



# Distributional Characteristics and Source Identification of Cadmium in Soils of the Pearl River Delta, China

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## Abstract

The results of the Multi-Purpose Geochemical Survey in the Pearl River Delta (PRD) show that the pollution is serious. In this study, the influence of geological genesis, soil-forming process, and human activities on soil quality in PRD is analyzed, and the influence factors, genesis and spatial distributional characteristics of cadmium (Cd) in different soil depths are studied by inverse distance weighted (IDW) and hot spot analysis. The results show that the spatial distribution of Cd is significantly different in PRD and high-value is mainly concentrated in the central cities of Guangzhou-Foshan-Jiangmen-Zhongshan-Zhuhai. Moreover, hot spots with higher Cd content in deep are mainly along Beijiang, Dongjiang, and Pearl River Estuary (PRE). Overall, our findings suggest that the high background value areas formed by marine-land and fluvial sediments as well as intensive human activities that make PRD become an area under the dual restriction of geological genesis and human activities, pollution control cannot be ignored.

**Keywords** Geological background · Human influence · Soil parent material · Spatial distribution difference · The Pearl River Delta

Natural geological origins and human activities are the main sources of heavy metals in soils. The soil consists of solid, liquid and gas three parts where the minerals that make up the solid part are mainly the primary and secondary mineral particles of rocks, so the content of heavy metals in soils is closely related to the original soil-forming parent materials, and parent materials directly affect the background value of heavy metals in soils (Facchinelli et al. 2001; Zhang et al. 2003; Zhou et al. 2007). The geological structure is complex, the weathering of rocks is intense, granites are widely exposed, and magmatic rocks dominated by intrusive rocks accounts for about 1/3 of the distribution area in PRD (Song et al. 2012). Furthermore, a large number of metal elements enter the river and soil in the form of point source (mining and smelting activities, etc.) and non-point source (fertilizers and pesticides, etc.), causing agricultural production affected (Christophoridis et al. 2009; Fu et al. 2014; Gao et al. 2016;

Liu et al. 2013; Lai et al. 2016). The pollution sources are diverse and complex, strongly influenced by human activities and geological origins in PRD, so it's imperative to strengthen the research on the sources of heavy metals in soils.

In the past, the data on soil pollution was always kept silent until Guangdong took the lead in publicizing the proportion of heavy metal polluted soil in PRD reaching 28% in 2013, which attracted public attention to the problem of soil pollution. Parent rock, primary minerals, and secondary minerals are the main intrinsic factors of heavy metals, the strata of PRD are relatively well-developed and diverse. Additionally, the quaternary loose sedimentary and the residual soil layer in the bedrock area are well developed, which limestone, granite, and sediment are widely distributed from the residual and transported deposits (Zhi et al. 2007; Chen et al. 2000). “Maternal rock and climate combination type” is the decisive intrinsic factor of trace element content in zonal soil and different parent soils have different effects (He et al. 2017). Estuarine sediments are an important sink for heavy metals and the dense river network provides conditions for heavy metal migration, forming Xijiang and Beijiang alluvial areas as the main enrichment areas of the Quaternary delta, and Tanjiang and Dongjiang delta alluvial areas

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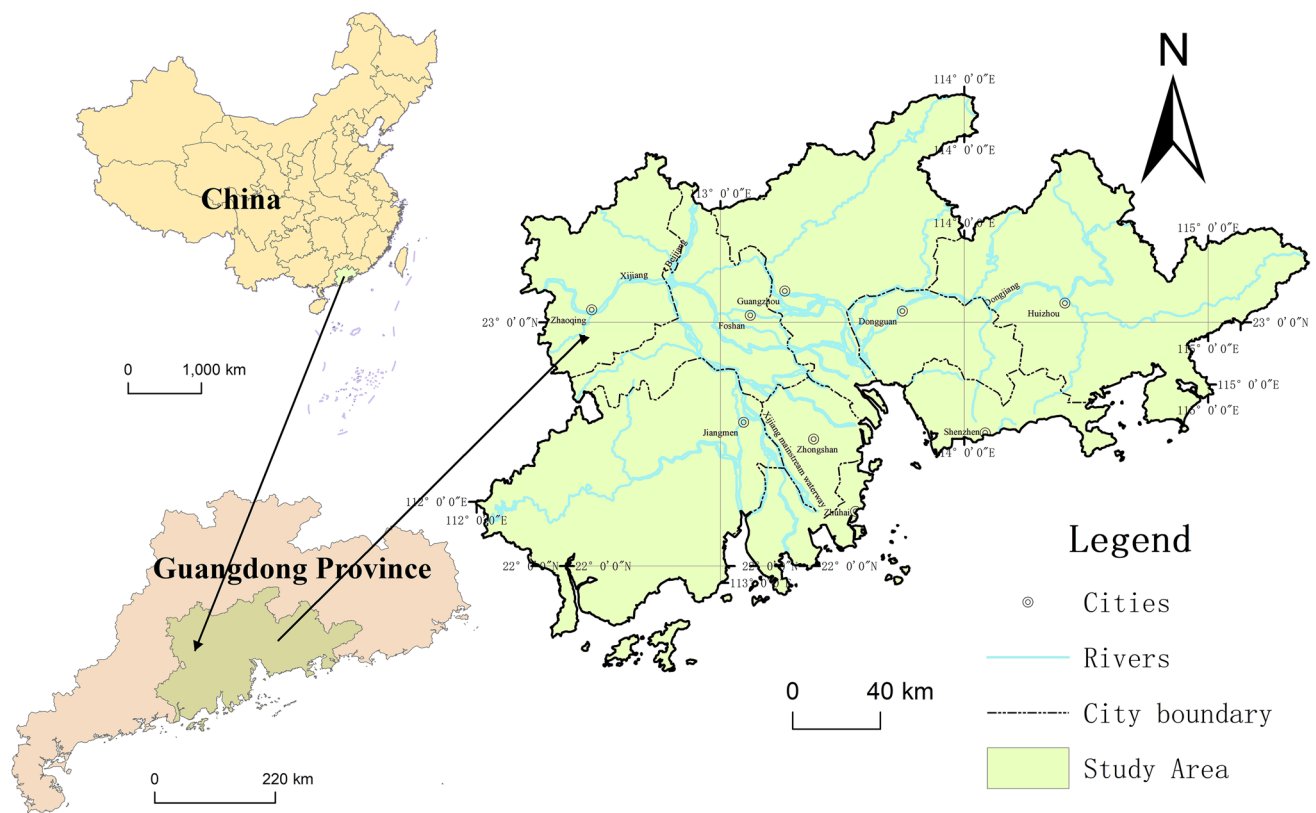
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as the distribution situation of high background areas (Ip et al. 2004; Liu et al. 2012; Hudson-Edwards et al. 2001; MacDonald et al. 2000; Sutherland 2000). The social and economic activities and the rapid development of urbanization in PRD have made the pollution problem more serious, some studies pointed out that PRD has gradually become the region with the fastest change of the ecological environment, the most serious resource depletion and serious environmental pollution in China (Zheng et al. 2008). Coal-burning, motor vehicles, and industry are important internal factors leading to the content of heavy metals increase in PRD, and the main sources of Cd in agricultural soil are agricultural farming, transportation activities, land-use types and soil management (Wong et al. 2003; Li et al. 2009; Cai et al. 2019). It has been pointed out that mining, manufacturing, and rock weathering are the main sources of heavy metals in the waters of Asia (Zhou et al. 2020), soil environment, especially heavy metal pollution, has become a hot topic of public concern, which seriously threatens the safety of soil-crop-human (Wang et al. 2019).

PRD is an important geographical and economic region of China where is located in the south-central part of Guangdong Province and at the mouth of the Pearl River, which includes Guangzhou, Shenzhen, Foshan, Dongguan, Zhongshan, Zhuhai, Huizhou, Jiangmen and Zhaoqing (Fig. 1).

Jiangmen and Zhaoqing are located in the west, Guangzhou and Foshan are located in the hinterland, Zhongshan and Zhuhai are located in the south, Dongguan is located in the northeast, Shenzhen is located in the East Bank, and Huizhou is located in the northeast. At present, the “Great PRD” metropolitan area with Guangzhou, Hong Kong and Shenzhen as the core has become the world’s largest super metropolitan area and important economic and industrial center of China (Chen et al. 2012), but the good interactive development of cities in PRD has also brought new impacts and challenges to environmental governance. PRD is the most important population gathering area in Guangdong Province. At present, many studies have shown that Cd is the most important heavy metal pollution element in PRD. Liu et al. (2019a, b) found that human activities have a significant impact on heavy metals, the content of heavy metals in water and sediment samples increases gradually from the upstream to the downstream, and Cd in sediment has a high activity and bioavailability, which have huge potential harm to the regional ecosystem. Zheng et al. (2020) assessed the potential risk of heavy metal pollution in soils and crops of urban agglomeration in PRD and obtained that Cd is the most common heavy metal pollutant. Therefore, combined with the above research, this paper analyzes the spatial distributional characteristics of Cd in surface and deep soils of



**Fig. 1** Location of the study area

PRD, strengthens the identification of pollution causes and influencing factors of Cd, as well as the regions affected by geological causes and human activities in PRD.

### Materials and Methods

According to the 6th census, the total population of PRD has reached 55.9409 million and the relatively concentrated population distribution area has gradually expanded from the middle of PED to the East (You et al. 2013). PRD mainly develops a comprehensive industry based on light industry and it is one of the densely populated areas of China’s township industry with a large number of industries, high density, and wide coverage, the demand for mineral resources and metals are huge, but the discharge of waste is relatively lagging. The original data are geochemical and pH distribution maps in surface and deep soils in the Multi-Purpose Regional Geochemical Survey Maps of PRD Economic Zone (1:250,000) by Guangdong Provincial Geological Survey Institute in 2011.

Besides the Multi-Purpose Regional Geochemical Survey Maps of PRD Economic Zone, the data of the 1:1 million SOTER terrane matrix factor in Guangdong Province is derived from the National Science and Technology Basic Condition Platform-National Earth System Science Data Sharing Platform (<http://www.geodata.cn>).

The spatial interpolation analysis method is widely used in the research of meteorology, agriculture, land, and other fields (Chen and Liu 2012). The paper uses the IDW to study the distribution of heavy metals. The formula is:

$$\hat{Z}(s_0) = \sum_{i=1}^N \alpha_i Z(s_1) \tag{1}$$

where  $\hat{Z}(s_0)$  is the predicted value at  $s_0$ ; N is the total number of samples;  $\alpha_i$  is the weight of each sample;  $Z(s_1)$  is the measured value obtained at  $s_1$ (Tang and Yang 2011), IDW has strict requirements on each input point, which will decrease as distance increases.

Hot spot analysis has significant significance for spatial distribution feature recognition. If the feature itself is a high-value (low-value) and surrounded by other elements that also have high-value (low-value), then the area will be identified as a high-value area (low-value area) (Xu et al. 2019). This paper uses the Hot spot analysis tool (Getis-ord  $G_i^*$ ) on the ArcGIS 10.2 platform to identify hot and cold spots. The formulas are:

$$G_i^*(d^2) = \sum_{j=1}^n W_{ij}(d)X / \sum_{j=1}^n X_{ij} \tag{2}$$

$$Z(G_i^*)^2 = G_i^* - E(G_i^*) / \sqrt{Var(G_i^*)} \tag{3}$$

where  $G_i^*$  is a commonly used indicator to describe cold and hot spots,  $Z(G_i^*)$  is used to identify the spatial distribution in different regions,  $W_{ij}$  is the spatial weight between elements i and j(the reciprocal of the distance between sampling points i and j),  $X_i$  and  $X_j$  are the values of variables x on attribute feature X on i and j, respectively, Var is the variance of random quantity, E is the expectation (Zhang et al. 2015a, b). In this paper, the natural discontinuous point grading method is used to divide spatial distribution areas into high-value accumulation areas, high-value dispersion areas, general area, low-value dispersion area, and low-value accumulation area.

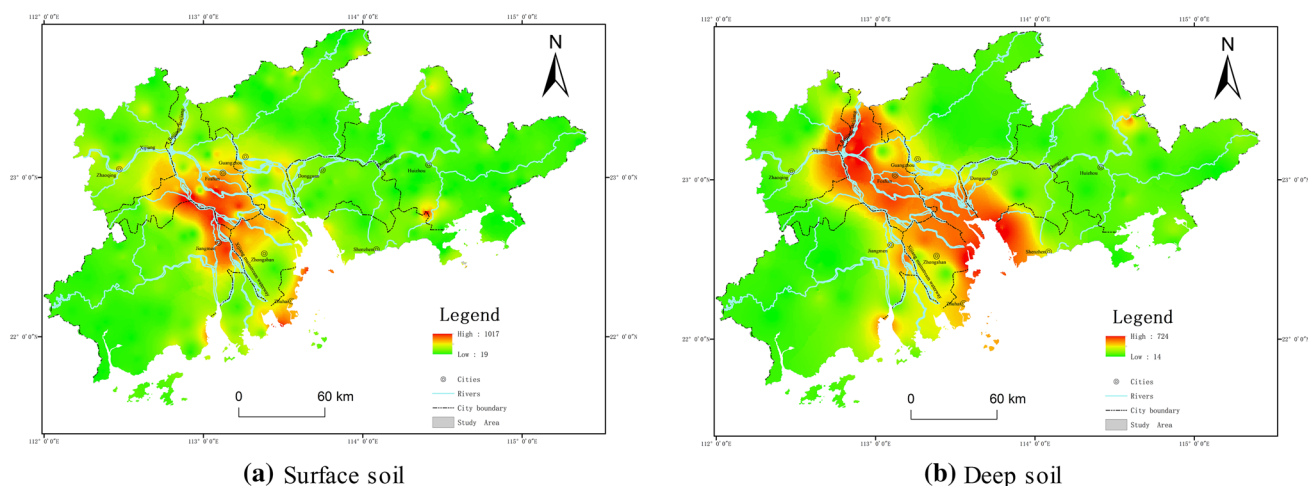
The content of heavy metals varies in different regions, it is necessary to divide pollution areas for pollution control. The pollution zone of this paper mainly uses the local soil environmental background value. The formula is:

$$R = C/B \tag{4}$$

where R is the ratio of the content of an element, C is the content of an element in the soil, and B is the background value of the soil environment of an element(Guo and Wu 2014). The background value is the normal content of elements in various natural bodies (rocks, weathering products, soils, sediments, et al.) under normal geographical conditions and geochemical conditions, where soil background value refers to the content of elements in the soil that hasn’t been or less polluted and destroyed. To provide suggestions for pollution control and treatment, discriminate pollution areas by Cd content distribution at different depths and pollution zoning methods are necessary. The latest authoritative data of soil environment background value in PRD is the “Soil Heavy Metal Risk Assessment Screening Value-PRD (DB 44/T1415-2014)” issued by the Guangdong Provincial Bureau of Quality and Technical Supervision in 2014, in which the background value of Cd is 0.11 mg/kg. The minimum environmental risk control value of soil Cd is 1.5 mg/kg (pH ≤ 5.5) in Soil Environmental Quality - Agricultural Land Soil Risk Control Standards (trial) (GB15618-2018), which can obtain surface and deep soils Cd content didn’t exceed the risk control-value. Comparing the content of Cd in surface and deep soil with the background value, if  $R \leq 1$ , it is a non-pollution area, otherwise it is a pollution area.

### Results and Discussion

Geochemical maps of Cd elements in surface and deep soils of PRD were vectorized and the spatial distribution of Cd in soils was obtained by IDW (Fig. 2a, b), which shows that the content of Cd in the surface soil of PRD is generally low and median, while high-value is mainly concentrated in the



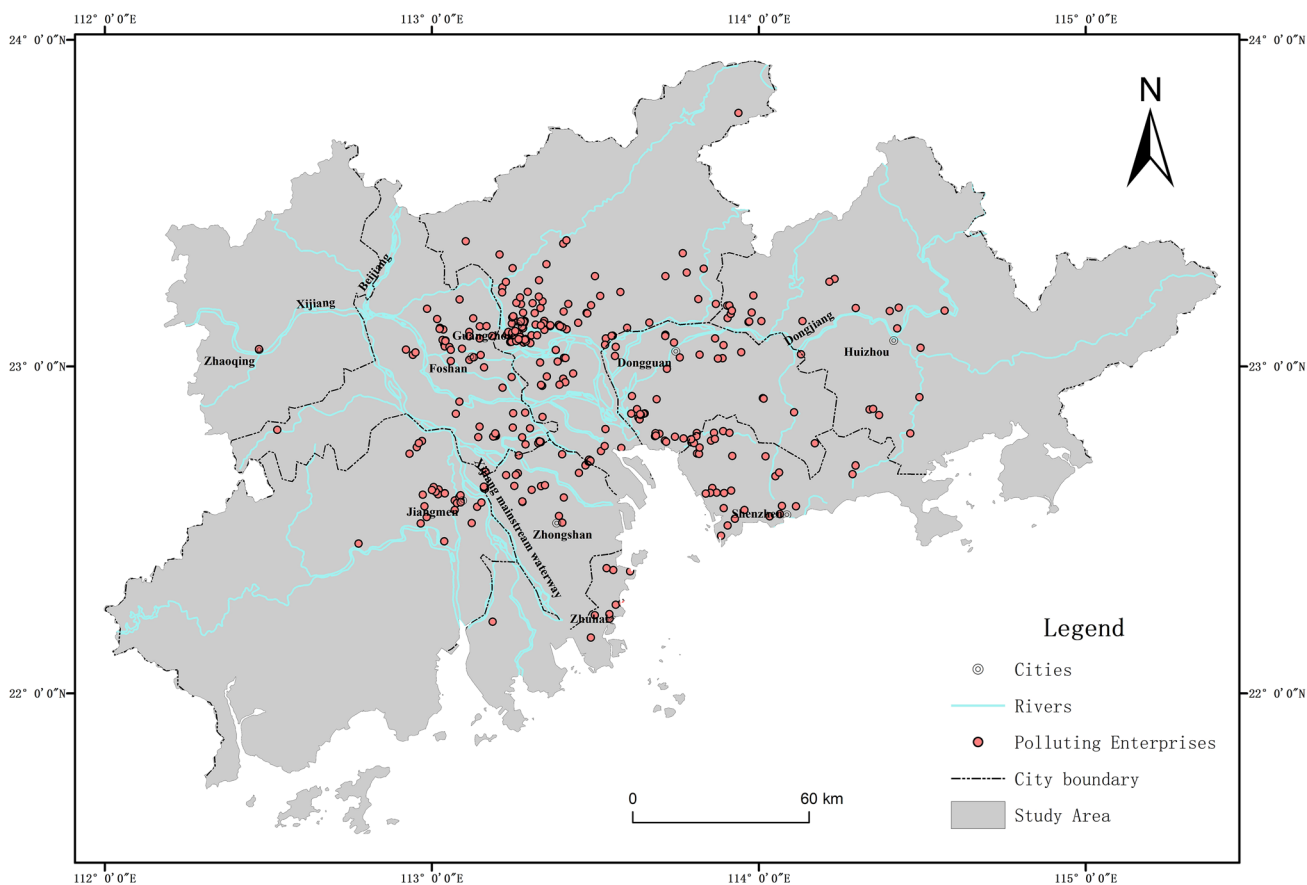
**Fig. 2** Spatial distribution of Cd in soils

area west of the central part, such as Foshan, Jiangmen and the area along the main watercourse of Xijiang River, while other areas are sporadic in the form of plaques. The degree of heavy metal pollution in surface sediments in most areas of PRE is medium to low, and the heavy pollution areas are mainly distributed in the vicinity of the Pearl River Eight Gates (Ma et al. 2016). Moreover, the Cd content in deep soil has a certain similarity with the surface soil which the low-value and the median-value occupy most of the area, the high-value mainly concentrates in the intersection of Xijiang and Beijiang and central cities, as well as some areas where Zhongshan, Zhuhai, and Shenzhen are located, while the overall coverage area is higher than the surface soil.

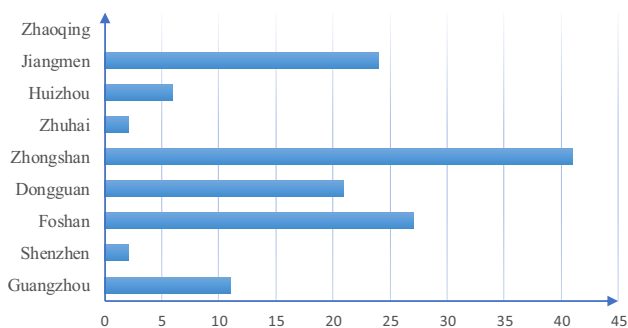
The change of heavy metal content in surface soil is closely related to human activities (industrial and mining, domestic garbage and agricultural materials, et al.), among which the enterprise is an important potential source of heavy metal pollution, which are the main reasons for the increase of heavy metal content, especially electronic factories and enterprises. Through the “Green Net”, we count enterprises in 9 cities of PRD that may cause pollution to soil (Fig. 3) which shows that polluting enterprises are mainly concentrated in Guangzhou-Foshan-Shenzhen-Dongguan-Jiangmen and other cities in the central of PRD, which is inextricably linked to the rapid development of PRD industry, the petrochemical, pharmaceutical, leather, and electronics factories are developing rapidly which causes many pollutants such as Cd, Pb, and Ni to increase that flow through the soil, the content of heavy metals will rise and soil environmental quality is threatened. Studies on heavy metals in sediments from the west coast of Shenzhen indicated that Cd mainly comes from industrial wastewater (Huang et al. 2018), there is a spatial consistency between the distribution of industrial enterprises and the heavy metal contaminated areas of soil, and irrigation channel is the transmission way

of soil Cd pollution, which affect the surrounding soil and groundwater (Wang et al. 2020; Yuan et al. 2019).

Different types of enterprises have obvious effects on heavy metals, and the types and contents of heavy metals in the surrounding soil may be different. The influence of enterprise type on Cd is obvious (electronic enterprise > furniture enterprise > hardware enterprise) (Wu et al. 2008), researchers pointed out that the Cd content of electroplating industry accounted for about 57%, and the main pollution areas were concentrated in the confluence of Xijiang River and its tributaries in PRD (Liu et al. 2019a, b). Zhao et al. (2019) found that the heavy pollution areas of Cd were basically concentrated around large and medium-sized electroplating enterprises in soils of typical industrialized and urbanized cities in South China, Ozaki et al. (2019) also proposed that the plant was the main emissions source. Figure 4 shows the number of concerned plots around the electronic and electroplating enterprises in the cities of PRD, which are concentrated in the south-central part, among which the number of Zhongshan, Foshan, Jiangmen, and Dongguan are large, which is also the most significant area of Cd spatial distribution. The enrichment of heavy metals is mainly controlled by human pollution (Sun et al. 2017), in recent years, the content of heavy metals has declined after treatment, but the environmental pollution problem is still very prominent. The major anthropogenic inputs of heavy metals to soils and the environment were metalliferous mining and smelting in the north of Guangdong Province. Heavy metal elements in PRD are widely distributed in Guangzhou-Foshan and its surrounding economically developed areas where soil heavy metal pollution is relatively serious are mainly in the vicinity of metal smelting processing and electroplating enterprises, what's more, the large-scale development of chemical and pharmaceutical industries are the main factor of heavy metals content high (Dou et al. 2015a, b; Lu and Feng 2012).



**Fig. 3** Distribution of polluting enterprises in PRD



**Fig. 4** Number of polluted plots around electronic/electroplating enterprises

Researchers point out that the Cd pollution caused by human activities, Cd generally stays and accumulates at 0–15 cm of the soil surface for a long time, and the content of pollution elements decreases with the deepening of the soil layer, so crops that absorb nutrients from the surface soil are vulnerable to the pollution which has a huge impact on the soil and crop safety in PRD (Li et al. 2009; Li and He 2016).

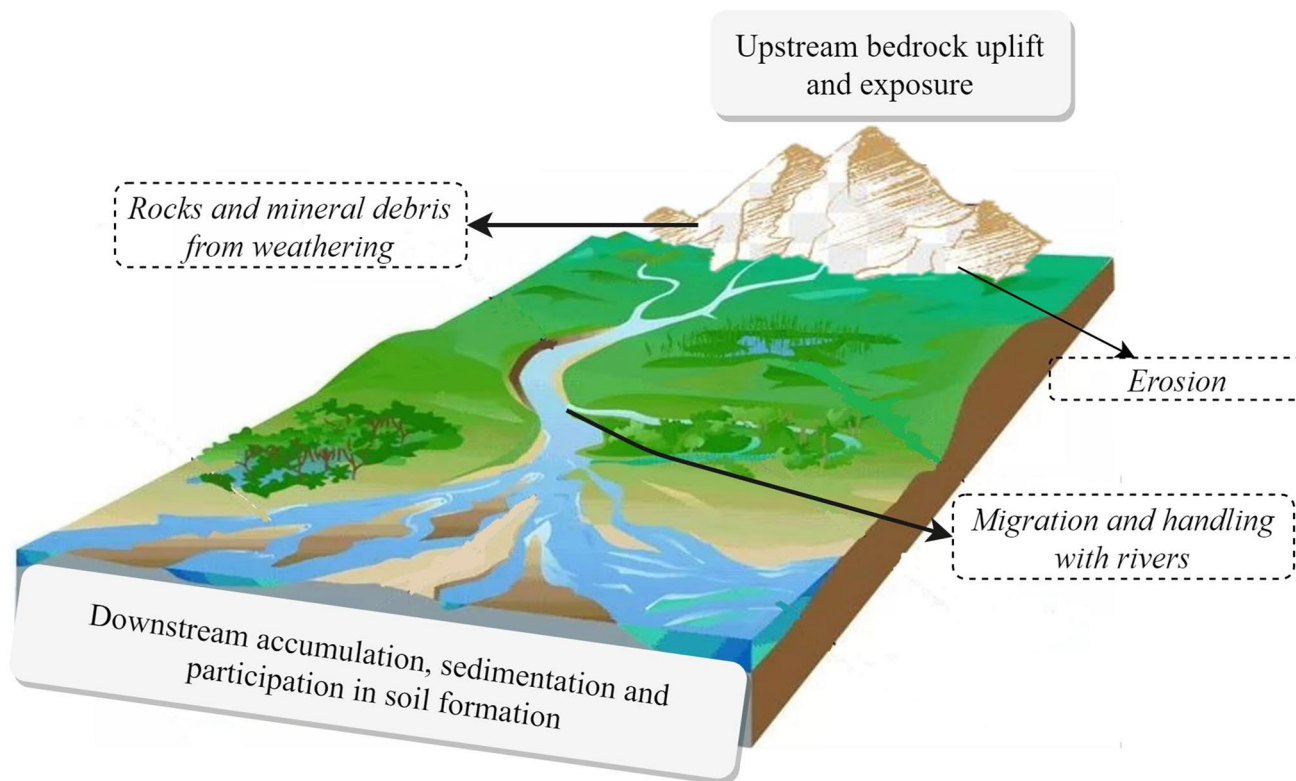
PRD has a variety of landforms where is a typical area of magmatic rocks dominated by acidic granite. There are

many rocks such as dolomite, marble, and granite in mountains, hills, and plains. The parent materials are the internal cause of heavy metal sources and different parent material types determine the initial heavy metals content in soils (Gu et al. 2014; Micó et al. 2006). The change of heavy metals content in deep soil is closely related to soil-forming parent materials and various primary and secondary minerals in soil-formation. Weathering causes the bedrock to break, the physical and chemical properties change, and form a loose weathered shell and a large amount of debris. The weathered material migrates under the influence of many environmental factors such as rivers and winds, and finally collects and deposits in the downstream. During the process, heavy metals carried in bedrock also migrate and deposit, which makes heavy metals content in downstream soil increase (Zhen et al. 2016). The river is an important thrust force for heavy metal migration, which will directly affect the environmental quality of the river coastal areas, the content of heavy metals in sediments is high in the west of PRE (Fan et al. 2002; Wang et al. 2010; Feng et al. 2012); Geng et al. (2015) pointed out that natural geology, especially surface erosion, is the main factor affecting heavy metals. The soil quality of PRD is controlled by geological and geochemical

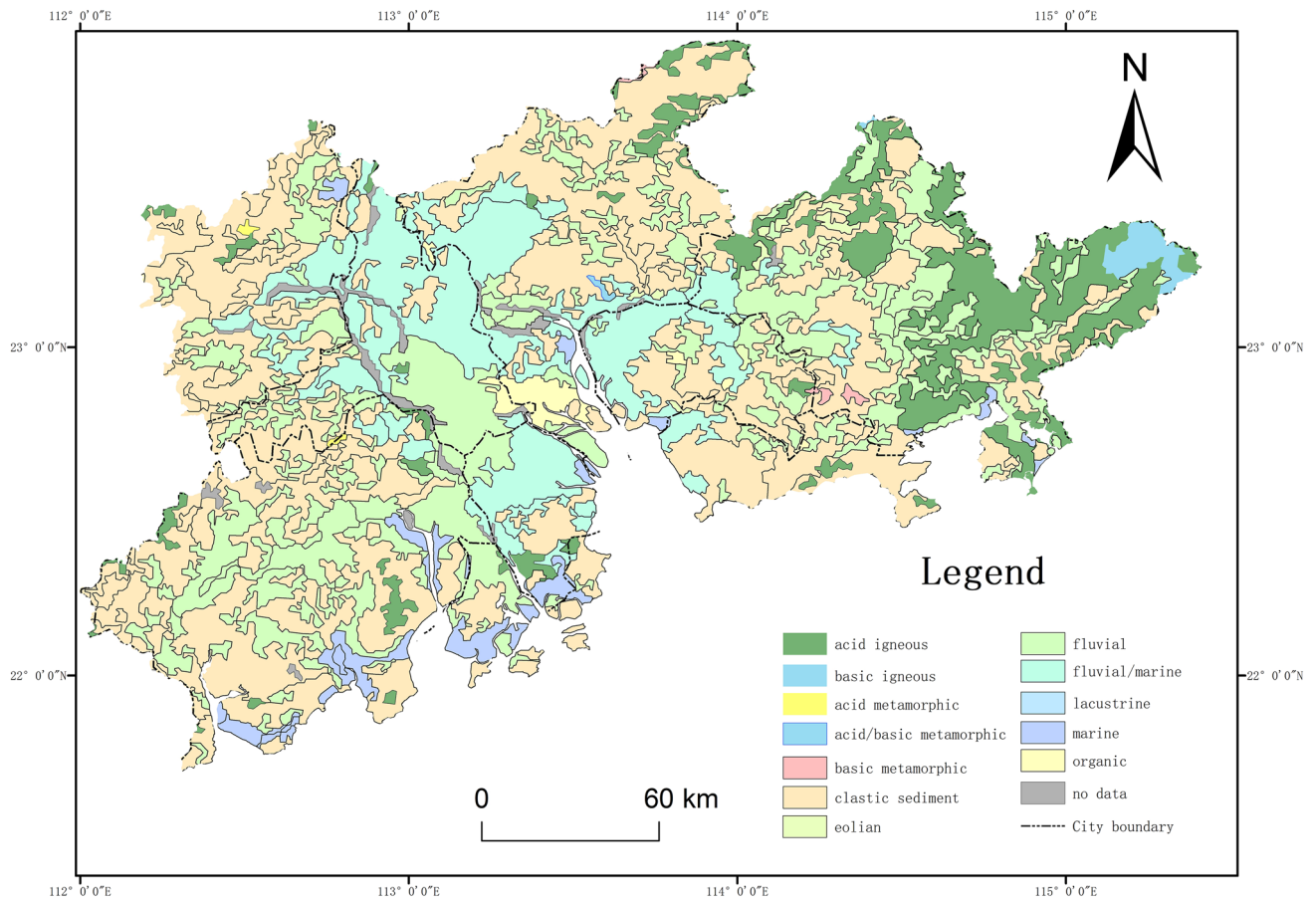
background where high background areas are directly related to the widely distributed metal deposits as well as marine, land, and fluvial sediments (Chen et al. 2011). The river network in PRD is densely distributed, and heavy metals migrate and transport along rivers, where the rivers along Xijiang and Beijiang are controlled by soil-formation and rock weathering, which makes the soil background value of heavy metals high (Fig. 5). The above analysis shows that the high Cd content in surface and deep soils are mainly concentrated in the south-central cities, forming the main gathering area of Cd. Spatially, Cd concentrations were higher in PRD and north of Guangdong Province, while the Cd content is at middle and low levels in other areas. The overall environmental quality of PRD is still good, and high Cd content areas are mainly concentrated in southeast coastal sedimentary areas with strong marine-land and land-marine interaction, and the high content of Cd are related to geological background and human activities (Dou et al. 2014; Liu et al. 2012; Han et al. 2018).

To further identify pollution sources and compare the distributional characteristics in soils, combining the parent material distribution of PRD with the content of heavy metals in surface and deep soils. Fig. 6 shows that the distribution of the parent materials is extensive in PRD where abundant fluvial parent materials, marine parent materials, and clastic sediments are accumulated in the

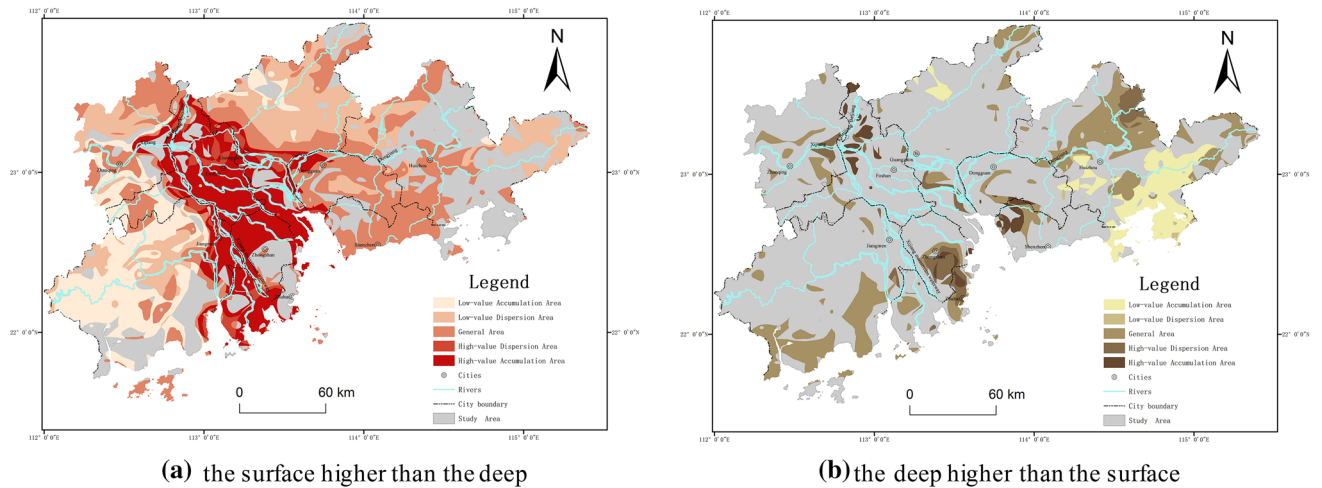
central part except the central and eastern parts, while granite is mainly distributed in the east. It can be seen from Fig. 7a that high-value areas of the surface higher than the deep are concentrated in the central, covering cities such as Guangzhou-Foshan-Jiangmen-Zhongshan-Zhuhai, where possess rapid economic development, prosperous handicrafts, and leading electronics industries, causing increased heavy metal content in surface soil. The low-value areas are related to the weaker human disturbance nearing the Dasha River and Jinjiang reservoirs in the west. Many hot spots existed throughout PRD region due to the transportation and pollution of the river, due to abundant sediments downstream of rivers and exceeding human activities, heavy metal pollution is aggravated and pollution areas are mainly distributed in the fluvial channel and outside the estuary (Sun et al. 2017; Bi et al. 2017). Figure 7b shows that high-value areas of the deep higher than the surface are mainly distributed along Dongjiang, Beijiang, and Shenzhen-Zhongshan-Zhuhai where soil parent material as well as sediments from a large number of fluvial and marine parent materials deposit and aggregate around rivers. High-value dispersion areas are distributed in the southwest and northeast, which are related to the variety of rocks in PRD, especially the extensive exposure of granite. Residual quartz sand in granitic soil enhances Cd activity (He et al. 2017).



**Fig. 5** Formation and migration process of parent rock (source: Zhijindong cave Global Geopark (<http://www.gzzjd.com/en/>))



**Fig. 6** Distribution of soil parent materials in PRD



**Fig. 7** High-value and low-value distributional characteristics of Cd

The result of the correlation between the surface and deep soils of PRD shows that the source of heavy metals is not caused by a single factor (Zhang et al. 2015a, b), thus to research the source must analyze the surface and deep

together. Lin et al. (2017) used principal component analysis to explain why fluvial migration in PRD is responsible for the higher content of heavy metals downstream of rivers, but industrial sources, natural and agricultural sources are

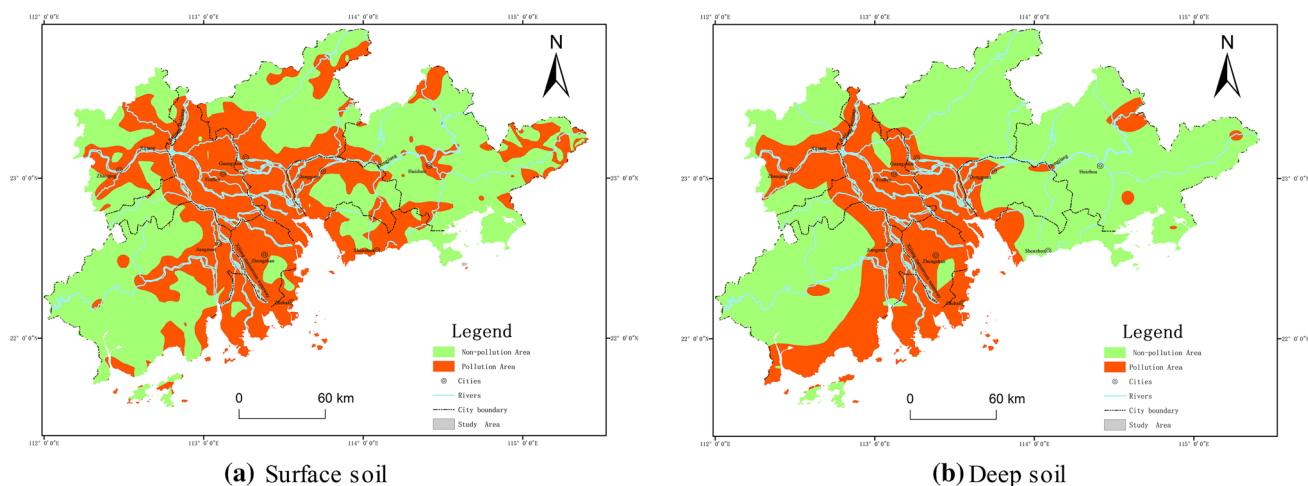
also cannot be ignored. Heavy metals in soils affected by geological genesis and human activities migrate, making the pollution areas more dispersed. In the hot spot analysis, there are similarities between the results of the surface soil of Cd content larger than that of the deep soil and the distribution of polluting enterprises, and the distribution of Cd in the deep layer is more than that in the surface layer, which is closely related to the distribution of the main river systems in PRD.

Combining the surface and deep Cd element identification results, and obtaining pollution zoning results in PRD (Fig. 8a, b), which shows that the surface pollution area is more than the deep pollution area. Cd content is lower than the soil environment background value, mainly in the southwest and northeast regions, and surface and deep pollution areas are concentrated in south-central cities such as Guangzhou-Foshan-Jiangmen-Zhongshan-Zhuhai, while other areas have scattered pollution. The formation of pollution areas is closely related to sediments and soil-forming parent materials which are frequently affected by fluvial migration and sedimentation as well as human activities, making PRD become a pollution area under the dual influence of geological genesis and human activities. Cd content in the southwest and northeast regions will be lower and high-value is distributed in the form of plaques in Foshan - Jiangmen - Zhongshan and other cities (Li et al. 2015).

In summary, the spatial distribution of Cd in PRD is constrained by geological conditions and human activities. Surface soil heavy metal content increasing and pollution are mainly caused by human activities, which are related to the industrialization and urbanization developing rapidly in PRD, causing pollution status cannot be ignored and emission of pollutants increasing, which is also confirmed in the distribution of polluting enterprises; the deep-pollution areas are concentrated in the central south and the intersection

of Xijiang and Beijiang which are closely related to the heavy metal content of soil background, which make the area show pollution characteristics. Research shows the Cd content is not only affected by the parent material and fluvial migration, but also various human activities and polluting enterprises are also important factors to aggravate pollution. There are obvious regional characteristics in the area where most of the high-value areas are distributed around the water system. The sedimentary materials migrate along rivers and accumulate downstream of rivers, but anomaly occurrence in some areas.

In this work, Cd is selected to analyze the sources and spatial distributional characteristics in different soil depths. The study shows that the high value of Cd content in the surface and deep soil of the Cd pollution in PRD are mainly concentrated in the middle, overlapping with the population distribution concentration area and urban areas with rapid economic development, high-value in deep soil also includes the surrounding area of PRE where is related to the movement and accumulation of sediments around rivers. Cd in surface soil is directly related to the rapid economic development and human activities, electronic/electroplating enterprises are the main sources of Cd, which makes the plots around the enterprises most vulnerable to pollution and reduces the soil quality, so it is an urgent problem to strengthen the pollution control of enterprises. Cd in deep soil is directly related to the heavy metal elements in the parent materials that carried by rivers in PRD, Cd pollution is concentrated at river confluence and downstream, where more Cd containing fluvial/marine and clastic sediments are accumulated. The sources of Cd are diverse and subject to the dual constraints of geochemical background as well as human activities, which shows a trend of high in central and southern surrounded by river intensive areas, and low in other regions.



**Fig. 8** Pollution area of Cd in soils



The change of soil Cd content in PRD is the result of the joint influence of geological genesis and human activity sources where heavy metal anomalies are affected by the upstream ore-forming geological background and sedimentary environment of marine-land interaction. Besides being affected by human activities (manufacturing factories, industrial, and mining enterprises), the impact of natural geological origins also accounts for an important reason in the high-value area of Cd in south-central PRD, making it a heavily polluted area (Liu et al. 2012; Wang et al. 2008; Lai et al. 2016). Furthermore, the surface Cd pollution is easy to accumulate in the surface layer, but with the long-term irrigation of the soil, the content of heavy metals migrating downward will increase and accumulate in the deep layer. Given the pollution situation in PRD, it is necessary to take pollution control measures (Dou et al. 2013), Pollution zoning is a common practice for pollution prevention, for pollution areas, on the one hand, the surface polluted soil is mainly controlled and supplemented by means of remediation to treat in time. On the other hand, to prevent the migration of pollutants and reduce the risk of soil environment, it should be combined with existing physical, chemical, biological and other technical measures, and agronomic measures should be taken first to reduce the concentration of heavy metals or change its state of attachment in soils for the effects of engineering measures to deep soil are limited.

The soil background value cannot be ignored, geological causes the content of heavy metals in the soil exceeds the standard. There is a great difference between the mechanism of heavy metal change influenced by geological genesis and human activities. It is difficult to identify the genesis of heavy metal because of the migration and transformation after enrichment in the complex system of soil. Moreover, the migration of heavy metals in soils leads to uncertainty in identifying pollution sources by correlation when heavy metals content in surface and deep soils are similar, at the same time, the identification of pollution areas will also have interference. Therefore, the pollution situation of surface and deep soils concentrated in the central region needs to be further clarified. Because the data in this work come from the surface (0–20 cm) and deep (150–200 cm) soils in the Multi-Purpose Regional Geochemical Survey, the analysis of heavy metal content in the middle of the soil is lacking and there is still room for research on the migration of heavy metals in soils and the force of specific factors, so the source identification of heavy metals needs to be further optimized.

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