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Heavy metal contamination in Shanghai agricultural soil

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A R T I C L E I N F O

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A B S T R A C T

As heavy metals in soil could enrich in biomass and pose health risk to human, it is vital to monitor their contaminations to ensure qualified agricultural production. In this study, we collected >4000 soil samples from agricultural fields in Shanghai during 2010~2020, and unveiled heavy metal contamination status in this metropolitan. We found that although Shanghai has a long industrialization history, the heavy metal levels in agricultural soil are within safe ranges according to national standard. Specifically, the median levels of Cd, Hg, As, Pb, Cr and Cu are 0.11, 0.13, 7.47, 23.80, 41.00 and 28.30 mg/kg, respectively, which are as good as, or even better than national averages. However, there are spatial and temporal heterogeneities for heavy metal contaminations in Shanghai. For example, the levels of Cd, Hg and Cr are relatively higher in some districts with high industry density, which should be further monitored in the future. Moreover, while the levels for Cd, Cr and Pb have decreased, the level for Hg has mildly increased during this period which needs counteractive measures. Correlation analysis of heavy metal levels and soil fertility parameters suggested overuse of fertilizers may be related to heavy metal contamination in some regions. In summary, our study present by far the largest and most comprehensive landscape of heavy metal contamination in Shanghai agricultural soil, which will be useful for future policy-design and land use planning to ensure safe agricultural production.

1. Introduction

Heavy metals in soil could enter agricultural products and pose both carcinogenic and non-carcinogenic health risks for human beings, and children are especially vulnerable [1–5]. The main types of heavy metal or metalloid in soil include Cd, Hg, As, Pb, Cr, Cu, Zn, Ni, *etc*. [6–11]. While industrialization has played important role in rapid economic development, it has also caused serious heavy metal pollution in soils or surface sediments [1,12,13]. Heavy metal contamination of agricultural soil has become a global issue [14–16], and it has also caused concerns in China for the past several decades [17–21]. According to report on the National Soil

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Contamination Status Investigation (2005–2013), 19.4 % of agricultural soils in China are contaminated, and industrial, mineral mining and agricultural activities are the major causes [22,23].

Shanghai is one of the earliest cities in China open to the world, and it has been leading the industrialization in China for more than a century [24–26]. Besides import of agricultural products from other regions, Shanghai is also providing local-produced agriculture products for the residents [27]. The agricultural products from local fields have the advantages of freshness and strict quality control, making them popular consumption choices. Considering the long history of industrialization in Shanghai, it is thus critical to monitor soil pollution to ensure safe agricultural production. As China is a large country with great spatial heterogeneity [28,29], a precise local landscape of heavy metal levels in Shanghai is the first step to assess contamination status and make proper policies.

During the past ~10 years, we have been collecting soil samples from agricultural fields in Shanghai for Green Food production monitoring. For the soil samples collected, we measured the pH values, the levels of key heavy metals and metalloid (Cd, Hg, As, Pb, Cr and Cu), as well as soil fertility parameters. With more than 4000 soil samples, in this study we present by far the most comprehensive landscape of heavy metal contamination in Shanghai agricultural soil.

2. Results

2.1. Overview of the agricultural soil samples from Shanghai

In this study, we collected a total of 4044 soil samples from 976 agricultural entities in Shanghai between 2010 and 2020 (Fig. 1A). There are ~5700 agricultural entities in Shanghai [30], and our sampling size is a good representation of them. The number of samples collected in 2018, 2019 and 2020 are relatively higher than early times like 2010–2014 and 2017 (Fig. 1B). About 88 % of soil samples were collected from 8 districts of Shanghai: Chongming, Pudong, Fengxian, Songjiang, Qingpu, Jinshan, Minhang, and Jiading, where most of the agricultural fields in Shanghai are located. We collected pH values and concentrations of key heavy metals and metalloid (Cd, Hg, As, Pb, Cr and Cu) for all 4044 samples. We also collected soil fertility data like organic material content, and N, P, K

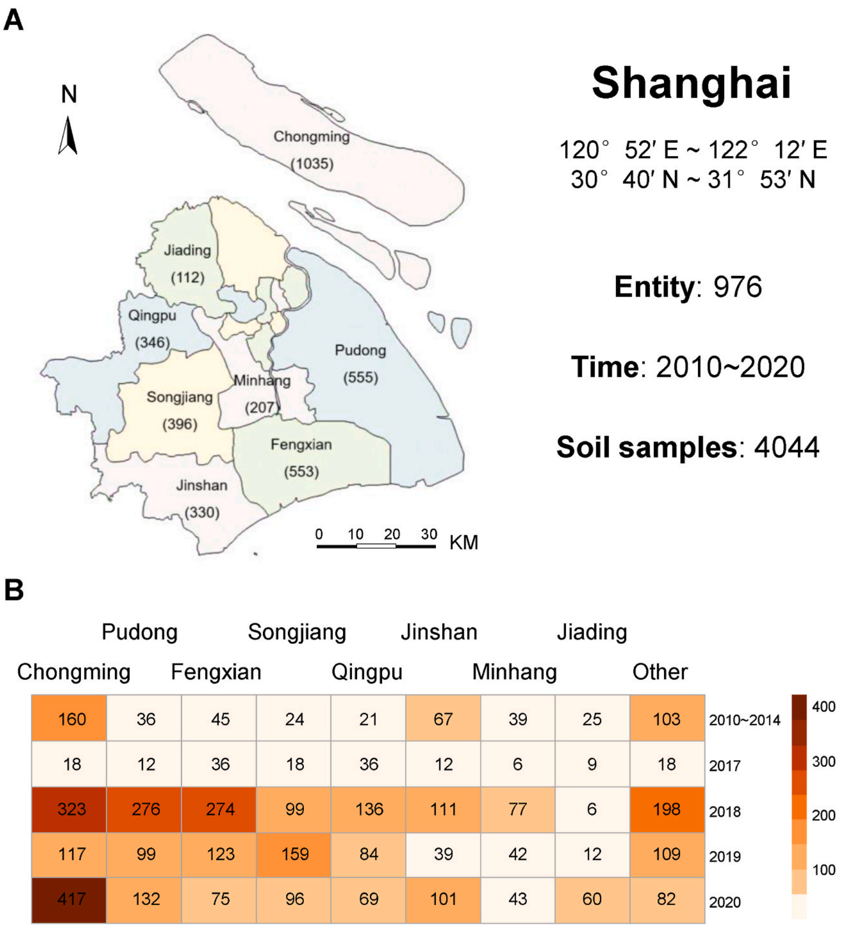

Fig. 1. Overview of agricultural soil samples from Shanghai. (A) The map of Shanghai showing the number of soil samples from 8 major agriculture-related districts. (B) Heatmap showing the number of soil samples from each district at each time point. The color key is showing ranges of the sample numbers.

concentrations from 3500 samples. The heavy metal contamination data collected in this study is by far the largest and sole source of agricultural fields for Green Food production in Shanghai.

The functional effects of heavy metals on crops are closely related to soil pH, and Green Food industry standards in China (Green Food-Environmental quality for production area, NY/T 391–2021, issued by the Ministry of Agriculture and Rural Affairs of China) are also set according to soil pH ranges. The pH values for most of soil samples from Shanghai agricultural fields are around 7, and most of districts in Shanghai exhibit similar pH ranges (Fig. S1). There are exceptions, with samples from Songjiang and Qingpu having lower

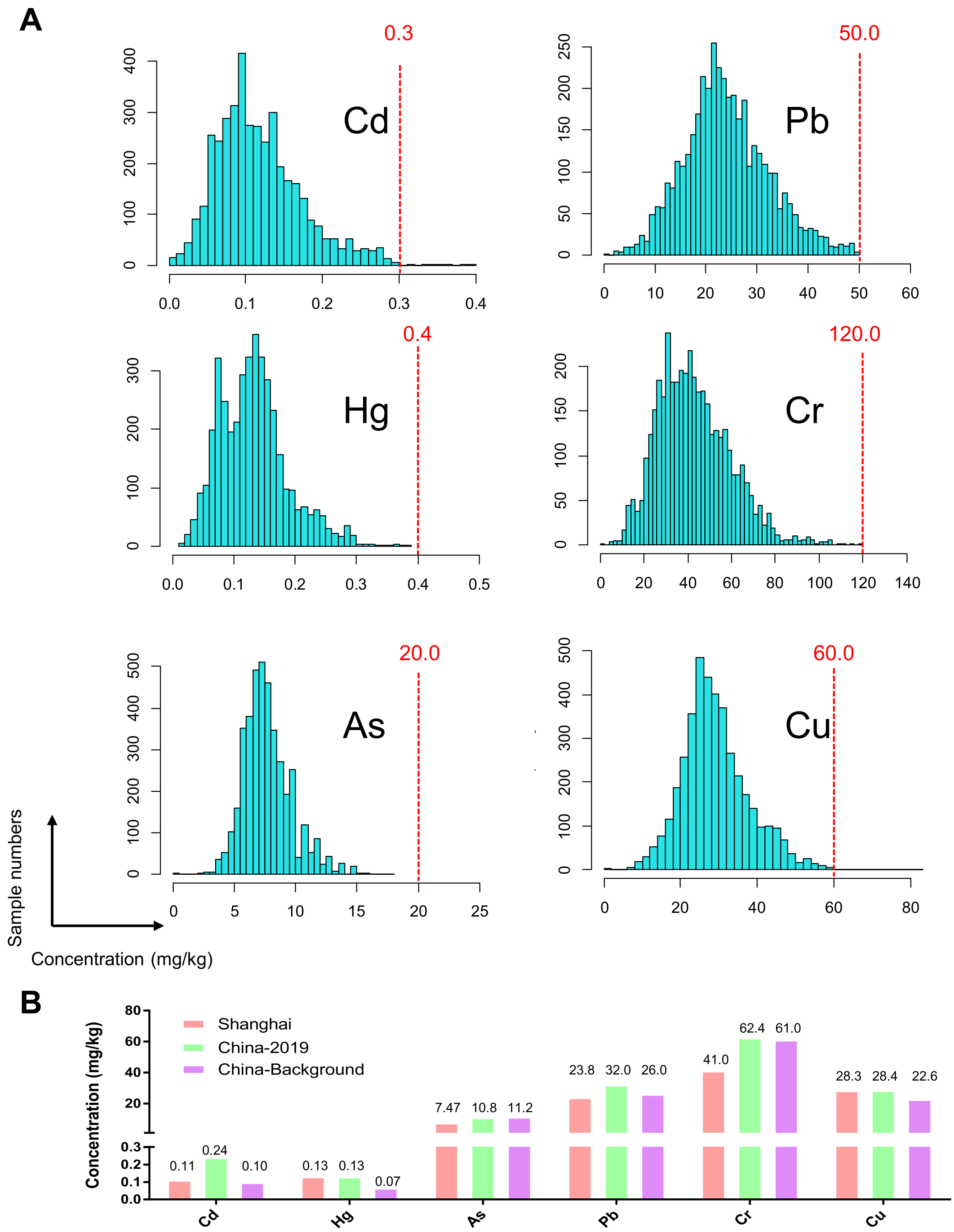

Fig. 2. Overview of heavy metal levels in Shanghai agricultural soil. (A) Histogram plot showing the statistical distribution patterns of six heavy metal contamination levels. The *y* axis represents the number of samples, and the red lines highlight upper limits of heavy metals for Green Food production in China. (B) Comparison of median levels of heavy metals in Shanghai agricultural soil and mean values from China (Huang et al., *Sci Total Environ* (2019), 651:3034–3042) and background values from China (China National Environmental Monitoring Center, 1990).

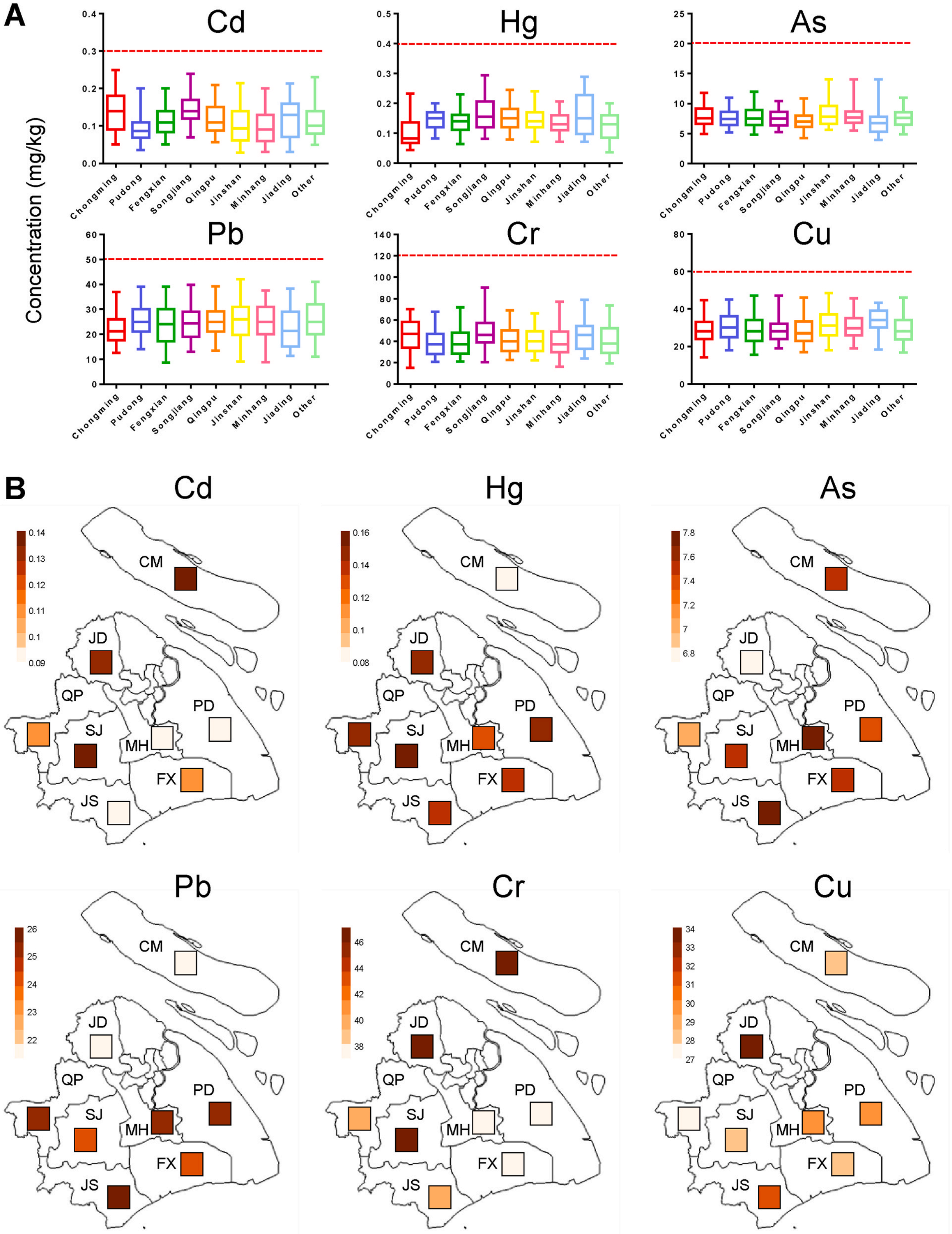

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Fig. 3. Spatial heterogeneity of heavy metal contaminations in Shanghai agricultural soil. (A) Box plot showing the concentrations of heavy metals from different districts of Shanghai. The boxes are showing the medians and the 1st and 3rd quartiles, and whiskers are showing the 5–95 % intervals. The red lines highlight the upper limit of heavy metals for Green Food production in China. (B) Spatial distribution maps for heavy metals in 8 major agriculture-based districts of Shanghai. Median values (mg/kg) are shown for each district, and the color keys are showing concentration ranges. Abbreviations: Chongming, CM; Pudong, PD; Fengxian, FX; Songjiang, SJ; Qingpu, QP; Jinshan, JS; Minhang, MH; Jiading, JD.

pH values and medians of 6.7 and 6.9, respectively, and samples from Chongming having higher pH values and median of 7.4. As an island formed inside the Yangtze River, Chongming has relatively younger history and is showing some unique soil features compared with other districts in Shanghai, which may explain its higher soil pH range.

2.2. Heavy metal contaminations are not severe in Shanghai agricultural soil compared with national standard

We checked the statistical distribution patterns of heavy metal concentrations in all soil samples from Shanghai (Fig. 2A). In general, the heavy metal levels in Shanghai agricultural soil are much lower than the national standard for agricultural soil in China (Soil environment quality-Risk control standard for soil contamination of agricultural land, GB15618-2018) [31], and also lower than the stricter Green Food industry standard (NY/T 391–2021) [32]. Specifically, the distribution of Cd has a single peak, with majority of values lower than industry standard of 0.3 mg/kg. The distribution of Hg levels has two peaks, with one peak at ~0.07 mg/kg and the other at ~0.14 mg/kg. The peak at ~0.07 mg/kg is close to the national background for Hg [33]. Although there are heterogeneities of Hg levels, all values are much lower than the Green Food industry standard limit of 0.4 mg/kg. Similarly, the distribution of As, Pb, Cd, and Cu also exhibit single peak distribution, and almost all of samples have levels much lower than the Green Food industry standards. The results suggested that the levels of heavy metals in Shanghai agricultural soils are lower than both national standard and Green Food industry standard, laying good foundation for safe agricultural production.

For all the soil samples analyzed, the median levels of Cd, Hg, As, Pb, Cr and Cu are 0.11, 0.13, 7.47, 23.80, 41.00 and 28.30 mg/kg, respectively (Fig. 2B). Compared with the national averages in China from a 2019 meta-analysis and national background values in 1990 [33,34], the soil quality in Shanghai is as good as, or even better than national averages. Recent studies indicated that Cd is the top polluting element in China, especially in South China [2,9], but the median level for Cd in Shanghai is lower than national average and similar as national background value. The levels for Hg and Cu in Shanghai are similar as national averages and higher than background values. The levels for As, Pb and Cr in Shanghai are also lower than national averages and not higher than background values. The results suggested that although Shanghai has a long history of industrialization, the soil quality for agricultural field is still better than national average. The soil quality in Shanghai is also not bad compared with other places with long industry history. For example, the median levels of Cd and Pb are ~0.3 mg/kg and ~40 mg/kg in UK [35], which are higher than that in Shanghai.

2.3. Spatial and temporal heterogeneity of heavy metal contaminations in Shanghai agricultural soil

We then compared the statistical distribution patterns of heavy metals from different regions of Shanghai (Fig. 3A and Fig. S2). For the distributions of Cd, Chongming and Songjiang have statistically higher levels of contamination compared with other regions. As the solubility of Cd will increase by lowering pH [36], the lower pH values of soil samples from Songjiang could be a possible reason behind the higher level of Cd. For the contamination by Hg, the levels from Chongming are the lowest among all districts, while Songjiang and Jiading have relatively higher levels of contamination. For As and Pb, all districts have similar levels of contamination much lower than the national and Green Food industrial standards. The distribution of Cr level is similar as that of Cd, with higher levels in Chongming, Songjiang and Jiading. For the distribution of Cu, the levels are higher in Jiading and Jinshan compared with other regions. Based on the results, Cd, Hg and Cr are showing relatively higher levels in some districts of Shanghai and needs more attention in the future, while As, Pb and Cu are homogenously distributed spatially in Shanghai within safe thresholds. Spatial distribution maps for the six heavy metals are showing a more straightforward view of the heterogeneity among different districts in Shanghai (Fig. 3B).

We then took into consideration the sample collection time to further check the temporal changes of heavy metal contaminations in Shanghai agricultural soil. The patterns of heavy metal contamination in different districts at different time points are generally consistent with above analysis with time points combined. Interestingly, different heavy metals are showing different dynamic trends. For example, the levels of Cd, Pb and Cr have decreased in majority of regions from 2010 to 2020, but the levels of Hg have moderately increased during this period (Fig. 4A). The decreasing trend of Cd levels in Chongming is especially obvious, while the increasing trend of Hg in Shanghai is mild. The levels of As and Cu have been fluctuating moderately without clear dynamic trends. A recent study observed declining trend of soil Cd level since 2012 in China [9], consistent with the data in Shanghai. Further grouping samples from different districts into different time points also provided new insight (Fig. 4B). For example, Hg level in Fengxian and As levels in Fengxian and Jinshan in 2020 are higher than other groups, which are not revealed in combined analysis of all time points. Although the sampling size may affect the statistical accuracy, the relatively higher levels of heavy metals in a specific region at given time points still raise alarm for close monitoring in the future and early regulatory intervention.

2.4. Comprehensive profile of heavy metal contamination in Shanghai agricultural soil

After separately analyzing each heavy metal, we then checked the global contamination feature of the soil samples by combined

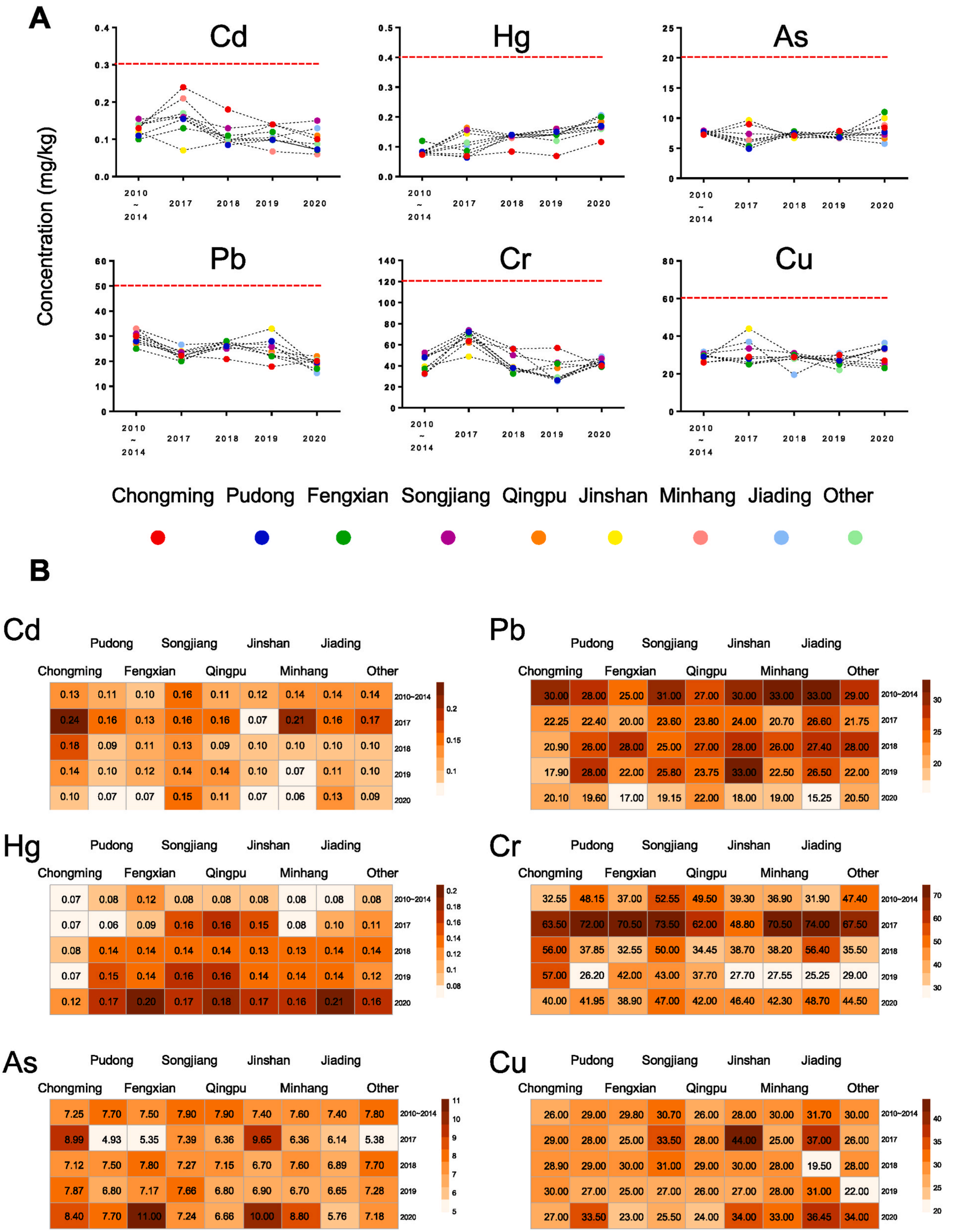

Fig. 4. Temporal heterogeneity of heavy metal contaminations in Shanghai agricultural soil. (A) Temporal changes of median values for each heavy metal in each district. (B) Heatmaps showing medians of soil heavy metal levels in each district at each time point in Shanghai. The color keys are showing the concentration ranges, and the median values are also provided for each group.

analyses of the six heavy metals. After normalization of each heavy metal, we used heatmap to view heavy metal contamination patterns by ranking samples according to collection times (Fig. 5A). Heterogeneity of contamination features could be observed, for example, Hg contaminations for samples collected between 2010–2014 are relatively lower compared with samples collected later, consistent with above analysis.

By employing Principal Component Analysis (PCA), we then had an easier visualization of the similarity of contamination features among the soil samples. For samples collected at different time points, there are temporal heterogeneity, especially between earlier samples (2010–2014, and 2017) and more recent samples (Fig. 5B). This also reflects dynamically evolving trends for those heavy metals as described above. The differences of heavy metal contamination profiles for samples from different districts, however, are not

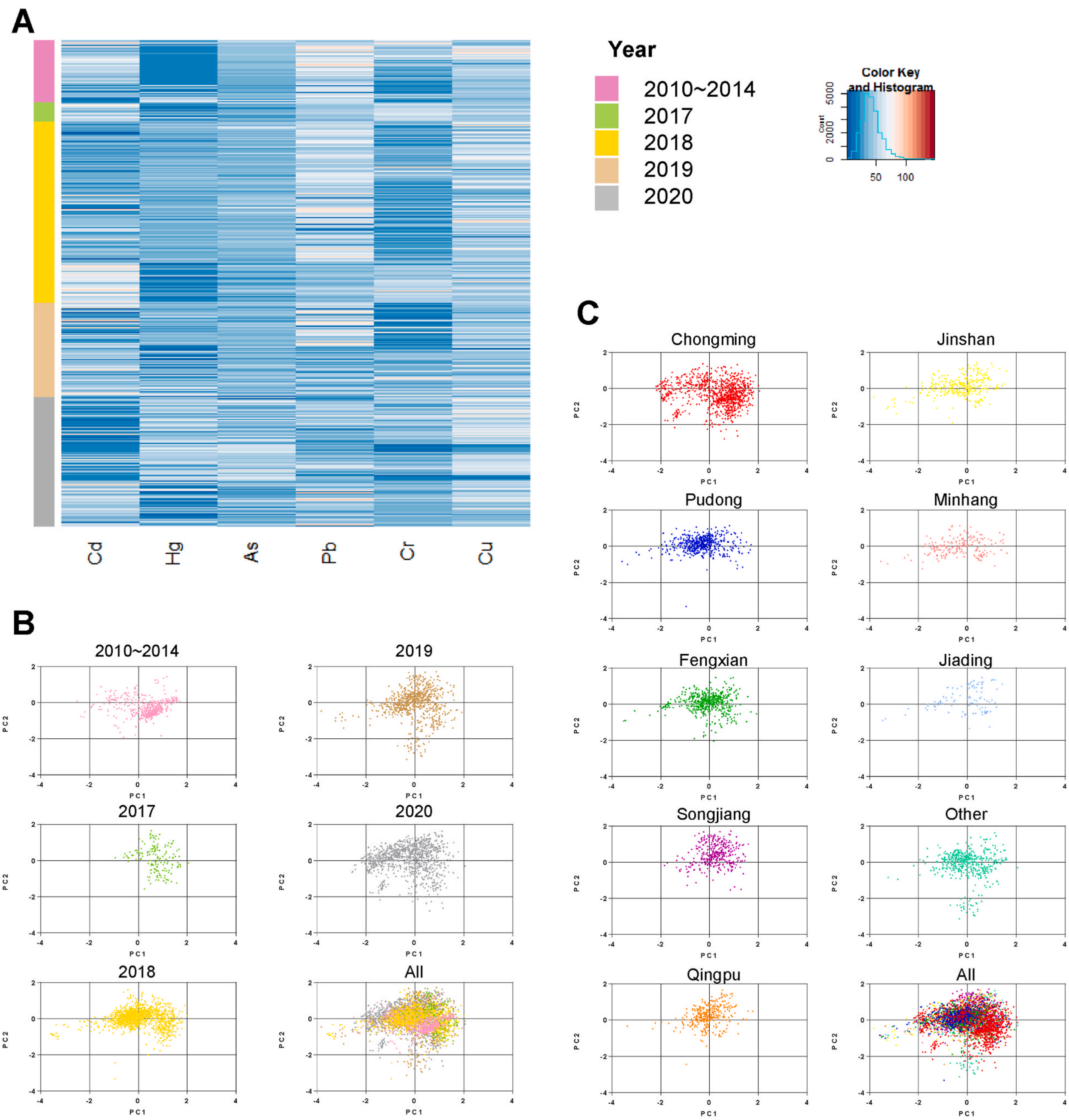

Fig. 5. Comprehensive global profile of heavy metal contamination in Shanghai agricultural soil. (A) Heatmap showing the global contamination profile for all soil samples in Shanghai. The normalized values for heavy metals are used in the plot, and the sample collection times are provided for each sample. (B) Principal Component Analysis (PCA) plot showing the similarity of samples from different time points. (C) PCA plot showing the similarity of samples collected from different regions.

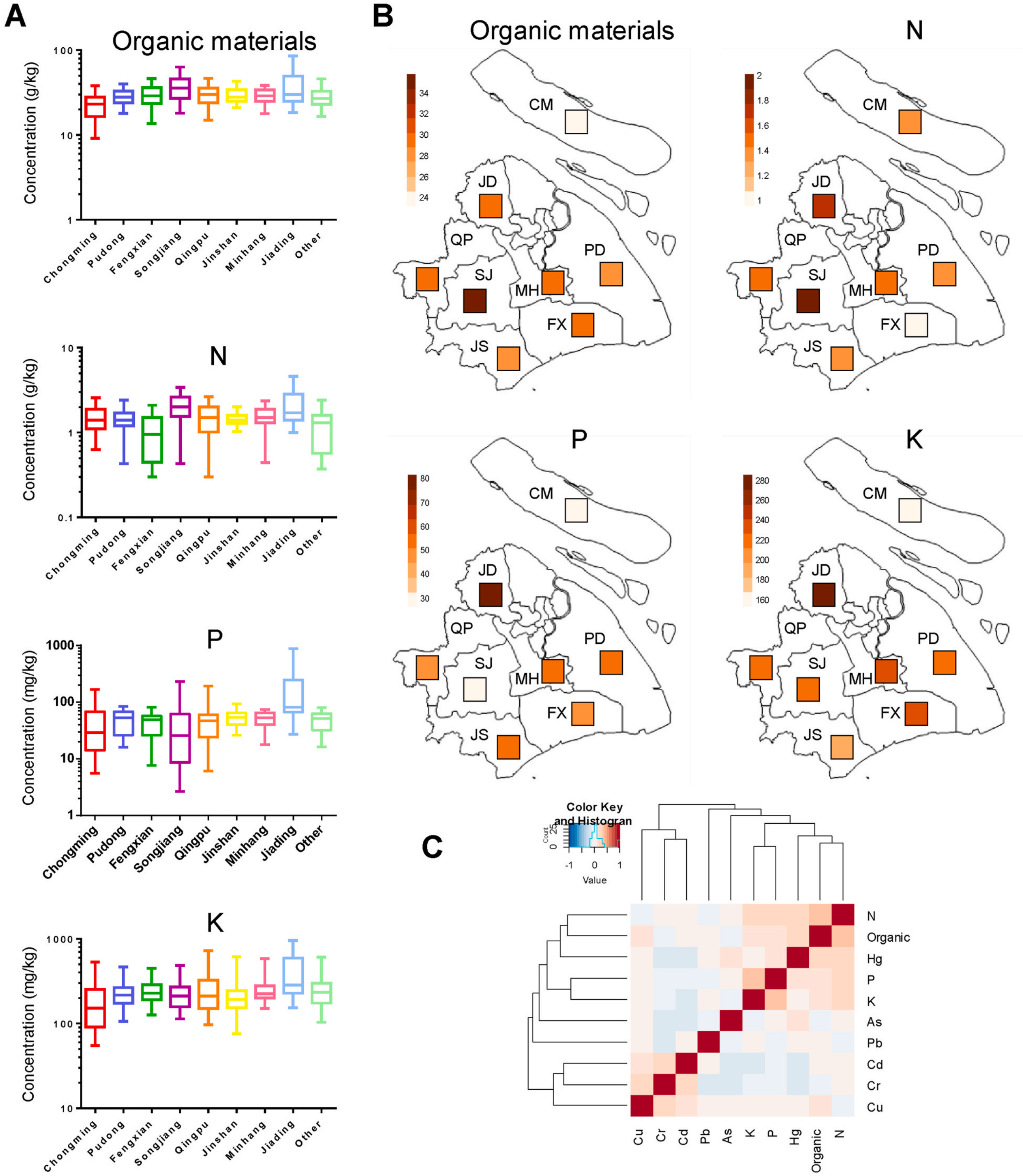

Fig. 6. Relationship between heavy metal contamination and soil fertility in Shanghai. (A) Box plot showing statistical distributions of five fertility parameters for soil samples from different districts of Shanghai. The boxes are showing the medians and the 1st and 3rd quartiles, and whiskers are showing the 5–95 % intervals. (B) Spatial distribution maps for soil fertility parameters in 8 major agriculture-based districts of Shanghai. Median values are shown for each district, and the color keys are showing parameter ranges. For parameter unit, (g/kg) is used for Organic materials and N, and (mg/kg) is used for P and K. Abbreviations: Chongming, CM; Pudong, PD; Fengxian, FX; Songjiang, SJ; Qingpu, QP; Jinshan, JS; Minhang, MH; Jiading, JD. (C) Hierarchical clustering of correlations scores between heavy metal levels and soil fertility parameters.

that dramatic (Fig. 5C). This is probably because only limited heavy metals exhibit difference (such as Cd, Pb, Hg) in limited districts (such as Songjiang and Jiading), while other heavy metals remain similar among majority of districts. It should be noted that there are relatively smaller numbers of samples from some districts, for example regions besides the 8 major agriculture-based districts, which may affect reliability of data interpretation. More soil samples from those under-represented regions are expected in the future to facilitate a sample-balanced analysis.

2.5. Heavy metal contamination in Shanghai is related to soil fertility

As we also collected soil fertility data from 3500 samples (~86.5 % of total soil samples in this study), we then checked their relationship with heavy metal contaminations. The fertility features are showing relatively higher heterogeneity compared with heavy metals (Fig. 6A), probably because they could be easily affected by human interventions such as application of fertilizers/pesticides. There are also spatial heterogeneities for soil fertility between different districts, and spatial distribution maps for soil fertility parameters are showing a more straightforward view of such heterogeneity (Fig. 6B). For example, the levels of N are relatively higher in Songjiang and Jiading, and levels of P and K are higher in Jiading. These results suggested there may be link between high soil fertility and high level of heavy metal contaminations in some regions.

We then performed correlation analysis of heavy metal levels and soil fertility features (Fig. 6C). Interestingly, Hg is clustered among fertility features like organic materials, N, P, and K, while Cd, Cr and Cu are clustered together. Pb and As are not showing high collection with other parameters. The correction analysis showed that the contamination of Hg may be related to soil fertility features. As we mentioned above that the heavy metal contamination in Shanghai is not severe compared with national averages, we also checked the fertilizer application in Shanghai. We could see a declining trend for the fertilizers used at Shanghai in during 2000~2020, which is now among the lowest in China (Fig. S3). This suggested that Shanghai is working towards more rational fertilizer utilization, which could help maintaining its relatively lower level of heavy metal contaminations. However, a few districts are showing relatively higher level of soil fertility parameters and heavy metal contaminations, which should get more attention in the future. For example, reduced fertilizer utilization in Jiading is recommended to check heavy metal contamination change in the future.

3. Discussion

Heavy metal contamination of agricultural soils pose great risk to food safety and human health [37,38], and it is technically challenging to remediate contaminated soils [39]. Our data indicated that the soil samples in Shanghai exhibit low level of heavy metal contaminations compared with the national and industrial standards for Green Food production in China, laying good foundation for safe agricultural production. This finding is especially precious considering the long history of industrialization in Shanghai. According to a recent investigation of industrial legacies in China and their relationship with soil heavy metal pollution, chemical manufacturing, ferrous metal and non-ferrous metal processing, and mines pose serious risks [1]. It is probably the far-away distance of Shanghai from mineral mines prevent serious heavy metal pollution in agricultural fields. Similarly, a recent analysis of soils in Pearl River Delta of China also suggested that although levels of heavy metals have increased during 1989~2019, most of the contamination levels are not posing hazard risk to food security and human health [12]. The findings thus give the hope of safe agricultural production in regions near big metropolitans.

Distribution heterogeneity is an important topic in heavy metal contamination analysis [13,40]. This study showed that while the levels of heavy metals are relatively safe in Shanghai agricultural soil, there are spatial and temporal heterogeneities, and it will be beneficial to closely monitor key industry-centered districts in the future to prevent hazardous-level heavy metal contamination of agricultural soils. An interesting district is Chongming, which has relatively higher levels of Cd and Cr, but lowest level of Hg in Shanghai. The results are generally consistent with a previous small-sized investigation of Chongming island [41]. As Chongming is planning to build into a world-class ecological island with modern agriculture [42,43], detailed investigation and assessment of heavy metal distribution within the island is recommended for proper land use planning. The spatial heterogeneities of heavy metals in other districts of Shanghai are also closely related to their industrial structures. For example, Jiading has relatively higher level of Hg, Cr and Cu, while Songjiang has relatively higher level of Cd, Hg and Cr. According to 2020 statistics, Jiading and Songjiang are among top 4 districts in Shanghai based on industrial output per capita area [44]. Jiading has large-scale auto industry, while Songjiang is focused on electronic industry, which may be related to the heavy metal contaminations in the two districts. Although heavy metal levels in agricultural soils from those districts are below the safety limits, their relatively higher levels among some districts in Shanghai still raise alarm for future close monitoring and proper environmental policy intervention.

Due to economic development requirement, the urban areas are expanding while agricultural fields are shrinking in Shanghai [45,46]. Shanghai will decrease and then maintain ~120,000 ha of agricultural fields between 2035 and 2050 according to the Shanghai Master Plan 2017–2035 [47], approximately half of the present size. The heavy metal contamination landscape presented in this study will then provide useful guidance to select the least contaminated fields for high-quality agriculture production. Continuous monitoring of heavy metal contaminations in agricultural soils, as well as from adjacent regions, are also required to assess current status and predict future trends [48,49].

For limitation of this study, one drawback is that Geographical Information System (GIS) format data for the soil samples is not available due to regulatory requirements. Although the lack of sampling location coordinates may hinder precise mapping more detailed investigation of spatial heterogeneity, our analysis at district level is still providing interesting results on heavy metal spatial distributions. This will be approximately at the resolution of ~20 km, and be still useful for future comparison purposes. Another limitation is the causes behind the higher level of heavy metals in some districts are not revealed. For example, the reason for increasing trend of soil Hg level is unknown. Considering that Hg could retain and enrich in biomass for long term and cause health risks [50,51], the results call for further investigation and strict control to avoid deterioration of soil quality. Another drawback is that the correlation analysis of the heavy metal levels and soil fertility parameters preliminarily imply rather confirm their direct linkages, which still need further validation.

In summary, here we present a by far the most comprehensive landscape of heavy metal contamination status of agricultural soil at Shanghai. Our results showed that the heavy metal contaminations are not severe in Shanghai compared with national average, providing basis for high quality agricultural production in this big metropolitan. The spatial and temporal heterogeneity of the contaminations, however, raise alarm for specific regions to closely monitor heavy metal levels in the future and take counteractive measures to prevent further deterioration for specific heavy metals.

4. Methods

4.1. Description of study area

Shanghai is located at estuary of the Yangtze River in China (E: 120°52′ to E: 122°12′, N: 30°40′ to N: 31°53′). It is the largest city in China with a population of ~25 million, and only 10.7 % of population are rural-based, making the rate lowest among all provinces in China [30,52]. Shanghai has established an industry structure covering major sectors, such as steel, petrochemical industry, shipbuilding, auto industry, modern electronics, and so on [44]. In 2021, 264,350 ha of land were used for agricultural production in Shanghai (41.7 % of the total areas), which produced 939,600 tons of grains and 2,486,400 tons of vegetables [44,53].

4.2. Collection of agricultural soil samples in Shanghai

A total of 4044 soil samples from agricultural fields in Shanghai were collected by Shanghai Center of Agri-Products Quality and Safety during 2010~2020. The sampling details, such as depth of sampling and sample location selection, were all in accordance with the Green Food industry standard in China (Green Food-Specification for field environmental investigation, monitoring and assessment, NY/T 1054–2021, issued by the Ministry of Agriculture and Rural Affairs of China). pH values and levels of six key heavy metal or metalloid, namely Cd, Hg, As, Pb, Cr and Cu, were measured for all samples, and soil fertility parameters including organic material content, and N, P, K concentrations were collected from 3500 samples. Strict quality control ensured all the required parameters were determined for the soil samples. Sample collection time and collection location were also used for the analysis. This dataset is by far the largest for Shanghai agricultural soil, and the sole source of contamination data for agricultural fields used for Green Food production in Shanghai.

4.3. Statistics of heavy metal levels in Shanghai agricultural soil

For statistical distribution patterns of heavy metal levels, histogram plot was used for each heavy metal. The Green Food industry standard (Green Food-Environmental quality for production area, NY/T 391–2021) issued by the Ministry of Agriculture and Rural Affairs of China was used as a reference for upper limit for heavy metal contaminations in agricultural soils. As pH values for most the soil samples fall within the pH 6.5–7.5 range, we used values from this condition as the reference. For comparison with heavy metal contamination levels in China, we used the national heavy metal pollution data from a 2019 meta-analysis of China between 2005 and 2015 [34], as well as the national background values issued by China National Environmental Monitoring Centre in 1990.

4.4. Spatial and temporal analysis of heavy metal levels in Shanghai agricultural soil

We then grouped the heavy metal contamination data according to their locations and collection times for spatial and temporal analysis. Box plot was used to give more straightforward visualization of the median, the quartiles, as well as the 5–95 % intervals for comparison purpose between different regions. Unpaired *t* tests were performed to check the statistical significance for comparisons of contamination levels between different districts using GraphPad Prism. We also plotted the median values for heavy metals in each district according to time point, to give temporal dynamic trend for each contaminant. Heatmaps for the medians of each heavy metal in each region at each time were also used to give temporal-spatial insights into the soil contamination status. For spatial distribution maps of heavy metal levels, pH levels, and soil fertility parameters, we used a square to represent the mean value for a specific parameter in each district, and the color of the square is based on heat-maps of those values. Color keys are used to show the ranges of the values.

4.5. Comprehensive view of heavy metal contamination status in Shanghai

After analyzing each heavy metal contamination separately, we then checked the contamination status of all heavy metals. As the levels for different heavy metals are not directly comparable, we used the Green Food industry standard upper limits to normalize each heavy metal. After that, we used a single heatmap to show all six heavy metal contamination together. Based on the normalized data, we also performed Principal Component Analysis (PCA) using Singular R package (Fluidigm) as previously described [54]. The PC1 and PC2 values were used to show the distribution of all soil samples, and the sample collection regions and times were mapped to this PCA graph to investigate spatial and temporal heterogeneities of heavy metal contaminations.

4.6. Relationship between heavy metal contamination and soil fertility

We also collected the soil fertility data for 3500 soil samples, including organic materials, and N, P, K levels. We used box plot to check the spatial heterogeneities of soil fertility parameters in different regions of Shanghai. We also performed correlation analysis of heavy metal levels and fertility parameters using R. The hierarchical clustering of correlation scores provided clues to the associations between heavy metals and soil fertility.

Data availability

The heavy metal contamination data and soil fertility data are not deposited into a publicly available repository due to regulatory restrictions, but they will be made available to the readers upon request.

CRediT authorship contribution statement

Ruihong Li: Data curation, Formal analysis, Investigation. **Jingzhi Wang:** Data curation, Formal analysis, Investigation. **Yuanfei Zhou:** Resources, Software. **Weiyi Zhang:** Conceptualization, Supervision, Funding acquisition. **Dongsheng Feng:** Conceptualization, Supervision. **Xianbin Su:** Conceptualization, Funding acquisition, Investigation, Supervision, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2023.e22824.

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