



Multivariate and geostatistical analyses of heavy metal pollution from different sources among farmlands in the Poyang Lake region, China

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Abstract

Purpose Intensive urbanization and industrialization activities have caused the continuous discharge of heavy metals into the soils of China's Poyang Lake region, where they pose a major threat to human health. Yet, the spatial characteristics of these heavy metals in farmland soils and their pollution sources in this region remain unclear.

Materials and methods We collected 115, 84, 26, and 987 farmland soil samples with heavy metal pollution from different sources (industrial and mining enterprises, sewage irrigation, urban, and general farming) in the Poyang Lake region, respectively.

Results and discussion Descriptive statistics revealed that 1.1% of As, 7.3% of Cd, 0.3% of Pb, and 0.2% of Hg concentrations did surpass China's Soil Environmental Quality Management Standard (GB 15618-2018). In addition, 32.8% of As, 74.4% of Cd, 89.2% of Pb, 45.0% of Cr, and 13.7% of Hg concentrations of soil samples exceeded the background soil concentrations of heavy metals in this region, notably Cd and Pb, whose maximum values were respectively 11.64- and 21.47-fold the background values. Compared with general farming areas, the other three pollution sources had significantly higher concentrations of As, Cd, Pb, Cr, or Hg in their soils. The results of principal component (PC) and geostatistical analyses showed that PC strongly related to mainly As and Cr was mainly influenced by natural characteristics while PC closely related to mainly Cd, Pb, and Hg was driven by mainly human activities in farmland soils from the four sources.

Conclusions Factor loadings for the different sources showed As and Cr on the same PCs, and Cd, Pb, and Hg on the other PCs, confirming the association of the heavy metal pollution with its sources.

Keywords Geostatistics · Heavy metal pollutant · Pollution source · Poyang Lake region

1 Introduction

There are two main origins of heavy metals in the soil (Li et al. 2013a): (1) natural characteristics, which represent the concentrations of heavy metals originating from the parent rock substrate, and (2) human activities, including the discharge of sewage around industrial and mining enterprises, irrigation of polluted water resources, discharge of pollutants in and around

cities, and the application of pesticides and fertilizers. Although the original or background concentration of soil heavy metals depends on local natural characteristics, undoubtedly their accumulation is generally attributed to humans (Sun et al. 2013). The heavy metals entering soils may originate from different pollution sources, which broadly consist of four types: industrial and mining enterprise areas, sewage irrigation areas, urban areas, and general farming areas.

Industrial and mining enterprises, such as power generation, mining, smelting, landfill leakage, or fossil fuel (Li et al. 2012; Rodríguez Martín et al. 2013), are the major sources of heavy metal inputs to the environment. Due to constraints on freshwater availability for agricultural irrigation, sewage water has been used instead as an important supplement and alternative water resource during the past three decades in developing countries (Rattan et al. 2002, 2005; Chen et al. 2016; Meng et al. 2016). Heavy metals can also migrate into farmland soils from the atmosphere (via dry and wet settlement)

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and sewage irrigation, most notably Cd, Pb, and Hg (Hani and Pazira 2011; Kim et al. 2015; Zeng et al. 2015; Chen et al. 2016). Urban areas, where most people now live and generate more human activities, can serve as an important indicator of human exposure to heavy metals in the urban terrestrial environment (Nriagu and Pacyna 1988; Li et al. 2013a). Soil heavy metals will usually manifest at high pollutant concentrations (or even higher), arising mainly from modern industries, transportation traffic, and municipal waste in urban areas (Wong et al. 2006; Gallagher et al. 2008). In general farming areas, soil heavy metals can be accumulated due to applications of agrochemicals, fertilizers, biosolids, and composts (Mantovi et al. 2003; Rodríguez Martín et al. 2013). Since related agricultural practices are also intensive, such as animal breeding and husbandry, these practices are also important sources of heavy metals into farmland soils (Rodríguez Martín et al. 2013).

Regardless of their origin or source of pollution, heavy metals can accumulate in plant crops and enter humans through the food chain (Aelion et al. 2009). Better knowledge about the potential hazards of soil heavy metal concentrations with respect to types of pollution sources undoubtedly has implications for utilizing agriculture resources and improving environmental quality (Rodríguez Martín et al. 2006). To do this, multivariate statistical analyses (e.g., Pearson or Spearman correlations and principal component analysis [PCA]) are particularly useful tools (Yu et al. 2018). Geostatistics combined with semivariograms (such as Kriging estimates) offer pronounced advantages in spatial prediction and uncertainty analysis; this approach not only simulates the spatial structure and variation of heavy metals in soil but also visualizes the spatial distributions of heavy metals in soil (Li et al. 2013a; Jiang et al. 2018).

China's Poyang Lake region is a commodity grain base, one that also contains the largest freshwater lake in China (Jiang et al. 2018). Soil metal pollution has occurred in this region because of many contributing factors, including the growing demand for food, the increased development of agricultural land, and the excessive use of agricultural chemicals (Yuan et al. 2011). Worsening the pollution load in some parts of this region is the long-term irrigation of cultivated soil with sewage containing a considerable amount of heavy metals (Ji et al. 2015; Zhang et al. 2017). Unfortunately, few studies have actually examined heavy metal pollution in farmland soils (Yuan et al. 2011; Shao et al. 2016; Zhang et al. 2017; Dai et al. 2018), especially around industrial and mining enterprise areas, irrigation areas, urban areas, and general farming areas. Heavy metal pollution in farmlands threatens the sustainability of food crop production in Poyang Lake region, and it may erect a potential barrier for the international trade of its food products (Sun et al. 2013).

In the present study, we investigated the spatial characteristics of As, Cd, Pb, Cr, and Hg distributions in farmland soils

and their pollution sources in the Poyang Lake region. The specific objectives were (1) to evaluate the spatial distributions in farmland soils of As, Cd, Pb, Cr, and Hg; (2) to assess heavy metal pollution of farmland soils for different sources (industrial and mining enterprises, sewage irrigation, urban, and general farming); and (3) to identify key factors driving the spatial variation in soil heavy metal pollution from different sources.

2 Materials and methods

2.1 Study site

This research was carried out in the Poyang Lake region, Jiangxi Province, China (Fig. 1a). The corresponding geographical coordinates are 114° 29' E–117° 42' E and 27° 30' N–30° 06' N, which include 14 counties (county-level cities or districts) of Nanchang, Shangrao, and Jiujiang City, covering a total area of 2.13×10^4 km². The terrain here is gentle and the elevation is generally < 50 m with dense river networks (Xie et al. 2016). The climate is subtropical-humid, with an annual average temperature of 16–20 °C, annual precipitation of ~ 1500 mm, and an average frost-free period of 240–300 days, all of which together provide favorable conditions for agricultural production (Guo and Jiang 2018). Parent materials are mainly quaternary red clay, river and lake deposits, weathered red sandstone, weathered argillaceous rock, weathered quartzite, and weathered acid crystalline rock (Fig. 1b). The major soil types are anthrosols, ferralsols, and primosols, with a small amount of argosols and gleyosols (Fig. 1c).

2.2 Soil sampling, laboratory analysis, and quality control

A total of 1212 farmland soil samples were collected from representative sites based on administrative regions, topographic conditions, soil types, parent materials, pollution sources, and natural sources from November 2015 to February 2016 (Fig. 1). Their sampling points were randomly distributed based on a 1 × 1-km regular grid, with each grid unit containing at least one sampling location (Fig. 1a). Following this sampling design, we collected 115, 84, 26, and 987 farmland soil samples from industrial and mining enterprise areas (collected where some polluting type of industrial and mining enterprises has occurred), sewage irrigation areas (collected from tillage soils that were/are sewage-irrigated), urban areas (collected from areas surrounding a high density of buildings and roads), and general farming areas (collected from soils inaccessible to industrial and mining enterprises, sewage irrigation, and urban activities), respectively. Each soil sample consisted of five subsample soil cores obtained from a depth of 0–20 cm using a stainless steel

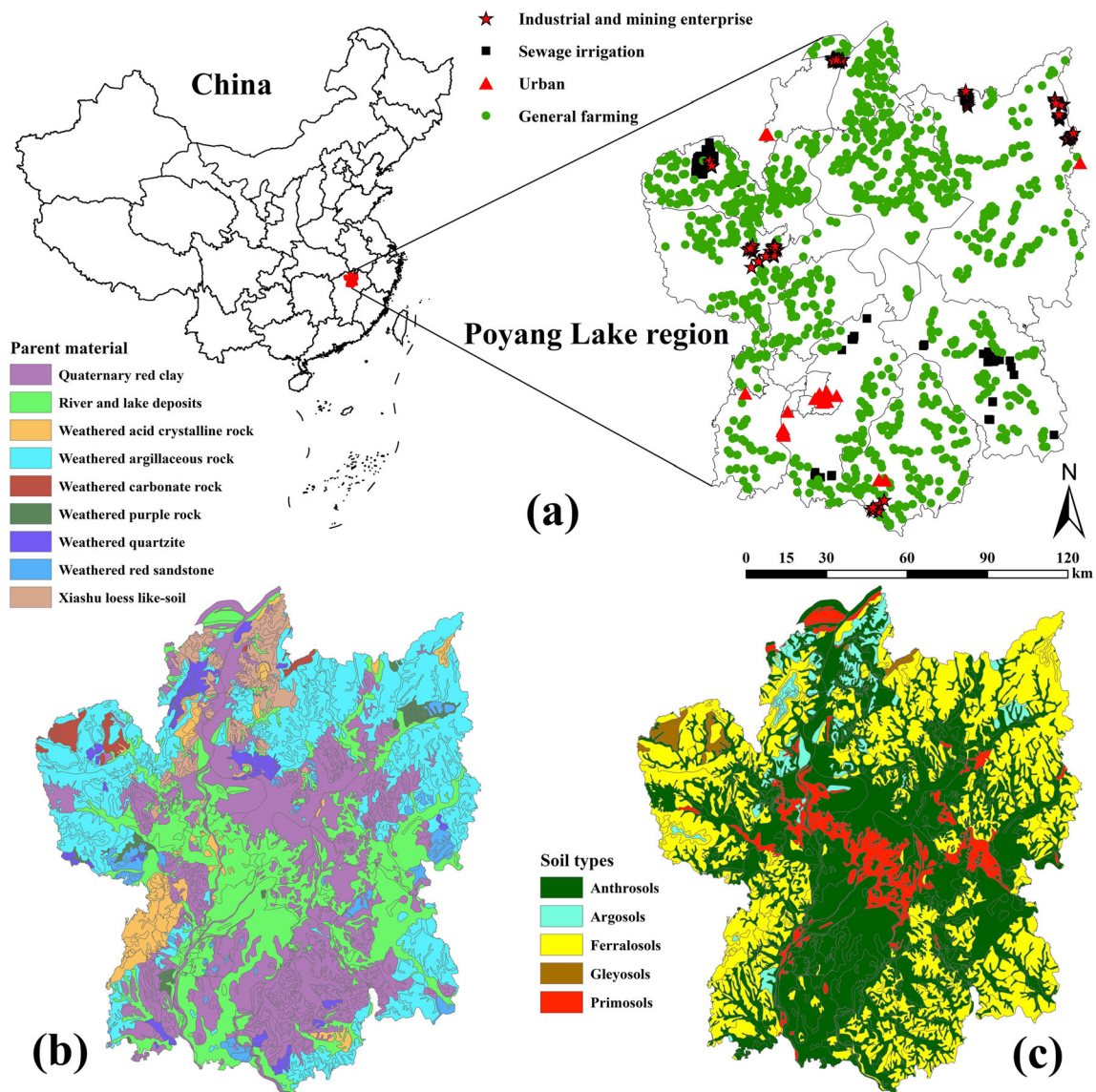


Fig. 1 Sampling locations of farmland soils exposed to different pollution sources of **a** and **b** parent material and **c** soil types in the Poyang Lake region of China

manual augur. The collected soil samples were stored separately in polyethylene bags for later analysis, and the location of all composite samples was recorded by a handheld global positioning system (HD8200X-GPS, Zhonghaida, Guangzhou, China).

Briefly, 0.5–1.0 g of air-dried soil samples were digested with a 2:5:3 mixture of HClO₄–HNO₃–HF (USEPA 1996), and the concentrations of As, Cd, Pb, Cr, and Hg in their extracts determined by inductively coupled plasma atomic emission spectrometry (ICPS-7500, Shimadzu, Kyoto, Japan). Quality control entailed the following: (1) an analysis of 15 random samples and six national standard samples and (2) a random selection of samples to ensure that the mean deviation was less than 3%. Detailed analytical procedures followed the national test standard's instructions for soil in

China, which were discussed by Li et al. (2013a). Soil pH was measured by potentiometry using a soil/water ratio of 1/2.5 (Guo et al. 2018).

2.3 Statistical analysis

Data analyses were performed in SPSS v22.0 software (IBM SPSS, Somers, NY, USA). Descriptive statistical analyses were used to describe the general conditions of soil heavy metal concentrations. One-way analysis of variance (ANOVA) tested for significant differences in the mean soil heavy metal concentrations between the four source types ($p = 0.05$). Pearson or Spearman correlations revealed the associations between soil heavy metal concentrations and pH. PCA was used to reduce the set of original variables and

thereby extract a small number of latent factors (principal components, PCs), to analyze the relationships among heavy metals. If the sample size of PCA > 5 times the number of variables, it is reasonable to conclude that the results are robust and accept (Rosenberg et al. 2000).

2.4 Geostatistical analysis

Geostatistics is currently the widest method used to visualize the soil distributions of heavy metals (Bai et al. 2010; Yu et al. 2018). This method uses field survey sample data from which semivariograms are derived; based on these, an unbiased estimation method for regional variables of sampled and unsampled points is performed (Li et al. 2013a, 2015). Under the second-order intrinsic assumption of stability, the semivariogram is calculated as follows:

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i + h)]^2$$

where $\gamma(h)$ is the semivariogram; h is the step length, namely the spatial interval of sampling points used by the classification to decrease the individual number of spatial distances of various sampling point assemblages; $N(h)$ is the logarithm of sampling points when the spacing is h ; and $Z(x_i)$ and $Z(x_i + h)$ are the values when the variable Z is at the x_i and $x_i + h$ positions, respectively.

The semivariance function was calculated and fitted to theoretical models in GS+ v7 (Gamma Design Software, Plainwell, MI, USA), and the best-fitting models were selected for PCs and heavy metals based on both the maximum determination coefficient and residual sum of squares of the model. Kriging is an established geostatistical interpolation method, which uses the semivariogram to quantify the spatial variation of regionalized variables and also provides the parameters for spatial interpolation. Maps of the heavy metal concentrations were generated via ordinary Kriging interpolation (Bai et al. 2010; Chen et al. 2016; Dai et al. 2018) with the

support of ArcGIS-Geostatistical Analyst (ESRI Inc., Redlands, CA, USA).

3 Results and discussion

3.1 Descriptive statistics of soil heavy metals

The soil pH range was 3.34–8.37, but the vast majority of soil samples (95.41%) were acidic; just 4.59% of them had a pH > 7.0, with only 1.24% having a pH > 8.0. The leading cause for this acidic soil pH is the large amount of fertilizer (especially nitrogen) applied in the study region we investigated (Guo et al. 2018), but some of it is partially attributable to the parent material, soil type, and topography (Jiang et al. 2018). The pH is a critical factor affecting the dissolution, precipitation, adsorption, and desorption of soil heavy metals (Martínez and Motto 2000). Acidic pH can certainly promote the mobility of heavy metals in soil (Rodríguez Martín et al. 2006), but further detailed studies are required to assess where and which heavy metals are accumulated or depleted in the polluted farmland soils.

Exploring the data revealed soil heavy metal concentrations (Table 1) with average concentrations (mg kg^{-1}) of As (9.28), Cd (0.19), Pb (28.51), Cr (62.14), and Hg (0.12), which did not exceed those corresponding China’s Soil Environmental Quality Management Standard. Nevertheless, for a percentage of soil samples, we also found that 1.1% of As, 7.3% of Cd, 0.3% of Pb, and 0.2% of Hg concentrations did surpass China’s Soil Environmental Quality Management Standard (GB 15618-2018); in addition, 32.7% of As, 74.4% of Cd, 89.2% of Pb, 45.0% of Cr, and 13.7% of Hg concentrations exceeded the background value of heavy metal concentrations in the Poyang Lake region (Table 1), notably the majority of those for Cd and Pb. Their maximum concentrations were respectively 11.64 and 21.47 times greater than the background values, indicating human interference underpinned the prominent accumulation of these two metals in soils. A conclusion was also drawn by

Table 1 Descriptive statistics of pH and heavy metal concentrations in farmland soils in the Poyang Lake region of China

| Property | Range (mg kg^{-1}) | Average (mg kg^{-1}) | Standard deviation (mg kg^{-1}) | Coefficient of variation | Standard level (mg kg^{-1}) ^a | Over-standard rate | Over-background value rate ^b |
|----------|-------------------------------|---------------------------------|--|--------------------------|---|--------------------|---|
| pH | 3.34–8.37 | 5.19 | 0.74 | 14.3% | – | – | – |
| As | 1.16–63.10 | 9.28 | 5.08 | 54.7% | 30 | 1.1% | 32.7% |
| Cd | 0.04–2.83 | 0.19 | 0.12 | 63.2% | 0.3 | 7.3% | 74.4% |
| Pb | 1.97–206.00 | 28.51 | 12.59 | 44.2% | 80 | 0.3% | 89.2% |
| Cr | 25.10–125.00 | 62.14 | 13.92 | 22.4% | 250 | 0.0% | 45.0% |
| Hg | 0.02–0.73 | 0.12 | 0.07 | 58.3% | 0.5 | 0.2% | 13.7% |

^a Standard level for heavy metals in agricultural soils in China (GB 15618-2018)

^b Background value by the report on farmland environmental quality in the Poyang Lake region, 1986

Sun et al. (2013) for urban soil in Shenyang, Northeast China. The coefficient of variation for the five heavy metal concentrations increased from 22.4% (Cr) to 63.2% (Cd) and was ranked as follows Cd > Hg > As > Pb > Cr, revealing the marked influence of human activity on both Cd and Hg in soil.

3.2 Relationships of soil heavy metals and pH

Correlation analysis (Table 2) showed that the correlation coefficient between As and Cr concentrations was significant and reached the largest in industrial and mining enterprises, sewage irrigation, urban, and general farming ($r = 0.301, 0.341, 0.253,$ and $0.473,$ respectively), suggesting the two heavy metals may come from similar origins of pollution. Since Cd and Pb versus Hg concentrations (except Hg and Cd in general farming) were significantly related in different sources, the associations of As and Cr versus Hg concentration were not, indicating Hg disparate sources for As and Cr while a similarity of pollution sources for Cd and Pb in the four sources. The correlation analysis results suggested Pb and

Cd concentrations may not be affected by pH in industrial and mining enterprises (Table 2), which is consistent with its main origin likely being past from vehicle emissions and other activities around urban areas (Rodríguez Martín et al. 2006).

Given their positive correlations with pH, the latter may promote Cr accumulation in industrial and mining enterprise soils; Cd, Pb, and Hg accumulation in urban soils; and Cd accumulation in general farming soils. Although more OH⁻ in solution fosters heavy metal precipitation as a hydroxide, 95.41% of soils in the Poyang Lake region were acidic, so the competition between organic matter, iron manganese oxides, and heavy metals in the soil is weakened by the still dominant H⁺ in solution, increasing the concentration of soluble and exchangeable heavy metals (Rodríguez Martín et al. 2006; Naidu et al. 2010). The negative correlation found between pH versus Hg concentration in industrial and mining enterprises, Pb concentration in sewage irrigation and general farming, and As concentration in urban, indicates that those heavy metals are easily precipitated as carbonate in an acid soil environment (Martínez and Motto 2000).

Table 2 Correlations among heavy metals and pH from different sources among farmland soils in the Poyang Lake region of China

| Sources | Heavy metals ^a | As | Cd | Pb | Cr | Hg | pH |
|---------|---------------------------|----------|---------|----------|---------|----------|-------|
| I | As | 1.000 | | | | | |
| | Cd | 0.079 | 1.000 | | | | |
| | Pb | 0.139 | 0.131 | 1.000 | | | |
| | Cr | 0.301** | 0.112 | 0.263** | 1.000 | | |
| | Hg | -0.027 | 0.253** | 0.201* | 0.121 | 1.000 | |
| | pH | 0.101 | 0.105 | 0.086 | 0.531** | -0.315** | 1.000 |
| II | As | 1.000 | | | | | |
| | Cd | 0.339** | 1.000 | | | | |
| | Pb | -0.056 | 0.385** | 1.000 | | | |
| | Cr | 0.341** | 0.148* | -0.181 | 1.000 | | |
| | Hg | -0.015 | 0.151* | 0.452** | 0.085 | 1.000 | |
| | pH | 0.055 | -0.020 | -0.286** | 0.213 | -0.056 | 1.000 |
| III | As | 1.000 | | | | | |
| | Cd | -0.183 | 1.000 | | | | |
| | Pb | -0.177 | 0.370 | 1.000 | | | |
| | Cr | 0.253* | 0.165 | 0.011 | 1.000 | | |
| | Hg | -0.186 | 0.424** | 0.327** | 0.083 | 1.000 | |
| | pH | -0.424** | 0.541** | 0.232* | 0.091 | 0.681** | 1.000 |
| IV | As | 1.000 | | | | | |
| | Cd | 0.137** | 1.000 | | | | |
| | Pb | 0.199** | 0.489** | 1.000 | | | |
| | Cr | 0.473** | 0.209** | 0.134** | 1.000 | | |
| | Hg | 0.062 | 0.058 | 0.093** | 0.046 | 1.000 | |
| | pH | 0.039 | 0.147** | -0.088** | 0.019 | -0.028 | 1.000 |

I industrial and mining enterprises, II sewage irrigation, III urban, IV general farming

* $p < 0.05$; ** $p < 0.01$

^a If the dataset had normal distribution, Pearson correlation analysis was used; otherwise, Spearman correlation analysis was used

3.3 Discriminating natural and human activities in different sources

The way the individual associations of heavy metal elements are distributed in the four different sources (industrial and mining enterprises, sewage irrigation, urban, and general farming) is determined by the PCA method. Kaiser-Meyer-Olkin (KMO) and Bartlett's tests in Table 3 indicate that the data set was reluctantly acceptable for principal component analysis (KMO > 0.5) and passed the significance test (Kaiser and Rice 1974). Based on the eigenvalues > 1, the first two main factors (PC1 and PC2) respectively accounted for 56.16%, 63.84%, 65.89%, and 59.68% of the total variance in the four sources, respectively (Table 4).

In industrial and mining enterprises, PC1, explaining 29.69% of total variance and closely relating to mainly As and Cr, shows positive factor loadings on As and Cr; PC2, explaining 26.47% of the total variance, strongly and positively relates to Cd, Pb, and Hg (Fig. 2a). In sewage irrigation, PC1, explaining 33.27% of total variance, closely relates to Cd, Pb, and Hg; PC2, explaining 30.57% of total variance, strongly relates to As and Cr and shows high positive factor loadings (Fig. 2b). In urban, PC1, explaining 39.89% of total variance, closely relates to Cd, Pb, and Hg; PC2, explaining 26.00% of total variance, closely relates to As and Cr and shows high positive factor loadings (Fig. 2c). In general farming, PC1, explaining 30.40% of total variance, closely relates to Cd, Pb, and Hg; PC2, explaining 29.28% of total variance, strongly relates to As and Cr and shows high positive factor loadings (Fig. 2d).

The factor loading plot (Fig. 2) shows the grouping of heavy metals from the four sources. According to previous studies (Rodríguez Martín et al. 2006; Krami et al. 2013), the association of elements with factors can be indicated by natural characteristics and human activities. In industrial and mining enterprises, sewage irrigation, urban, and general farming, several hotspots of high metal concentration were identified by the geochemical maps, the spatial distribution of PCs (industrial and mining enterprises: PC2; sewage irrigation, urban, and general farming: PC1) indicates that Cd, Pb, and Hg were point source pollutants while other PCs on behalf of As and Cr in the four sources were relatively smooth

(Fig. 3), revealing the obvious artificial inputs of Cd, Pb, and Hg in the study region. The likely sources for this are industrial, including chemical, petrochemical, and agricultural fertilizers (e.g., iron, copper, arsenic, cadmium, stone) activities, as well as the agricultural use of chemical fertilizers and municipal waste. Specifically, the delivery, storage, and handling of raw materials, the process of manufacturing, and the disposal of wastes are the three main stages of most industrial manufacturing processes driving heavy metal pollution (Syms 2004). Sewage irrigation can increase the environmental loading of heavy metals due to the increased organic carbon content and lowered pH, further modifying the ensuing pollution that occurs in soil (Rattan et al. 2005). Urban sites are concentrated areas for human activities, with relatively high population sizes and traffic densities (Li et al. 2013a).

There were also some polluting enterprises found around these hotspot areas. According to previous reports (EPRIJP 1986; Deng et al. 2014), there was a Jiujiang glass chemical fiber plant and oil refinery, a Jinxian dyeing and weaving and food factory, a Lidu winery, and a Poyang chemical fertilizer plant and shipyard, in addition to De'an wineries and building material factories. It is tempting to believe that the Cd, Pb, and Hg in these soils mainly came from the "three wastes" (waste gases, waters, and residues) in areas around the industrial and mining enterprises (Rodríguez Martín et al. 2006; Deng et al. 2014). From 2010 to 2012, the sewage treatment rate in this area was 50.16–57.21%, while the exhaust emission was 4.309–5.369 × 10⁹ t, while the utilization rate of industrial solid waste was only 5.5–14.73% in Shangrao City. Furthermore, the Environmental Protection Department of Jiangxi Province reported in 2017 that 60.68% of urban sewage was untreated, with only 36.36% of industrial solid waste comprehensively utilized. Those greatly increase the concentration of Cd, Pb, and Hg in farmland soils (Bai et al. 2010; Krami et al. 2013; Rodríguez Martín et al. 2013), and some hotspots were identified.

However, this does not necessarily mean that natural characteristics are not important contributors. In fact, neither of these factors can be ignored, although they are not equally important. Nonetheless, soil Cr concentration depends primarily on natural characteristics (Rodríguez Martín et al. 2006, 2008) while the origins of Pb are long well known (automobile exhaust and industrial smog). Across almost all areas of farmland, both As and Cr came from similar origins, while Cd, Pb, and Hg came from the same origins. The PCA and correlation analysis results for the soil heavy metals from different sources further support it (Fig. 2 and Table 2). Lee et al. (2006) reported PCAs of soil heavy metals in three different regions (urban, suburban, and country parks) of Hong Kong, China, which showed soil Cd and Pb in the same PC. Recently, Yu et al. (2018) showed that the origins of soil As and Cr are highly similar in Gaoqing County, China. Rodríguez Martín et al. (2006) also showed that Cd, Pb, and

Table 3 Kaiser-Meyer-Olkin and Bartlett's tests in the principal component analysis in the Poyang Lake region of China

| Index | Kaiser-Meyer-Olkin | Significance of Bartlett's test |
|-----------------------------------|--------------------|---------------------------------|
| Industrial and mining enterprises | 0.569 | 0.000 |
| Sewage irrigation | 0.579 | 0.000 |
| Urban | 0.552 | 0.004 |
| General farming | 0.550 | 0.000 |

Table 4 Total variance explained and matrixes of the principal component analysis loadings of heavy metals in farmland soil in the Poyang Lake region of China

| Sources | Index | As | Cd | Pb | Cr | Hg | % of variance | Cumulative % |
|---------|-------|--------|-------|--------|--------|--------|---------------|--------------|
| I | PC1 | 0.636 | 0.151 | 0.340 | 0.850 | -0.467 | 29.69 | 29.69 |
| | PC2 | 0.177 | 0.550 | 0.669 | 0.036 | 0.736 | 26.47 | 56.16 |
| II | PC1 | 0.025 | 0.587 | 0.884 | -0.117 | 0.723 | 33.27 | 33.27 |
| | PC2 | 0.825 | 0.519 | -0.145 | 0.746 | -0.015 | 30.57 | 63.84 |
| III | PC1 | -0.517 | 0.767 | 0.652 | 0.246 | 0.808 | 39.89 | 39.89 |
| | PC2 | 0.716 | 0.207 | 0.028 | 0.818 | -0.217 | 26.00 | 65.89 |
| IV | PC1 | 0.112 | 0.842 | 0.854 | 0.112 | 0.238 | 30.40 | 30.40 |
| | PC2 | 0.850 | 0.093 | 0.078 | 0.851 | 0.048 | 29.28 | 59.68 |

I industrial and mining enterprises, *II* sewage irrigation, *III* urban, *IV* general farming

Hg have similar origins in agricultural topsoils in Spain. From research elsewhere and the results of this study, we may infer that the association patterns of heavy metal pollutants from different sources seem to be the same in the agricultural soils in China and around the world. Generally, PCs closely related to As and Cr were controlled by natural characteristics, while

PCs closely related to Cd, Pb, and Hg were subject to human activities (Fig. 2 and Fig. 3).

Further, it is difficult to distinguish the effects of each PC. In other words, not all heavy metals were independently distributed on a fixed PC. For instance, Cd and Hg were mainly related to PC1 yet also partly related to PC2 in sewage

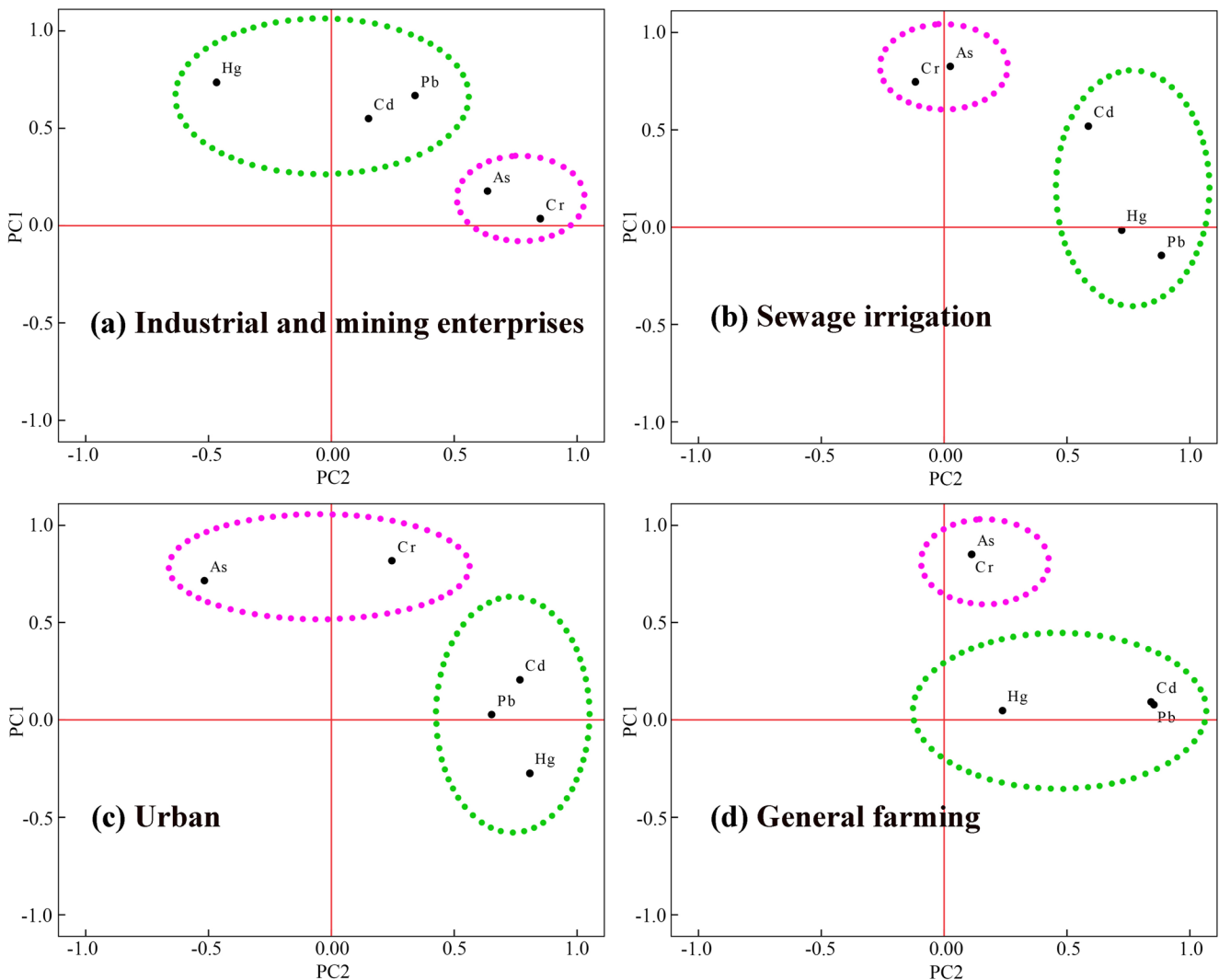
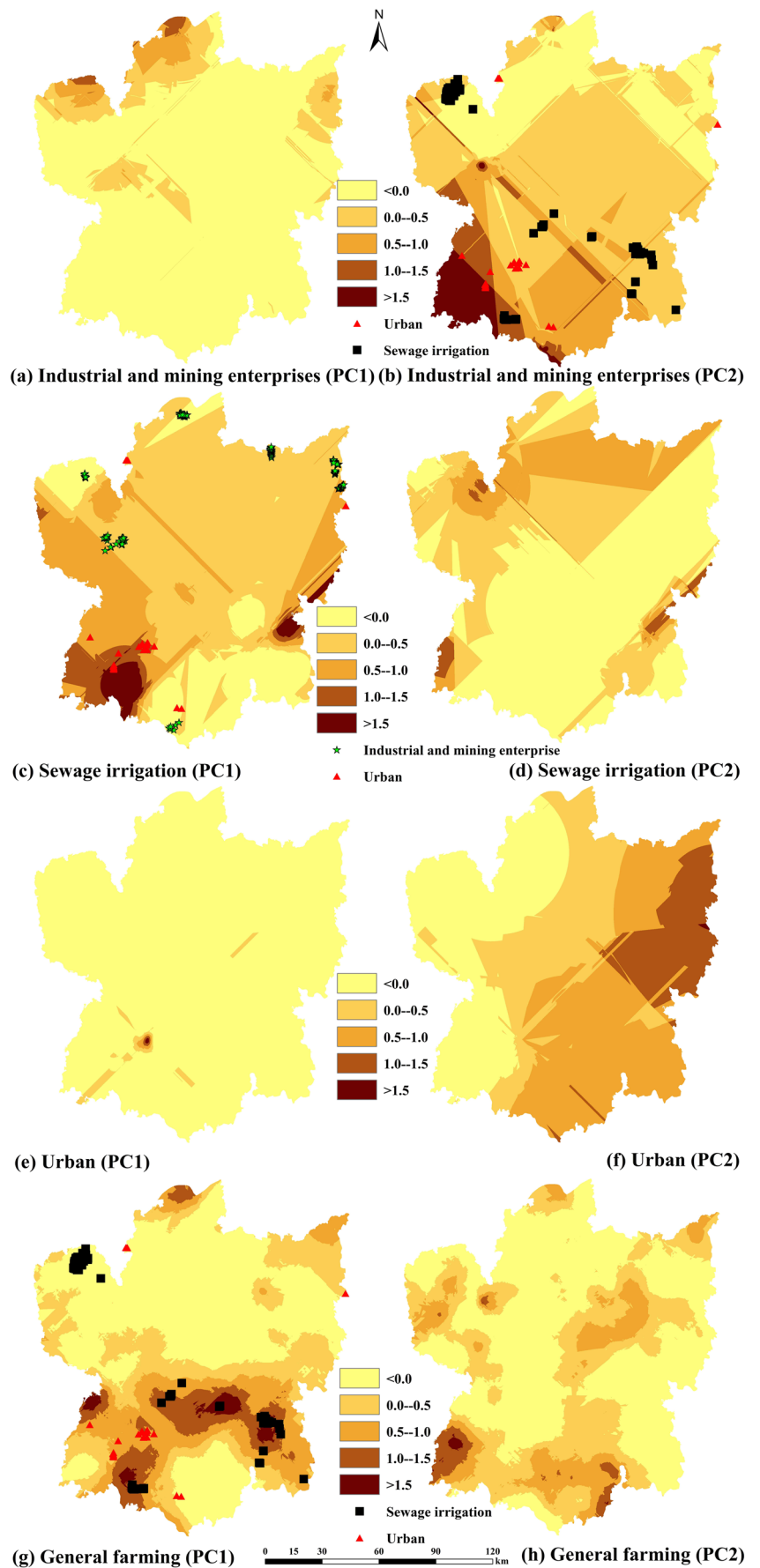


Fig. 2 Factor loadings for the principal components of heavy metal pollution in farmland soils from different sources in the Poyang Lake region of China

Fig. 3 Maps of the principal component factors for heavy metal pollution in farmland soils from different sources in the Poyang Lake region of China



irrigation and general farming, respectively, demonstrating that heavy metals respond synergistically to a variety of factors (Rodríguez Martín et al. 2013; Li et al. 2013b). In addition, the greater Cd concentration may have been due to more fertilizer applied, yet another source may also be the metal-processing industry (Lin 2002). As Table 5 shows, all the As, Cd, Pb, Cr, and Hg soil concentrations were often strongly affected by human activities (Rodríguez Martín et al. 2008). The PCA simply indicates the hypothetical origins of these heavy metals in the soil (natural characteristics, human activity, or mixing).

3.4 Spatial distribution of heavy metals

Table 6 shows the semivariogram parameters for As, Cd, Pb, Cr, and Hg, for which the best-fitting models were linear, linear, exponential, exponential, and spherical, respectively. Natural characteristics reduce the spatial variation of soil heavy metals, while human activities enhance its spatial variation (Guo and Jiang 2018). According to Cambardella et al. (1994), the spatial variation of soil heavy metals is classified as follows: < 25%, strong; 25–75%, moderate; and > 75%, weak. Herein, the nugget/sill ratios were 100.00% for As and Cr indicating that these two heavy metal elements have a pure nugget effect and a constant variation on the whole. The

nugget/sill ratios of Pb, Cr, and Hg show that the spatial variation was strong in Pb and moderate in Cr and Hg (Guo and Jiang 2018; Jiang et al. 2018). For Pb and Cr, the stochastic variation was less than the spatial structural variation; Hg is the opposite of Pb and Cr, indicating that the variation of Pb, Cr, and Hg was under the control of natural characteristics and human activities at the same time. The distance parameters of As and Cr were 69.31 km and 237.30 km, respectively. Normally, a wide range expresses a major area of influence on heavy metal values in soil and is attributed to natural characteristics (Goovaerts 1997; Rodríguez Martín et al. 2013), such as parent material. In our study, this general aspect also reflected a longer scale of influence, indicating that the influence of human activities on Cd, Pb, and Hg resulted in a smaller spatial correlation.

Based on the best-fitting models of the semivariogram, ordinary kriging was used to interpolate and obtain the spatial distributions of As, Cd, Pb, Cr, and Hg, which are shown in Fig. 4. Pearson correlation analysis was conducted, and the mean square deviation ratio was determined between the measured and predicted values (Table 7). The Pearson correlation coefficients were greater than 0.564 ($p < 0.01$), with the mean square deviation ratio close to 1 (Li et al. 2015; Guo and Jiang 2018). This indicates that the interpolation results of ordinary kriging were relatively reliable.

Table 5 Descriptive statistics of heavy metal concentrations in farmland soils for the different pollution sources sampled in the Poyang Lake region of China

| Heavy metal | Sources | Range (mg kg ⁻¹) | Average (mg kg ⁻¹) | Background value (mg kg ⁻¹) | Standard deviation (mg kg ⁻¹) | Coefficient of variation |
|-------------|---------|------------------------------|--------------------------------|---|---|--------------------------|
| As | I | 1.26–63.10 | 10.32a | 10.10 | 8.80 | 85.3% |
| | II | 1.16–43.20 | 10.01a | | 8.73 | 87.2% |
| | III | 2.06–19.50 | 7.09c | | 4.13 | 58.6% |
| | IV | 1.27–35.20 | 9.16b | | 3.93 | 42.9% |
| Cd | I | 0.07–2.83 | 0.24a | 0.13 | 0.27 | 112.5% |
| | II | 0.06–0.89 | 0.21b | | 0.12 | 57.1% |
| | III | 0.13–0.50 | 0.24a | | 0.11 | 45.8% |
| | IV | 0.04–0.73 | 0.18c | | 0.08 | 44.4% |
| Pb | I | 12.10–58.50 | 28.39c | 17.7 | 7.47 | 26.3% |
| | II | 15.90–68.80 | 31.86b | | 13.18 | 41.4% |
| | III | 18.20–206.00 | 48.83a | | 36.87 | 75.5% |
| | IV | 1.97–98.80 | 27.70c | | 11.18 | 40.4% |
| Cr | I | 33.70–109.00 | 66.69a | 63.20 | 17.02 | 25.5% |
| | II | 31.00–96.90 | 55.70c | | 13.05 | 23.4% |
| | III | 42.70–125.00 | 67.83a | | 18.72 | 27.6% |
| | IV | 25.10–123.00 | 62.00b | | 13.17 | 21.2% |
| Hg | I | 0.03–0.29 | 0.09b | 0.173 | 0.05 | 55.6% |
| | II | 0.04–0.39 | 0.10b | | 0.06 | 60.0% |
| | III | 0.02–0.37 | 0.14a | | 0.09 | 64.3% |
| | IV | 0.03–0.73 | 0.12a | | 0.08 | 66.7% |

For a metal, different letters indicate significant differences between concentrations ($p < 0.05$)

I industrial and mining enterprises, *II* sewage irrigation, *III* urban, *IV* general farming

Table 6 Semivariogram models for the first two principal component (PC) factors in the Poyang Lake region of China

| Index | Index | Fitting model | Nugget | Sill | Nugget/sill (%) | Range (km) | R^2 | RSS |
|-------------------|-------|---------------|--------|--------|-----------------|------------|-------|-------|
| I | PC1 | Gaussian | 0.402 | 2.814 | 14.29 | 88.33 | 0.742 | 0.287 |
| | PC2 | Exponential | 0.698 | 1.397 | 49.96 | 27.39 | 0.999 | 0.000 |
| Sewage irrigation | PC1 | Gaussian | 0.171 | 2.186 | 7.82 | 30.31 | 0.990 | 0.003 |
| | PC2 | Gaussian | 0.555 | 1.898 | 29.24 | 19.88 | 0.730 | 0.032 |
| Urban | PC1 | Gaussian | 0.332 | 1.826 | 18.18 | 34.16 | 0.890 | 0.270 |
| | PC2 | Gaussian | 0.391 | 2.792 | 14.00 | 107.73 | 0.622 | 1.170 |
| General farming | PC1 | Spherical | 0.489 | 1.109 | 44.09 | 85.90 | 0.977 | 0.013 |
| | PC2 | Exponential | 0.607 | 0.963 | 63.03 | 11.70 | 0.778 | 0.041 |
| As | | Linear | 25.86 | 25.86 | 100.00 | 69.31 | 0.260 | 0.221 |
| Cd | | Linear | 0.015 | 0.015 | 100.00 | 69.13 | 0.247 | 0.000 |
| Pb | | Exponential | 60.10 | 249.30 | 24.11 | 42.00 | 0.922 | 0.868 |
| Cr | | Exponential | 77.70 | 192.70 | 40.32 | 237.30 | 0.943 | 0.525 |
| Hg | | Spherical | 0.0030 | 0.0045 | 66.67 | 7.00 | 0.604 | 0.000 |

R^2 determination coefficient, RSS residual sum of squares, I industrial and mining enterprises

In these metals, the spatial distributions of Cd, Pb, and Hg were point source pollutants and showed a very similar spatial pattern, with contamination hotspots located simultaneously in the south of the study area, while the spatial distributions of As and Cr were similar and relatively smooth (Fig. 4), indicating that they came from the same sources, respectively. Hotspots emerged in places, and these were mainly distributed in the south around the urban (Fig. 4b, c, e). The best explanation for this pattern is that the urbanization process is faster and transportation is more developed in the south than in the north of the Poyang Lake region, which included the administrative center of the Jiangxi Province (Nanchang) and traffic routes extending in all directions (expressway and railway) (Fig. 4).

It is well known that both of these aspects of human activities can contribute considerably to raising Pb concentrations (Rodríguez Martín et al. 2006; Hani and Pazira 2011). It is reasonable to think of air and water dispersing industrial pollutants and spreading them over a much wider area than the manufacturing sites themselves, which contributes to the accumulation of heavy metals in farmland soils (Söderström 1998). Pb can be found in the exhaust gas of motor vehicles, oil spills, vehicle brake pads, and tire wear. This pollutant is deposited with atmospheric dust or surface residuals and accumulates with the rain on both roadsides of the soil, but it seems to decrease farther from hotspots. Sanchidrian and Mariño (1980) also found that Pb concentrations increased with greater traffic lines, largely because industrial fumes and combustion exhaust gases can travel long distances and eventually the Pb accumulates in farmland via atmospheric deposition. Previous studies have also shown that Cd and Pb are closely related to traffic and related activities, so traffic emissions may be a common origin of controlling the accumulation of Cd and Pb in soil (Dai et al. 2018; Yu et al. 2018). Finally, with respect to Hg, it can have multiple origins, with

agricultural production activities usually making a low contribution and urban waste and sewage irrigation more likely to be the main origins (Meng et al. 2016; Chen et al. 2016). Table 5 also shows that Hg reached its maximum concentration in farmland soils from urban areas.

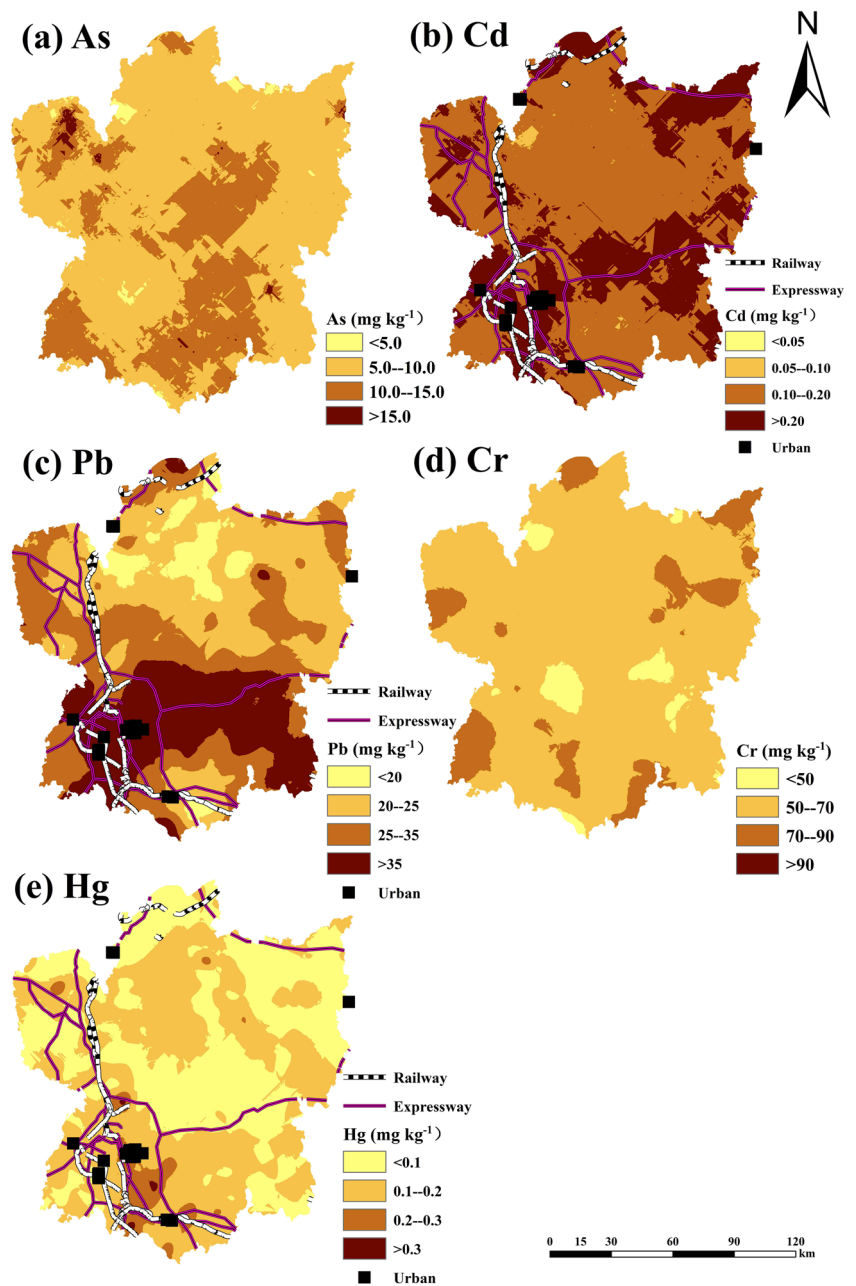
Figure 4 also shows that the concentrations of As and Cr are mainly affected by natural characteristics; hence, their spatial correlation held across the study region; the concentrations of Cd, Pb, and Hg are mainly affected by human activities as several hotspots of high metal concentration were identified by the geochemical maps, as Fig. 2 and Fig. 3 also further proved. In fact, they were strongly correlated in the statistical and PCA results for the four potential sources, in which strong associations were found between As and Cr or Cd and Pb (Table 2).

3.5 Heavy metal pollution from different sources

There are different ways and sources that could lead to heavy metal pollution in farmland soils (Rattan et al. 2005; Lee et al. 2006; Li et al. 2013b). Although many pollution sources have moved away from farmland areas, historical legacies persist since the reform and opening up of China in 1978 (Jiang et al. 2018). According to Fig. 4, the soil pollution by As, Cd, Pb, Cr, and Hg in the Poyang Lake region does not present an optimistic scenario. This implies that human activities have caused serious pollution to farmland soils, because natural characteristics hardly change over a short period of time in this study region.

Table 5 summarizes the heavy metal concentrations in farmland soils with respect to the pollution sources investigated. Compared with general farming areas, the soil concentrations of As, Cd, and Pb were significantly higher, on average, in the industrial and mining enterprise and sewage irrigation

Fig. 4 Spatial distribution maps of soil heavy metal concentration in the Poyang Lake region of China



areas in Poyang Lake region, clearly demonstrating the latter's role in accumulating these pollutants in soil (Zeng et al. 2015; Meng et al. 2016). Both Cd and Pb, but not As, in the urban areas were also significantly higher than in general farming areas, demonstrating that human activities considerably increase soil Cd and Pb concentrations but have a negligible

influence on As (Lee et al. 2006; Sun et al. 2013). Among the metals we analyzed, Pb had the highest average concentration in urban areas, in spite of the Chinese government's ban on the sale and use of leaded gasoline since 2000. Because of the extremely stable property of Pb, the pollution caused by leaded gasoline used in the last century clearly persists in the

Table 7 Cross-validation results of the ordinary kriging interpolation for heavy metals in the Poyang Lake region of China

| Index | As | Cd | Pb | Cr | Hg |
|---------------------------------|---------|---------|---------|---------|---------|
| Pearson correlation coefficient | 0.661** | 0.564** | 0.802** | 0.704** | 0.614** |
| Mean square deviation ratio | 0.775 | 0.621 | 0.863 | 0.663 | 0.642 |

** $p < 0.01$

environment (Chen et al. 2012; Lv et al. 2013). Lee et al. (2006) reached similar conclusions for Cd and Pb in urban soils from Hong Kong. The soil concentration of Hg was significantly lower in the industrial and mining enterprises and sewage irrigation areas when compared with the general farming areas. This suggests that neither industrial and mining enterprises nor sewage irrigation enhanced Hg accumulation (Li et al. 2013b). Hg accumulation is mainly caused by human activities in urban areas (since it is significantly higher than in the general farming areas). In order to reduce Cd, Pb, and Hg pollution, corresponding effective management and mitigating measures should be implemented in the urban areas of the study region immediately.

In the industrial and mining enterprises, As and Cr concentrations significantly exceeded those of general farming areas, illustrating the industrial and mining enterprises' contribution to augmenting As and Cr in soil (Table 5). Chen et al. (2016) showed that Cr was mainly from industrial activities in Xi'an City, China. In addition, As concentrations in sewage irrigation areas significantly exceeded those in general farming areas, illustrating sewage irrigation's contribution to augmenting As in soil (Table 5). Meng et al. (2016) showed that sewage-irrigated soil contained much higher concentration of As compared with clean water-irrigated soil in Tianjin, China. Compared with general farming areas, the concentrations of Cr were significantly greater in urban areas (maximum 67.83); this indicates urban activities promoted the accumulation of Cr in soil (Lee et al. 2006; Chen et al. 2016). In order to ensure food safety, continuous monitoring of the phenomenon of excessive As and Cr concentrations in industrial and mining enterprises and sewage irrigation near farmlands is needed.

4 Conclusions

Multivariate statistical and geostatistical methods were used to assess the soil pollution levels of five heavy metals and their spatial variation in Poyang Lake region of China. The soil concentrations of As, Cd, Pb, Cr, and Hg showed characteristics of accumulation near industrial and mining enterprise areas, sewage irrigation areas, or urban areas compared with general farming areas. In these metals, the spatial distributions of Cd, Pb, and Hg were point source pollutants and showed a very similar spatial pattern while the spatial distributions of As and Cr were similar and relatively smooth, indicating that they came from the same sources, respectively. The hotspot areas of metal contamination were mainly concentrated in the southern part of the Poyang Lake region and strongly related to industrial and mining enterprises, sewage irrigation, and urban activities. Interestingly, the origin of particular heavy metals from different pollution sources seemed to be similar (natural characteristics, human activity, or mixing). Overall, this study

provides a basic understanding of the origin of soil heavy metals from different pollution sources.

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