Soil physiochemical properties and landscape patterns control trace metal contamination at the urban-rural interface in southern China[☆]

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ABSTRACT

This study examined the influences of three subsets of environmental factors (i.e. soil physicochemical properties including pH, organic matters and soil texture, landscape patterns, and parent materials) on the spatial variations and sources of soil trace metal contamination across an urban-rural environmental gradient in Guangzhou City, southern China. We collected 318 surface soil samples from forests, orchards, farmlands, and urban lawns using a random tessellation design for selecting sample sites. The geo-accumulation indices showed that 18%—88% of soil samples were contaminated: moderate to high contamination with Cd and Hg, low to moderate contamination with Cu, Pb, Zn and Ni, and low contamination with As and Cr. However, less than 13% of soil samples were considered to have exceeded the national standards causing environmental and human health concerns. The mean geo-accumulation indices increased in the order of forest, paddy field/orchard, vegetable, road/residential, and park/residential areas for As, Cd, Ni, Pb, Zn, closely following a land disturbance gradient. Spearman Correlation and Cluster Analyses showed that Pb-Cu-Zn had traffic-related origins, Cd-Hg were mainly influenced by fertilization or industrial emissions, and As-Cr-Ni had geogenic origins for agricultural soils. In contrast, the Ni, Hg and Cd contamination sources for urban soils included both anthropogenic and geogenic origins. The Stepwise Regression and Partial Redundancy Analyses showed that three subsets of environmental factors explained 43%—87% of variations of soil contamination for both agricultural and urban soils. We concluded that soil contamination was mainly controlled by soil physiochemical properties followed by landscape patterns. Soil absorption of aerial loads of trace metal pollutants dominated the soil contamination processes. Our findings implied that improving soil physiochemical properties and landscape designs can strengthen environmental buffering and carrying capacity, thus alleviating soil contamination and reducing non-point-source pollution in the study region.

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1. Introduction

Soils directly and indirectly provide key ecosystem services such as food supplies and water quality improvement (Pavao-Zuckerman and Pouyat, 2017). Accumulations of trace metals in

soils pose great risk to human health (Sun et al., 2016). Excessive intakes of trace metals (e.g. Pb, Cd) may damage human nervous, circulatory, and immune systems (Pan et al., 2018). Soil contamination also threatens groundwater quality and water supply (Defo et al., 2015), ecosystem productivity, and ectomycorrhizal fungi (Evans et al., 2015).

Globally, the rapid urbanization and industrialization have caused serious soil contamination with trace metals due to air and water pollution in Europe (Rodriguez-Bocanegra et al., 2018), Asia (Tepanosyan et al., 2017; Pan et al., 2018), and America (Nezat et al., 2017). Both urban and agricultural soils have showed elevated trace

metals during the past three decades around Chinese cities (Luo et al., 2009; Wei and Yang, 2010; Luo et al., 2015), especially in the more developed and industrialized regions (Pan et al., 2018). Identifying factors influencing the distribution and sources of trace metals is a critical step for soil management and pollution control decision makings in a rapid urbanizing environment (Weissmannova et al., 2015; Yuswir et al., 2015).

Numerous studies on spatial distributions and sources of trace metals have focused on both agricultural soils in peri-urban areas (Huang et al., 2015; Sharma et al., 2015; Hu et al., 2018) and urban core soils (Gu et al., 2016; Rozanski et al., 2018). Many studies lumped the major soil trace metal pollutants (As, Cd, Cr, Cu, Hg, Ni, Pb and Zn) into two groups (Pb-Zn-Cu-Cd-Hg and As-Cr-Ni) according to their emission sources (Karim et al., 2014; Vince et al., 2014; Weissmannova et al., 2015). Group Pb-Zn-Cu-Cd-Hg was believed to come mainly from vehicle/transportation (Karim et al., 2014) and industrial emissions (Weissmannova et al., 2015; Xiao et al., 2015). The group As-Cr-Ni was primarily originated from natural sources (Vince et al., 2014; Li et al., 2017a). However, studies also showed that Cr and Ni were attributed to the industrial and transportation sources (Nazzal et al., 2015; Liu et al., 2016b). Other trace metal emission sources include urban legacies, previous agricultural activities, and soil management also influenced soil trace metal accumulations (Clarke et al., 2015). Furthermore, the existing urban soil studies mainly focused on lands for recreational, sporting or economic activities, such as playgrounds, shopping plazas and sport facilities (Khan et al., 2016; Rozanski et al., 2018), residential areas, public parks (Nezat et al., 2017; Gu and Gao, 2018), brownfields (Qian et al., 2017), industrial lands (Wu et al., 2018), and roadsides (De Silva et al., 2016).

Several environmental factors are believed to be responsible for elevated soil trace metal concentrations in an urban environment. These include trace metal deposition, retention and solubility at different scales (Fritsch et al., 2010; Wu et al., 2017), climate, parent materials, human population activities (Liu et al., 2016c), soil physiochemical properties (Navarrete et al., 2017), landscape patterns (Li et al., 2017a), urban legacies (Nezat et al., 2017), previous agricultural activities and soil management (Clarke et al., 2015). The dominated factors affecting the soil contamination process vary greatly in both time and space (Li et al., 2015; Liu et al., 2016b; Navarrete et al., 2017). For example, soil organic carbon content, pH, available phosphorous, soil texture, and cation exchange capacity (CEC) as well as human population played an important role in urban soil trace metal accumulation (Liu et al., 2016b; Navarrete et al., 2017). Other factors such as land use pattern, urban landscape design, and wind speed contributed little (Liu et al., 2016b).

The urban-rural interface areas are increasingly suffering from trace metal pollution originated from both point (e.g. industrial emissions, power plants) and non-point sources (e.g. vehicle traffic) (Zuo et al., 2018). The spatial distributions of trace metals generally decreased from the central urban area to the suburban or exurban areas with most accumulations in areas with anthropogenic activities or disturbance (Li et al., 2017b; Solgi and Oshvandi, 2018). Most existing studies on soil contamination focused on a single land use and cover type or a single environmental factor for soil remediation purposes. Few studies attempted to comprehensively examine both anthropogenic and background factors that affect soil contamination patterns and processes across a holistic urbanization gradient to address landscape management questions. Previous individual studies in core urban area or agricultural soils in peri-urban areas do not provide a whole picture of soil contamination for an "urban environment". In order to fully understand soil contamination processes in a highly urbanizing environment, a landscape approach is needed and both the agricultural and urban soils should be examined to provide information for landscape management purposes.

Therefore, our overall objectives are to comprehensively evaluate the variations, sources and environmental drivers of trace metals in a rapidly urbanizing environment in Guangzhou City, southern China. We asked the following two specific research questions: (1) whether the soil contamination, sources and driving forces in agricultural soils differed with those of urban soils in the same urban landscape matrix, and (2) which environmental factors including soil physicochemical properties, landscape patterns, and parent materials mainly controlled the soil contamination across an anthropogenic disturbance gradient? We hypothesized that the magnitude of soil trace metal contamination and distribution differ across an urban-rural environmental gradient as influenced by soil physiochemical properties as well as landscape patterns. The two factors represent the sources of trace metal pollutants and how these trace metal pollutants interact with the environments, surface soils in particular.

2. Materials and methods

2.1. Study area

The Guangzhou City (112°57′-114°3′E, 22°26′-23°56′N) is the capital of Guangdong Province located in the warm and humid southern China (Fig. 1). The region is characterized as having a typical subtropical monsoon climate with an annual mean temperature of 20–22 °C and average annual precipitation of about 1,700 mm. It is one of most populous (about 14 million) and developed (Gross Domestic Production about 294 billion US dollars in 2016) cities in China (National Bureau of statistics, 2017). This study focuses on the Guangzhou City (about 1470 km^2) (Fig. 1), which includes six administrative districts: Liwan (LW), Haizhu (HZ), Tianhe (TH), Baiyun (BY), and Huangpu (HP).

2.2. Soil sampling and trace metal extraction

The main soil parent materials in the study region are developed from geological formations of alluvial deposits (48%), granite (32%) and sandshale (12%). The probability-based soil sampling method (Hope et al., 2003) was employed to obtain the representative and unbiased soil samples within the study region that covers 1470 km^2. A randomly tessellation design (2 km × 2 km grids) was used for soil sample site selection. In total, there were 318 samples, including forest (10), orchard (103), farmland (128), urban lawn (74) and others (3) (Fig. 1). The soil samples were collected between October 14–16, 2015 and November 2–30, 2016. For the remote locations not accessible (e.g. forests on the steep slopes), the surrogates of the same land use type within 100–200 m of the sample site were used. Each soil sample included 5–8 sub-samples of topsoil (0–20 cm) around the sampling point. Soil samples were kept in sealed kraft packages after blending evenly for further chemical analysis in a laboratory. The geographical coordinates (latitude and longitude) and altitude for all the sample sites were obtained from GPS, and the land use types and possible pollution sources were also recorded in the field.

Soil samples were first air dried at a room temperature between 20 °C and 23 °C, then small rocks and debris were removed, and sieved with 10 (for determining pH and texture) and 100 mesh (for organic matter and trace metal measurement) nylon sieve, respectively. All of the soil samples were stored in polyethylene bags for further chemical analysis. Soil pH was measured by potentiometers after extraction by soil-water suspension (soil: water = 1/2.5), while the organic matter content and soil texture were determined by the potassium dichromate oxidation volumetric method and hydrometer method, respectively (Araujo et al., 2017).
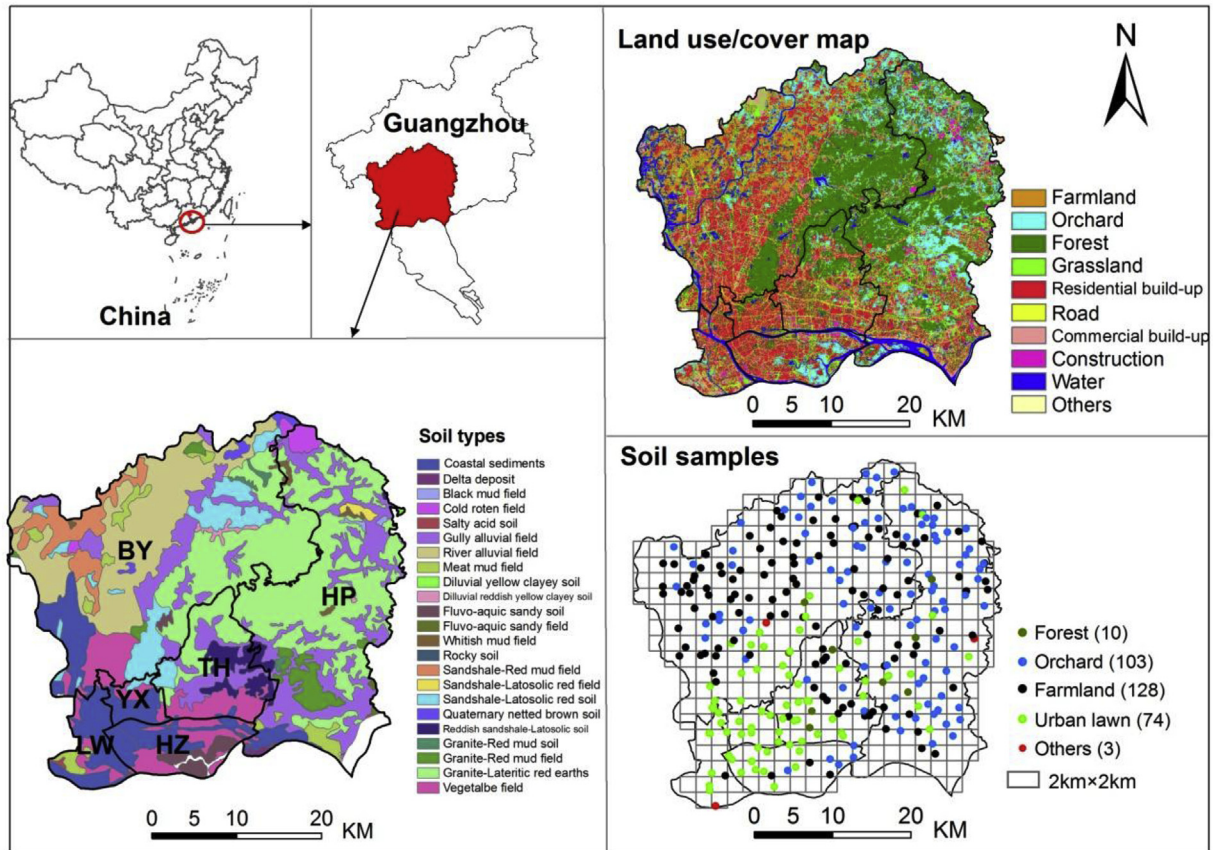
Fig. 1. Study location, soil type, land use/cover, and soil sampling scheme including number of samples by land use across an urban-rural gradient in Guangzhou, southern China. BY= Baiyun District, HP=Huangpu District, TH = Tianhe District, YX=Yuexiu District, LW = Liwan District, HZ=Haizhu District.

For trace metal concentration measurement, see detailed formation in our previous study (Li et al., 2015). The quality assurance and quality control (QA/QC) procedures were conducted by using Chinese standardized reference material (GSS-24), obtained from the China National Center for Standard Reference Materials. Replicates and blank-corrections were also analyzed simultaneously to check the accuracy of the results with a standard deviation within 4%. The recoveries for all the eight trace metals ranged from 96% to 102%.

2.3. Assessment of soil contamination with trace metals

The soil trace metal contamination level was evaluated by the Geo-accumulation Index (I_{geo}) as:

$$I_{geo} = log_2(C_i/1.5B_i) \tag{1}$$

Where C_i refers to the measured trace metal concentrations, while B_i refers to the background values of soil trace metals for Guangdong Province in China (CEMC-China Environmetal Mointoring Center, 1990; Li et al., 2015). According to the values of I_{geo}, soil contamination is classified as follows (Li et al., 2015):

0. No contamination ($I_{geo} \leq 0$)
1. Slight contamination ($0 < I_{geo} \leq 1$)
2. Moderate contamination ($1 < I_{geo} \leq 2$)
3. Moderate to heavy contamination ($2 < I_{geo} \leq 3$)
4. Heavy contamination ($3 < I_{geo} \leq 4$)
5-6. Heavy to extreme contamination ($4 < I_{geo} \leq 5$; 5) and extreme contamination ($I_{geo} > 5$; 6)

We adopted the modified Nemerow synthetic contamination index (I_{nm}) (Li et al., 2017a) to comprehensively evaluate pollution:

$$I_{nm} = power((I_{geomax}^2 + I_{geoave}^2)/2, \ 0.5) \tag{2}$$

Where I_{geomax} and I_{geoave} refer to the maximum and average contamination levels of the eight trace metals in the top soils, respectively.

To identify whether soil contamination negatively affects environmental quality or human health, we also calculated the exceeding standard rate (ES_i):

$$ES_i = C_i/S_i \tag{3}$$

Where C_i refers to the measured trace metal concentrations, while S_i refers to the standard soil screening values for soil contamination for both agricultural (GB15618-2018) or development land (GB36600-2018) as set in China.

The modified Nemerow Synthetic Standard Exceeding Rate derived from ES_i is:

$$ES_{nm} = power((ES_{max}^2 + ES_{ave}^2)/2, \ 0.5) \tag{4}$$

Where ES_{max} and ES_{ave} refer to the maximum and average exceeding standard rates of trace metals (i.e. As, Cd, Cr, Cu, Hg, Ni, Pb, Zn), respectively. When ES_i or ES_{nm} is greater than 1, the soil contamination poses risk to food production or human health.

2.4. Data processing and analysis

2.4.1. Land use and land cover

The land use and cover map of 2013 (1:2,000) (Zhang et al., 2018) was developed from the 1:2000 aerial photos with a spatial resolution of 0.1 m with geometric and terrain (1:500) correction. The original 64 land use types were aggregated into nine including forest, farmland, water, residential built-up, commercial built-up, road, constructions, orchard, grassland, and others (Fig. 1). The 1,470 km^2 study area was dominated by urban built up and farm lands.

2.4.2. Deriving landscape metrics

Landscape pattern is characterized by both a landscape's composition and configuration (Forman and Godron, 1986). We here chose percentage of landscape (PLAND), mean patch area (AREA-MN), patch density (PD), largest patch index (LPI), mean fractal dimension index (FRAC-MN), and aggregation index (AI) to represent the main landscape pattern (Zhang et al., 2018). These metrics were calculated by Fragstats 4.2 (Mcgarigal et al., 2012) based on land use and cover map of 2013 with s spatial resolution of 2.5 m.

2.4.3. Multivariate statistical analyses

Based on literature (Li et al., 2015; Khalilova and Mammadov, 2016; Liu et al., 2016c; Li et al., 2017a; Navarrete et al., 2017), three types of environmental drivers of soil trace metal contamination were selected: (1) soil physicochemical properties including soil pH, organic matter (OM) and texture (silt, sand, and clay contents); (2) landscape patterns as determined by common landscape metrics; and (3) main parent material composition (i.e. %deposit, % granite, %sandshale). In addition, we also considered building characteristics such as floor number, area, height and density in densely urbanized areas (Kosheleva et al., 2018). We did not include such analysis for other rural areas due to data availability.

ANOVA analysis with post-hoc test (Tukey method for variance homogeneity variables, otherwise using Dunnett's T3 method) was used to compare soil contamination among different land use types. Spearman Correlation Analysis and Hierarchical Cluster Analysis by the Ward method were conducted to explore sources of soil contamination with trace metals. We used the mean values of geo-accumulation indices (I_{geo}) within the 2-km by 2-km sampling unit as the dependent variables. The mean values of soil physiochemical properties, landscape patterns indicated by landscape metrics and parent material composition (% deposit, % granite, % sandshale) within each grid cell were used as the independent factors.

To determine which environmental factor controlled the soil contamination, we used the standardized stepwise regression coefficients to compare the significance of individual environmental variables (Ma et al., 2016b). In addition, we used partial redundancy analysis (PRDA) through variation partitioning to determine the importance of three groups of environmental variables (i.e. SPP, LP, and PM) (Li et al., 2015). All the multivariate statistical analyses including spearman correlation, cluster analysis and stepwise regression were conducted by SPSS (ver. 22). PRDA was conducted by CANOCO Windows (version 5) and the significant variables were selected using the built-in module of "iterative-forward-selection".

3. Results

3.1. Soil contamination by land use and special variability

The I_{geo} indices indicated that moderate to high contamination with Cd and Hg, low to moderated contamination with Cu, Ni, Pb

and Zn and for the comprehensive contamination level. Low contaminations were found from As and Cr for majority of 318 soil samples (Table A.1). The spatial distributions of soil samples with high contamination levels concentrated in the urban core region of Tianhe, Yuexiu, Liwan and Haizhu and the southern part of Baiyun and Huangpu districts (Fig. 2). However, compared with the soil screening levels of GB15618-2018 for agricultural soils and GB36600-2018 for development land, less than 12.6% of soil samples exceeded the standards and the rates exceeding the standards decreased in the order of Cd > Pb > As > Cu > Zn > Hg > Cr (Fig. A1). Furthermore, soil samples with high exceeding rates were scattered in Baiyun, Huangpu, Tianhe, and Haizhu Districts.

The mean values of I_{geo} indices increased in the order of forest, paddy field/orchard, vegetable, road/residential, park/residential for As, Cd, Ni, Pb, Zn and the comprehensive pollution index (P_{nm}) showed an increase with a land disturbance gradient (Fig. A2). The ANOVA analysis showed that the mean I_{geo} values of forest were significantly lower than that of vegetable for Cr and Zn, while Orchard < Vegetable for As, Cr, Ni, Cd, Hg contamination and P_{nm} (p < 0.05). Likewise, the contamination level of agricultural soil in paddy field, orchard and forest was significantly lower than that of urban soils in park, residential and road especially for As, Cd, Cr and Zn. Soil contamination with Hg in vegetable fields was significantly higher than that of road, and contamination for park was higher than road for Hg and Pb. Soil contamination level increased with the urbanized gradient for all the trace metals except Cr, Ni and Cu (p < 0.05; Fig. 3).

3.2. Sources of soil contamination

The I_{geo} indices of the eight trace metals in both agricultural and urban soils generally had significant correlations with each other (p < 0.05; Table A.2). For agricultural soils, the high correlation coefficients between trace metal pairs of Cu-Zn (r = 0.72), Cr-Ni (0.68), Cr-Cu (0.66), Zn-Ni (0.66) and Cu-Ni (0.65), indicated their common pollutant sources. The results of hierarchical cluster analyses demonstrated three major groups: (1) Cu-Zn-Pb-Ni, (2) Cd-Hg, and (3) As-Cr (Fig. A3). The majority of soil samples had low levels of contamination with As, Cr and Ni (Fig. 2), and there were high correlations between Cr and Ni. Thus, the third group (i.e. As-Cr-Ni) had a lithologic origin. Cu-Zn-Pb and Cd-Hg might originate from two different anthropogenic sources (e.g. traffic or industrial emissions, fertilization) as indicated by their high contamination levels. For urban soils, the dendrograms by cluster analyses for I_{geo} indices showed three clusters of Cu-Zn-Pb, As-Ni, and Hg-Cr-Cd (Fig. A.4). Similar to agricultural soils, Cu-Zn-Pb and As-Ni for urban soils might originate from anthropogenic and natural sources, respectively. However, Hg-Cr-Cd were probably a result of the mixed sources due to both lithology and traffic emissions because there were moderate correlations among Hg-Ni (r = 0.44), Hg-Zn (r = 0.56), Cr-Cu (r = 0.66), Cr-Ni (r = 0.58), Cd-Cu (r = 0.49), Cd-Hg (r = 0.55), and Cd-Zn (r = 0.50).

3.3. Environmental driving forces of soil contamination

3.3.1. The stepwise regression

Selected environmental variables explained 43%—87% of variations of soil contamination for all the soil samples (Tables A.3-A.5). For agricultural soils, Cu-Pb-Zn-Ni generally showed significantly positive correlations with pH, OM, percentage of orchard and commercial buildings (PLAND), and largest patch index (LPI) or aggregation index (AI) of water, while significant negative correlations with sand contents and PLAND or LPI of farmland. In addition, Cu and Pb contamination was significantly influenced by the landscape patterns of residential built-up (LPI, FRAC-MN) and road
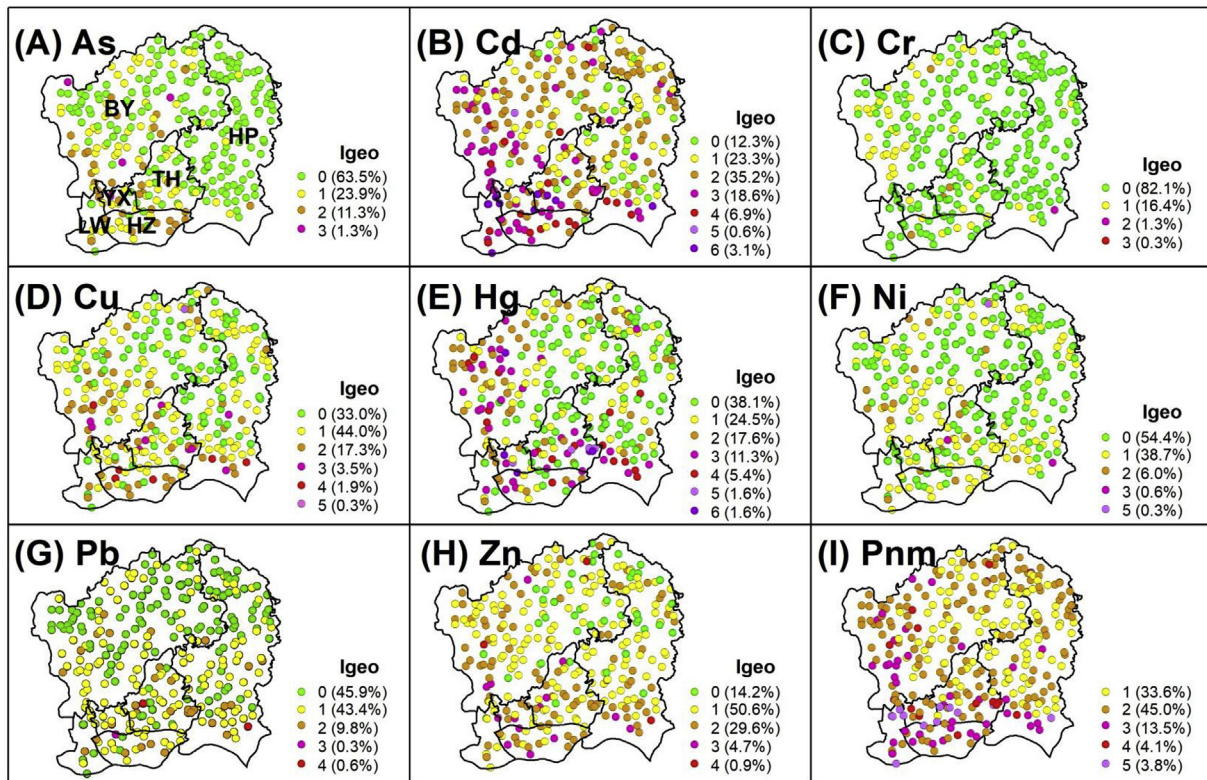
Fig. 2. Spatial distributions of the geo-accumulation indices (I_{geo}) for As (A), Cd (B), Cr (C), Cu (D), Hg (E), Ni (F), Pb (G), Zn (H) and the comprehensive contamination index (P_{nm}). The I_{geo} and P_{nm} are classified into 6 contamination levels (0 = no contamination, 1 = slight contamination, 2 = moderate contamination, 3 = moderate to heavy contamination, 4 = heavy contamination, 5 = heavy to extreme contamination, 6 = extreme contamination). The values in the parenthesis represent the percentage of samples in each of the contamination category.
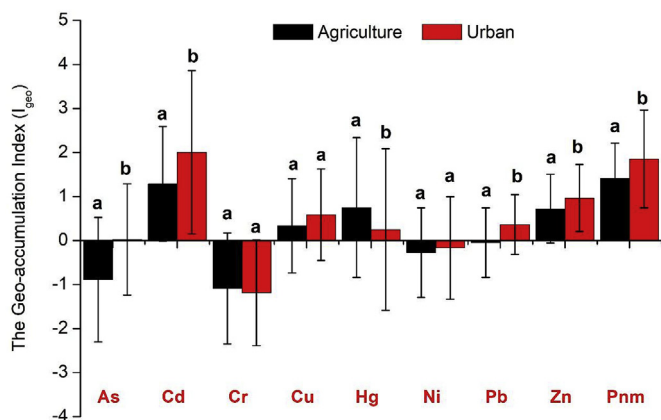

Fig. 3. Comparison of the mean values of geo-accumulation indices (I_{geo}) and comprehensive pollution index (P_{nm}) between agricultural (including forest, paddy field, orchard and vegetable filed) and urban lawn soils including parks, residential build-up, commercial build-up, and roads. Different letters over the bars means significant differences, while the same letter means no significant differences by ANOVA analysis with post-hoc test (Tukey method for variance homogeneity variables, otherwise using Dunnett's T3 method) ($p < 0.05$).

(fragmentation degree-PD and AI), respectively. Furthermore, parent materials showed significant correlations with Cu and Zn. Contamination by As-Cr and Cd-Hg had significant correlations with OM and part of landscape patterns of residential/commercial build-up or road (LPI, AREA-MN), while significantly negative correlations with part of landscape patterns of forest/orchard/grassland (PLAND/PD). According to the standardized stepwise

regression coefficients, OM explained most of the variations of soil contamination for majority of the trace metals and the comprehensive contamination level (P_{nm}).

For urban soils, contaminations for all the trace metals showed significant correlations with soil physiochemical properties (e.g. OM, sand, clay) and landscape patterns. However, the regression model could only explain 8% and 27% of variations of Pb and P_{nm}. Similar to agricultural soils, soil physicochemical properties played a more important role in explaining variations of soil contamination for nearly all the trace metals. For Cd and Zn contamination, fragmentation of commercial build-up and/or building density was the dominant environmental driving forces. When pooling all data together including both agricultural and urban soil samples, the environmental factors explained 43%–57% of variations and the comprehensive contamination level, which was mainly controlled by the soil physicochemical properties.

3.3.2. Partial redundancy analysis

Three subsets of environmental variables (i.e. soil physicochemical properties-SPP, landscape pattern of different land use and cover type-LP, parent materials-PM) explained 45.7%, 48.6% and 44.3% of the variations of soil contamination for all the samples, agricultural soils, and urban soils, respectively. The SPP alone, LP alone and PM alone could explain 46.7%–85%, 15.3%–34.8% and 2.0%–5.6% of the explained variance. The most shared variance was between SPP and LP (15.3%–34.8%) and SPP, LP and PM (12.4%–18.8% except for urban soils. SPP alone and shared with other variables explained 49.6%–76.8% of the explained variations of soil contamination ([a]+[d]+[f]+[g]), indicating a strong influence of SPP. LP was the second most important factor influencing soil

contamination. LP alone and shared with other variables explained 5%–51.6% of the explained variations of soil contamination. SPP alone, LP alone and their interactions explained 73.6%–88.1% of the explained variance of soil contamination. PM played the least influence on soil contamination as PM alone and shared with other variables accounted for only 11.9%–26.3% of explained variance.

4. Discussion

4.1. Soil contamination differed among different land use types and urbanization gradient

We found that Cd and Hg were the top two soil pollutants with moderate to high contamination levels among all samples (Fig. 2). This finding was consistent with previous studies for agricultural soils (Guan et al., 2001; Chai et al., 2003; Chen et al., 2016), urban soils (Gu et al., 2016), and both agricultural and urban soils (Chen et al., 2016) in the study region. However, regarding pollution levels for human health concerns, in references to national standards (GB15618-2018 for agricultural soils and GB36600-2018 for developed lands), less than 13% of total soil samples reached the hazard level affecting the local environment or human health. This was considered rather lower than previously found (Li et al., 2009; Lei et al., 2013; Chen et al., 2016; Gu et al., 2016). Chen et al. (2016) found that the excess portion (1.4%–33.2%) of the eight trace metals exceeded national soil quality standards for agricultural soils in Guangzhou. These exceeding rates had a range of 15.4%–30.8% for As, Ni and Cu in Nansha district (Lei et al., 2013), 30%–52% for Zn and Hg in farmland near petrochemical industries (Li et al., 2009), and 61%–96% for Pb, Ni, Cu and Cd in urban parks (Gu et al., 2016). The soil contamination differed among trace metals and across different areas in Guangzhou (Chen et al., 2016). Our work and other studies all show that there is a large variability of soil contamination across different land use types, local areas, and sampling sites. Therefore, sampling sizes from existing studies may cause bias to represent general spatial patterns.

Soil contamination levels for nearly all the trace metals followed an urbanization gradient (i.e. urban soil > agricultural soil) similar to the pollution patterns in forest soils across the urban-rural gradient reported by Pouyat and McDonnell (1991). The probable reason might be due to the fact that anthropogenic disturbances such as industrial activities and vehicle exhausts delivered considerably more trace metals to the atmosphere (Bermudez et al., 2012), water (Sun and Lockaby, 2012) and soils (Solgi and Oshvandi, 2018) as compared with less disturbed areas. Furthermore, the urban regions with high soil contamination levels were mainly found in the urban core areas and coincided with high building density and road pavements (Fig. 2). For agricultural soils, the localized high level pollution areas were mainly found in Baiyun and Huangpu districts (Fig. 2).

4.2. Complex sources of trace metals

Trace metals in Group 1 (i.e. Cu-Pb-Zn) and Group 2 (i.e. As-Ni) determined by the hierarchical cluster analyses appeared to have the same origins for both agricultural and urban soils. Group 1 could be of anthropogenic origins primarily from vehicle emissions, as the traffic-related characteristics of Pb, Zn and Cu were commonly found in the urban environment (Milenkovic et al., 2015; Yuswir et al., 2015; De Silva et al., 2016). In Group 2, As and Ni were dominated by lithogenic origins with low soil contamination levels, consistent with previous findings (Vince et al., 2014; Li et al., 2017b). The lithogenic origin of Cr and Ni were well-known and had been clearly identified in cities across China (Zhou et al., 2015) and elsewhere (Rodriguez Martin et al., 2013; Esmaeili

et al., 2014; Karim et al., 2014). A few previous case studies suggested that As was originated from mining and industries (Mehr et al., 2017) or coal combustion and water incineration (Ma et al., 2016a).

The sources of trace metal in Group 3 (i.e. Cd-Hg) and Group 4 (Cr) were different across the urbanization gradient. For example, our study showed that Cr in agricultural soils had a lithologic origin, while it had a mixed source of both natural and anthropogenic origins in urban soils. Previous studies did find that Cr and Ni in the urban environment were of anthropogenic origins such as traffic or industrial emissions (Milenkovic et al., 2015; Kim et al., 2017) or of the mixed source (Gu et al., 2016). In addition, Cd and Hg in agricultural soils in our study region were dominated by anthropogenic factors such as fertilization, traffic or industrial emissions. Our findings were consistent with reports by Liu et al. (2016b) who conducted a study in northeastern China. However, our study suggested that Cd and Hg in urban soils were of mixed sources. This was slightly different from Weissmannova et al. (2015), Xiao et al. (2015), and Liu et al. (2016c) who suggested that anthropogenic origins especially the industrial emissions dominated the pollution sources. Consistent with our study, Zuo et al. (2018) also found that the sources of the same trace metals in river banks changed across an urban-exurb gradient.

4.3. Key environmental factors influencing soil contamination

Soil physiochemical properties, landscape patterns, parent materials explained 43%–87% of variations of the contamination in agricultural and urban soils (Table A.3-A.5). Soil physiochemical properties had significantly positive (e.g. pH, OM and clay content) or negative (sand content) correlations with soil contamination for majority of trace metals. These findings are consistent with findings of Kosheleva et al. (2014, 2015), indicating that trace metals tended to accumulate in soils with high pH and organic matter content with a heavier texture. One probable reason might due to the low mobility of trace metals in the clayey and alkaline soils (Khorshid and Thiele-Bruhn, 2016).

Concentrations of Pb, Cu, Cd and Hg for both agricultural and urban soils showed significant correlations with landscape patterns of residential/commercial buildings and road (Tables A.3-A.5). This implied that the potential sources of trace metals were industrial/commercial activities and vehicle exhausts. Our study also found the areas with high soil contamination were coincided with industrial centers and transport hubs with high anthropogenic activities. However, surprisingly, the explanatory power for Pb contamination in urban soils was rather low (8%). Although previous studies indicated Pb accumulation in soils was traffic-related (Milenkovic et al., 2015; Liu et al., 2016a), we did not find its correlations with the distance to roads. Thus, the mechanisms of Pb contamination in urban soils were unclear and further studies are warrant. Furthermore, parent materials (i.e. %Granite, % Sandshale) also influenced soil contamination especially for Cd, Hg, Cu and Zn in the study region and any assessment of trace metal pollution in an urban environment must consider background soil chemistry.

Consistent with stepwise regression analyses, the PRD analyses showed that soil contamination levels were primarily controlled by soil physiochemical properties, followed by landscape patterns and parent materials (Table 1). Previous studies also documented similar notable influences of soil physicochemical properties (i.e. pH, OM, CEC, soil texture and available phosphorous) on trace metal accumulation in an urban environment (Liu et al., 2016c; Navarrete et al., 2017). Other factors such as land use patterns, urban landscape, and wind speed contributed less than soil properties (Liu et al., 2016c). The probable mechanisms might be that the physicochemical properties of surface soils directly determined their

Table 1

Variation partitioning of soil contamination among three subsets of environmental factors: (a) soil physicochemical properties (SPP), (b) landscape pattern (LP), and (c) parent materials (PM). The total variance of soil contamination is partitioned into the unique fractions of the variance explained by each set ([a], [b], [c]) and the common fraction shared by two ([d], [e], [f]) and three sets ([g]) of environmental subsets.

| Fraction | | All soil samples (N = 220) | | | | | Agricultural soil (N = 171) | | | | | Urban soil (N = 44) | | | | |
| | | Variation | | | Tested Fraction | | Variation | | | Tested Fraction | | Variation | | | Tested Fraction | |
		Eigenvalue	% of all	% of explained	F	P	Eigenvalue	% of all	% of explained	F	P	Eigenvalue	% of all	% of explained	F	P
A	SPP	0.229	22.9	50.0	15.8	0.0	0.227	22.7	46.7	10.9	0.0	0.376	37.6	85.0	5.0	0.0
b	LP	0.075	7.5	16.5	1.7	0.0	0.074	7.4	15.3	1.7	0.0	0.154	15.4	34.8	1.4	0.1
c	PM	0.017	1.7	3.7	3.7	0.0	0.010	1.0	2.0	2.2	0.0	0.025	2.5	5.6	1.3	0.3
d	SPP + LP	0.049	4.9	10.8	/	/	0.057	5.7	11.6	/	/	−0.140	−14	−31.7	/	/
e	LP + PM	0.032	3.2	7.0	/	/	0.029	2.9	5.9	/	/	0.044	4.4	10.0	/	/
f	SPP + PM	−0.002	−0.2	−0.4	/	/	−0.002	−0.2	−0.4	/	/	0.020	2.0	4.4	/	/
G	SPP + LP + PM	0.057	5.7	12.4	/	/	0.091	9.1	18.8	/	/	−0.036	−3.6	−8.1	/	/
	Total Explained	0.457	45.7	100.0	4.8	0.0	0.486	48.6	100.0	4.8	0.0	0.443	44.3	100.0	2.0	0.0

absorption capacity of aerial loads of pollutants. In this case, the atmospheric deposition was the dominant source of trace metals in both urban and agricultural soils (Luo et al., 2009; Nanos and Rodriguez Martin, 2012).

Surface soils offer important buffers to pollutants leaching to deep soil profiles. This implied that improving soil properties could be an effective way of preventing deep soil and water contamination. Kosheleva et al. (2014, 2015) found that altering physicochemical properties of urban soils by elevating pH, OM, Fe-Mn oxides and clay contents elevated the concentrations of trace metals by 33%–99%. Furthermore, changing the farming practices and promoting no-till and fellow might be another way to improve soil physicochemical properties (i.e., increasing OM) to facilitate buffering the soil pollutants.

5. Conclusions

About 18%–88% of soil samples were considered to be moderately to highly contaminated with Cd and Hg, low to moderately contaminated with Cu, Pb, Zn and Ni, while As and Cr were similar to background values. However, less than 13% of soil samples were considered to cause threats to environmental and human health. In addition, soil contamination levels increased along an anthropogenic disturbance gradient. All of the eight soil trace metals except As, Cr and Ni were of anthropogenic origins. Three subsets of environmental variables (i.e. soil physicochemical properties including pH, organic matters and texture, landscape pattern, and parent materials) explained nearly half or more variations of soil contamination. Soil properties were identified as the most critical factors influencing soil contamination for both agricultural and urban soils.

Our findings provide new insight into environment-soil contamination relationships in a highly heterogeneous landscape matrix in the urban-rural interface. Effective watershed management to control non-point source water pollution, improve soil and water quality, and minimize soil contamination risk to human health must consider improving soil properties and landscape designs that maximizes environmental buffering and carrying capacity of trace metals in southern China.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.04.065.

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