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Spatiotemporal patterns and drivers of soil contamination with heavy metals during an intensive urbanization period (1989–2018) in southern China☆

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A B S T R A C T

This three-decade long study was conducted in the Pearl River Delta (PRD), a rapidly urbanizing region in southern China. Extensive soil samples for a diverse land uses were collected in 1989 (113), 2005 (1384), 2009 (521), and 2018 (421) for heavy metals of As, Cr, Cd, Cu, Hg, Ni, Pb and Zn. Multiple pollution indices and Structural Equation Models (SEMs) were used in attribution analysis and comprehensive assessments. Data showed that majority of the sampling sites was contaminated by one or more heavy metals, but pollutant concentrations had not reached levels of concerns for food security or human health. There was an increasing trend in heavy metal contamination over time and the variations of soil contamination were site-, time- and pollutant-dependent. Areas with high concentrations of heavy metals overlapped with highly industrialized and populated areas in western part of the study region. A dozen SEMs path analyses were used to compare the relative influences of key environmental factors on soil contamination across space and time. The high or elevated soil contaminations by As, Cr, Ni, Cu and Zn were primarily affected by soil properties during the study period, except 1989–2005, followed by land use patterns. Parent materials had a significant effect on elevated soil contamination of Cd, Cr, Ni, Pb and overall soil pollution during 1989–2005. We hypothesized that other factors not considered in the present study, such as atmospheric deposition, sewage irrigation, and agrochemical uses, may be also important to explain the variability of soil contamination. This study implied that strategies to improve soil physiochemical properties and optimize landscape structures are viable methods to mitigate soil contamination. Future studies should monitor pollutant sources identified by this study to fully understand the causes of heavy metal contamination in rapidly industrialized regions in southern China.

1. Introduction

Soil quality is critical to food safety, human health, and ecosystem sustainability. The intensification of urban, industrial, and agricultural activities degrades soil quality through soil contamination as measured by concentration of heavy metals (Zeng et al., 2018). For example, soil heavy metal contamination has become a serious environmental concern in southern China amid the rapid industrialization (Pan et al., 2018; Zhang et al., 2019). The latest national soil pollution survey showed that 16.1% of lands exceeded China's maximum pollution limits (GB15168-1995; SEPA, 1995). About 19.4% of farmland sample sites were considered polluted while 29.4%–36.3% polluted sample sites were for

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commercial, industrial and mining areas (MEP and MLR, 2014). The soil contamination hotspots concentrated across southern China such as the Yangtze River Delta and the Pearl River Delta where rapid industrialization and urbanization were most found in these regions (MEP and MLR, 2014). Local estimates of pollution rates for heavy metal concentrations were reported to be even greater than national average (e.g., 43% for Mercury and 91% for Cadmium in Shunde city, southeast China) (Cai et al., 2015). Zhang et al. (2015b) estimated that 13.9% of grain production in China were affected by high levels of soil heavy metal concentrations in farmlands across the country. Site-specific soil pollution management and prevention measures require information on where, when, and why soil contamination threatens crop and human health.

Global studies suggested that accumulations of heavy metals were site-dependent and spatially heterogeneous, with higher concentrations in areas with intense human disturbances such as mining and smelting (Jamal et al., 2019; Li et al., 2014), industrial/traffic activities (Cai et al., 2019; Kim et al., 2017; Song et al., 2015), sewage irrigation (Meng et al., 2016), urban development (Duzgoren-Aydin et al., 2006; Song et al., 2018), and fertilizer and pesticide applications (Shi et al., 2019b). Due to the concerns of environmental risks and human health, studies have investigated the impacts of sewage irrigation (Ai et al., 2018; Delibacak, 2009), degraded water reuse (Corwin and Ahmadb, 2015), mining activities (Chandra et al., 2014; Ordóñez Fernandez et al., 2007), intensive agrochemical applications (Gil et al., 2018) and reservoir water level fluctuations (Pei et al., 2018) through time. However, a common limitation among these studies is limited by spatial extent and temporal duration (e.g. < 7 years). Most of these studies show increased concentrations of heavy metals in areas experiencing human disturbances over time. However, a few studies also reported a consistent decreased or fluctuating response in agricultural soils for 15–20 years (Li et al., 2015b; Liu et al., 2019a; Shao et al., 2016). The decrease in soil contamination was generally associated with enhanced environmental protection efforts such as land closure or technology upgrade by industries or wastewater treatment plants, introduction of clean energy production, and greater regulations of agrochemical uses (Li et al., 2015b; Liu et al., 2019a; Shao et al., 2016). A few regional or national synthesis studies using published papers (Shao et al., 2016; Shi et al., 2019a; Shi et al., 2019b; Yang et al., 2018; Zhang et al., 2019) suggested there was a large uncertainty in trend of soil pollution due to discrepancies in sampling methods, sample size, and study objectives. Overall, few studies have addressed the spatiotemporal changes in soil contamination at a regional scale. An unbiased soil sampling over a large spatial extent and long-term *in-situ* soil monitoring are still lacking due to high soil heterogeneity and high cost and labor-intensive implementations of large-scale sampling.

Accurate and detailed inventory of soil heavy metals is rather challenging due to the complex processes of soil contamination and a wide range of pollutant sources. Multivariate statistical (e.g., principal component analysis, cluster analysis, redundancy analysis, factor analysis, multivariate linear regression) and geostatistical analyses (e.g., geographically weighted regression) have been widely used to explore the proxies or drivers of soil contamination with heavy metals across space and/or time (Cai et al., 2019; Li et al., 2017; Song et al., 2016; Wang, 2016). Previous studies suggested that atmospheric deposition (Feng et al., 2019), soil physiochemical properties (Ye et al., 2019), land use and/or its patterns (Li et al., 2017; Li et al., 2019; Shao et al., 2016), elevation (Nickel et al., 2014; Zhang et al., 2019), precipitation and temperature (Zupancic, 2017), and socioeconomic conditions (Lin et al., 2018; Liu et al., 2016; Song et al., 2016) affect soil contamination to varying degrees. However, the traditional correlation analysis has limitations in dealing with latent variables (i.e., not directly observable) and exploring the causations between multiple dependent variables and independent variables. Structural Equation Modeling (SEM) is an alternative analytical technique (Grace et al., 2012) to correlation analysis to overcome some deficiency of the latter technique. SEM is an established causal model (Grace and Keeley, 2006; Grace et al., 2012) that is built on the priori hypothesis of the relationships of variables. SEM integrates regression, path and factor analyses, and has potentials to explore the direct and indirect effects of environmental variables on soil contamination. SEM has been widely applied in many fields, such as soil science (Chen et al., 2015a), disease ecology (Cobb et al., 2010), health science (Kusurkar et al., 2013), hydrology (Sanchez et al., 2015), and sustainability science (Liu et al., 2019b). The SEM model is well suited to reveal the causes of soil contamination in spite of its limited application at present. The identification of causations between soil contamination and environmental variables and the key environmental drivers is beneficial for best management decisions for soil contamination control and remediation.

In this study, using a SEM framework, we examined the relationship between spatiotemporal patterns of soil heavy metal contamination and environmental factors from 1989 to 2018 in the Pearl River Delate, in southern China. We address two research questions: 1) How does the soil heavy metal contamination change during the past three decades in PRD? and 2) What are the causations between soil contamination (Ys) and environmental variables (Xs), especially the relative contributions and direct vs. indirect effects of Xs on Ys. We hypothesize that “human activities are the key drivers for elevated soil contamination across both space and over time” in the study region. Management decisions for mitigating soil pollution require process-based understanding of the pollution sources and controlling factors over space and time.

2. Materials and methods

2.1. Study area

The Pearl River Delta (PRD) is located in the south-central Guangdong Province, adjacent to Hong Kong and Macao in southern China (Fig. 1). Dominated by a hilly and mountainous topography, the region resembles a horseshoe-shaped low-lying harbor crisscrossed by a network of Pearl River tributaries that drain into the South China Sea. Sediment deposits come from three main branches of the Pearl River, Xijiang and Beijiang draining from the west and the north sides and Dongjiang draining from the east side, forming the alluvial delta (i.e., PRD). Granite, river/sea alluvial deposit and sandshale are the three main types of parent rock materials. A southern subtropical monsoon climate dominates in this area with mean annual temperature and precipitation of 22°Cand 1900 mm, respectively (Li et al., 2015a).

The PRD is a megalopolis consisting nine major cities: Guangzhou (GZ), Shenzhen (SZ), Jiangmen (JM), Foshan (FS), Dongguan (DG), Zhongshan (ZS), Zhuhai (ZH), and part of regions of Huizhou (HZ) and Zhaoqing (ZQ). The PRD, dominated by farmlands and small rural villages in the 1980s, has experienced rapid economic development, population growth and urban expansion in the past four decades. By 2017, population and GDP across all nine core cities, excluding counties and prefectural cities, had reached about 34 million or 37.1% of the provincial total and 90.4 billion US dollars (or 85.8% of provincial total), respectively (National Bureau of statistics, 2018). Consequently, urban land cover across the PRD was notable in extent, increasing from 4346 km^2 (% 10.5 of the PRD total) in 1990 to 7239 km^2 (17.6%) today (2017).

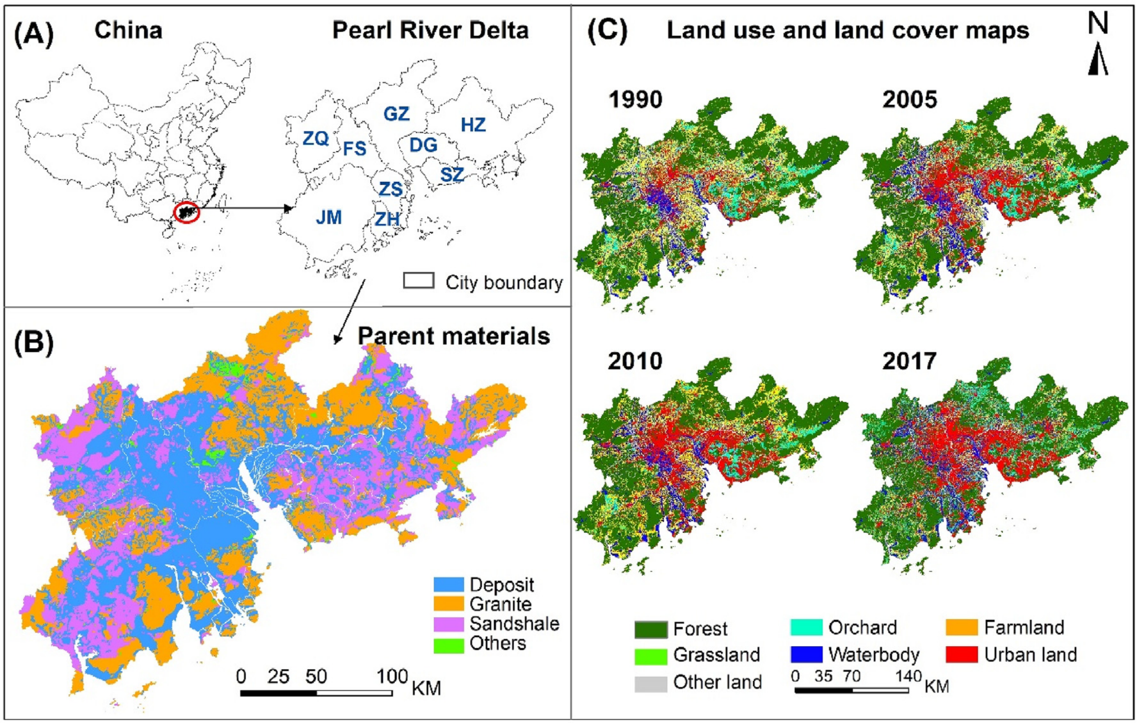

Fig. 1. Geographic location (A), map of parent materials (B), and land use and land cover maps (C) of the Pearl River Delta, south China.

2.2. Soil sampling and heavy metal content extraction

To characterize soil contamination dynamics through space and time, we used an extensive dataset of topsoil (0–20 cm) samples collected in 1989, 2005, 2009, and 2018 (Table S1; Fig. 2). The soil properties and pollutant parameters in 1989 (Chen et al., 2011), 2005 (Li et al., 2015a), and 2009 (Zhang et al., 2012) are historical data. We obtained them from Guangdong Institute of Eco-environmental Science & Technology. The data in 2018 were collected and chemically analyzed by our research group. The study purposes and strategies of soil sampling varied over the four periods. To facilitate the data comparison across different sampling schemes, we used the tessellation design of soil sampling in 2018 (i.e., 5 km × 5 km grid for urban core areas and 10 km × 10 km for suburban or rural areas) as a basis. The soil samples in 1989, 2005, and 2009 were projected into the tessellation (5 km × 5 km or 10 km × 10 km grid). Both the dependent and independent variables were summarized per soil sampling unit (5 km × 5 km or 10 km × 10 km).

Each soil sample constituted a mixed of 5–15 subsamples at each site (0–20 cm). Soil samples were pretreated with procedures including air-drying at room temperature (20°C–23 °C), removing rocks and debris, sieving with 10 or 100 mesh nylon sieve, and storing in polyethylene bags before delivering to the laboratory for chemical analyses. Soil samples were measured for pH with the potentiometer method, organic matter with the potassium dichromate oxidation volumetric method, and soil structure with the hydrometer method. Soil trace metal concentrations were determined by Hydrogen-Atomic Fluorescence Spectroscopy (HG-AFS), Flame Atomic Absorption Spectrophotometry (FAAS), Atomic Fluorescence Spectroscopy (AFS), and Inductively Coupled Plasma-Mass Spectrometry, ICP-MS; Table S2). The Chinese standardized reference materials (GSS-8 for 1989; GSS-1 for 2005; ESS-1 and GSS-1 for 2009; GSS-24 for 2018), replicates and blank corrections, were used for quality assurance and quality control (QA/QC) to ensure high accuracy (i.e., standard deviations < 5%). The recoveries for the eight heavy metals (five heavy metals in 2005) ranged from 92% to 108% (Cai et al., 2012; CEMC, 1990; Li et al., 2019; Zhang et al., 2006; Zhang et al., 2012). Detailed soil sampling strategy, heavy metal concentration extraction, and quality control methods are found in previous studies (Li et al., 2015a, 2017).

2.3. Data analyses

2.3.1. Soil contamination assessment

Single pollution indices (I_{geo}) and exceeding standard rate (ES_i) and modified Nemerow synthetic pollution indices of P_{nm} and ES_{nm} were calculated as follows:

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

$$ES_i = C_i/S_i \tag{2}$$

$$P_{nm} = \mathrm{power}((I_{geomax}{}^2 + I_{geoave}{}^2) \Big/ 2,\ 0.5) \tag{3}$$

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

where C_i represent soil heavy metal concentrations, B_i background soil heavy metals concentrations of Guangdong Province, China (CEMC-China Environmetal Mointoring Center, 1990), and S_i standard soil screening concentrations of heavy metals for agricultural (GB15618-2018) or development land (GB36600-2018) in China (Li et al., 2019). I_{geomax}, I_{geoave}, ES_{max} and ES_{ave} refer to the maximum and average values of I_{geo} and ES_i, respectively. The 1.5 in Equation (1) is a correction factor that considers the changes in background values due to diagenesis (e.g. sedimentary characteristics and rock properties). If I_{geo} or P_{nm} is larger than zero or ES_i or ES_{nm} larger than one, the soil is determined as polluted (I_{geo}) or harmful to food

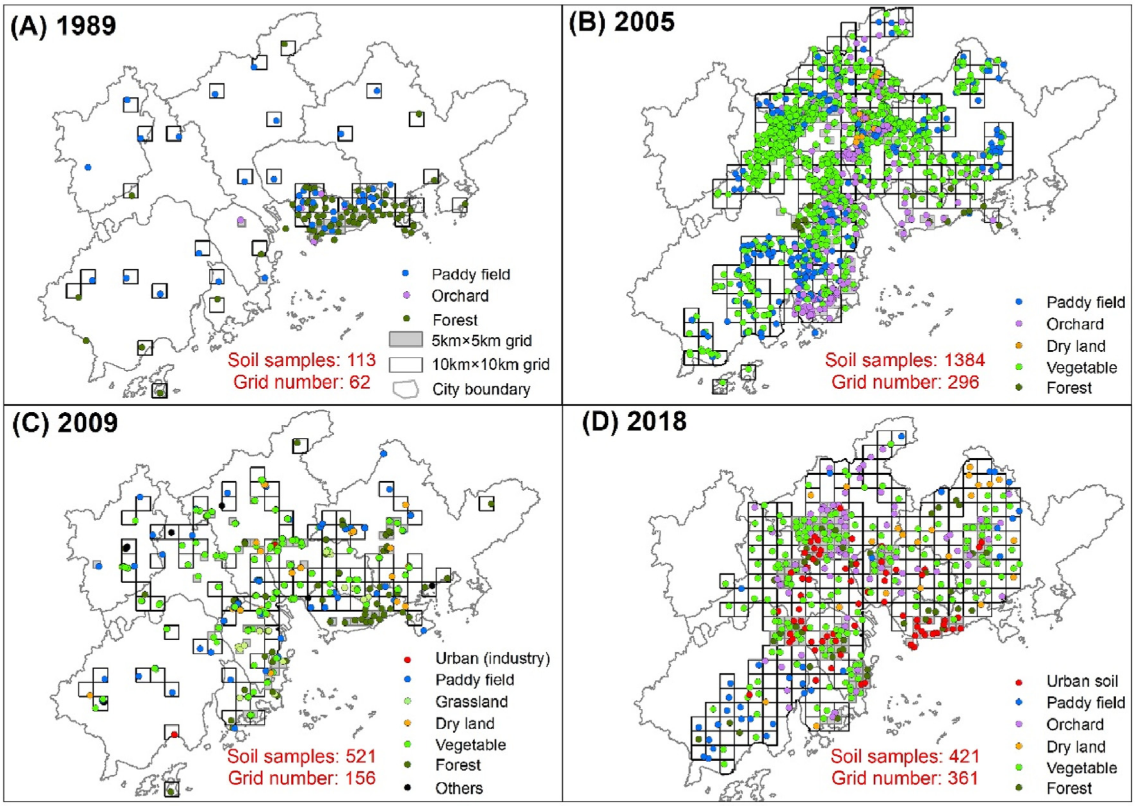

Fig. 2. Soil samples of the Pearl River Delta region in 1989(A), 2005(B), 2009(C) and 2018 (D).

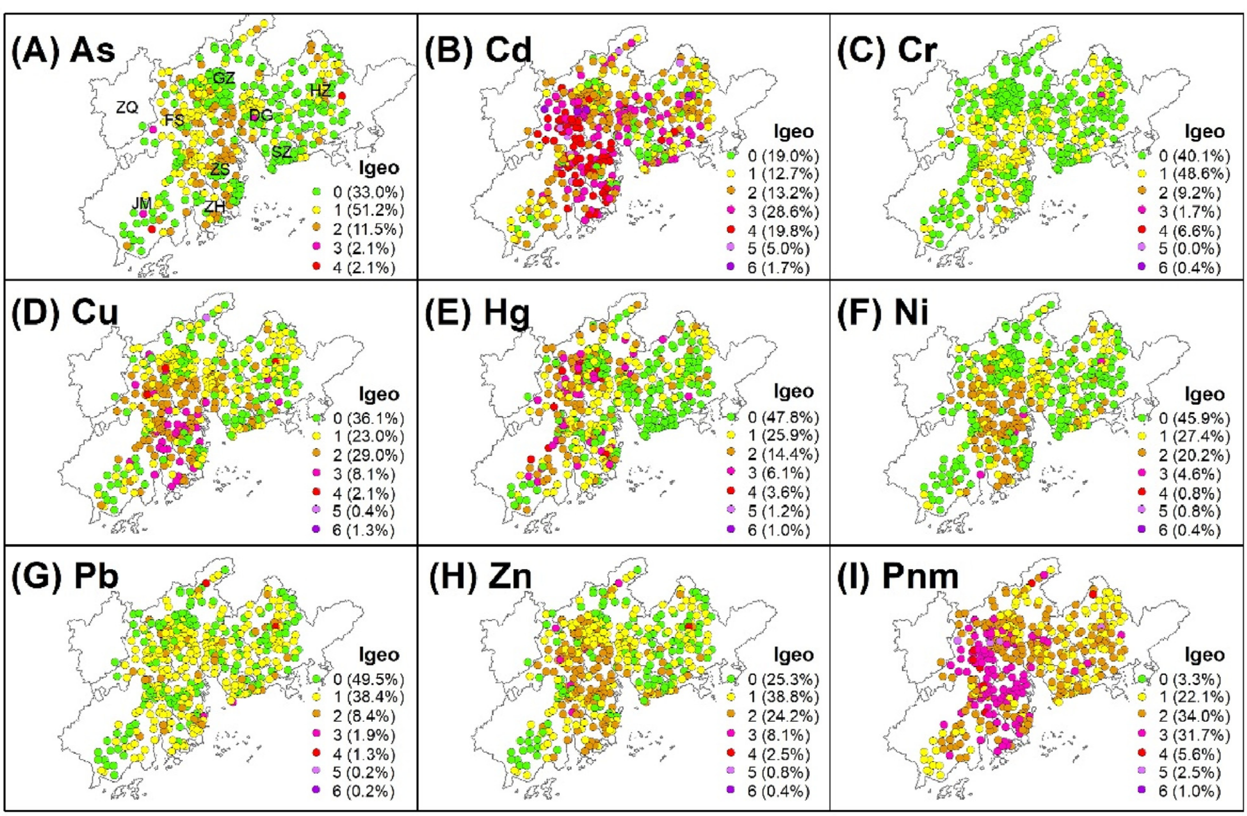

Fig. 3. Spatial distributions of geo-accumulation indices (I_{geo}) for As (A), Cd (B), Cr (C), Cu (D), Hg (E), Ni (F), Pb (G), Zn (H), and comprehensive pollution index (P_{nm}) (I) of Pearl River Delta region in 2018.

security or human health (ES). The values of I_{geo} from low to high are classified into 6 groups: no pollution ($I_{geo} \leq 0$), slight pollution ($0 < I_{geo} \leq 1$), moderate pollution ($1 < I_{geo} \leq 2$), moderate to heavy pollution ($2 < I_{geo} \leq 3$), heavy pollution ($3 < I_{geo} \leq 4$), heavy to extreme pollution ($4 < I_{geo} \leq 5$), and extreme pollution ($5 < I_{geo} \leq 6$) (Li et al., 2019).

2.3.2. Environmental variable selection and quantification

We selected a pool of 59 commonly used independent variables (Table 1) and grouped them into six latent variables (i.e., parent materials, topography/greenness, soil properties, socioeconomic factors, pollutant sources, and landscape patterns) to examine their relationship with soil contamination (Chandra et al., 2014; Jiang et al., 2019; Li et al., 2015a, 2015b; Li et al., 2019; Lin et al., 2018; Liu et al., 2016; Qiao et al., 2019; Zhou and Wang, 2019). A suite of landscape pattern metrics such as patch area percentage (i.e., PLAND), mean patch area (i.e., AREA), patch density (i.e., PD, ED), largest patch index (LPI), mean fractal dimension index (i.e., FRAC), and aggregation (i.e., AI) were computed for forest, farmland, waterbody, orchard, grassland, and urban land for each sampling grid across the region (Table 1; Li et al., 2019). The landscape pattern metrics were quantified using Fragstats 4.2 (Mcgarigal et al., 2012); while the other environmental variables were quantified using spatial analysis tools in ArcGIS 10.5.

2.3.3. Structural equation modeling

We used structural equation modeling (SEM) to examine a suite of conceptual pathway models describing direct and indirect effects of environmental variables on soil contamination. The model combines regression, path and factor analyses to examine the causality between different variables (Grace and Keeley, 2006). The SEM was conputed in IBM SPSS Statistics Amos (version 24). We have formulated and tested dosens of SEM models for each contaminant and each year. We performed multiple SEM models for the following response variables:1) soil pollution indices (I_{geo}, P_{nm}) at a specific year of 1989, 2005, 2009, and 2018, respectively, and 2) changes in soil pollution indices (ΔI_{geo}, ΔP_{nm}) during 1989–2005, 2005–2009, and 2009–2018, respectively. We used the six latent environmental variables (at a specific year) and their changes (between the two years; except parent materials) as the predictors in these SEMs: parent materials, topography/greenness, soil properties, socioeconomic factors, pollutant sources, and landscape patterns of land use. Because parent materials change little through time, this latent variable were kept the same in the SEMs at each year or time period. All of the response and predictor variables were summarized using means per sampling unit (5 km × 5 km or 10 km × 10 km).

Prior to conducting SEM modeling, both independent (X) and dependent (Y) variables were transformed using formulas of $\log_{10}(X + 1 - X_{min})$ or $\log_{10}(Y + 1 - Y_{min})$ to make sure that Xs or Ys approximately approached a normal distribution. We conducted preliminary stepwise regressions using original 59 independent variables to determine which of our candidate variables from each category to include in the final path analyses. For the number of candidate variables in a category more than two, we further ran dimension reduction analyses to create principal components if necessary, to represent the latent variable of this category. The stepwise regression and dimension reduction analyses were conducted in IMB SPSS Statistics (version 25).

3. Results

3.1. Spatiotemporal patterns of heavy metal contaminants

In 1989, both single (I_{geo}) and comprehensive (P_{nm}) contamination indices suggested that the majority of the region's soils were not polluted when compared to background values of Guangdong Province (Fig. S1). More than half of soil samples suggested high levels of As and Cd by 2005, and nearly all indices of heavy metals suggested high levels of pollution by 2009 and 2018 (Fig. 3, S2, S3). The compounded P_{nm} index indicated that over 96% of soil samples were contaminated by several heavy metals by 2005, 2009 and 2018. Although the pollution level for any single heavy metal was at slight to high levels, the comprehensive contamination (P_{nm}) was moderate to high levels. The exceeding rate indices (ES_i, ES_{nm}) indicated that less than 30% of collected soil samples in the PRD were harmful to food security and human health during the past three decades. For example, less than 28% of the 421 soil samples collected in 2018 exceeded the standards with the exceeding rates from high to low of Cd, ES_{nm}, Cu/Pb, As, Zn, Ni, and Cr/Hg (Fig. S4). Soil samples with high pollution level or exceeding rates were mainly distributed in the western PRD including Guangzhou (GZ), Foshan (FS), Zhongshan (ZS), Zhuhai (ZH) and Shenzhen (SZ) (Fig. 3, S1, S2, S3, S4).

Table 1
A summary of environmental variables selected for structural equation models.

Data	Temporal and spatial resolution	Variable	Source
Parent materials	Vector map	Percentages of deposit (%Depo), granite (%Gran) and sandshale (%Sandsh)	Guangdong Institute of Eco-environmental Science & Technology (GIEST) (Li et al., 2015a; Li et al., 2019)
Topography/Greenness	DEM, 30 m × 30 m; NDVI, 2005, 2009, and 2018 (1 km × 1 km)	Elevation from digital elevation model (DEM) or normalized difference vegetation index (NDVI)	DEM from GIEST; NDVI from Institute of Geosciences and Resources, Chinese Academy of Sciences (IGR-CAS) (http://www.resdc.cn)
Soil properties	1989, 2005, 2009, and 2018; soil samples	Organic matter (OM), pH, soil humus content (SHC), and contents of sand (Sand), silt (Silt) and clay (Clay)	GIEST
Socioeconomic factors	GDP and POP, 1990/1995, 2005, 2010 and 2015 (1 km × 1 km)	GDP and population density (POP)	IGR-CAS (http://www.resdc.cn); (Liu et al., 2005; Yi et al., 2006)
Pollution source	1990, 2005, 2010, and 2018; 30 m × 30 m	Distances to the nearest road (Dis_Rd), mine (Dis_Mine), industry (Dis_Ind) and waterdody (Dis_Wat)	Land use or land cover data from Guangdong Academy of Sciences in China (1990, 2005 and 2010) and Guangzhou University (2017); road and river maps in 2005 and road map in 2018 by IGR-CAS (http://www.resdc.cn)
Landscape patterns	1990, 2005, 2010, 2017; 30 m × 30 m	Landscape metrics including Percentage (PLAND), patch density (PD), edge density (ED), largest patch index (LPI), mean patch area (AREA), mean fraction dimension index (FRAC), and aggregation index (AI) for six types of land uses (i.e. Forest, Orchard, Farmland, Grassland, Waterbody, and Urban land)	Calculated by Fragstats 4.2 based on land use or land cover data (Li et al., 2017, 2019)

Soil contamination levels also changed over the study period. Both the mean single (I_{geo}) and comprehensive (P_{nm}) soil contamination indices increased over time when compared to regional background values (Figs. S5, S6). The mean comprehensive pollution indices (P_{nm}) for the common sampling grids increased from 0.82 ± 0.29 (mean ± Std. Dev.) in 1989, to 0.88 ± 0.37 in 2005, to 1.10 ± 0.67 in 2009, and to 1.34 ± 0.60 in 2018, resulting in a gradual change from slight to moderate pollution levels (Fig. S5). The soil contamination level increased for all the heavy metals from 1989 to 2018 with a different magnitude of increase ranging from 0 to 1.2 (Fig. S6). Although concentrations increased over time, the majority of soil samples did not reach a hazard level especially for As, Cr, Ni and Pb (Fig. S4).

3.2. Driving forces of soil contamination

The SEM path analyses showed variable impacts of environmental factors on soil contamination across the study region (Tables S3, S4). Because not all computed SEMs fitted to the data well ($R^2 < 0.45$) (Table S3), we mainly focused on SEMs with a high ($R^2 > 0.45$) explanatory power (Fig. 4, S7-S10). The best-performed models of 1989, 2005, 2009, and 2018 (As, Cr, Ni, Cu, and Hg in 1989, Cu and Zn in 2009, and As, Cr, Ni, Hg, and Zn in 2018; Fig. 4 and S7) explained from 44% to 68% of variations in soil contamination for each respective heavy metal ($p < 0.05$). The standardized path coefficients indicated that soil properties had the greatest significant direct effect on soil contamination with As, Cr, Ni, Cu, and Zn, followed by landscape patterns (Tables S3, S4; Fig. 4, S7). For example, the soil contamination level tended to be lower in land use dominated by forest, orchard and grassland as represented by higher land percentage, (PLAND) and mean path area (AREA) and the largest patch index (LPI). These factors had significant direct effects (Fig. 4, S7). For waterbody, farmland and urban land, higher PLAND, AREA and patch density (PD/ED) tended to have higher soil contamination levels (Fig. 4, S7). However, the parent materials and landscape patterns for farmland, forest and orchard land showed large and direct impacts on soil contamination with Hg in 1989, with the greatest effects from parent materials (Fig. 4). Topography, greenness, socioeconomic factors, and pollutant sources generally had a weak or no significant ($p > 0.05$) relationship with soil heavy metal contamination (Fig. 4, S7). Compared with landscape pattern of land uses, pollutant source, and soil property, indirect effects of parent materials, topography/greenness, and socioeconomic factors on soil contamination with As, Cr, and Ni (e.g., 1989, 2018), Cu and Zn (e.g., 2009, 2018) are relatively high (Table S4). However, similar to the direct effects, these indirect effects were not statistically significant ($p > 0.05$; Table S4).

The dominated factors influencing soil contamination also changed with time (Fig. S8, S9, S10). For example, at the early stage of the study period (i.e., 1989–2005), parent materials showed the greatest direct effect among all the environmental variables on the changes in comprehensive index of soil contamination (P_{nm}), while landscape patterns showed the greatest effect during the recent period of 2005–2009 (Fig. S8). Similarly, the parent material characteristics and/or landscape patterns had the largest impacts on soil contamination with Cr and Ni during 1989–2005 while soil properties dominated the impacts during 2009–2018 (Fig. S9). Parent material characteristics had the greatest effects on elevated

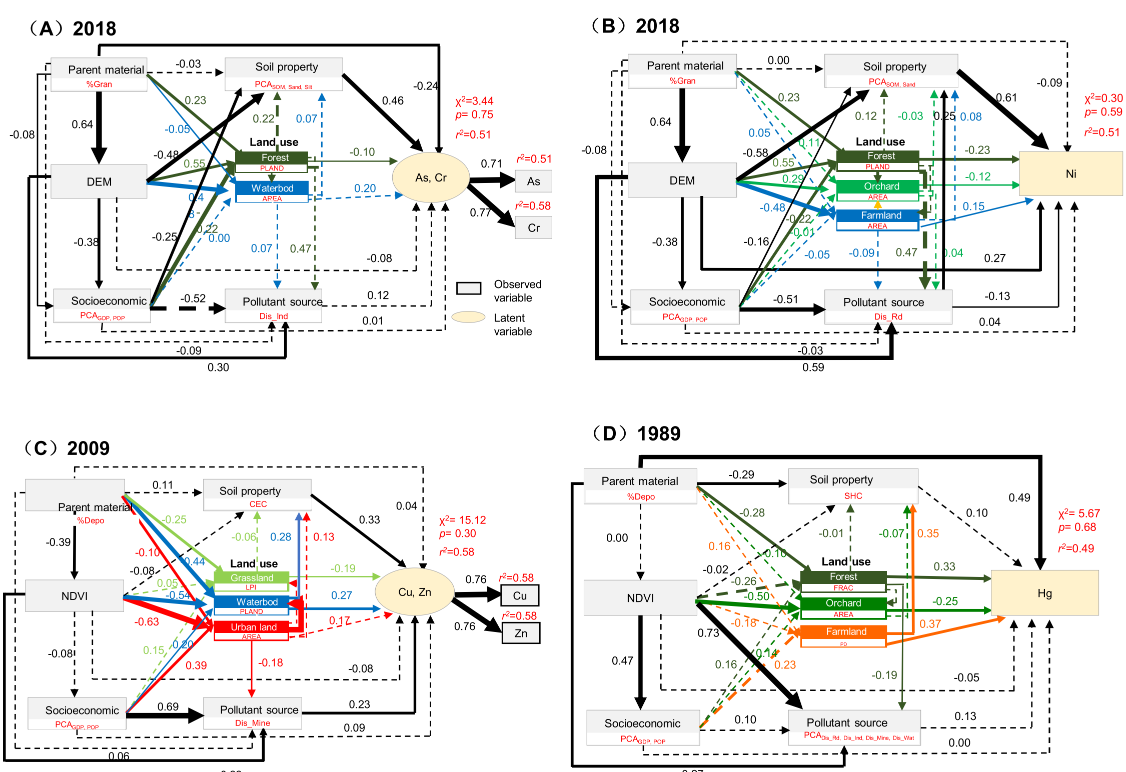

Fig. 4. The structure of Structure Equation Model (SEM) for assessing the impacts of five groups of independent factors (i.e. parent materials, topography/greenness, socioeconomic factors, soil property, and pollutant source) on soil contamination of As and Cr (A) and Ni (B) in 2018, Cu and Zn in 2009 (C), and Hg in 1989 (D). Solid line and dash line represent the impacts of independent factors on soil contamination are significant ($p < 0.05$) and not significant, respectively; numbers on the line represents the direct impacts. The thicker the line is, the greater the impact is. $PCA_{SOM,\ Sand,\ Silt}$, $PCA_{SOM,\ Silt}$, $PCA_{GDP,\ POP}$, $PCA_{Dis\_Rd,\ Dis\_Ind,\ Dis\_Mine,\ Dis\_Wat}$ represent the first principal component of the original independent factors denoted in subscript (the explained variance of the component larger than 60%). NDVI and DEM represent the normalized difference vegetation index and elevation derived from digital elevation model, respectively.

soil contamination of Cd and Pb during 1989–2005, while soil properties or landscape patterns showed most effects on elevated Zn and Hg during 2009–2018 (Fig. S10). Overall, in the early stage of urbanization/industrialization (1989–2005), the natural lithology of parent materials generally accounted for the elevated concentrations of heavy metals especially for Cr, Ni, Cd and Pb. However, during the recent time period (2009–2018), soil physiochemical properties and/or urban landscape had pronounced impacts and led to higher levels of Cr, Ni, Zn and Hg. Landscape patterns were secondary in terms of direct and indirect effects on changes in soil contamination during 2009–2018 (Figs. S9, S10). All the independent variables had lower indirect effects than the direct effects on soil contamination with heavy metals except As, Cr, Ni, Cu and Zn during 2009–2018 (Table S4).

4. Discussion

4.1. Soil contamination elevated during the past three decades

Our data showed that the distributions and variations of soil contamination were space-, time- and heavy metal pollutant-dependent. High pollution levels were found in the western part of PRD, including Foshan, Zhongshan, Zhuhai, and Shenzhen. This pattern was consistent with previous findings (Chen et al., 2012; Li et al., 2015a; Li et al., 2017; Zhang et al., 2014) and was likely explained by the combined effect of river alluvium from the West and North Rivers and possibly also by high levels of atmospheric depositions (Sha et al., 2019; Yang et al., 2007; Zhang and Wang, 2001). The soils in western part of the study region partially inherited the feature of high concentrations of heavy metals (especially for Cr, Ni, Cd and Pb; Fig. 1B) from the parent materials of river alluvium (Yang et al., 2007; Zhang and Wang, 2001). Human activities, such as mining and smelting, industrial and traffic emissions, and sewage irrigation might release additional heavy metals into the soils, especially for Pb and partial Cd, Cu, Zn and Hg (Zhang et al., 2014; Zhou and Wang, 2019).

Overall, the single pollution level (I_{geo}) for each heavy metal was not high as expected. Most of soil sampling grids were classified as uncontaminated or slightly contaminated for As, Cr, Ni and Pb across all the four time periods, while Cd levels were classified as slight to moderate pollution. The soil contamination in the region was compounded with multiple heavy metals as indicated by the comprehensive pollution indices (P_{nm}). Approximately 59.3%–99.3% of soil samples were contaminated by one or more heavy metals during the period of analysis and the mean comprehensive pollution level increased from slight to moderate across the region. However, compared with China's maximum allowable concentration levels of heavy metals in agricultural soils (GB15618-2018) and urban soils (GB36600-2018), less than 30% of soil samples (e.g. less than 26.4% in 2018) reached the hazard level to influence food security or human health. This finding was consistent with recent studies that reported overall low exposure of heavy metals in food production systems and human settlements across the study region (Chang et al., 2014; Li et al., 2014; Zhang et al., 2014).

The Cd, Hg and Cu were the top three pollutants with both a large magnitude and changes in I_{geo}, especially during 2005–2018. The pattern was somewhat consistent with previous literature in China (Chen et al., 2015b; Pan et al., 2018; Zhang et al., 2015b). Cd, Hg and Cu are generally associated to anthropogenic sources such as industrial emissions or fertilization (Liu et al., 2016; Weissmannova et al., 2015). Rapid urbanization and industrialization have elevated soil heavy mental concentrations considerably especially for Cd, Hg, Cu, Pb and Zn in the study region during the past three decades (Lin et al., 2018; Zhang et al., 2015a; Zhou and Wang, 2019). Similarly, our study also found that human activities that modified soil properties or landscape patterns for different land uses were pronounced for Zn in 1989, 2009, 2018, and Cu in 2009, and Hg in 1989. In contrast, previous studies found that heavy metals such as As, Cr and Ni were primarily derived from natural sources (i.e., parent materials) (Hu and Cheng, 2013; Li et al., 2015a). However, in some cases, anthropogenic sources of industries and transportation were found to be the sources for As, Cr and Ni (Li et al., 2019). Our study also found that the sources of As, Cr, and Ni were mixed. The changes in soil contamination with Cr and Ni during 1989–2005 mainly originated from natural sources of parent materials (Table S3). For soil contamination with As, Cr, and Ni in 1989, 2018 and changes in their concentrations during 2009–2018, landscape patterns and soil properties as influenced by human activities were the main drivers (Table S3).

4.2. Environmental drivers of soil contamination in both space and time

As hypothesized, our SEM analyses indicated that human activities that altered landscape patterns and/or soil physiochemical properties were the key factors affecting soil contamination, especially for heavy metals of As, Cr, Ni, Cu, Zn, Hg over both space and time. This temporally extended analysis using data from multiple sampling time periods was consistent with recent findings that observed larger contributions of soil properties and landscape pattern to the variations of soil contamination across the space at local scale using similar methods (Li et al., 2019). Likewise, Jiao et al. (2018) showed that anthropogenic activities could contribute about 41% of the variations of heavy metal accumulation in farmland soil. A few other studies also suggested that soil properties such as pH, OM, CEC and soil texture had notable influences on soil contamination with heavy metals (Kosheleva et al., 2014, 2015; Liu et al., 2016; Navarrete et al., 2017). One probable mechanism is that soil conditions affect metal mobility and retention capacity (Mojid et al., 2016). High pH and organic matters favor the release of most heavy metals from contaminated soil colloids or heavy metal immobilization (Gil et al., 2018; Ma et al., 2019; Nedelescu et al., 2017; Wiatrowska and Komisarek, 2019). Khorshid and Thiele-Bruhn (2016) and Li et al. (2019) observed higher heavy metal concentrations in locations where soils had higher pH, and OM and heavier soil textures. In addition, manganese (Mn) and iron oxides in soil have strong adsorptions on As and Cd (Ma et al., 2019; Suda and Makino, 2016). Land uses reflect the intensity of human activities. Consistent with previous studies (Li et al., 2017; Li et al., 2019), our findings suggested that landscape patterns are significant in explaining variance in soil contamination, but contributions of landscape structure were less pronounced than that of soil properties. Furthermore, our study found that parent materials had the greatest effect on the elevated comprehensive and single soil contamination with Ni, Cd and Pb during 1989–2005. This implied that, at the early stage of urbanization, the pedogenisis of previous parent materials mainly controlled the increase in soil contamination. However, other factors such as landscape patterns of land uses and soil properties were more pronounced in affecting soil contamination through time. Similar to our findings, Jiang et al. (2019) showed the dominant factors influencing soil contamination with As, Cr and Hg changed from geological environment in 1983 to human activities in 2010.

Soil contamination involves complex both natural and anthropogenic processes (Karim et al., 2014; Mihaljevič et al., 2019). The SEMs explained well about the environmental effects on variations of soil contamination for most heavy metals in 1989 and 2018, and the temporal changes during 1989–2005 and 2009–2018 (Table S3). However, the SEMs apparently were not able to explain the all the spatial variabilities ($R^2 < 0.45$) of soil contamination for most heavy metals in 2005 and 2009, and during 2005–2009 (Table S3). The poor model performances in 2005 and 2009, and 2005–2009 were likely due to uncaptured inventory variables that represent the entire sources of heavy metal inputs. For example, atmospheric deposition, fertilization and agrochemical uses and irrigation of wastewater, were not included in our analysis due to data availability. The atmospheric deposition and sewage sludge were documented to be the main sources of most heavy metals in agricultural soils especially in western PRD (Li et al., 2015a, 2017). Sha et al. (2019) found high levels of atmospheric depositions in the western part of the study region based on emission inventory data. Although we could not include the monitored atmospheric deposition data, we observed the strong effects of landscape patterns and soil physicochemical properties on soil contamination through both space and time. The underlying mechanism might be because land use distributions and soil properties reflected the amount and absorption capacity of atmospheric deposition, respectively. Meantime, it is well known that pedogenisis of parent materials is a long process and heavy metals in parent materials rarely move to the top soil layer under natural conditions. However, heavy metals with a source of human activities such as atmospheric deposition often accumulate into the top layer by soil leaching.

4.3. Implications to soil contamination control measures

Our long-term data show pronounced effects of landscape patterns and soil properties on the distribution and magnitude of soil pollution. Thus, effective strategies and measures to control regional soil contamination must reduce soil contamination from both point (e.g., industry) and non-point sources (e.g., atmospheric, fertilizers). It is critical to establish a long-term *in-situ* monitoring network identify pollution sources and evaluate effectiveness of land management practices in controlling soil contamination. Proper reforestation or greening and other urban forestry efforts that improve soil properties, enhance nutrient and water cycles and capture or intercept atmospheric deposition may help mitigating soil contamination problems (Curran-Cournane et al., 2015; Trammell et al., 2011). Heavy metals are usually high in alluvial deposition areas due to historical deposition and river sediment transport. In such areas, restoring natural vegetation covers may be most effective for soil contamination control. Dou et al. (2017) suggested that factories that release pollutants should be kept away at least 50 km away from these parent materials or tidal river networks. In addition, altering soil properties such as application of lime-rick materials and organic amendments increasing organic matters (OM) and clay contents (Khan et al., 2017; Kosheleva et al., 2014, 2015; Obiora et al., 2019) is helpful to mitigate soil contamination. Recently, biochar has been widely used to immobilize heavy metals in contaminated soils by significantly changing the soil pH, OM, CEC and soil redox potential and microbial community (Meng et al., 2018).

5. Conclusions

Our three-decade (1989–2018) comprehensive regional assessment of spatiotemporal changes of soil contamination in southern China found that soil contamination was slightly elevated for all the heavy metals examined (i.e., As, Cd, Cr, Ni, Cu, Zn, Pb and Hg) except Cd, Hg and Cu that had moderate contamination. However, most soils did not reach hazard levels to food security and human health. Soil samples with high contamination level were mainly located in the western Pearl River Delta region, coincided with densely populated areas known for high pollutant emissions and locations where alluvial deposition and elevated background concentrations.

Our study generally supports the hypothesis that “human activities influencing landscape patterns and/or soil properties are the key factors affecting soil contamination”. Other factors such as atmospheric deposition, polluted water from irrigation, and agrochemical uses are identified that may also explain the large variability of soil contamination. In addition, parent materials are important to explain the changes of soil contamination during 1989–2005. Future research on deeper understanding of soil contamination dynamics should regularly monitor atmospheric deposition, soil properties, and land use changes, especially in the western part of the study region, where high background concentrations of heavy metals in soils were found.

Communities aiming at reducing pollutants exposure could adopt comprehensive management practices such as installing forest vegetation buffers around heavily polluted factories while reducing industrial emissions and wastewater discharge and managing sewage irrigation and cropland chemical uses in agricultural lands. Optimizing landscape patterns through land use change (e.g., increasing percentage of forest and grassland, reducing patch density of farmland) and improving soil physiochemical properties (e.g., increasing pH and organic matter) might be the most effective management strategies in alleviating soil contamination especially in western Pearl River Delta region.

Declaration of competing interest

The authors declare no competing financial interest.

CRediT authorship contribution statement

Cheng Li: Methodology, Formal analysis, Writing - original draft. **Georgina M. Sanchez:** Methodology, Formal analysis, Writing - review & editing. **Zhifeng Wu:** Writing - review & editing. **Jiong Cheng:** Writing - review & editing. **Siyi Zhang:** Writing - review & editing. **Qi Wang:** Writing - review & editing. **Fangbai Li:** Writing - review & editing. **Ge Sun:** Writing - review & editing. **Ross K. Meentemeyer:** Writing - review & editing.

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Appendix A. Supplementary data

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