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Current Situation of Agricultural Soil Pollution in Jiangsu Province: A Meta-Analysis

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Abstract: In recent years, heavy metal contamination in agricultural soils in Jiangsu Province has attracted more and more attention. However, most studies have been characterized by their small scale, few samples, and short-term monitoring. The overall status and temporal accumulation characteristics of heavy metals have not been fully reflected. Therefore, this paper attempted to provide a comprehensive analysis of the current status of heavy metals and provide accurate information for soil pollution management in Jiangsu Province. This paper collected available data in the literature (1993–2021) on heavy metal-polluted agricultural soils in Jiangsu Province. Based on these available data, the weighted mean values of each heavy metal were obtained by meta-analysis. Then, the ecological risks in soils were evaluated and spatiotemporal variations in the accumulation of heavy metals were explored. In addition, suggestions for pollution prevention and control were made by predicting future heavy metal concentrations. The results showed that Cd and Hg were the major polluting elements in Jiangsu Province. The spatial enrichment of heavy metals followed the pattern of southern > northern > central. Heavy metal concentrations in Nanjing, Suzhou, and Xuzhou should be paid special attention. The ecological risk level of heavy metals in agricultural soils in Jiangsu Province was high, predominantly contributed by Hg and Cd. The accumulation of most heavy metals gradually decreased after 2010, while the opposite happened with Cd. Jiangsu Province should continue to take active pollution control measures in order to maintain the decreasing trend of heavy metal concentrations in farmland soils. This study could provide a scientific and theoretical basis for the development of pollution control and soil remediation measures.

Keywords: heavy metal pollution; meta-analysis; spatiotemporal accumulation trend; pollution control measures

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1. Introduction

The soil environment is facing the negative effects of rapid industrialization and urbanization [1]. Heavy metal pollution of agricultural soils has become one of the common environmental problems faced worldwide [2–4]. A wide range of heavy metal pollutants from rock weathering, industrial activities, mineral smelting, transport, and agricultural activities have been absorbed into worldwide farmland soils [3,5–7]. The physical and chemical properties, and the biological characteristics of the soil have been negatively affected by heavy metal contaminants. The severe heavy metal contamination could endanger the stability of soil ecosystem function and structure, and destroy species richness and diversity [8,9]. In addition, heavy metals could be absorbed into the human body through oral, skin, or respiratory contact, and endanger human health through replacement and synergistic reactions [10]. China is a typical agricultural country with insufficient per capita farmland and reserve arable land resources [11]. The National Soil Pollution Census Report published in 2014 showed that over 19% of farmland soil sites in China were polluted, and the primary pollutants were Cd, Cu, and Hg. It was estimated that more than 10 million tons of food in China were contaminated by heavy metals, resulting in economic losses of over CNY 20 billion [2,3]. Therefore, the harm caused by farmland soil heavy metals should not be underestimated.

A large number of studies on soil heavy metal pollution have been carried out to date in China. However, due to limitations of human and material resources, the studies on soil heavy metals have been mostly concentrated in typical polluted areas or a limited range of sampling sites, such as the soil pollution study in an industrial park (including 62 samples) [1], the study on soil pollution in agricultural fields of Pb–Zn smelters (25 samples were collected) [12]. The large-scale heavy metal studies have mostly focused on a single heavy metal element, such as Hg [13], and Cd [14]. Furthermore, most studies on soil heavy metal pollution lack long-term monitoring to reflect changes in heavy metal accumulation over time [15,16]. These limitations hinder the researchers in forming a comprehensive understanding of the spatiotemporal variations and driving factors of heavy metal pollution in farmland soils. Therefore, characterizing the spatiotemporal variations of multiple heavy metals in agricultural soil is an urgent problem to be solved.

Meta-analysis is a retrospective empirical approach that allows data to be extracted from a large number of examples in the literature and results to be integrated and compared [17]. Due to its advantages of integrity and reliability, it is widely used in many fields, such as economics, medicine, psychology, etc. [18–20]. In recent years, meta-analyses have been gradually applied to describe the relationship between pollutants and the environment [21–24]. Huang et al. (2019) reviewed the studies from 2005 to 2017 and mapped the spatiotemporal distribution of soil heavy metals in different cropping systems at the provincial level in China [4]. Li et al. (2014) used a meta-analysis method to collect available data from 72 mining areas in China during 2005–2012, to identify heavy metal mining and pollution areas that need to be controlled urgently [22]. She et al. (2021) successfully evaluated the current situation and temporal accumulation trends of soil heavy metal contamination in the Yangtze River Delta region using meta-analysis [25]. All these studies demonstrated that meta-analysis could collect reliable studies and data from a large amount of literature and overcome the limitations of large-scale studies on soil heavy metal contamination. Furthermore, the combination of meta-analysis and subgroup analysis could effectively characterize the spatial and temporal variation of heavy metals.

Jiangsu Province is located in the Yangtze River Delta economic belt and is densely populated [26]. Jiangsu is industrially developed, with a large number of metal smelting, chemical, pharmaceutical, microelectronics, and other factories [27,28]. Obviously, the ecological environment has been seriously disturbed by human activities [4]. Jiangsu Province has a long history of planting wheat, rice, maize, and other crops. The agricultural soil heavy metal pollution has attracted the attention of an increasing number of researchers [25,29]. However, most studies of the pollution in agricultural soils in Jiangsu have focused on the pollution status of single cities or areas close to pollution sources, such as Nanjing, Wuxi, and Kunshan [30–32]. In addition, there have been few studies on the long-term monitoring of soil heavy metals across Jiangsu Province.

Therefore, this paper conducted a meta-analysis based on 129 primary publications on agricultural soil pollution in Jiangsu Province published from 1993 to 2021, to evaluate the current status and spatiotemporal accumulation trend of heavy metal pollution. The main purposes of this study were: (1) to evaluate the pollution status and ecological risk level of Jiangsu agricultural soil heavy metals; (2) to analyze the spatial and temporal distribution of agricultural soil heavy metal enrichment; (3) to explore the influencing factors of the spatial and temporal distribution of agricultural soil heavy metals. This study can provide a scientific theoretical basis for pollution source control and soil remediation in Jiangsu Province. In addition, the results of this study may also have valuable implications for soil pollution research in similar rapidly urbanizing and industrializing areas, such as Zhejiang Province and the Pearl River Delta.

2. Materials and Methods

2.1. Study Area

Jiangsu Province is situated in the lower reaches of the Yangtze River and the Huaihe River, between 116°18′–121°57′ E and 30°45′–35°20′ N (Figure 1). The average annual temperature in Jiangsu is 13–17 °C with a frost-free period of 200–240 days a year. The average annual precipitation is 800–1300 mm, decreasing from south to north. Jiangsu Province provides favorable natural conditions and an economic base, and it is an important agricultural production area in China. In 2020, the total sown area of farm crops in Jiangsu Province reached 77,400 km^2. Most of the crops in Jiangsu Province are two-harvests-a-year crops, and the grain crops are mainly rice and wheat, followed by corn and sorghum. Jiangsu Province spans three natural zones from north to south, with a warm temperate zone, a northern subtropical zone, and a central subtropical zone [33]. The soil types of the three natural zones include brown loam, yellow-brown loam, brown-red loam, swampy soil, paddy soil, etc. The seven geomorphic units are divided from northern to southern Jiangsu, and the distribution of soil heavy metal background values in different geomorphic units showed significant differences [33,34].

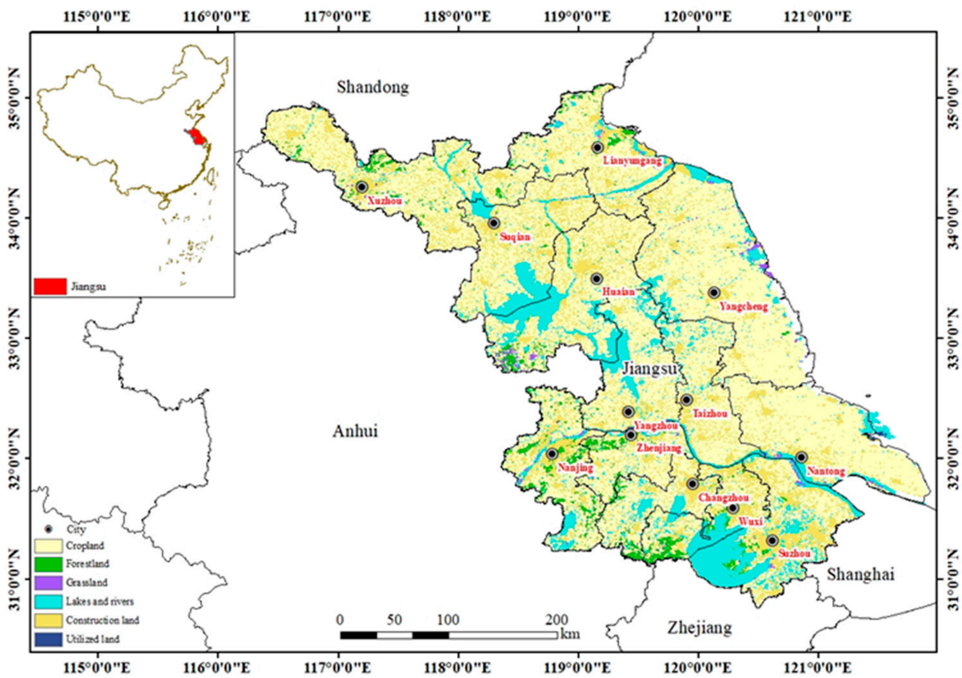

Figure 1. Location of the study area.

2.2. Literature Selection

A comprehensive search was performed of primary research publications published between 1993 and 2021 that reported heavy metals in agricultural soils in Jiangsu Province. The bibliographic searches were carried out in the Web of Science (WOS) and China National Knowledge Infrastructure (CNKI) databases, using the keywords "toxic element", "heavy metal", or individual elements (Cd, Cr, Cu, Hg, Ni, As, Pb, and Zn) and "agricultural soil", or "farmland soil" and "Jiangsu", or individual cities (Nanjing, Suzhou, Wuxi, Zhenjiang, Yangzhou, Nan-tong, Taizhou, Changzhou, Xuzhou, Yancheng, Huaian, Suqian, and Lianyungang).

To be included in subsequent analyses, these preliminary studies had to meet the following criteria: (i) only field experiments monitoring topsoil (0–20 cm, or 0–15 cm) in Jiangsu agricultural soils were selected; (ii) selected studies should indicate the number of sampling sites and the size of the research area; (iii) soil sample collection complied with technical specifications (NY/T395-2012; HJ/T166-2004). The distribution of sample points should be representative and uniform. Sampling should follow the principle of equal amount and multi-point mixing; (iv) acceptable methods should be used for soil sample preparation and analysis. The quality control and insurance processes should be included in the analysis. Soils should be digested with mixed acids (e.g., HNO_3-$HClO_4$-HF, HNO_3-$HClO_4$-HCL, or HCl-HNO_3-HF-$HClO_4$), and samples determined by ICP-MS, ICP-AES, ICPOES, CAAS, or AAS. The sample duplicates, reagent blanks, and standard controls should be included, and the limits of determination and recoveries reported; (v) the means, standard deviations, and ranges of samples can be extracted directly from the graphs, tables, or calculated from the primary studies.

2.3. Meta-Analysis

2.3.1. Data Extraction

The data extracted from each article include: (i) article information (title, first author, and year of publication); (ii) the location (address, administrative division, size of the study area, and the number of samples); (iii) emission sources (according to the location of the study area and the anthropogenic sources mentioned in the article), wherein the area was divided into "NF (normal farmland)", and "RF (risk farmland)" for further subgroups analysis; (iv) cropping system, categorized as paddy, upland, vegetable, and other; (v) the summary statistics of contaminations (mean, standard deviation, range, coefficient of variation, the median of Cd, Cr, Cu, Hg, Ni, As, Pb, and Zn). The extracted data are presented in Supplementary 2.

2.3.2. Heterogeneity and Sensitivity Analysis

The extracted data were preliminarily organized, and the missing data were filled in (standard deviation, size of the study area, etc.). Due to the variability of data collection regions and analysis methods, follow-up analysis cannot be carried out immediately after finishing data extraction. To ensure the statistical significance of the results, the collated data were divided into eight element files for extreme values detection. Publication bias and low-quality studies could introduce outliers into the database. Publication bias means that researchers focused more on soil status in typical contaminated areas, such as around industrial parks or mines. These results were easily prioritized for publication, but outliers could still have been added to the database [4]. The extreme values of outliers may distort the weighted mean results in the study [35]. Subgroup analyses in a meta-analysis are usually used to manage publication bias. Sensitivity analysis can be used to trim and filter out the outliers. The outlier determination selected common diagnostic indicators in this paper, such as studentized deleted residual, DFFITS, Cook's distance, and COVRATIO [36].

2.3.3. Weighted Mean Calculation

In the study area, soil samples are usually collected according to certain rules and then heavy metal concentrations are determined by the same assay method. The arithmetic mean is calculated to represent the mean value of heavy metal content. The amount of information contained in the heavy metal concentration data obtained by this determination method is essentially the same. However, the amount of information carried in the data is inconsistent between different studies due to different sampling methods and determination methods. The spatial distribution of the heavy metal concentration data is also uneven. The arithmetic average obtained by directly calculating these data may result in a large deviation from the true level. The basic idea of meta-analysis is to assign different weight coefficients to the data from different studies to balance the degree of influence of each study on the calculation. This makes the calculated final effect value closer to the true level [37]. The effect value in meta-analysis is a weighted mean value [4]. There are two models to determine weights, the fixed-effects model and the random-effects model [38]. In the fixed model, the weights are the inverse of the variance. Since there are unavoidable differences in environmental conditions and methods, heterogeneity is introduced into the random-effect model to balance variation, and weight is calculated by "heterogeneity" and variance. Based on the characteristics of the field experiment, the study area, the number of sampling points, and the variance are essential reference values for weight calculations. It can be established that the data from a large research area with more sampling sites and smaller variances are a more reliable basis for conclusions and should be given bigger weight. According to this, the weight Wi can be calculated as [4]:

$$W_i = A_i \times \frac{N_i}{Sd_i} \tag{1}$$

where Wi is the weight of each individual study, A_i, N_i, and Sd_i are the size of the study area, the number of soil samples, and the standard deviation of the heavy metals in each study, respectively.

Then the weighted mean content (C) refers to:

$$C = C_i \times \frac{W_i}{\sum_{i=1}^{n} W_i} \tag{2}$$

where C is the weighted mean, and C_i and W_i represent the calculated mean concentrations of individual heavy metals and weight in each study, respectively. The distribution of weights showed non-normality and appeared skewed (Supplementary 1, Table S1). This could lead to over-reliance on studies with extremely high weights in calculating the weighted mean. To solve the problem of high weights affecting the fitting mean, the natural logarithm of the weight was calculated using Equation (3). After logarithm conversion, the distribution of transformed weights (W_i^*) was closer to the normal distribution (Supplementary 1, Table S1). The logarithm weighted mean (C^*) should be recalculated with W_i^* according to Equation (4).

$$W_i^* = \lg\left(A_i \times \frac{N_i}{Sd_i}\right) \tag{3}$$

$$C^* = C_i \times \frac{W_i^*}{\sum_{i=1}^{n} W_i^*} \tag{4}$$

where W_i^* and C^* are the natural logarithm of the weight and recalculated mean, respectively. A_i, N_i, Sd_i, and C_i have the same meaning as in Equations (1) and (2).

Data statistical analysis and management were performed with Microsoft Excel 2010 (v.2010, Microsoft Corporation, Redmond, WA, USA). Spatial analysis and mapping were conducted with ArcGIS (v10.3, ESRI Inc., Redlands, CA, USA). Meta-analysis was performed in RStudio with the Metafor package (v.3.5.3, AT&T, Murray, NJ, USA) [36].

3. Results

3.1. Study Area Publication Bias and Sensitivity Analysis

The data heterogeneity test showed that the concentration distributions of all elements presented non-normal distributions. The preliminary exclusion was performed for the extreme values caused by the low data accuracy of studies, and the result was recalculated. Figure S1 shows the result of outlier selection by sensitivity analysis (refer to the Supplementary 1). The data were divided into "NF (normal farmland)", and "RF (risk farmland)" for subgroup analysis, according to the existence of pollution sources around sampling sites. Table S2 showed the result of the weighted average calculated by the "all data" (all data collected from the publications), "EO (all data excluding outliers)", "NF (normal farmland)", and "RF (risk farmland)" in the Supplementary 1. By comparing the calculation results of each group, it was found that the weighted means of all elements calculated for the "RF" group were higher than those of the "NF" group, which means that publication bias does exist in this study. Since the contents were higher in the "RF" group, the "all data" group has a higher value than the "NF" and "EO" groups, especially for Cd, Hg, and Zn. However, there were no remarkable differences observed between the results of the "EO" and "NF" groups. It can be believed that follow-up analysis using data from the "EO" group can provide reliable results for the overall situation in this study. Therefore, sensitivity analysis could effectively test outliers in this study, and subgroup analysis could also reduce publication bias.

3.2. Overall Status of Heavy Metals in Agricultural Soil

The summary of eight heavy metals in agricultural soil in Jiangsu is shown in Table 1. The most studies were conducted on the pollution of Pb (121), Cd (119), and Cr (104), and the fewest studies were on Zn (88) and Ni (58). The weighted mean values calculated in the "EO" group provided the overall status of heavy metals in agricultural soil in Jiangsu. Compared with the Jiangsu agricultural soil background values, the concentration of Hg and Cd were six and two times higher than their background values, showing the highest degree of accumulation, followed by Cu, Pb, and Zn. The contents of Cr and As were below their background values. In addition, only As was still within the safe range of the national soil background value (CNEMC, 1990), while other elements exceeded this background value. However, compared with the national standard value of agricultural soil environmental quality (GB 15618-2018), only 6.47% of studies showed that the concentration of Cd exceeded the standard, while other elements did not exceed the standard.

Table 1. Statistics on the weighted mean values of heavy metals in Jiangsu agricultural soil (mg/kg).

	Cd	Cr	As	Cu	Hg	Ni	Pb	Zn
Number of studies sampled	119	104	100	100	93	58	121	88
Standard values [a]	0.6	300	25	100	0.5	50	140	250
All sampling number	97,659	54,047	57,044	83,628	57,969	64,265	60,563	83,238
Samples exceeded	6	0	0	0	0	0	0	0
Number of outliers	5	1	4	3	6	0	8	6
Weight mean values [b]	0.18	64.66	8.98	30.19	0.15	31.25	29.15	80.85
Jiangsu background values [c]	0.09	75.60	10.00	22.70	0.025	30.80	24.90	62.90
National background value [d]	0.10	61.00	11.20	22.60	0.07	26.98	26.00	74.26

[a]: Standard values 6.5 < pH < 7.5 (GB 15618-2018); [b]: weight values of heavy metals, with outliers deleted; [c]: Jiangsu background values [29]; [d]: the soil background value in China (CNEMC 1990).

3.3. The Spatial Distribution of Heavy Metals

In order to quantify the variations between regions and cities, subgroup analyses based on geographical location and city level were conducted (Table 2). More studies were carried out in southern Jiangsu (including Nanjing, Suzhou, Changzhou, Wuxi, and Zhenjiang), followed by the central region (Yangzhou, Nantong, and Taizhou), and the northern region (including Xuzhou, Yancheng, Suqian, Lianyungang, and Huaian). The eight high-concentration elements were prevalent in southern cities, while northern cities showed high concentrations of three elements (Cd, As, and Ni). The central region only showed a slight accumulation of heavy metals. In summary, the accumulation of heavy metals was most severe in southern Jiangsu Province. The result agreed with previous reports, that socio-economic activities were more prevalent in southern Jiangsu, and the enrichment of heavy metals was more significant [29].

Table 2. The weighted mean values (mg/kg) and potential ecological risk indexes of heavy metals in each city.

	Cd	Cr	As	Cu	Hg	Ni	Pb	Zn	RI
Suzhou	0.15	60.72	9.01	32.85	0.28	31.82	32.20	89.41	532.98
Wuxi	0.19	61.44	8.98	33.21	0.16	27.18	36.68	82.00	346.86
Changzhou	0.16	75.99	8.02	29.27	0.18	29.75	26.04	71.20	371.64
Nanjing	0.29	82.38	10.63	37.00	0.13	37.54	33.96	100.20	341.29
Zhenjiang	0.21	75.17	9.38	30.69	0.13	33.51	30.06	88.82	306.48
Southern	0.15	68.77	9.26	32.88	0.22	33.15	33.09	87.59	424.29
Nantong	0.19	53.78	7.24	21.97	0.14	23.13	27.29	73.90	299.67
Yangzhou	0.14	67.80	9.62	22.87	0.01	26.43	25.75	79.14	239.65
Taizhou	0.13	72.63	7.18	24.39	0.07	30.30	26.53	73.60	181.34
Central	0.16	59.24	7.90	11.16	0.10	26.30	26.47	77.60	240.78
Xuzhou	0.31	66.82	11.28	26.94	0.08	31.28	25.71	72.867	266.25
Lianyungang	0.14	58.61	9.65	31.52	0.10	39.98	29.29	76.700	243.24
Yancheng	0.14	67.27	8.00	22.08	0.04	30.61	20.57	72.51	141.68
Huaian	0.13	70.89	10.32	26.00	0.03	38.00	23.95	67.20	127.13
Suqian	0.09	75.10	12.00	23.80	0.02	34.90	24.70	59.50	87.93
Northern	0.29	64.75	10.29	27.06	0.09	33.14	25.59	69.81	265.47

The weighted mean of heavy metal contents in each urban area was compared with the background values of farmland soil in Jiangsu Province to better understand the accumulation of soil heavy metals in each city. The weighted mean values of eight elements calculated by subgroup analysis are shown in Figure 2 and Table 2. There were significant differences in the accumulation of farmland soil heavy metals among different cities. The average Cd content in surface soil in 12 cities (except Suqian) was higher than the Jiangsu background value. Xuzhou had the highest Cd content (0.31 mg/kg) in topsoil, while Suqian and Taizhou had the lowest Cd content (0.13 mg/kg). This result did not agree with previous reports claiming that Cd pollution was more severe in Nanjing [25]. The soil reports in Xuzhou were fewer than in Nanjing, and the uneven distribution of sampling sites may cause the discrepancy. The average Cr content in surface soil in two cities (Nanjing and Changzhou) was higher than the Jiangsu background value. Nanjing had the highest Cr content (82.38 mg/kg) in topsoil, while Nantong had the lowest Cr content. The average As content in surface soil in four cities (Nanjing, Xuzhou, Huaian, and Suqian) was higher than the Jiangsu background value. Suqian had the highest As content (12.00 mg/kg), while Taizhou had the lowest As content (7.18 mg/kg). The average Cu content in surface soil in 12 cities (all except Yancheng) was higher than the Jiangsu background value. Nanjing had the highest content of Cu (37.01 mg/kg), while Nantong had the lowest Cu content (21.98 mg/kg). The average Hg content in surface soil in 11 cities (all except Yangzhou and Suqian) was higher than the Jiangsu background value. Suzhou had the highest Hg content (0.29 mg/kg), while Yangzhou had the lowest Hg content (0.01 mg/kg). The average Ni content in surface soil in seven cities (all except Wuxi, Changzhou, Nantong, Yangzhou, Taizhou, and Yancheng) was higher than the Jiangsu background value. Lianyungang had the highest Ni content (39.98 mg/kg), while Nantong had the lowest Ni content (23.13 mg/kg). The average Pb content in surface soil in 10 cities (all except Yancheng, Suqian, and Huaian) was higher than the Jiangsu background value. Wuxi had the highest Pb concentration (36.68 mg/kg), while Suqian had the lowest content. The average Zn content in surface soil in 12 cities (all except Suqian) was higher than the Jiangsu background value. Nanjing had the highest Zn content (100.20 mg/kg), while Suqian had the lowest Zn content (59.50 mg/kg). Overall, the soil heavy metal concentrations in Jiangsu province showed a spatial trend of south Jiangsu > north Jiangsu > central Jiangsu.

Figure 2. The weighted mean values of heavy metals in different cities.

The ecological risks in each city and the entire Jiangsu were assessed using Hakanson's ecological risk index (Table 2). According to Hakanson's ecological risk grading rules (the methods are detailed in the Supplementary 1) [39], the ecological risk level of heavy metals in the agricultural soil of Jiangsu was high, predominantly due to Hg and Cd. The other six elements showed no significant ecological risk. The potential risk of heavy metals in the agricultural soils of Jiangsu showed a spatial trend of southern > northern > central (Figure 3). In terms of cities, Huaian, Suqian, and Yancheng showed a slight risk, five cities (Lianyungang, Xuzhou, Nantong, Yangzhou, and Taizhou) were medium risk, and five cities (Suzhou, Wuxi, Changzhou, Nanjing, and Zhenjiang) showed a high risk.

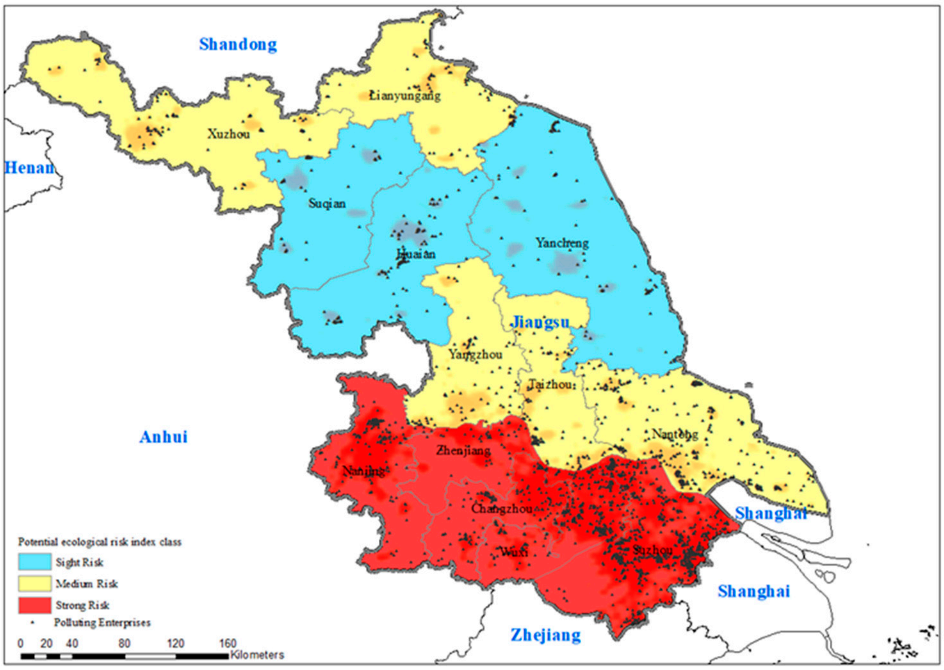

Figure 3. Spatial distribution of potential ecological risk index for heavy metals.

3.4. Temporal Variation of Heavy Metals

In order to observe the cumulative change over time from 1993 to 2021, an annual subgroup analysis was performed. The results showed that there were fluctuations in the accumulation of soil heavy metals over time, but the differences were not significant. Soil is the sink for various pollutants in the environment, and contaminants continue to accumulate when contaminant input consistently exceeds output [40]. Therefore, cumulative meta-analysis and linear regression explore the temporal trend in heavy metal accumulation (Figure 4). During the study period, the accumulation of Cu, Zn, and Hg continued to decrease, with Hg decreasing most significantly. The accumulation of Cd, Pb, Cr, Ni, and As fluctuated. From 1993 to 2021, the inputs of Cd, Hg, Pb, Zn, and Cu were greater than the outputs, whereas As, Cr, and Ni remained in balance with background values. The accumulation of Pb increased until 2010 and then decreased, while Cd did the opposite. Apparently, the cumulative change point is close to the year 2010.

The results of the annual subgroup analysis of the ecological risk index showed that the ecological risk in agricultural soil was mainly contributed by Hg and Cd during the study period in Jiangsu Province (Supplementary 1, Figure S2). The ecological risks of other elements (Cu, Pb, Cr, Zn, As, and Ni) were not significant. The ecological risk of Hg was extremely high, but the overall trend was downward. The risk level of Hg decreases from extremely high to high risk. The ecological risk of Cd showed an overall upward trend from slight to medium risk. The ecological risk of the other elements (Cu, Pb, Cr, Zn, As, and Ni) fluctuated but all were at slight risk level. Since 2010, the change trend of ecological risk of all heavy metal elements has gradually slowed down.

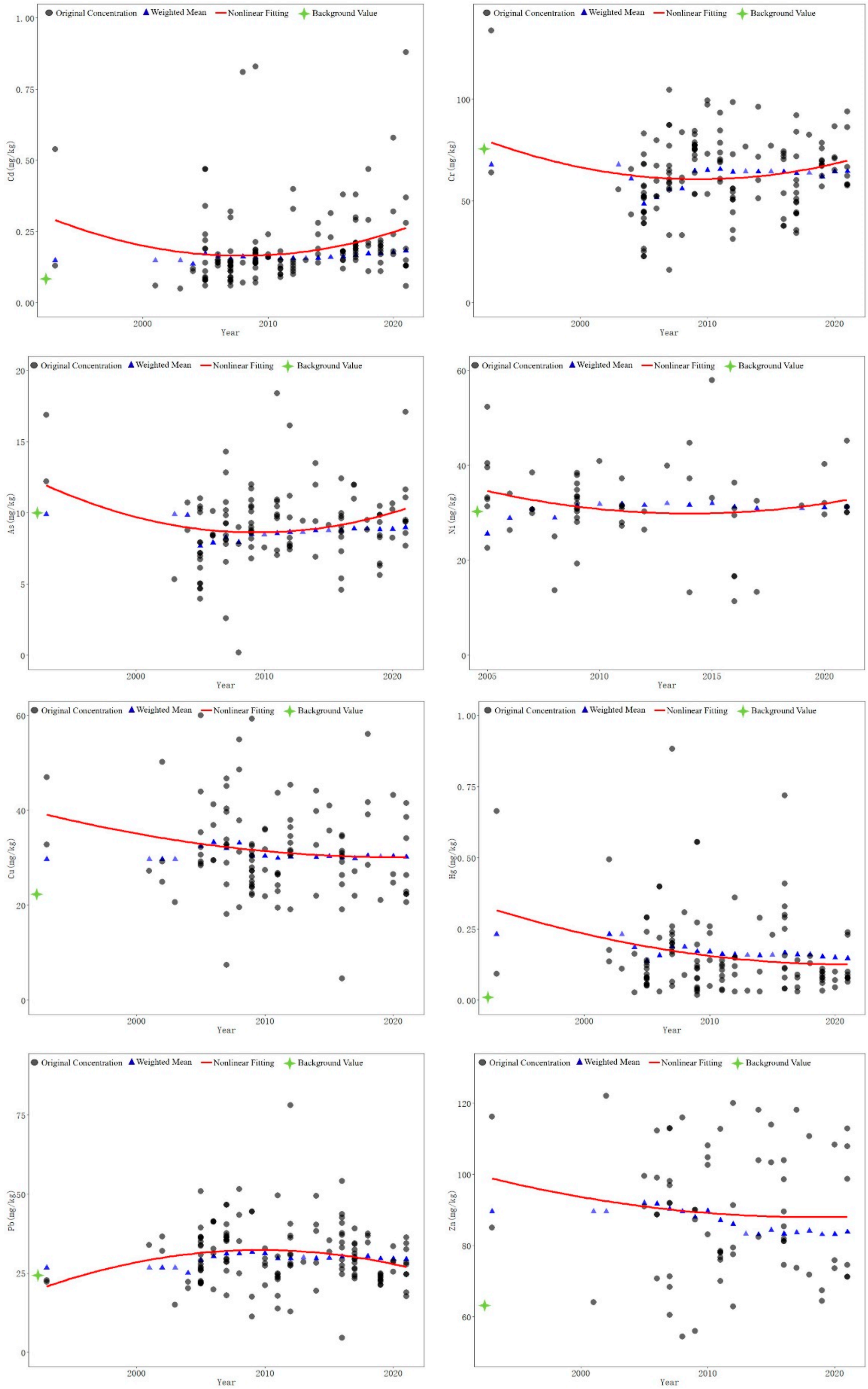

Figure 4. The temporal variation of heavy metal accumulation. The green points represent the agricultural soil background values in Jiangsu.

4. Discussion

4.1. Comparison with Related Studies

Table 3 shows the results of this study compared with related studies on heavy metal content [41]. The finding of this study was generally consistent with those of Liao et al. (2009), She et al. (2021), and Huang et al. (2019) indicating that the pollutants were Cd, Hg, Zn, and Pb in Jiangsu Province, especially Cd, and Hg [4,25,41]. The weighted mean of Hg in this study was higher than that of Zhejiang Province and Yangtze River Delta whole farmland soil, while the concentration of Cd was lower [25,28]. Moreover, as an economically developed region, the PRD showed more accumulation of heavy metal pollutants than Jiangsu [42]. The Jiangsu agricultural soil heavy metal contents of this study were close to the national average levels [4]. This indicated that the results of the meta-analysis of this study were further confirmed to be reliable.

Table 3. The comparison between this study and other studies.

	Cd	Cr	As	Cu	Hg	Ni	Pb	Zn	Reference	Period	Sites
Jiangsu Province agricultural soil	0.19	61.48	8.90	31.12	0.16	30.24	29.41	83.83	This study	1993–2021	149 cases
Jiangsu Province soil	0.15	76	9.40	26	0.09	32.90	26.80	73	[29]	2009	24,186 sites
Zhejiang Province soil	0.23	47.84	7.25	23.96	0.12	21.31	36.79	91.39	[28]	2016–2017	125 sites
YRD, agricultural soil	0.25	68.84	8.14	32.58	0.14	29.30	32.32	92.35	[25]	1993–2020	118 cases
Pearl River Delta Vegetable soil	0.35	58.14	14.29	/	0.15	/	50.82	/	[43]	2012	232 sites
China agricultural soil	0.24	62.20	10.70	28.30	0.13	28.20	32.10	83.30	[4]	2005–2017	336 cases
USA soil	0.34	37	5.20	17	0.06	/	16	48	[44]	1981	1318 sites
Europe agricultural soil	0.18	20	5.50	15	0.03	/	16	45	[45]	2008	2108 sites
India	/	147.05	9.99	79.05	/	52.46	41.80	178.50	[46]	2019	32 sites
Iran	1.53	147.05	/	60.15	/	35.53	46.59	94.09	[47]	2015	-
World soil	0.40	40.00	7.20	30.00	0.1	20.00	2–300	90.00	[48]	/	/

Compared with other regions in the world, the concentrations of Cr, Cu, Hg, and Zn in this study were more than twice those reported for the USA and Europe, but less than half those of Iran and India [44–47,49]. The As concentrations were slightly higher than in the US and Europe. This may be related to the higher natural background values of soils in Jiangsu Province than in the United States and Europe. Many studies have shown that economic development and human activities in Jiangsu Province have significant negative effects on the soil environment [29,41]. Therefore, there is still much room for progress in agricultural soil governance in Jiangsu Province, compared with European countries and the United States.

Interestingly, although Cd was a relatively serious presence in Jiangsu agricultural soil, it was lower than the world average, similar to Europe [45,48]. The concentration of Cd in the United States and Iran was also higher than in Jiangsu Province [44,47]. Although the Cd concentration of heavy metals in Jiangsu Province was not exceptional compared with other areas of the world, the severity of the pollution has been noted by many reports [48]. This may be related to the over-concentration of publications due to researchers' excessive focus on certain contaminated regions. For example, 39% of the Cd pollution studies were based on encrypted sampling sites in highly contaminated areas, which provided values several times higher than normal farmland. This made plenty of studies on Cd pollution in agricultural soil in Jiangsu Province easier to publish but resulted in an exaggeration of the real situation. Although the negative impact of publication bias was attenuated in the previous analysis, it was difficult to eliminate completely.

On the other hand, more stringent standards were used for heavy metal control in Chinese agricultural soils than in most parts of the world, especially for Cd, Hg, and Pb. The farmland soil risk control standards in this study were derived from "The Risk Control Standard for Environmental Quality of Soil Contaminated Agricultural Land" (GB 15618-2018, risk screening value 6.5 < pH ≤ 7.5). The standard value for Cd and Hg was 0.6 mg/kg (GB 15618-2018). The risk control standard for the environmental quality of other countries is shown in Table S3 (refer to the Supplementary 1). The risk control values for Cd and Hg were 1.8 mg/kg and 2.6 mg/kg in the UK, and even 3 mg/kg and 0.8 mg/kg in Canada [50]. The enrichment level of Cd and Hg in Jiangsu Province was 6.2% and 15% of the Japanese control standards, and only 0.12% and 0.10% of those in Australia [50,51]. The Cd and Hg showed high concentration levels in this study but were still far below the risk control standards of most countries in the world. Although the enrichment of Cd and Hg needs to be controlled urgently, the public need not be too alarmed by pollution levels in agricultural soil in Jiangsu Province. Overall, the content of heavy metals in agricultural soil in Jiangsu Province was in a medium state of enrichment. Meanwhile, Jiangsu Province, as an important agricultural and industrial province in the Yangtze River Delta region, could not relax control over polluting enterprises close to farmland.

4.2. The Causes of Spatial Distribution

Rock weathering and soil formation processes retain heavy metals in the soil, which was not only the original source of heavy metals in agricultural soils, but also one of the reasons for the differences in the spatial distribution of heavy metals [52]. The warm temperate soil development north of the Huaihe River in Jiangsu Province was mostly brown loam, with high concentrations of Cd, Pb, and Ni in the background values [33]. There were four main soil types of upland soil, among which swamp soils had high background values of Hg, Zn, and Ni [34]. In addition, the background values of heavy metals in different geomorphic units in Jiangsu Province also reflected the difference. The background value of Cd was the highest in the Yangtze River Delta plain. The contents of Hg and Zn were the highest in the plain soil of Taihu Basin [53]. However, rapid socioeconomic development, industrial production, and human activities have led to more and more pollutants from anthropogenic sources being enriched in agricultural soil [54]. Regional differences in economic level and industrial structure have become another important reason for differences in the soil enrichment status of heavy metals [2]. Jiangsu Province is an important province for grain and vegetable production in the Yangtze River Delta region. Effectively identifying the spatial distribution and the potential influencing factors of agricultural soil heavy metal enrichment is vitally important for regulating heavy metal pollution and reducing the negative impact on the agricultural soil environment. Numerous studies have already proven that industrial production, mining activities, transportation, and agricultural activities were the primary anthropogenic sources of heavy metal contamination in agricultural soil [51,55,56].

Jiangsu Province is one of the fastest-developing provinces in China with developed secondary and tertiary industries. Industrial manufacturing, transportation, chemical raw material production, and other industries produce a lot of waste gas, soot, wastewater, and waste residue. This means that the soil environment may carry a huge amount of anthropogenic waste discharge and accumulate heavy metal pollutants. In this study, the cities of Jiangsu Province were divided into three groups according to the level of potential ecological risk (Figure 3).

The first group was located in southern Jiangsu, and the ecological risk class was high. These cities (Nanjing, Suzhou, Wuxi, Changzhou, and Zhenjiang) had high concentrations of eight heavy metals in agricultural soils, particularly Cd, Hg, Pb, Zn, and Cu. Compared with other geomorphic units, the background values of heavy metals were generally high in the Yangtze Delta Plain and Taihu Basin Plain in southern Jiangsu, especially Cd, Hg, and Zn [34,53]. Previous studies have shown that the background value of heavy metals in the parent material of soil formation was also higher in southern Jiangsu [33,53]. Southern

Jiangsu had the highest night lighting index and showed extremely high levels of human activities (RESDC, http://www.resdc.cn, accessed on 10 December 2022). From Figure 2, more than 60% of the key polluting enterprises of Jiangsu in 2021 were located in southern Jiangsu, of which more than 50% could cause soil pollution. The studies of soil exceedance background values were mainly concentrated in southern Jiangsu, especially in Suzhou and Nanjing (Supplementary 2). It is worth mentioning that in these studies, more than 50% of the sampling sites in Nanjing and 30% in Suzhou showed obvious pollution sources within 5 km. These pollution sources cover various fields, including mining and smelting, industrial manufacturing, metal processing, apparel manufacturing, chemical raw material production, etc. For example, there was a significant enrichment of Cd (0.38 mg/kg) in the soil of southeastern Baguazhou, where many large factory enterprises were distributed [57]. Due to industrial emissions from a lead battery plant in Suzhou, the sampling sites of serious Pb pollution accounted for 11.3%. According to statistics, Suzhou had the most polluting enterprises, and also showed the highest ecological risk index. In addition, many heavy metal pollutants piled up in factories could form acid rain through the oxidation of the air and then return to the ground through the rainfall process, which may cause secondary pollution of the soil [58]. The annual cumulative precipitation in southern Jiangsu Province was the highest in the province, which provided favorable conditions for forming a local acid rain climate near pollution sources [59]. In summary, the soil parent materials and natural conditions lead to higher soil background values in cities in southern Jiangsu than in other regions, and the complex and intensive industrial activities further aggravate the enrichment of heavy metals.

The second group included Nantong, Yangzhou, and Taizhou in the central region and Xuzhou and Lianyungang in the northern. The Cd, Hg, Pb, and Zn were enriched in the farmland soil in this urban agglomeration. The background value of Ni was high in the geomorphic units of Lianyungang [34]. As described in Section 3.3, Lianyungang had the highest Ni concentration. Previous study has indicated that Ni in the soil was highly dependent on the soil parent material, and the human input of Ni was lower than the background contents in the soil [60]. Therefore, the Ni concentration in this study was dominated by the soil parent material. Xuzhou city had the highest Cd concentration, deserving special attention. More than 30% of the sampling sites where Cd exceeded background values were surrounded by mining areas and transportation hubs (Supplementary 2). The mining area with rich resources and a superior combination of conditions located in Xuzhou has large mineral reserves and high grades [23]. With an annual raw coal production capacity of more than 10 million tons, Xuzhou is an important coal-producing area and electric power base in eastern China [61]. Large amounts of waste gas, wastewater, and residue containing heavy metals were produced in the process of mining and smelting. These pollutants entered farmland soil through atmospheric deposition and surface runoff, accumulated in the soil environment, causing contamination at 30% of the sampling sites. The cumulative annual precipitation of this group was lower than that of southern Jiangsu [59]. The abundant precipitation–evaporation and groundwater coupling also affected the migration of heavy metal pollutants into the soil [62]. For example, many heavy metal pollutants produced by the Xuzhou mining area without being recycled in time could combine with the air to form acid rain. Acid rain and acidic wastewater from the mine pits enter the ground through the runoff action of surface water, causing secondary soil pollution. The heavy metal pollutants, under acidic conditions, were more easily leached out in the rainwater [58]. This was another important reason for local heavy metal enrichment in this group of cities. The central region of Jiangsu is one of the important food production bases in Jiangsu Province. Agricultural activities also contributed to the enrichment of soil heavy metals in the region, including the use of pesticides, fertilizers, and agricultural plastic films [63,64]. In addition, the economic level of central Jiangsu was lower than that in southern Jiangsu, and all indicators were in the second gradient. Industrial activities and transportation affected the accumulation of farmland soil heavy metals. In summary, industrial activities and agricultural activities significantly affected the accumulation of farmland soil heavy metals in the region, causing medium ecological risk.

The third group of cities included Suqian, Yancheng, and Huaian, which were located in northern Jiangsu. The soil environment showed slight ecological risk. Compared with other cities, these cities were characterized by lower regional GDP and lower car ownership, but higher consumption of agricultural fertilizers (http://stats.jiangsu.gov.cn, accessed on 20 December 2022). In addition, this study found no obvious industrial or traffic pollution sources near the agricultural soil sampling sites in these cities. This meant that agricultural activities may contribute more to the accumulation of Cd, Hg, and Zn in these cities' farmland soils. The background value of Ni was high in the geomorphic units of Lianyungang. Although Suqian had the highest As content (12.00 mg/kg), it only slightly exceeded the background soil value in Jiangsu province. Previous studies have shown that the As background value of the Suqian geomorphic unit was 12 mg/kg [34]. Therefore, geomorphic structure and agricultural activities significantly dominated the distribution of heavy metals in the soil of this group. Another influencing factor may be that relatively fewer studies on this group of cities were collected in this study compared to other cities, which influenced the results.

4.3. The Causes of Temporal Variation

Farmland soil is an open buffer material system that exchanges energy with the external environment [40]. Heavy metals in agricultural soils follow the law of mass balance: when the heavy metal inputs are greater than the output, a surplus of soil heavy metals occurs, resulting in accumulation, otherwise, heavy metals are in a reduced state [25]. Although Cd, Pb, Zn, Hg, and Cu had accumulated in farmland soil in Jiangsu Province, the accumulation of most heavy metals had decreased after 2010. Considering the continuous urbanization and industrialization of Jiangsu, temporal data on industrial and agricultural production were collected (Figure 4).

During the study period, the GDP in Jiangsu Province grew year by year, with the most prominent growth trend in southern Jiangsu. The proportion of primary industry and secondary industry output declined continuously (Figure 5a). Obviously, the industrial structure of Jiangsu was being constantly restructured. The consumption of agricultural chemicals and chemical fertilizers decreased yearly. Although the consumption of agricultural plastic film increased, since 2010 it has shown negative growth (Figure 5b). The annual growth rate of livestock production also slowed down, especially for hogs and poultry [4]. Jiangsu comprehensively promoted agricultural prevention and control technologies and promoted the real-name purchase of pesticides and other measures to effectively reduce the input of agricultural materials. It made an important contribution to improving the soil environment and reducing heavy metal concentration. The emissions of industrial sulfur dioxide and wastewater decreased continuously. The downward trend was more pronounced after 2010 (Figure 5c,d). Industrial soot emissions have been falling since 2014. Although waste residue emissions continue to increase, the growth rate slowed down year by year (Figure 5d). The average annual growth rate of waste residue showed negative growth after 2010. China promotes a circular economy, reduces resource waste, and controls high-polluting enterprises [3]. Jiangsu strictly implemented national policies, changed its industrial structure and energy consumption, and effectively limited pollutant emissions. These measures reduced heavy metal input, causing the concentration to show a downward trend.

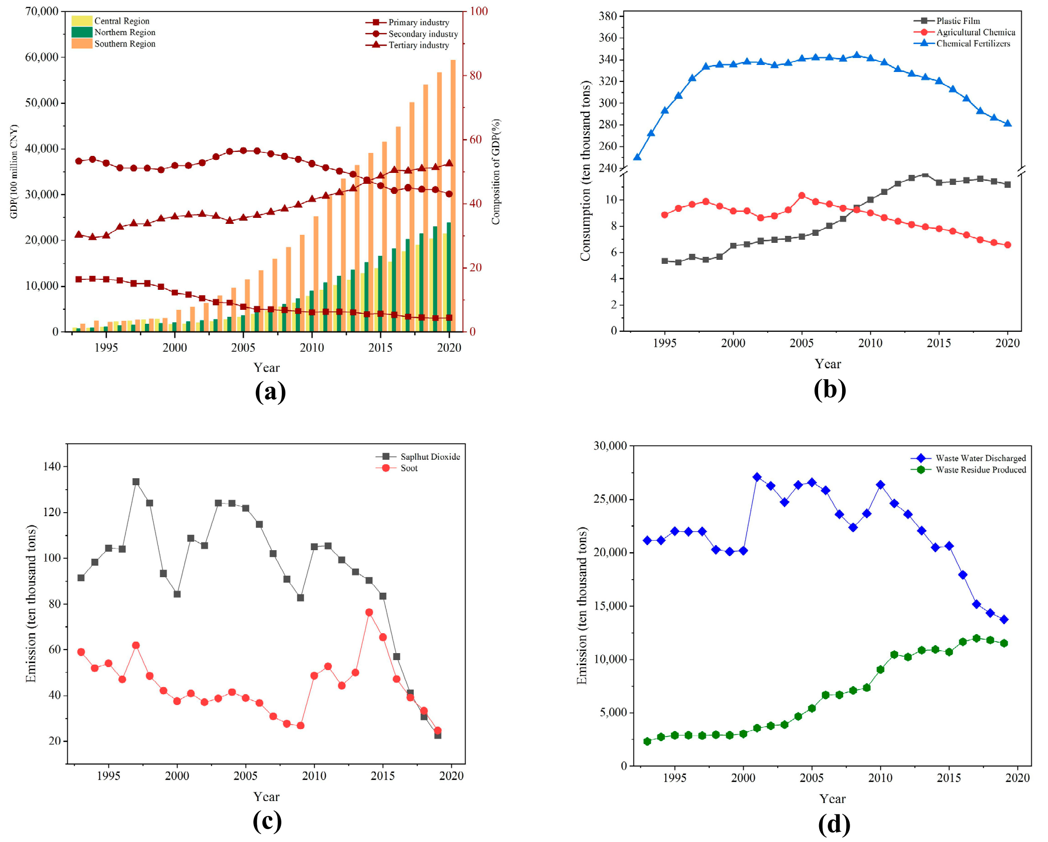

Figure 5. Temporal trends in economic and emission data: (**a**) Annual GDP and composition of different regions in Jiangsu Province; (**b**) annual consumption of agricultural materials; (**c**) annual emissions of sulfur dioxide and soot; (**d**) annual emissions of wastewater and residue.

Due to the long-term accumulation and concealment of heavy metals, the accumulation of heavy metals in agricultural soil could not be rapidly eliminated only by controlling the emissions of pollution sources [63,65]. Therefore, it is essential to explain the accumulation of heavy metals over time from the perspective of output. The heavy metals in farmland soil were mainly exported through crop harvest, leaching, and surface runoff [66]. Hou et al. (2014) showed that the annual flux of agricultural soil Cd from Suzhou was 1.44 g/ha and 0.66 g/ha through crop harvest and leaching, respectively, while the annual flux from Nanjing was 0.47 g/ha and 1.02 g/ha [67]. It was proved that crop harvest and leaching were the primary output pathways of soil heavy metals in southern Jiangsu. However, previous studies have demonstrated that the output of heavy metals through crop harvest and leaching losses was significantly lower than the heavy metal input from various anthropogenic sources [67,68]. Another reason for the declining trend of heavy metals was the remediation and treatment efforts for contaminated agricultural soil. Jiangsu is one of the pilot provinces for ecological and environmental governance. A list of polluting enterprises in all cities has been established and their location, type of pollution, and emission status are monitored (http://sthjt.jiangsu.gov.cn, accessed on 25 December 2022). In addition, a large number of environmental remediation enterprises have emerged in the last decade, using chemical, physical, and biological technologies to remediate the contaminated soil environment [69]. With the development of soil remediation efforts, the trend of soil heavy metal enrichment in Jiangsu gradually weakened, but the remediation effect needs long-term monitoring.

4.4. Prediction and Recommendation

The soil heavy metal pollution control in Jiangsu has achieved specific results. In this study, the cumulative prediction model was used to predict the heavy metal concentration in agricultural soils of Jiangsu under optimistic and no-variation scenarios in 2030 and 2050, respectively (the methods are detailed in the Supplementary 1). Suggestions are put forward for further prevention and control of soil heavy metal pollution in Jiangsu.

The optimistic scenario assumes that Jiangsu shuts down all high-polluting industrial enterprises and bans the unreasonable use of pesticides and fertilizers [70]. Under this scenario, the concentration of heavy metals in Jiangsu farmland soil would decrease by 2030 (Supplementary 1, Table S4). The number of cities with extreme heavy metal concentrations would also decrease. By 2050, the concentration of heavy metals in the soil would continue to decrease to close to the soil background value. Nevertheless, the Cd and Hg would still show a high excess standard rate. Clearly, even with the strict control measures, the current heavy metal concentrations in the soil in Jiangsu Province may take decades to clean up sufficiently to equal the background values.

The no-mutation scenario assumes that the current accumulation and removal rates of heavy metals remain unchanged [71]. The results showed that the concentrations of Cd and Hg would increase while other elements decrease in 2030 (Supplementary 1, Table S5). Under this scenario, heavy metal concentrations predicted for 2050 are higher than in the optimistic scenario. The rate of heavy metal depuration is slower than the optimistic scenario. Clearly, maintaining the current rate of soil depuration of heavy metals may result in a longer time being required to remove accumulated heavy metals than in the optimistic scenario. Therefore, it is necessary to take a series of preventive and remedial measures to accelerate soil depuration and maintain the current trend of sustained decline of heavy metals.

First, the monitoring of pollution sources should be strengthened to control anthropogenic emissions. To achieve this goal, a dynamic and effective monitoring network and emission inventories for Jiangsu agricultural soil heavy metals should be established to identify the exact pollution sources [2]. Prevention and control measures should be taken to cut off the flow of pollutants into farmland completely. For example, closing polluting industrial enterprises around farmland, and avoiding excessive use of fertilizers and pesticides [72]. In addition, the circulation of heavy metals could be prevented by stopping the means of transmission, such as reducing the dry and wet deposition of atmospheric pollutants and prohibiting untreated industrial mining-polluted water for agricultural irrigation.

Second, screening for low accumulation crops should be implemented and cropping systems changed. The concentration of Cd and Hg showed significant enrichment in Jiangsu farmland soil. It is recommended to select crops with a low accumulation of Cd and Hg for planting. Previous studies have shown that the accumulation of Cd and Hg content in japonica-type crops was lower than that in indicia-type, and the Hg content in the edible portion of low-Hg accumulating crops could be reduced by more than 90% [73,74]. In addition, the cropping system in contaminated areas could be changed to reduce soil heavy metal contamination by replacing crops with cash crops, such as converting wheat land to orchards.

Third, effective soil remediation measures should be adopted. Many new physical, chemical, and biological soil remediation technologies have emerged in the last decade, including biomass carbon remediation materials and deep pendant barrier technologies [69]. Remediation of Cd-contaminated rice soils using iron-based biochar has been studied and is effective in passivating Cd [75]. The results of soil remediation studies in Yixing showed that the greater the input of biochar, the more pronounced the weakening effect. When the input of biochar reaches 3%, the weakening effect exceeds 20% [76]. In addition, hyperaccumulator plants could be planted to transfer heavy metals from underground to the plants and harvest the plants to remove the heavy metals. Some studies have shown that planting Sedum can effectively transfer elements such as soil Cd and Zn [25]. Meanwhile, a low-cost and highly efficient passivator could be added to prevent secondary pollution of the soil environment.

Finally, attention should be paid to the negative impact of soil acidification and further soil acidification prevented. Soil acidification has resulted in soil hardening, nutrient loss, and ultimately serious degradation of soil quality [77–79]. Rapid industrial development and extensive use of chemical fertilizers accelerated soil acidification and indirectly increased heavy metal enrichment in farmland soil [77]. Atmospheric deposition was another significant cause of soil acidification [80]. Soil acidification increased heavy metal activity, leading to enhanced uptake of heavy metals by crops and thus increasing the risk of human consumption [81]. Soil pH in Jiangsu showed a spatial pattern of acidity in the south and alkalinity in the north, and heavy metal concentration was higher in the south than in the north. Therefore, measures should be taken urgently to prevent further soil acidification. Measures such as strict control of SO_2 and NO_x emissions, reduction in fossil fuel use, and expansion of clean energy production and consumption could be used to control acid deposition [82]. Sound nutrient input management systems and tillage practices should be developed to reduce nitrogen fertilizer application. According to the nitrogen absorption rule of crops, the method and time of nitrogen fertilizer application should be reasonably selected [83]. The application of organic fertilizers should be increased, especially organic substances such as slightly alkaline soil fertilizers, to improve the base fertility of the soil, and thus improve the soil nitrogen supply capacity [84]. In addition, the application of lime and straw ash could further improve soil pH value [85].

5. Conclusions

Based on 129 publications on soil heavy metal pollution in Jiangsu Province, we summarized and analyzed the farmland soil heavy metal content, spatiotemporal accumulation trend, and influencing factors in all the cities of Jiangsu. This study has a specific reference value for the comprehensive understanding of the current situation and spatiotemporal characteristics of heavy metal accumulation in farmland soil in Jiangsu. Meanwhile, it could provide the scientific basis for soil pollution prevention and control, and soil management in Jiangsu.

The results showed that heavy metals were enriched in Jiangsu agricultural soil, especially Cd and Hg. The spatial pattern of heavy metals in Jiangsu agricultural soil was southern> northern> central. The concentrations of Cd and Zn in Nanjing, Hg in Suzhou, and Cd in Xuzhou were urgently controlled. The agricultural soil in Jiangsu as a whole showed a high ecological risk level, due to the strong potential ecological risk of Hg and Cd. During the study period, the accumulation of Cu, Zn, and Hg decreased continually, while the accumulation of Cd, Pb, Cr, Ni, and As fluctuated. The inputs of Cd, Hg, Pb, Zn, and Cu were greater than the outputs, whereas As, Cr, and Ni remained in balance with background values. There were obvious anthropogenic sources in agricultural soil in Jiangsu. The intensive industrial activities, transportation, and agricultural production contributed significantly to the enrichment of farmland soil heavy metals. In addition, more aggressive pollution control measures are still needed to maintain the current sustained decline in heavy metal concentrations in farmland soils.

This study collected a large amount of data from publications on Jiangsu agricultural soil heavy metals, which ensured timeliness and comprehensiveness. However, several limitations still existed. First, the publications were unevenly distributed spatially, and there were more studies on the southern than on other parts of Jiangsu Province, which led to a deviation in pollution identification. Second, the existence of special sampling points in the literature, such as the encrypted sampling points in mining areas, showed an overestimation of heavy metal pollution levels in some cities. In addition, this study combined economic, demographic, industrial, and agricultural data to explore the causes of spatial and temporal variation of heavy metals in different cities' agricultural soils. Soil types and geological conditions have also been proven to be associated with heavy metal enrichment in agricultural soils and could be included in further studies. However, the results of this study still advance our knowledge of pollution status and characteristics of agricultural soil heavy metals in Jiangsu, which could help in the development of pollution policies.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/land12020455/s1, Figure S1: The Outlier selection by sensitivity analysis. Figure S2: The annual subgroup analysis of the ecological risk index. Table S1: The Shapiro-Wilk test of weights and mean concentrations. Table S2: The weighted mean values (mg/kg) of heavy metals calculated by the four datasets. Table S3: The risk control standard for the environmental quality of other countries (mg/kg). Table S4: Concentration projections in optimistic scenario. Table S5: Concentration projections in no-mutation scenario. References [86–92] are cited in the supplementary materials.

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