. Multiple methods for the identification of heavy metal sources in cropland soils from a resource-based region. *Sci. Total Environ.* **2019**, *651*, 3127–3138. [CrossRef]
30. Liu, C.; Wei, B.K.; Bao, J.S.; Wang, Y.; Hu, J.C.; Tang, Y.E.; Chen, T.; Jin, J. Polychlorinated biphenyls in the soil–crop–atmosphere system in e-waste dismantling areas in Taizhou: Concentrations, congener profiles, uptake, and translocation. *Environ. Pollut.* **2020**, *257*, 113622. [CrossRef]
31. Xu, L.; Dai, H.P.; Skuza, L.; Xu, J.M.; Shi, J.C.; Shentu, J.L.; Wei, S.H. Integrated survey on the heavy metal distribution, sources and risk assessment of soil in a commonly developed industrial area. *Ecotoxicol. Environ. Saf.* **2022**, *236*, 113462. [CrossRef]
32. Chuai, X.W.; Huang, X.J.; Wang, W.J.; Zhang, M.; Lai, L.; Liao, Q.L. Spatial variability of soil organic carbon and related factors in Jiangsu Province, China. *Pedosphere* **2012**, *22*, 404–414. [CrossRef]
33. Oduor, A.M.O.; Yang, B.F.; Li, J.M. Alien ornamental plant species cultivated in Taizhou, southeastern China, may experience greater range expansions than native species under future climates. *Glob. Ecol. Conserv.* **2023**, *41*, e02371. [CrossRef]
34. DARAJP (Department of Agriculture and Rural Affairs of Jiangsu Province). *Food Production Announcement*; DARAJP: Nanjing, China, 2020. (In Chinese)
35. Batool, F.; Hussain, M.I.; Nazar, S.; Bashir, H.; Khan, Z.I.; Ahmad, K.; Alnuwaiser, M.A.; Yang, H.H. Potential of sewage irrigation for heavy metal contamination in soil–wheat grain system: Ecological risk and environmental fate. *Agr. Watter Manage.* **2023**, *278*, 108144. [CrossRef]
36. Kaya, F.; Başayiğit, L.; Keshavarzi, A.; Francaviglia, R. Digital mapping for soil texture class prediction in northwestern Türkiye by different machine learning algorithms. *Geoderma Reg.* **2022**, *31*, e00584. [CrossRef]
37. MAC (Ministry of Agriculture of China). *Technical Specification for Monitoring Farmland Soil Environmental Quality (NY/T 395–2012)*; MAC: Beijing, China, 2012. (In Chinese)
38. MAC (Ministry of Agriculture of China). *Technical Specification for Contamination Monitoring of Agricultural Products (NY/T 398–2000)*; MAC: Beijing, China, 2000. (In Chinese)
39. Duane, M.J.; Facchetti, S.; Pigozzi, G. Site characterization of polluted soils and comparison of screening techniques for heavy metals by mobile ICP-MS (fixed laboratory) and EDXRF (fixed laboratory). *Sci. Total Environ.* **1996**, *177*, 195–214. [CrossRef]
40. Abdelhafiz, M.A.; Liu, J.; Jiang, T.; Pu, Q.; Aslam, M.W.; Zhang, K.; Meng, B.; Feng, X.B. DOM influences Hg methylation in paddy soils across a Hg contamination gradient. *Environ. Pollut.* **2023**, *322*, 121237. [CrossRef]
41. Li, Y.Y.; Wang, Y.J.; Zhang, Q.J.; Hu, W.J.; Zhao, J.T.; Chen, Y.H.; Zhong, H.; Wang, G.; Zhang, Z.Y.; Gao, Y.X. Elemental sulfur amendment enhance methylmercury accumulation in rice (*Oryza sativa* L.) grown in Hg mining polluted soil. *J. Hazard. Mater.* **2019**, *379*, 120701. [CrossRef]
42. MEEC (Ministry of Ecological Environment of China). *Risk Control Standard for Soil Contamination of Agricultural Land (GB15618-2018)*; MEE: Beijing, China, 2018. (In Chinese)

43. AFDC (Administration of Food and Drug of China). *Limit for Contaminants in Food (GB2762-2017)*; AFDC: Beijing, China, 2017. (In Chinese)
44. SCOC (Soil Census Office of China). *Second National Soil Survey of China*; SCOC: Beijing, China, 2009. (In Chinese)
45. Soleimani, H.; Mansouri, B.; Kiani, A.; Omer, A.K.; Tazik, M.; Ebrahimzadeh, G.; Sharafi, K. Ecological risk assessment and heavy metals accumulation in agriculture soils irrigated with treated wastewater effluent, river water, and well water combined with chemical fertilizers. *Heliyon* **2023**, *9*, e14580. [CrossRef]
46. Zhang, H.H.; Chen, J.J.; Zhu, L.; Li, F.B.; Wu, Z.F.; Yu, W.M.; Liu, J.M. Spatial patterns and variation of soil cadmium in Guangdong Province, China. *J. Geochem. Explor.* **2011**, *109*, 86–91. [CrossRef]
47. Zhou, X.Y.; Wang, X.R. Cd contamination status and cost-benefits analysis in agriculture soils of Yangtze River basin. *Environ. Pollut.* **2019**, *254*, 112962. [CrossRef]
48. Fei, X.F.; Xiao, R.; Christakos, G.; Langousis, A.; Ren, Z.Q.; Tian, Y.; Lv, X.N. Comprehensive assessment and source apportionment of heavy metals in Shanghai agricultural soils with different fertility levels. *Ecol. Indic.* **2019**, *106*, 105508. [CrossRef]
49. Li, X.Y.; Zhang, J.R.; Gong, Y.W.; Yang, S.H.; Ye, M.; Yu, X.; Ma, J. Status of mercury accumulation in agricultural soils across China (1976–2016). *Ecotoxicol. Environ. Saf.* **2020**, *197*, 110564. [CrossRef] [PubMed]
50. Liu, S.J.; Wang, X.D.; Guo, G.L.; Yan, Z.G. Status and environmental management of soil mercury pollution in China: A review. *J. Environ. Manage.* **2021**, *277*, 111442. [CrossRef] [PubMed]
51. Zhu, K.W.; Chen, Y.C.; Zhang, S.; Yang, Z.M.; Huang, L.; Lei, B.; Li, L.; Zhou, Z.B.; Xiong, H.L.; Li, X.X. Identification and prevention of agricultural non-point source pollution risk based on the minimum cumulative resistance model. *Global Ecol. Conserv.* **2020**, *23*, e01149. [CrossRef]
52. Gmochowska, W.; Pietranik, A.; Tyszka, R.; Ettler, V.; Mihaljevič, M.; Długosz, M.; Walenczak, K. Sources of pollution and distribution of Pb, Cd and Hg in Wrocław soils: Insight from chemical and Pb isotope composition. *Geochemistry* **2019**, *79*, 434–445. [CrossRef]
53. Pescatore, A.; Grassi, C.; Rizzo, A.M.; Orlandini, S.; Napoli, M. Effects of biochar on berseem clover (*Trifolium alexandrinum*, L.) growth and heavy metal (Cd, Cr, Cu, Ni, Pb, and Zn) accumulation. *Chemosphere* **2022**, *287 Pt 1*, 131986. [CrossRef]
54. Bur, T.; Crouau, Y.; Bianco, A.; Gandois, L.; Probst, A. Toxicity of Pb and of Pb/Cd combination on the springtail Folsomia candida in natural soils: Reproduction, growth and bioaccumulation as indicators. *Sci. Total Environ.* **2012**, *414*, 187–197. [CrossRef]
55. Park, J.H. Contrasting effects of Cr(III) and Cr(VI) on lettuce grown in hydroponics and soil: Chromium and manganese speciation. *Environ. Pollut.* **2020**, *266 Pt 2*, 115073. [CrossRef]
56. Deng, M.H.; Malik, A.; Zhang, Q.; Sadeghpour, A.; Zhu, Y.W.; Li, Q.R. Improving Cd risk managements of rice cropping system by integrating source-soil-rice-human chain for a typical intensive industrial and agricultural region. *J. Clean. Prod.* **2021**, *313*, 127883. [CrossRef]
57. Rashid, I.; Murtaza, G.; Dar, A.A.; Wang, Z.Y. The influence of humic and fulvic acids on Cd bioavailability to wheat cultivars grown on sewage irrigated Cd-contaminated soils. *Ecotoxicol. Environ. Saf.* **2020**, *205*, 111347. [CrossRef]
58. Cai, Y.M.; Wang, M.E.; Chen, B.D.; Chen, W.P.; Xu, W.B.; Xie, H.W.; Long, Q.Z.; Cai, Y.H. Effects of external $Mn^{2+}$ activities on *OsNRAMP5* expression level and Cd accumulation in indica rice. *Environ. Pollut.* **2020**, *260*, 113941. [CrossRef]
59. Bari, M.A.; El-Shehawi, A.M.; Elseehy, M.M.; Naheen, N.N.; Rahman, M.M.; Kabir, A.H. Molecular characterization and bioinformatics analysis of transporter genes associated with Cd-induced phytotoxicity in rice (*Oryza sativa* L.). *Plant Physiol. Bioch.* **2021**, *167*, 438–448. [CrossRef]
60. Riaz, M.; Kamran, M.; Rizwan, M.; Ali, S.; Parveen, A.; Malik, Z.; Wang, X.R. Cadmium uptake and translocation: Selenium and silicon roles in Cd detoxification for the production of low Cd crops: A critical review. *Chemosphere* **2021**, *273*, 129690. [CrossRef] [PubMed]
61. Li, C.; Sanchez, G.M.; Wu, Z.F.; Cheng, J.; Zhang, S.Y.; Wang, Q.; Li, F.B.; Sun, G.; Meentemeyer, R.K. Spatiotemporal patterns and drivers of soil contamination with heavy metals during an intensive urbanization period (1989–2018) in southern China. *Environ. Pollut.* **2020**, *260*, 114075. [CrossRef] [PubMed]
62. Bidar, G.; Pelfrêne, A.; Schwartz, C.; Waterlot, C.; Sahmer, K.; Marot, F.; Douay, F. Urban kitchen gardens: Effect of the soil contamination and parameters on the trace element accumulation in vegetables–A review. *Sci. Total Environ.* **2020**, *738*, 139569. [CrossRef] [PubMed]
63. Feng, W.L.; Guo, Z.H.; Xiao, X.Y.; Peng, C.; Shi, L.; Ran, H.Z.; Xu, W.X. Atmospheric deposition as a source of cadmium and lead to soil-rice system and associated risk assessment. *Ecotoxicol. Environ. Saf.* **2019**, *180*, 60–167. [CrossRef] [PubMed]
64. Rahman, S.U.; Wang, X.J.; Shahzad, M.; Bashir, O.; Li, Y.L.; Cheng, H.F. A review of the influence of nanoparticles on the physiological and biochemical attributes of plants with a focus on the absorption and translocation of toxic trace elements. *Environ. Pollut.* **2022**, *310*, 119916. [CrossRef] [PubMed]
65. Ahmed, R.S.; Abuarab, M.E.; Ibrahim, M.M.; Baioumy, M.; Mokhtar, A. Assessment of environmental and toxicity impacts and potential health hazards of heavy metals pollution of agricultural drainage adjacent to industrial zones in Egypt. *Chemosphere* **2023**, *318*, 137872. [CrossRef] [PubMed]
66. Viczek, S.A.; Aldrian, A.; Pomberger, R.; Sarc, R. Origins and carriers of Sb, As, Cd, Cl, Cr, Co, Pb, Hg, and Ni in mixed solid waste–A literature-based evaluation. *Waste Manag.* **2020**, *103*, 87–112. [CrossRef]
67. Wu, B.; Li, L.L.; Guo, S.H.; Li, Y. Source apportionment of heavy metals in the soil at the regional scale based on soil-forming processes. *J. Hazard. Mater.* **2023**, *448*, 130910. [CrossRef]

68. Mehmood, A.; Mirza, M.A.; Choudhary, M.A.; Kim, K.H.; Raza, W.; Raza, N.; Lee, S.S.; Zhang, M.; Lee, J.H.; Sarfraz, M. Spatial distribution of heavy metals in crops in a wastewater irrigated zone and health risk assessment. *Environ. Res.* **2019**, *168*, 382–388. [CrossRef]
69. Zhang, J.; Yang, R.D.; Li, Y.C.; Peng, Y.S.; Wen, X.F.; Ni, X.R. Distribution, accumulation, and potential risks of heavy metals in soil and tea leaves from geologically different plantations. *Ecotoxicol. Environ. Saf.* **2020**, *195*, 110475. [CrossRef] [PubMed]
70. Vaidya, B.P.; Hagmann, D.F.; Haramuniz, J.; Krumins, J.A.; Goodey, N.M. Artificial root exudates restore microbial functioning in a metal contaminated, barren, inactive soil. *Environ. Pollut.* **2022**, *312*, 120007. [CrossRef] [PubMed]
71. Pan, Y.P.; Wang, Y.S. Atmospheric wet and dry deposition of trace elements at 10 sites in Northern China. *Atmos. Chem. Phys.* **2015**, *15*, 951–972. [CrossRef]
72. Sharma, P.; Tripathi, S.; Sirohi, R.; Kim, S.H.; Ngo, H.H.; Pande, A. Uptake and mobilization of heavy metals through phytoremediation process from native plants species growing on complex pollutants: Antioxidant enzymes and photosynthetic pigments response. *Environ. Technol. Innov.* **2021**, *23*, 101629. [CrossRef]
73. MLRC (Ministry of Land and Resources of China). *Communique on the Quality and Grade of Cultivated Land in China*; MLRC: Beijing, China, 2019. (In Chinese)
74. Lavado, R.S.; Porcelli, C.A.; Alvarez, R. Nutrient and heavy metal concentration and distribution in corn, soybean and wheat as affected by different tillage systems in the Argentine Pampas. *Soil Tillage Res.* **2001**, *62*, 55–60. [CrossRef]
75. Kim, H.T.; Lee, T.G. A simultaneous stabilization and solidification of the top five most toxic heavy metals (Hg, Pb, As, Cr, and Cd). *Chemosphere* **2017**, *178*, 479–485. [CrossRef] [PubMed]
76. Tian, K.; Wu, Q.M.; Liu, P.; Hu, W.Y.; Huang, B.; Shi, B.; Zhou, Y.Q.; Kwon, B.O.; Choi, K.S.; Ryu, J.S.; et al. Ecological risk assessment of heavy metals in sediments and water from the coastal areas of the Bohai Sea and the Yellow Sea. *Environ. Int.* **2020**, *136*, 105512. [CrossRef]


**Disclaimer/Publisher's Note:** The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.