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Heavy metal and nutrient concentrations in top- and sub-soils of greenhouses and arable fields in East China – Effects of cultivation years, management, and shelter☆

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ABSTRACT

Although greenhouse vegetable production in China is rapidly changing, consumers are concerned about food quality and safety. Studies have shown that greenhouse soils are highly eutrophicated and potentially contaminated by heavy metals. However, to date, no regional study has assessed whether greenhouse soils differ significantly in their heavy metal and nutrient loads compared to adjacent arable land. Our study was conducted in Shouguang County, a key region of greenhouse vegetable production in China. Soil samples down to soil depths of 3 m were taken from 60 greenhouse vegetable fields of three different ages (5, 10, and 20 years) and from 20 adjacent arable fields to analyze the concentrations of heavy metals, nutrients, and soil physio-chemical parameters. A comparison of greenhouse soils with adjacent arable fields revealed that for greenhouses, (a) micro (heavy metals: Cu, Zn, and Mn) and macronutrients (Nmin, Olsen-P, available K) were significantly higher by a factor of about five, (b) N:P:K ratios were significantly imbalanced towards P and K, and (c) topsoil (0–30 cm) concentrations of the above-mentioned micro- and macronutrients increased with years of vegetable cultivation. In contrast, the soil concentrations of the heavy metals Cr and Pb were lower in greenhouse soils. Heavy metal concentrations did not vary significantly with soil depth, except for the micronutrients Cu and Zn, which were between 1- and 3-fold higher in the topsoil (0–30 cm) than in the subsoil (30–300 cm). The Nemerow pollution index (P_N) was 0.37, which was below the recommended environmental threshold value ($P_N < 1$). Structural equation model analysis revealed that soil nutrient concentrations in greenhouse soils are directly related to the input of fertilizers and agrochemicals. Lower values of soil Pb and Cr concentrations in greenhouses were due to the sheltering effect of the greenhouse roof, which protected soils from atmospheric deposition due to emissions from nearby industrial complexes.

1. Introduction

In the last two decades, China has seen a significant shift in vegetable production from open fields to greenhouses, largely driven by increasing consumer demand for vegetables, as well as by high profit margins for greenhouse vegetable producers (Xu et al., 2015). Currently, approximately 1/3 of all vegetables grown in China are produced in greenhouses (Liang et al., 2019; Zhao et al., 2021b). In addition, data on the

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use of greenhouses for vegetable production in China are impressive on a global scale. With a globally estimated 5.6 million ha of greenhouses in 2018 (Cuesta Roble, 2019), approximately 83% of this area is located in China (Fei et al., 2018). Most greenhouses in China are constructed in the East, particularly in Shandong Province (Yang et al., 2014), where many of the arable fields previously cropped with maize, or used for the production of rice and wheat in rotation, were converted to greenhouse vegetable fields for economic reasons (Wu et al., 2017).

The excessive use of fertilizers, pesticides, and irrigation water is a common practice in greenhouse vegetable production (Qasim et al., 2021; Zhao et al., 2021a, b), leading to soil nutrient imbalances and the accumulation of heavy metals (Bai et al., 2015). Several studies have highlighted a significant decline in soil pH (Bai et al., 2020; Lv et al., 2020), which is already notable not only in the topsoil (0–30 cm) but also in the subsoil, as far down as 3 m (Lv et al., 2020). As soil acidification strongly affects the bioavailability and mobility of heavy metals in greenhouse vegetable production systems (Kashem and Singh, 2001), heavy metals may be leached into the groundwater or taken up by vegetable crops. In addition to pH, soil organic matter (SOM) concentrations also affect the availability and mobility of trace elements (Holm, 2003; Xu et al., 2013), with SOM levels largely affected by residue management. However, to avoid the spread of plant diseases, greenhouse vegetable residues are not incorporated into soils (Ren et al., 2014; Zhao et al., 2020). Furthermore, as the incorporation of maize straw or other organic residues in addition to chicken manure is uncommon, greenhouse soils are often depleted in SOM compared to adjacent arable fields (Li et al., 2017; Liu et al., 2018). Moreover, greenhouse soils often have SOM concentrations of approximately 1% in the topsoil (Liu et al., 2018), which lowers the retention capacity not only for macro- but also for micronutrients and increases the risk of nutrient leaching. To compensate for leaching losses, farmers usually apply the above crop demand rates for micronutrients, which may also lead to the further deterioration of greenhouse vegetable soils.

The contamination of agricultural soils with heavy metals negatively affects plant growth and the soil microbiome, and is hazardous for food safety and human health (Wang et al., 2019). If the concentrations of heavy metals, such as copper (Cu), zinc (Zn), nickel (Ni), iron (Fe), and molybdenum (Mo), which are also important micronutrients, exceed plant demands, they become a serious threat to the environment (Chen et al., 2014). Although other heavy metals, such as lead (Pb), chromium (Cr), mercury (Hg), cadmium (Cd), and arsenic (As), are not essential for plant growth, they can negatively affect soil function, plant growth, and human health (Edelstein and Ben-Hur, 2018; Vardhan et al., 2019). The main sources of heavy metals in soils are phosphate and organic fertilizers, pesticides, and fungicides, although other sources include major industries, mining agglomerations, and atmospheric deposition (Timothy and Tagui Williams, 2019). Previous studies have shown that heavy metals accumulate at high concentrations in agricultural soils near metal mines (Kong et al., 2018; Pu et al., 2019; Xu et al., 2013). With an increase in the atmospheric deposition of heavy metals due to industrialization and urbanization, it is unsurprising that heavy metal concentrations in agricultural soils in China have increased in recent decades (Hu et al., 2014). The average atmospheric deposition fluxes of Cr, Ni, As, Cd, and Pb for typical agro-ecosystems in China are 8.3, 4.25, 2.02, 0.73, and 13.07 μg m^{-2} day^{-1}, respectively (Zhang et al., 2018). However, whether the soils of greenhouse vegetable production systems, wherein the soil is protected by a roof, have lower heavy metal concentrations than those of open fields has yet to be investigated. To date, previous studies have only considered fertilizers and pesticides as sources of heavy metals in greenhouse soils and adjacent arable fields (Bai et al., 2015; Hu et al., 2017b; Li et al., 2017), and the literature has yet to consider irrigation water or atmospheric deposition as additional input pathways for heavy metals.

The main objectives of this study were as follows: (i) to quantify the magnitude and vertical distribution of heavy metals and nutrients along the soil profile (0–3 m soil depth) in greenhouses of different ages as compared to adjacent open fields grown with maize (summer crop) and wheat (winter crop) in rotation; (ii) to assess the level and drivers of heavy metal pollution in the soil of greenhouses and identify the soil factors that promote heavy metal mobility in greenhouse soils compared to adjacent maize-wheat fields.

2. Materials and methods

2.1. Description of soil sampling region and site selection

The study area is located in the region of Shouguang County, Shandong Province (36°55′02″ to 36°58′59″ N, 118°43′37″ to 118°46′45″ E; Fig. S1a), which is a pioneering area for greenhouse vegetable production in China. The region has a typical continental monsoon climate, with a mean annual air temperature and precipitation (30 years average) of about 12.4 °C and 592 mm, respectively (Liang et al., 2015). Analyses of publicly available wind direction data showed that the prevailing daily mean wind directions in the last 11 years were distributed as follows: north (25%), south (15%), southwest (12%), southeast (11%), and northeast (7%) (https://lishi.tianqi.com/shouguang/index.html). It should be noted that southerly winds (annual mean: 38%) predominate in the wet summer period with monsoon rains, while northerly winds (annual mean: 32%) mainly occur during the dry winter. Soils in our study regions were mainly sandy loam in texture (on average 57% sand, 39% silt, and 4% clay; Table S2).

The study sites were selected in close cooperation with scientists from Qingdao Agricultural University and as a result of discussions with local farmers to ensure that the management of greenhouses and adjacent field sites chosen were typical for the study region. Sites were selected using criteria including crop growth, crop management practice, and the year of greenhouse establishment (see also Supplementary Information). With regards to the latter, we used a time series of remote sensing images (using Google Earth) to identify the year of greenhouse establishment in former arable fields. Once identified, we confirmed the findings on greenhouse establishment and management in discussion with landowners. In the study region, tomatoes are mainly produced during two cropping seasons per year. Typically, as farmers specialize, greenhouses built in the last 5–20 years were only used to grow tomatoes. In our study, we sampled the soils of greenhouses and adjacent open arable fields in five villages (Yangjiazhuang, Luojiazhuang, Houtuan, Beiling, and Shaojiazhuang village; Fig. S1a) in Shouguang County. In total, 60 greenhouses and 20 adjacent arable fields were sampled, or 12 greenhouses (four in each class of 5, 10, and 20 years since establishment) and four maize-wheat fields per village. Information on land use history, fertilization regimes, soil, and crop management was obtained from individual farmers. The geographical coordinates of each solar greenhouse and arable field were recorded using GPS (Garmin Colorado 300; USA).

2.2. Management of arable land and greenhouse vegetable production

According to our survey, farmers usually grow tomatoes in greenhouses twice a year. For example, tomatoes are transplanted in early January and harvested in mid-June (winter-spring season), followed by a fallow period of 4–6 weeks before a new cropping season starts in early August. The final harvest takes place towards the end of December (autumn-winter season). Conventional flooding (furrow) irrigation is the primary irrigation method used in the region by all farmers in the study area. For greenhouse vegetable production, the total amount of irrigation water used is approximately 1400 mm per year. The total annual N, P, and K inputs in the form of chemical fertilizer are approximately 1000, 700, and 800 kg N/P/K ha^{-1}, respectively. In addition, 800 kg N ha^{-1} in the form of organic fertilizer was added (Fan et al., 2014; own survey). Compound fertilizers, urea, and chicken manure are the most common fertilizer types used by farmers. Chicken manure is usually applied in combination with synthetic fertilizer as a basal fertilizer before transplantation. Moreover, synthetic fertilizers are applied as top dressings with flooding irrigation during the vegetable-growing period. In the study region, the dominant crop rotation on arable land was wheat during the winter season (mid--October to the end of June), followed by maize in the summer season (June to September). The total amount of irrigation water is approximately 300 mm (mainly for wheat), whereas the total fertilizer N application rate is approximately 500 kg N ha^{-1} per year for both winter wheat and summer maize (Zhang et al., 2019).

2.3. Sampling and analysis of soil, fungicide, pesticide, fertilizer, and irrigation water

Soil samples from six different depths (0–30, 30–60, 60–90, 90–150, 150–210, and 210–300 cm) were collected during the fallow period from July 23 to August 13, 2018. Soil samples were obtained using a soil auger (3.5 cm diameter) from a soil depth of up to 90 cm. For deeper soil layers (90–300 cm), a gasoline engine drill with an inner plastic tube (Transdrill Inc., Canada) was used. At each greenhouse, soil samples for depths down to 90 cm were taken along a grid, which consisted of nine individual sampling points. Soil samples from the three sampling points were pooled for individual soil layers (Fig. S1b). Because of the low ceiling of greenhouses towards the south-facing side (60–80 cm only, while the central part is 3 m high), the sampling of soils from 90 cm down to 300 cm depth was restricted to the central area of the greenhouse (a schematic representation of the soil sampling is shown in Fig. S1b). A similar soil sampling strategy was followed for adjacent maize-wheat fields, although three soil pits were dug to retrieve soil samples from 90 to 300 cm.

Following the removal of stones and roots, soil samples were air-dried at ambient temperature and sieved through a 2-mm mesh. For soil mineral N determination, subsamples were extracted with a 1:10 soil:$CaCl_2$ solution (0.01 M) and the soil suspension was shaken on a rotational shaker for 30 min. Following filtering, the NO_3^--N and NH_4^+-N concentrations were measured using an autoanalyzer (AA3; Nordstadt Hamburg, Germany). Soil Olsen-P was extracted using a 1:20 soil: $NaHCO_3$ solution (0.5 M at pH 8.5) for 30 min. Following extraction, the P concentrations were determined with an ultraviolet spectrophotometer (722s; Yidian, China) at a wavelength of 880 nm. Available soil K was extracted in a 1:10 soil:NH_4OAC soil suspension (1.0 M, pH 7.0) for 30 min, and the K concentrations in the filtrate were analyzed using a flame photometer (FP6431; Yidian, China). The pH and EC were measured in a 1:5 soil:water solution using a pH meter (FE20; Mettler Toledo, Switzerland) and an EC meter (FE30; Mettler Toledo, Switzerland), respectively. The heavy metal concentrations were analyzed using an acid digestion method, determined by digestion of reverse aqua regia (Lu et al., 2007), with 6 mL of a mixed solution of HNO_3 (68%) and HCl (38%) (volume ratio of 3:1). The heavy metal concentrations were measured using inductively coupled plasma-optical emission spectroscopy (7300DV; PerkinElmer, USA). The standard soil materials (GBW07403 and GBW07451) were used for quality control in the analysis of heavy metals, the detected values of Cr, Pb, Cu, Zn, and Mn in the samples were within their certified concentration ranges. Soil bulk density was determined using the method described by Blake and Hartge (1986). The soil texture was determined using a laser grain-size analyzer (Mastersizer, 2000; Malvern, England).

To identify potential sources of heavy metals in soils, eleven different fungicide products, three pesticides, and six chemical and organic fertilizers commonly used by local farmers were sampled. In addition, we also analyzed the irrigation water of nine different groundwater wells that access water from aquifers at depths of 80–150 m. All individual samples were measured at least in triplicate, using the methods outlined above.

2.4. Assessment of soil pollution

To assess soil pollution by heavy metals, we calculated a pollution index. The monomial pollution index (P_i) was calculated using the following equation (National Environmental Protection Agency of China, 2006):

$$P_i = C_i/S_i$$

where C_i (mg kg^{-1}) is the measured concentration of a metal in a soil sample and S_i (mg kg^{-1}) is the standard concentration of a metal in soils, which is commonly found in a specific region of China. With regards to the latter, we referred to the environmental quality evaluation for farmland of greenhouse vegetable production in China (National Environmental Protection Agency of China, 2006) for standard metal concentrations in soils of the Shandong region. In our study, $P_i \le 1$ indicates no contamination, $1 < P_i \le 2$ indicates low contamination, $2 < P_i \le 5$ indicates moderate contamination, and $P_i > 5$ indicates severe contamination with a given metal.

From this P_i, we calculated a pollution index using the approach described by Nemerow (1974), that is, the Nemerow pollution index (P_N). The following formula was used:

$$P_N = \sqrt{\frac{P_{im}^2 + P_{ia}^2}{2}}$$

where P_N is the Nemerow pollution index, P_{im} is the maximum monomial pollution index value of a metal, and P_{ia} is the average monomial pollution index for the study region. A $P_N \le 0.7$ indicates safe soils (unpolluted), $0.7 < P_N \le 1.0$ borderline safe soils, $1.0 < P_N \le 2.0$ low pollution, $2.0 < P_N \le 3.0$ moderate pollution, and $P_N > 3.0$ severe pollution (Nemerow, 1974).

2.5. Statistical analysis

Statistical analyses were conducted using SAS 8.2 (SAS Institute Inc., Cary, NC, USA). One-way and two-way ANOVA with a least significant difference test (LSD) ($p < 0.05$) were conducted to assess the effect of cultivation years and soil depth on the results. Correlation analysis was applied to reveal the relationships among soil nutrients, electrical conductivity (EC), pH, and heavy metals using Origin software (2020b, OriginLab). Structural equation models (SEM) were used to further analyze the relationships among the concentrations of soil mineral N, Olsen-P, available K, pH, EC, sand content, and heavy metals. SEM analyses were carried out using AMOS 22.0 (Amos Development Corporation, Chicago, IL, USA).

3. Results

In this study, the soil profiles of 60 greenhouses and 20 adjacent arable fields cropped with a rotation of summer maize and winter wheat in five villages (Yangjiazhuang, Luojiazhuang, Houtuan, Beiling, and Shaojiazhuang village) (Fig. S1a) from Shouguang County, Shandong Province, China, sampled to a soil depth of 3 m, were evaluated. We distinguished between six soil layers (0–30, 30–60, 60–90, 90–150, 150–210, and 210–300 cm).

3.1. Concentrations of nutrients and heavy metals along the soil profile

The contents of the heavy metals Cr and Pb were generally and significantly lower in the topsoil layer (0–30 cm) in greenhouse soils than in soils of adjacent arable fields, with contents decreasing with time since the establishment of the greenhouse (Fig. 1a and b). The concentrations of the heavy metals Cd and Ni were below the detection limit (Fig. 1c and d). In contrast to Cr and Pb, the contents of the micronutrients Cu, Zn, and Mn (Fig. 1e, f, h), as well as of the macronutrients

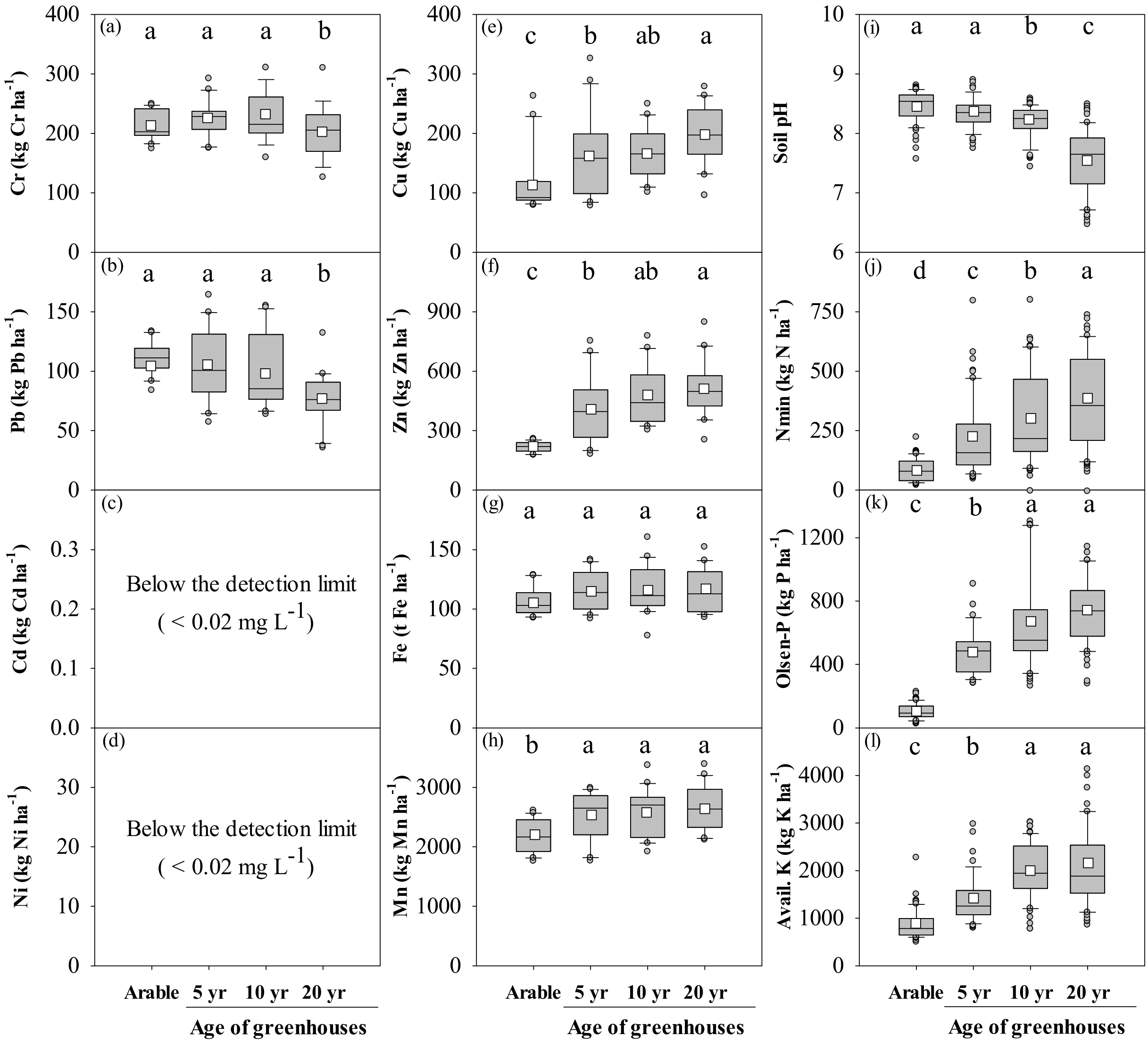

Fig. 1. Average contents (kg ha^{-1}) of the heavy metals Cr, Pb, Cu, and Zn and of the nutrients N (mineral N or Nmin), P (Olsen-P), K (available K), Fe, and Mn (plant available) in the topsoil (0–30 cm) of greenhouses of varying age and in adjacent arable fields ($n = 20$). Different letters refer to significant differences in element contents between the topsoil of arable fields and greenhouses of different ages ($p < 0.05$). The boundaries of the boxes indicate the first and third quartiles; lines and squares within the box represent the median and mean contents, respectively. Whiskers denote the 10th and 90th percentiles, with outliers shown as dots. Error bars represent the standard error of the mean.

Nmin, Olsen-P, and available K (Fig. 1j, k, l), were significantly higher in the 0–30 cm layer of greenhouse soils than in the adjacent arable fields. Moreover, the content of these elements increased with the years of greenhouse establishment. Topsoil pH values were lower in greenhouse soils than in arable fields and also decreased with time since the establishment of greenhouses (Fig. 1i). Similar trends, although often not statistically significant, were found for soil samples taken from the 30–60 cm soil layer (Fig. S2).

Both cultivation years and soil depth were found to have significant effects on the concentrations of heavy metals and soil nutrients (Table S1), except for the interactive effect of the age of greenhouses (year) and soil depth, which was not significant for Pb, Fe, and Mn. Generally, soil micro- and macronutrients along the soil profile (0–300 cm) increased with years of greenhouse establishment, not only in the topsoil, but also in the subsoil (Fig. 2c–i). Across all depths, soil Olsen-P was observed in the range of 3–22 mg kg^{-1} soil dry weight (SDW) in adjacent arable fields and 6–103 mg kg^{-1} SDW, 5–145 mg kg^{-1} SDW, and 15–162 mg kg^{-1} SDW in soils samples from 5-, 10-, and 20-year-old greenhouses, respectively. Similar differences between arable fields and greenhouse soils were also observed for Nmin and available K. However, the concentrations of Cu, Zn, Fe, and Mn in soils of arable fields were found to be lower than those in soils of greenhouses, and the age of greenhouses did not significantly affect concentrations of these elements in soil layers at a depth of >30 cm (Fig. 2c–f).

With regards to the Cr concentrations in the soil, regional differences were observed between sampling locations. Soil samples obtained from open fields in three of the five villages had significantly higher topsoil Cr concentrations (data not shown). These three villages (Houtuan, Beiling, and Shaojiazhuang) are located to the north to northwest of a steel rolling mill, while the other two villages (Yangjiazhuang, Luojiazhuang) are located to the southwest of this mill (Fig. S1a).

3.2. Nemerow pollution index and its correlation to soil physico-chemical properties

The average Nemerow pollution index, an index used to assess the pollution load of soils of greenhouses and arable fields with heavy metals (e.g. Cr, Pb, Cu, and Zn), ranged from 0.26 to 0.38 in the sites sampled (Fig. 3). The highest values were calculated for the topsoil, whereas the subsoil values were up to 30% lower. Notably, the index

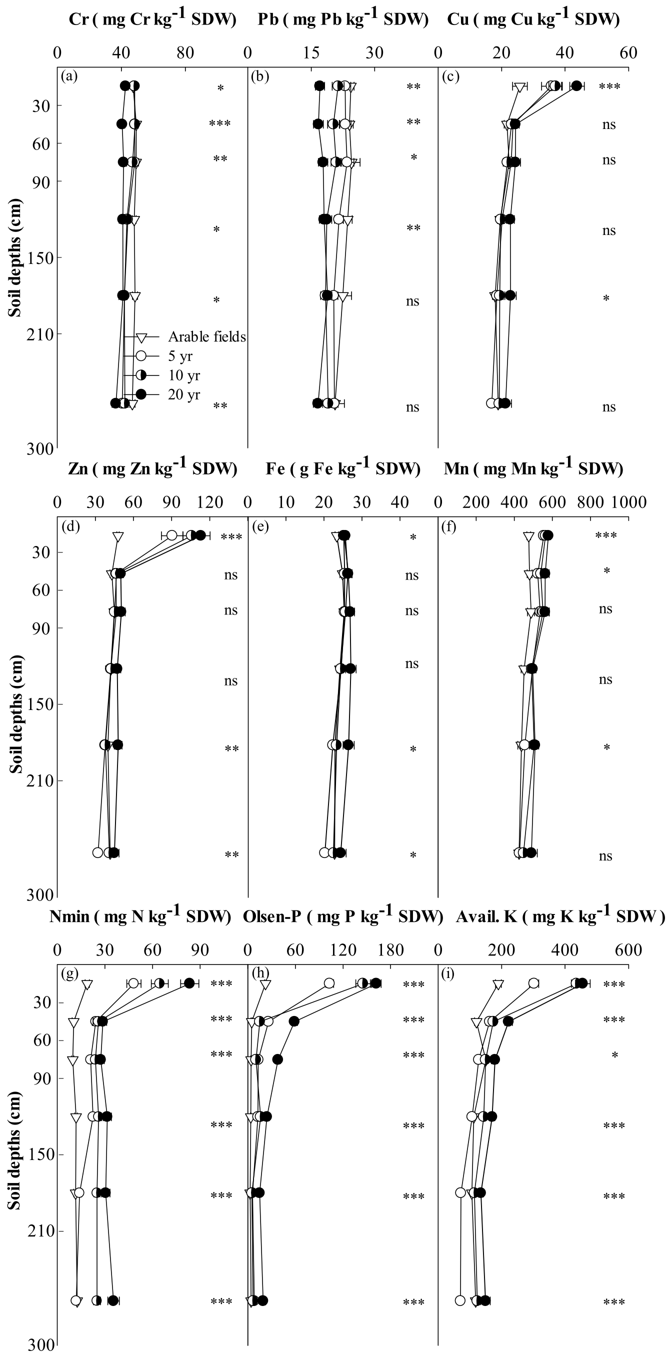

Fig. 2. Mean ± standard error (SE) concentration of heavy metals and soil nutrients in different soil depths for soils of greenhouses (differentiated age since establishment of the greenhouses) and adjacent arable fields (n = 20 for each group and soil depths). *, **, and *** indicate significant differences at p = 0.05, 0.01 or 0.001 comparing the soils of arable fields and greenhouses (all age classes) for different soil layers. SDW, soil dry weight.

was lower for greenhouse soils than for arable field soils. Moreover, the index values declined with increasing years since the establishment of greenhouses (Fig. 3).

Across all greenhouse and arable soils, soil Pb, but not Cr concentrations, were found to be negatively correlated with Nmin and Olsen-P concentrations and positively correlated with soil pH (Fig. 4). In contrast, the soil concentrations of Mn, Cu, and Zn were significantly positively correlated with soil Nmin, Olsen-P, available K, and EC, and negatively correlated with soil pH and sand content (Fig. 4). It should be noted that soil heavy metal concentrations did not correlate with any of the investigated soil physicochemical parameters when the conversion of arable land to greenhouse production was considered (Fig. S3).

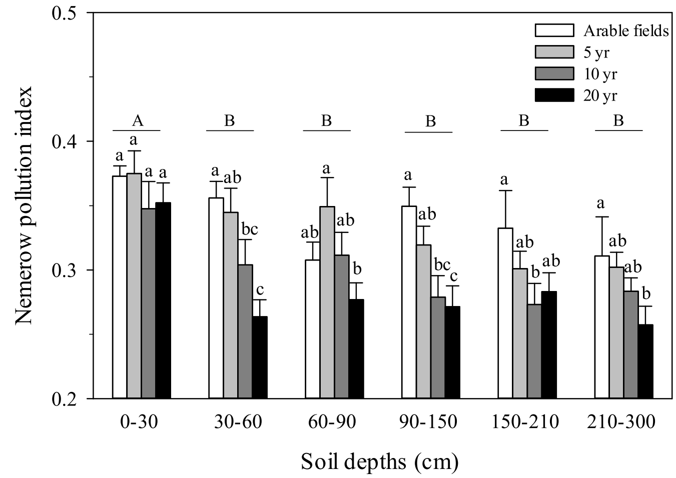

Fig. 3. Mean ± SE values of the Nemerow pollution index, which indicates the pollution load of soils of greenhouses and arable fields with heavy metals (e.g. Cr, Pb, Cu, Zn), for different soil depths and age of greenhouses since establishment (n = 20 for each group). Different capital letters represent significant differences among soil depths. Different lower case letters indicate significant differences between the soils of greenhouses (differentiated by age since establishment of the greenhouse) and arable fields.

3.3. *Concentrations of heavy metals in fertilizers, fungicides, and pesticides*

The concentrations of Cr in the fungicides ranged from 0 to 47 mg kg^{-1}, with an average of 15 mg kg^{-1} (Table S4). The average concentrations of Cr, Pb, Cu, Zn, Fe, and Mn were 15, 151, 27847, 2735, 1423, and 5616 mg kg^{-1} in fungicides; 3, 0, 24, 18, 189, and 54 mg kg^{-1} in pesticides; and 25, 88, 68, 370, 4973, and 351 mg kg^{-1} in fertilizers, respectively (Table S4). The concentrations of heavy metals in the irrigation water were below the detection limit in all samples. The total annual inputs of heavy metals to the soils of greenhouses due to the application of fungicides, pesticides, organic, and synthetic fertilizers are provided in Table S5.

Structural equation modeling (SEM) was used to identify the drivers and factors influencing the heavy metal concentration in the soils in the greenhouse and arable soils studied. With regards to the concentrations of Cr and Pb in the soil, SEM only explained 21% (Cr) and 9% (Pb) of the observed variability, respectively. Only soil texture ("Sand," Fig. 5a) significantly affected the soil Cr concentrations. In addition, for the other heavy metals (i.e. Cu, Zn, Fe and Mn), soil texture was identified as an important factor affecting its concentrations; with an increasing "Sand" texture, the concentrations of these heavy metals was reduced. However, in contrast to Pb and Cr, significant correlations were also observed for Cu, Zn, Fe, and Mn with soil Olsen P concentrations and soil pH, indicating that the soil concentrations of these elements were likely affected by fertilization and fungicide/pesticide management. For example, differences in the Fe concentrations of the topsoil (0–30 cm) were mainly correlated with differences in sand content ($\beta = -0.64$, $p < 0.001$; Fig. 5e), soil mineral N (Nmin) concentrations ($\beta = -0.40$, $p < 0.01$), and soil pH ($\beta = -0.27$, $p < 0.01$). Site differences in soil Mn concentrations were best described by differences in sand content ($\beta = -0.40$, $p < 0.001$; Fig. 5f) and soil pH ($\beta = -0.31$, $p < 0.05$).

4. Discussion

To our knowledge, this study is the first to assess the effects of time since the establishment of greenhouses on the concentrations of heavy

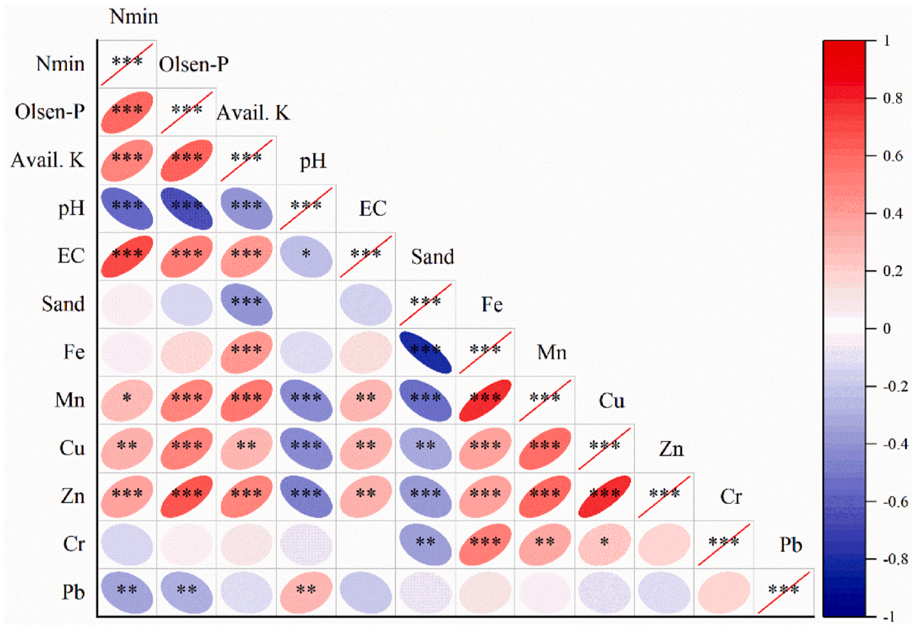

Fig. 4. Correlation matrix showing the relationship between soil parameters and soil heavy metal and soil nutrient concentrations for the soil layer at 0–30 cm. All data were used for this analysis (n = 80). Red and blue indicate positive and negative correlations, respectively. *, **, and *** indicate significant effects at p = 0.05, 0.01, and 0.001. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

metals in soil in previously arable fields. A total of 60 greenhouse soils to a depth of 3 m were sampled in five villages in Shouguang County, Shandong Province, China, wherein tomatoes had been continuously planted for a range of years, from 5, 10, to 20 years. Moreover, soils from 20 adjacent arable fields managed in rotation, with maize production in summer and wheat production in winter, were also analyzed. Surprisingly, in contrast to other reports (Kong et al., 2018), we found that in the study region, the soil concentrations of the heavy metals Pb and Cr were higher in arable fields than in greenhouses. However, in contrast, the soil concentrations of heavy metals and important plant micronutrients Cu, Zn, and Mn were found to be significantly higher in greenhouse soils than in the soils of arable fields. The same was observed for the concentrations of mineral N, Olsen-P, and available K.

4.1. *Pb and Cr concentrations are lower in greenhouse soils than in arable soils*

In our study, the topsoil Cr and Pb concentrations were significantly higher in adjacent arable fields than in greenhouses (Fig. 1 a, b and Fig. 2 a, b), which is in sharp contrast to other studies (Bai et al., 2010; Kong et al., 2018; Wang et al., 2018b). As shown in Fig. S1a, out of the five villages where greenhouses and arable soils were sampled, three villages were located a few kilometers north and two villages were located a few kilometers south of a steel rolling mill. Such industries are known to emit Cr and Pb (as well as Fe and Mn) into the atmosphere via waste gases and dust emissions (Kong et al., 2018; Pu et al., 2019). In our study, the highest Cr concentrations were found in topsoil samples from the three northern villages, whereas soil Cr concentrations at sampling sites in the south were significantly lower. This difference was attributed to the seasonal shifts in wind directions, with southerly winds dominating during the wet monsoon period from April to September and northerly winds dominating during winter. However, during the monsoon season, the atmospheric deposition of Cr and Pb will be higher in the vicinity of the steel mill as in winter, as wet deposition likely dominates the total atmospheric deposition during the monsoon season. In contrast, during the dry winter season, dry deposition prevails and pollutants may be transported over longer distances. Similar observations of increased soil concentrations of Cr and Pb close to steel mills have been previously reported by Qing et al. (2015), who analyzed urban soils in Anshan, a city with a large steel plant in northeast China. In addition, Yang et al. (2018) reported an elevated Pb and Cr contamination in the arable soils of a rural area close to Tangshan, China, due to emissions from nearby steel mills. As a result, we concluded that the lower Pb and Cr topsoil concentrations of greenhouses compared to adjacent arable fields resulted from the protective function of the greenhouse roof, which shelters soils from atmospheric deposition of heavy metals.

4.2. *The concentrations of micro- and macronutrients were higher in greenhouse soils compared to adjacent arable soils*

The concentrations of micro- and macronutrients in the soils of greenhouses were significantly affected by the time passed since the establishment of the greenhouses (Fig. 1). The topsoil concentrations of the micronutrients Cu and Zn were 2- to 3-fold higher in greenhouses than in arable fields (Fig. 1 e, f). In addition, higher topsoil concentrations of Fe and Mn were observed in 20-year-old greenhouses compared to arable fields (Fig. 1g and h). The greenhouse effect on the concentrations of these micronutrients, which are also heavy metals, can be explained by the significantly higher rates of the application of fertilizers, pesticides, and fungicides in greenhouse vegetable production systems compared to that in open field crop production (Table S4). However, the atmospheric deposition of Fe and Mn, which are also released by steel mills, partly obscures the results. Nevertheless, the close positive correlation of the soil Fe and Mn concentrations with those of Cr, Cu, and Zn (Fig. 4) indicated that fertilizer and pesticide/fungicide management in greenhouse systems are not only pivotal for the soil concentrations of Cr, Cu, and Zn, but also for those of Fe and Mn. Except for Cu and Zn, the concentrations of the heavy metals did not vary significantly with soil depth. For both elements, the average soil concentrations were 1–3 times higher in the 0–30 cm than in the 30–300 cm soil samples. The subsoil concentrations of Cu, Zn, Cr, and Pb in this study were comparable to those found for open fields in East China, as reported by Chen et al. (2016).

The concentrations of macronutrients in the topsoil, that is, those of mineral N, Olsen-P, and available K, were generally higher in

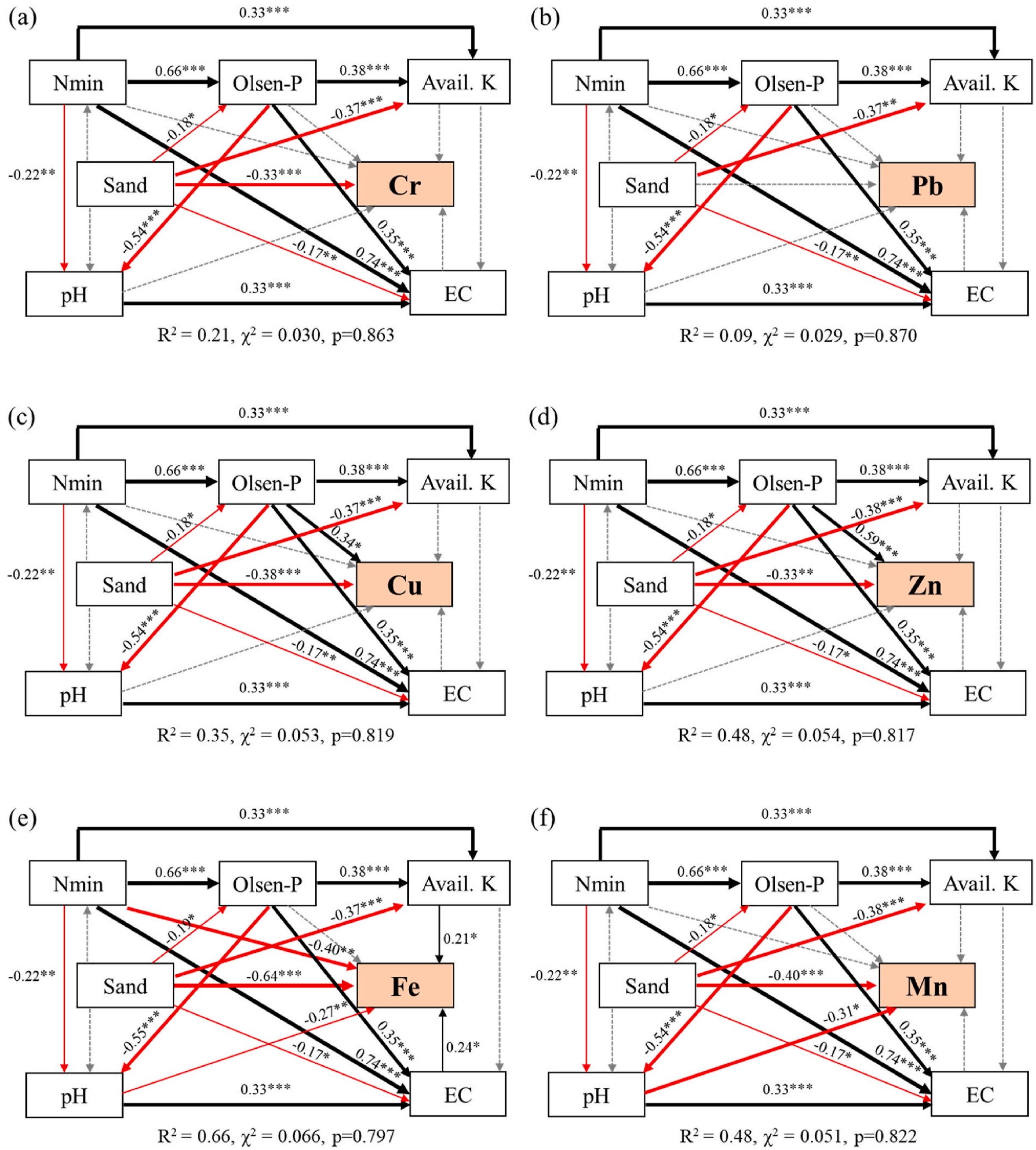

Fig. 5. The results of SEM to assess the effects of soil mineral N (Nmin), Olsen-P, and available K (Avail. K) concentration, pH, electric conductivity (EC), and soil texture (sand content) on the concentration of heavy metals in topsoil (0–30 cm) (n = 80). Solid black and red arrows represent significant ($p < 0.05$) positive and negative effects, respectively, while gray dashed lines indicate insignificant pathways. Arrow widths are proportional to the strength of the relationship. The numbers near the lines are standardized path coefficients, which show the significance of the variables in the model. R^2 represents the amount of variation of the soil heavy metal explained by all paths from each given panel. *, **, and *** indicate significant effects at p = 0.05, 0.01, and 0.001. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

greenhouse soils than in soils of arable fields, with these concentrations increasing significantly with the passing of time since the establishment of the greenhouses (Fig. 2). This observation is in line with previous studies on the concentrations of macronutrients in greenhouse soils, which also reported time-dependent increases in macronutrient concentrations in greenhouse soils (Bai et al., 2010; Chen et al., 2016; Wang et al., 2018a). The average concentrations of Nmin, Olsen-P, and available K in the topsoil (0–30 cm) of 5- to 10-year-old greenhouses were in the range of 48–65, 103–145, and 303–436 mg kg^{-1}, respectively, which are comparable to the ranges reported by Lv et al. (2020) and Chen et al. (2016) for greenhouses elsewhere in China. It should be noted that Olsen-P concentrations were >100 mg P kg^{-1} SDW, above the recommended critical values for achieving optimum yields (recommendation: 46–58 mg P kg^{-1} SDW) (Yan et al., 2013).

4.3. *Nutrient imbalances in greenhouse soils*

In greenhouse vegetable production systems in China, excessive amounts of fertilizers are often used by farmers, along with high rates of irrigation (Qasim et al., 2021; Zhao et al., 2021b). However, as the soil retention capacity for cations is much higher for anions, and as inorganic and organic N fertilizers are rapidly converted to nitrate (NO_3^-), the rates of N leaching are much higher for anions, such as NO_3^-, compared to the leaching rates for the cations P and K. Because farmers are unlikely to change the application ratios of N:P:K, differences in the leaching of N, P, and K result in large nutrient imbalances (Hong et al., 2014; Lv et al., 2019). The optimal N:P:K ratio for tomato production in China is considered to be approximately 1:0.2:1.4 (Yu et al., 2010). In our study, we found that the N:P:K ratios in the topsoil were 1:2.3:6.8 (after 20 years: 1:1.9:5.5). This indicates an oversupply of soils with P:K, and suggests that farmers may choose to not apply P:K fertilizer in the years to come. Similar observations regarding the imbalances of N:P:K in greenhouse soils were previously reported by Lv et al. (2020).

Our findings also highlight the significant leaching of K and P, in addition to N. The subsoil (30–300 cm) concentrations of Nmin, Olsen-P, and available K were 12–35 mg N kg^{-1}, 3–59 mg P kg^{-1}, and 71–221 mg K kg^{-1}, respectively, and, thus, at least a factor of 2–3 lower than the corresponding topsoil concentrations. However, the concentrations of these macronutrients were much higher than in adjacent arable soils and increased with years since the establishment of the greenhouses. Given the large area covered by greenhouses in the study region (8% of total land; Chen et al., 2021) and in agreement with many earlier studies (Lv et al., 2020), our results show that greenhouses are major sources of eutrophication and pollution of groundwater resources. Several studies have shown that the high rates of N fertilization used by farmers for greenhouse vegetable production in conjunction with the removal of plant biomass after harvesting result in a decline in SOM stocks, resulting in high rates of N leaching, the depletion of soil cations, and a decline in soil pH (Alves et al., 2019; Berthrong et al., 2009; Song et al., 2012; Tian and Niu, 2015). This was confirmed in the present study, where significant negative correlations were found between topsoil pH and the concentrations of soil nutrients and heavy metals (except Pb) (Fig. 4).

4.4. *Sources and pollution status of heavy metals in the study area*

Structural equation modeling (SEM) is an *a priori* approach that provides an intuitive graphical representation of complex networks of relationships (Hu et al., 2017a; Lv et al., 2020). SEM was used in this study to identify the causal relationships between soil physical and chemical parameters and soil heavy metal concentrations to understand the driving processes and potential sources of soil heavy metal contamination. Generally, there are two potential sources of heavy metals in agricultural soil: (1) heavy metals derived from the pedogenetic processes of the weathering of parent materials (Tian et al., 2016), and (2) heavy metals derived from anthropogenic activities related to (a) the excessive application of fertilizers, manures, agrochemicals, or heavy metal-contaminated irrigation water, or (b) atmospheric deposition (Bai et al., 2015; Gil et al., 2004).

The SEM results revealed that lighter-textured soils, that is sandy soils, showed lower concentrations of heavy metals. This was remarkable, as in our study, the topsoil sand content only varied within a rather narrow range of 53–61%. The effect of texture on soil heavy metal concentrations may be explained by the increased leaching losses with increasing sand content (Fig. 5). The positive correlation between the topsoil concentrations of Zn, Cu, and Fe to the Olsen-P (Zn, Cu) and Nmin concentrations provides insights into the main source of these heavy metals, that is, agrochemicals and N fertilizers. Moreover, SEM showed no significant effect of soil macronutrients and pH on Cr and Pb, with the exception of sand on Cr. The observation that no significant correlations existed between soil physicochemical parameters, such as pH, EC, or nutrient concentrations (N, P, K), and the topsoil concentrations of the heavy metals Cr and Pb further supports our hypothesis that these elements originate from atmospheric deposition.

The precise evaluation of the levels of heavy metals in soils is of great importance for pollution control. Our results show that the monomial pollution index was significantly higher in the topsoil than in the subsoil (Table S3). However, the Nemerow pollution index, which is used to calculate the pollution load of soils with multiple heavy metals (Nemerow, 1974), was below 0.7 for all sampling sites in our study region. In other words, the concentrations of heavy metals in the topsoil did not exceed critical levels for food production in China (Fig. 3). That being said, it is worth noting that other countries use lower thresholds for heavy metal pollution (Table S6). However, this is clearly a positive finding, as the mean Nemerow pollution index of farmland in China is currently approximately 2.15, indicating a severe contamination of soils with heavy metals (Yuan et al., 2021).

5. Conclusions

By comparing the soils of arable fields and intensively managed vegetable greenhouses in the study region, we found that greenhouse vegetable production has resulted in strong increases in soil nutrients and heavy metals. However, the contamination of greenhouse soils with heavy metals in the study region appears to remain below the level considered to jeopardize food production, most likely as a result of the sandy texture of the soils studied. In addition, a significant shift in the heavy metals (and nutrients) downward in the soil was also observed. Analyses of irrigation water, which in the study region was sourced from aquifers at 80–150 m soil depths, did not show increased heavy metal concentrations; therefore, it is likely that heavy metals continue to accumulate in the deeper soil and sediment layers. Although heavy metal sources are mostly linked to the use of agrochemicals and fertilizers, compelling evidence for atmospheric deposition was found for Cr and Pb in this study, likely due to emissions from a nearby steel mill.

Credit author statement

Li Wan: data acquisition and analysis, writing of the first draft, reviewing, and editing. Klaus Butterbach-Bahl and Shan Lin: study design and conceptualization, funding acquisition and project administration, supervision, reviewing, and editing. Haofeng Lv, Waqas Qasim, Longlong Xia, Zhisheng Yao, Xiadong Ding, and Xunhua Zheng: data analysis and review.

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

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