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Effects of Urbanization Intensity on the Distribution of Black Carbon in Urban Surface Soil in South China

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Abstract: Rapid urbanization causes the accumulation of large amounts of pollutants, including heavy metals, organic pollutants, and black carbon (BC). BC is the carbonaceous residue generated from the incomplete combustion of fossil fuels and biomass. It plays an important role on the migration of heavy metals and organic pollutants, as well as soil carbon sequestration. BC accumulation due to human activities greatly affects the global carbon budget, helps to drive climate change, and damages human health. To date, few studies have examined how the intensity of urbanization affects the distribution of BC in soils in urban areas. Therefore, the objective of this study is to determine the effects of urbanization intensity on the spatial distribution and content of BC in urban surface soil. We collected samples from 55 sites in South China and used a multi-scale geographical regression model to evaluate the impact of the interference intensity of urbanization on the amount and distribution of BC. Our results showed that the BC content was significantly higher in urban areas (9.74 $\pm$ 1.18 g kg^{-1}) than in rural areas (2.94 $\pm$ 0.89 g kg^{-1}) and that several urban parks with a higher interference intensity were hotspots of BC accumulation, suggesting that urbanization promoted BC accumulation. Our model revealed that road density was significantly and positively correlated with BC accumulation. Because there are more cars driving in areas with high road density, vehicle emissions may be one of the causes of BC accumulation. Our results also indicated that the impact of urbanization intensity on the BC distribution was sensitive to sampling density.

Keywords: urban soil; black carbon; urbanization; multi-scale geographically weighted regression

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1. Introduction

Black carbon (BC) is a component of fine particulate matter (PM $\leq$ 2.5 µm in aerodynamic diameter). Black carbon consists of pure carbon in several linked forms. It is formed through the incomplete combustion of fossil fuels, biofuel, and biomass, and is one of the main types of particles in both anthropogenic and naturally occurring soot [1,2]. Human activities, such as fossil fuel combustion, vehicle emissions, and garbage incineration, significantly increase BC accumulation [3]. Most BC eventually enters the soil and becomes part of the soil carbon pool [4]. It is estimated that BC accounts for 5–45% of surface soil organic carbon (SOC) and, in some places, as much as 60% of SOC, making it an important part of the soil carbon pool [5,6]. In addition, BC accumulation caused by human activities greatly impacts the global carbon budget, helps to drive climate change, and are deleterious to human health [4,7–10].

BC is common in soil disturbed by humans, including urban soil, and serves as a record of human activity in an area [11–13]. Previous studies showed that the amount of BC in the urban surface soil was strongly affected by human activities, such as agricultural activities, industrial production, fossil fuel burning, and vehicle emissions [14–16], and that its content was significantly higher in urban soil than in desert and agricultural soil [17]. In addition, there were significant differences in the soil BC content in different urban functional areas, and the amount of BC was significantly higher in industrial areas and in road green belts (which refer to the median, or the grassy area that separates two lanes of traffic going in opposite directions) than in other areas [18,19]. Some studies demonstrated that industrial activities and vehicle emissions were critical factors affecting BC accumulation [20,21].

Although many previous studies have examined the soil BC content, most of them focused on forest, agricultural, and desert soils, rather than on BC in urban soil with a large human population [11,17,22–28]. In addition, there are limited reports on the soil BC content in cities in China; research has mainly focused on the old industrial cities of Xuzhou and Anshan in north China [4,29] and Shanghai and Nanjing in east China [13,18]. However, different urbanization processes significantly affect the distribution of urban soil BC. South China, the area that has pioneered China's economic development, has a high degree of urbanization, and a great deal of human activity will inevitably cause BC accumulation. Therefore, it is imperative to explore the content and distribution of BC in urban soils in south China.

Urban soil BC may come from the natural and anthropogenic combustion of biomass and fossil fuels [6]. The properties and sources of BC affect its role in the biogeochemical cycle, thus affecting its environmental behaviour. Therefore, analysing the BC sources in urban soil will help us to understand further the effect of urbanization on soil BC accumulation and its environmental behaviour. The BC/OC ratio is often used to distinguish the sources of BC because of its high credibility and low cost [11]. The BC/OC ratio in soil reflects the degree of soil pollution and is related to specific human activities. A BC/OC value of approximately 0.1 indicates that BC mainly comes from biomass combustion; a value of approximately 0.5 indicates that BC mainly comes from mineral combustion [13,30,31].

The intensity of urbanization affects the storage of urban SOC and the accumulation of heavy metals and other pollutants in the soil [32,33]. Previous studies showed that the accumulation of heavy metals in urban soils was mainly affected by the SOC content, soil cation exchange capacity, and human population growth. In addition, land-use patterns and the urban landscape contributed to its accumulation [34]. Due to the strong adsorption of soil BC to soil heavy metals and organic pollutants, there was a significant correlation between the urban soil BC and the heavy metal pollution index [35–37]. However, there were limited reports on the effects of urbanization intensity on the distribution and accumulation of BC [38]. We sought to study the relationship between urbanization intensity and soil BC distribution to provide a reference basis for evaluating the environmental implications of urbanization for soil BC. We sampled soils in Guangzhou, south China, a typical urban environment of the region, in order to determine: (1) the spatial distribution of BC in urban surface soil with different urbanization intensity, and (2) the effects of urbanization intensity on the content of BC in the urban surface soil.

2. Materials and Methods

2.1. Research Area and Soil Sampling

Samples were collected in Guangzhou, Guangdong Province, south China, which is one of the central cities in the Guangdong–Hong Kong–Macao Greater Bay Area and has the characteristics of a typical urban area. The climate is a subtropical marine monsoon climate with an annual average precipitation of 1623.6–1899.8 mm and an annual mean temperature of 28.6 °C. The soil is classified as latosolic red soil in natural ecosystems.

In order to determine the long-term effects of urbanization intensity accurately, it is necessary to sample areas with relatively stable soil environments. We made a preliminary selection of urban ecosystems based on satellite imagery along the arterial roads across

the study area [39]. Finally, we chose 55 urban parks and rural forests as sampling plots (Figure 1). The 55 sites were classified into four ecosystem types according to the location, vegetation cover, amount of anthropogenic solid waste, dominant plant type, and management intensity. The four typical ecosystem types with increasing urban disturbance intensity are rural forest (9 sites), urban forest (19 sites), urban woodland (15 sites), and urban park (12 sites). Rural forests usually refer to woodlands far from urban roads and buildings, while urban forests refer to woodlands surrounded by human structures and isolated from other soil ecosystems. Urban forests are mostly located in hilly areas and are less disturbed by human beings. Urban woodlands are usually small in area and close to urban buildings, so the soil of urban woodland is seriously polluted by human activities wastes (plastics, wires, paper, bricks, clothing, etc.). Urban parks are managed ecosystems in which vegetation is often trimmed, watered, and fertilized, with litter removed from the ground, and soil may be turned over and contaminated with solid waste. Specific classifications are shown in Table 1. Latitude and longitude coordinates of all sample sites were recorded. At each sampling site, topsoil (0–10 cm) was collected with a steel auger (5-cm diameter) from five randomly selected spots and then mixed as a single composite sample. Soil samples were brought to the laboratory and passed through a 2-mm sieve within one week. Roots, rocks, and visible residues were removed manually. After the samples were air-dried under ventilated and dry conditions, they were ground and sieved into 100-mesh (0.149-mm) particles for future analysis.

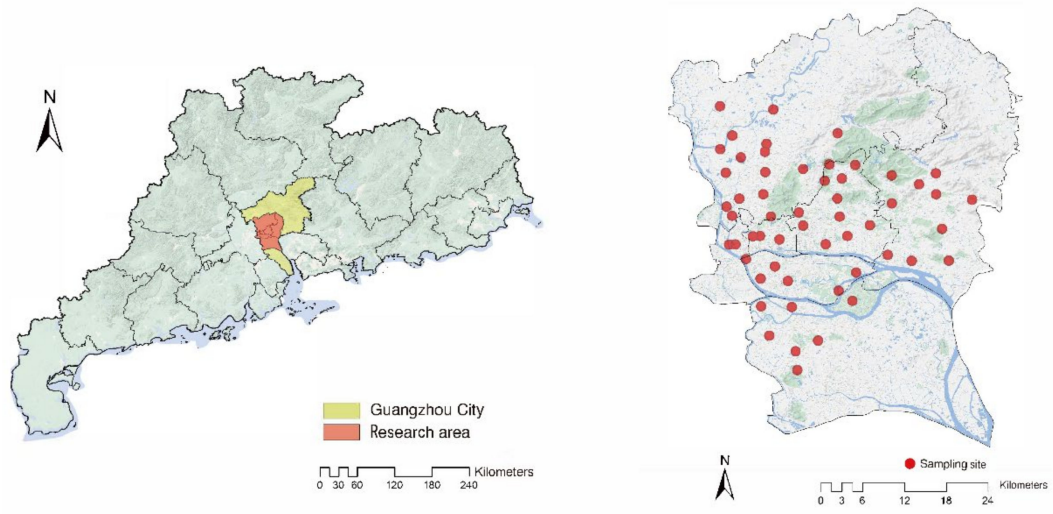

Figure 1. Study area and sampling sites.

Table 1. Classification based on ecosystem types and disturbance intensity.

	Location	Anthropogenic Wastes	Aboveground Litter Layer	Management	Disturbance Intensity
Rural forest (RF, 9 sites)	Rural	No	completed	No	Very low
Urban forest (UF, 19 sites)	Urban	Little	completed	No	Low
Urban woodland (UW, 15 sites)	Urban	Much	completed	No	High
Urban park (UP, 12 sites)	Urban	Little	absent	Yes	High

2.2. Laboratory Analysis

SOC was measured using the H_2SO_4–$K_2Cr_2O_7$ oxidation method [40]. Briefly, 0.5 g air-dried soil (0.149 mm) was acidified by a dichromate (H_2SO_4–$K_2Cr_2O_7$) solution for

30 min in a Pyrex digestion tube in a 40-tube block digester preheated to 170 °C. Then, the unreacted dichromate was estimated by the titration of the cooled digest with an acidified solution of ferrous ammonium sulfate with the use of *N*-phenylanthranilic acid as an indicator. BC content in soils was determined using the wet chemical oxidation method proposed by Lim and Cachier [41]. Briefly, 3 g of air-dried soil (0.149 mm) was added to a centrifugal tube and treated with 15 mL of 3 mol L^{-1} HCl for 24 h to remove the inorganic carbon. After centrifugation, the supernatant was decanted, and 15 mL of 10 mol L^{-1} HCl was added for 24 h to remove the CaF_2. Then the residue was oxidized with 15 mL of an acid potassium dichromate solution (0.1 mol L^{-1} $K_2Cr_2O_7$ and 2 mol L^{-1} H_2SO_4, 1:1, *v*:*v*) at 55 ± 1 °C for 60 h. During the reaction, $K_2Cr_2O_7$ was added until the colour changed to keep the oxidant in excess, and deionized water was added several times to maintain the volume of the solution. The residue carbon after this treatment was regarded as BC. The BC content of the residues was determined using an element analyser (FLASH EA-DELTA V, Thermo Fisher, Waltham, MA, USA).

2.3. Calculations and Statistical Analysis

The ratio of BC to OC reflects the sources of BC, calculated as:

BC/OC = the content of BC in soil/the content of OC content in soil.

The enrichment factor (EF) reflects the addition of BC to the soil from anthropogenic activity, calculated as:

$$EF = C_{sample}/C_{ref}$$

where C_{sample} and C_{ref} are the BC content in the soil sample and the background value of BC, respectively. In the present study, the lowest content of BC was used as the background value, 0.47 g kg^{-1}.

We used 2020 satellite images, a land-use status map, 2020 National Census Data, and Open Street Map to calculate urban built environment indicators that reflect the intensity of urbanization. These indicators were land-use diversity (LUD), the proportion of industrial land (PI), the proportion of green space in parks (PPG), the proportion of impervious surfaces (PIS), population density (PD), and road density (RD). All these indicators were calculated on three scales, with the sampling points as the centre and three different radii (500 m, 1000 m, and 1500 m).

$$LUD = \sum_{1}^{n} Pi \times \ln(Pi)$$

where *Pi* is the area of the *i*th land-use type, and *n* is the number of land-use types. In addition, PI = the area of industrial land/the total area, PPG = the area of park green space/the total park area, PIS = the area of impervious surface/the total area, PD = the population size/the total area, and RD = the length of road/the total area.

Multi-scale geographically weighted regression (MGWR) was used to analyse the relationship between urbanization intensity and the spatial distribution of soil BC using MGWR 2.2.1 [42].

Cluster analysis was performed using IBM SPSS statistics v. 21 (© 1989–2012 International Business Machines Corp., Armonk, NY, USA). Spatial distribution maps of BC content and the BC/OC ratio were obtained using geostatistical analysis of geography information system software (ArcGIS version 10.2, ArcGIS, Redlands, CA, USA). Figures were drawn with OriginPro 2016 (© 1991–2015 OriginLab Corporation, Northampton, MA, USA).

3. Results

3.1. Spatial Distribution of BC in Surface Soils

The spatial distribution of BC content and the BC/OC ratio in soils are shown with different colours and peak heights in Figure 2. Blue indicates a low BC content, and red

indicates a high BC content. The maps show that the BC content is higher in the urban areas, especially in several urban parks, than it is in the rural areas. The ratio of BC/OC is also higher in urban areas than in rural areas.

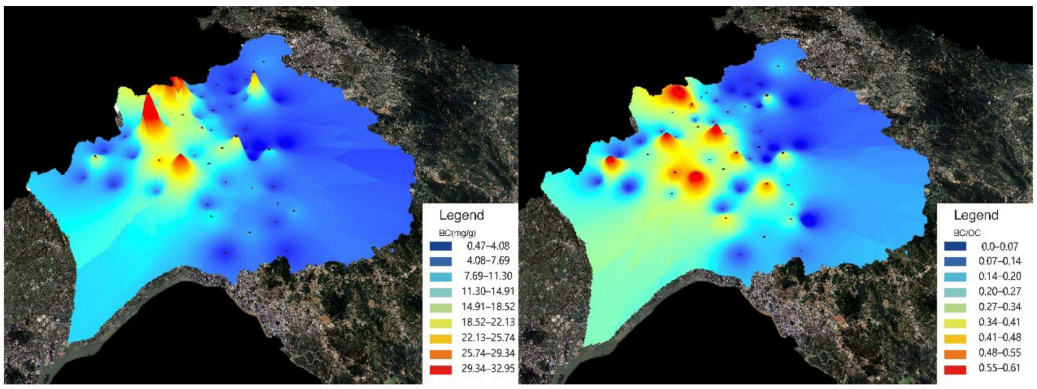

Figure 2. Spatial distribution maps of black carbon (BC) content and black carbon/organic carbon (BC/OC) ratio in soils of Guangzhou, south China.

Using hierarchical cluster analysis of the 55 soil samples, we generated a dendrogram (Figure 3). The soil samples were divided into two groups; group 1 contained all rural forest and urban forest sites with a low interference intensity, while group 2 contained urban parks with a high interference intensity. This result was consistent with our previous artificial grouping of sites.

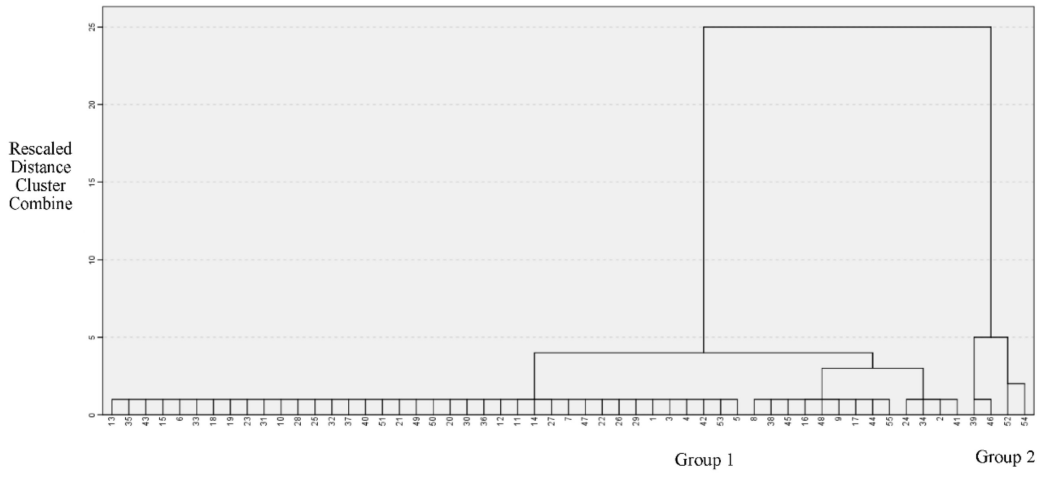

Figure 3. Dendrogram derived from the hierarchical cluster analysis of black carbon (BC) content from 55 soil samples.

3.2. Effects of Urbanization Intensity of Environmental Gradient on the Content of BC in Surface Soils

The content of BC in soils ranged from 0.47 to 33.12 g kg^{-1}, with a mean value of 7.36 ± 0.90 g kg^{-1} (Figure 4a). The content of BC in the soils varied with the interference intensity: urban park and urban woodland soils with a high interference intensity had much higher amounts of BC (9.74 ± 1.18 g kg^{-1}) than rural forest soils (2.94 ± 0.89 g kg^{-1}), which had a very low interference intensity (Figure 4a, $F = 2.98$, $p = 0.04$). The BC/OC ratio of the soils ranged from 0.03 to 0.62, with a mean value of 0.23 ± 0.02 (Figure 4b). The BC/OC ratio followed a similar trend as the BC content and was higher in the urban park and urban woodland soils than in the rural forest soils (Figure 4b, $F = 2.93$, $p = 0.04$).

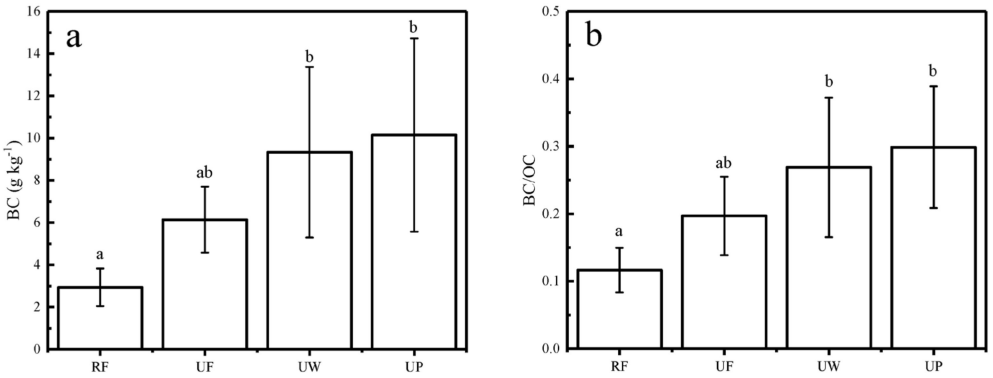

Figure 4. (**a**) Black carbon (BC) content in soils and (**b**) black carbon/organic carbon (BC/OC) ratio in soils. RF = rural forest, UF = urban forest, UW = urban woodland, and UP = urban park. Different letters indicate significant differences at different interference intensities at the level of $\alpha = 0.05$.

The EF of the BC in our study ranged from 1.0 to 70.9, with a mean value of 15.7 ± 1.9 (Figure 5). The EF of BC in the soils also varied with the interference intensity. The urban park and urban woodland soils, which had a high interference intensity, had much higher EF values than the rural forest soils, which had a very low interference intensity (Figure 5, $F = 2.98$, $p = 0.04$).

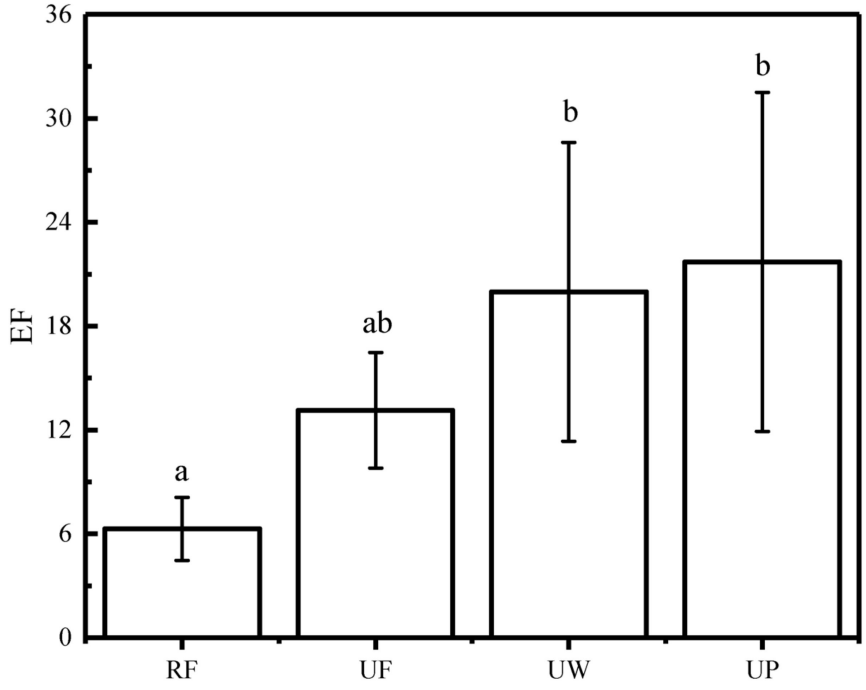

Figure 5. Enrichment factor (EF) of black carbon (BC) in soils. EF values are calculated from the BC content in soils divided by the background value of soils. RF = rural forest, UF = urban forest, UW = urban woodland, and UP = urban park. Different letters indicate significant differences among different interference intensities at the level of $\alpha = 0.05$.

3.3. Effects of Urbanization Intensity of the Urban Built Environment on the Content of BC in Surface Soils

The MGWR results (R^2, Table 2) showed that the urban built environment at a distance of 1000 or 1500 m from the sampling plots was more closely associated with the content of the BC in the sampling plot than the environment within 500 m. PPG, RD, and LUD were most closely associated with the BC content of the sampling plots at a distance of 1000 m from the plot, while PD, PI, and PIS were most closely associated with the BC content at a

distance of 1500 m (Figures S1–S5 and 6). This result indicated that the effects of the urban built environment on the distribution of BC might need to be evaluated at an appropriate scale. Except for PIS, all the factors were positively correlated with the amount of BC in the samples, which indicated that anthropogenic activities had a great influence on the BC content. The influence intensity of RD and PD decreased from west to east, while the influence of the other factors decreased from south to north (Figure 6). The western region is a central urban area with a large population and dense roads, indicating that human activities have a great influence on the BC content. Of the various factors, only RD had a significant effect on the BC content. Because there are more cars driving in the areas with a high RD, vehicle emissions are a vital source for BC accumulation.

Table 2. The statistical description of multi-scale geographically weighted regression (MGWR) coefficient. Bold numbers represent significant differences.

Scales	Variable	Mean	STD	Min	Median	Max	*p*
500 m R^2 = 0.19	LUD	0.015	0.047	−0.050	0.005	0.101	0.608
	PI	−0.052	0.035	−0.098	−0.058	0.014	0.701
	PPG	0.106	0.063	−0.006	0.103	0.204	0.829
	PD	0.097	0.014	0.061	0.098	0.122	0.356
	PIS	−0.101	0.079	−0.237	−0.103	0.024	0.681
	RD	0.367	0.121	0.111	0.416	0.494	**0.009**
1000 m R^2 = 0.28	LUD	0.166	0.149	−0.075	0.166	0.590	0.394
	PI	−0.024	0.021	−0.077	−0.022	0.003	0.777
	PPG	0.16	0.023	−0.121	0.158	0.213	0.686
	PD	0.18	0.016	0.145	0.182	0.212	0.186
	PIS	−0.086	0.025	−0.153	−0.083	−0.044	0.669
	RD	0.376	0.118	0.135	0.430	0.501	**0.032**
1500 m R^2 = 0.24	LUD	0.086	0.050	0.013	0.078	0.186	0.588
	PI	0.158	0.061	0.068	0.140	0.318	0.579
	PPG	−0.110	0.015	−0.138	−0.111	−0.071	0.888
	PD	0.397	0.024	0.339	0.401	0.431	0.064
	PIS	−0.272	0.024	−0. 327	−0.266	−0.233	0.313
	RD	0.240	0.090	0.061	0.293	0.331	0.259

LUD: land-use diversity, PI: the proportion of industrial land, PPG: the proportion of park green land, PD: population density, PIS: the proportion of impervious surface, and RD: road density.

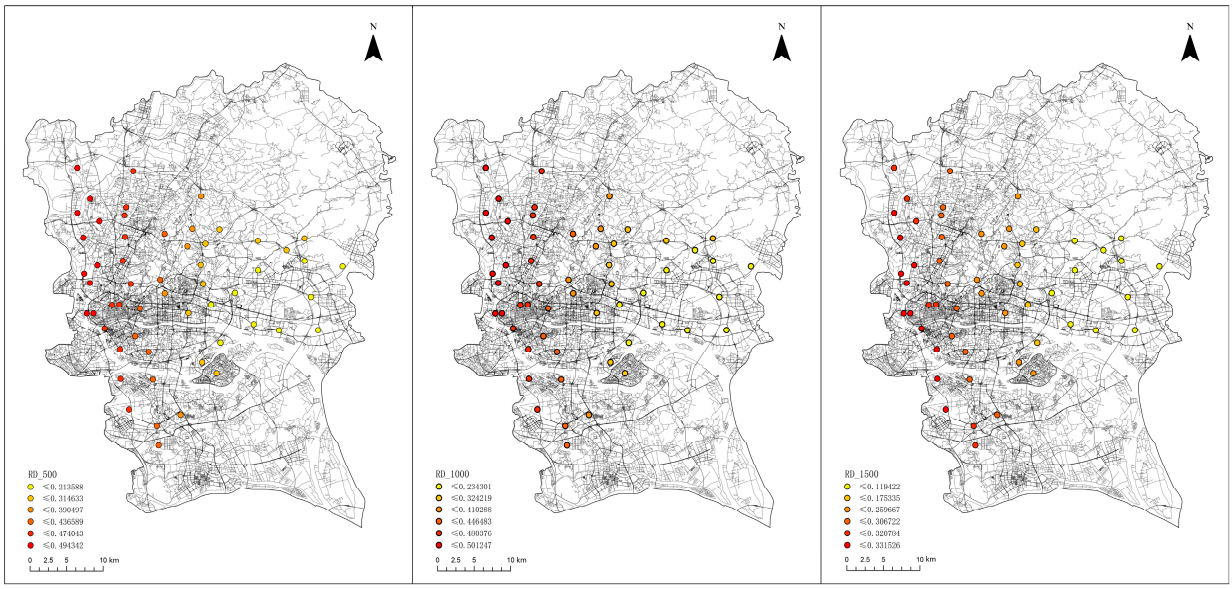

Figure 6. Spatial patterns of coefficients in the multi-scale geographically weighted regression analysis for road density (RD).

4. Discussion

4.1. Impact of Urbanization as Reflected by BC

The BC content in the soil from Guangzhou was highest in the urban forest soils and lowest in the rural forest soils; in accordance with previous studies, the BC content was higher in urban areas than suburban areas [21,29]. This result demonstrated that human activities (including anthropogenic wastes, management action, and human disturbance intensity) contributed to BC accumulation in Guangzhou city. The average content of the soil BC from Guangzhou was comparable with that from Beijing [21], Nanjing [13], Shanghai [18], and Xi'an [12], but much lower than that from the urban soils of Xuzhou [20] and Anshan [29]. The high content of the soil BC in urban areas dominated by the steel industry suggests that human industrial activity promotes the accumulation of BC. Many previous studies confirmed that the BC content in urban soils is correlated with urbanization [17,29]. For example, BC in older layers of soil resulted from historical coal use, while BC in the surface layer of soil was mainly from traffic emissions, especially emissions from diesel vehicles [13].

The BC content was significantly different in the various urban functional areas, which indicated that the type of urban landscape had a significant impact on BC accumulation [34,43]. The spatial distribution maps of the BC content and the BC/OC ratio in the soils showed that several urban parks are BC enrichment points. This indicated that urban parks have a lot of human activities, a high management intensity, and heavy traffic, resulting in the accumulation of BC. In the present study, road density also had a significant impact on the spatial distribution of BC. High road density reflects that there would be more cars driving on the road. Therefore, this indicated that vehicle emissions were an important source of BC accumulation. The previous studies also found that local traffic contributed to BC accumulation in compact urban areas [6,44,45].

According to the results of Sections 3.2 and 3.3, the content of BC in the various urban functional areas was affected by both the urbanization intensity of the environmental gradient and the urbanization intensity of the urban built environment, especially the anthropogenic wastes, management action, and vehicle emissions, which was in line with our ecosystem classifications and expected results. In addition, areas with a high population density inevitably led to more urban waste discharge and may have caused an increase in BC accumulation [7]. However, in the present study, the population density only had a slight effect on BC accumulation. A high population density does not necessarily produce BC substances, but low values of BC in highly populated areas may reflect an insufficient number of samples rather than a true relationship. Previous research that used a geographically weighted regression model to predict the SOC content reported that the sampling density had a great impact on the accuracy of the model and that an appropriate sampling density was 0.041 or 0.082 sites/km^2 [46]. Therefore, in future research, an appropriate sampling density should be used to increase the accuracy of the model and to clarify how the accumulation of BC reflects urbanization.

4.2. Correlation between Different BC Indexes and Its Sources

The EF was commonly used to evaluate the degree of human influence on the BC amounts. An EF value greater than 1 indicates that the BC content is higher than the background value, while an EF value closer to 1 indicates that the BC mainly comes from the soil. The EF value was higher at the sample sites with high levels of human activity in Guangzhou, a finding that was consistent with previous studies [20,29]. The EF values, once again, demonstrated that human activity promotes BC accumulation. In conclusion, BC accumulates to some degree in most urban soils in China; thus, the urban surface soil is an essential source of the sink of BC through dry and/or wet deposition [19].

The BC/OC ratio is correlated with the sources of BC accumulation. If the BC/OC value in the soil is around 0.1, BC is mainly derived from the combustion of biomass; while if this value is around 0.5, BC mainly comes from fossil fuels [18,21,44]. The BC/OC ratio was higher in the urban soil (0.30 ± 0.09) than in the rural soils (0.12 ± 0.03) in Guangzhou.

This is mainly because urban soils often receive large amounts of atmospheric deposition and pollutants. Therefore, fossil fuel combustion makes a significant contribution to BC accumulation in urban soils, whereas biomass combustion is the main source of BC in rural forest soils. This result was consistent with previous studies reporting sources of BC in Nanjing and Shanghai, China [13,18]. The previous studies also confirmed that the spatial heterogeneity of the BC/OC ratio was mainly linked to human population growth and an increase in the number of vehicles [19,20]. BC is considered quite refractory and has a long turnover time in soil [22,47]. Therefore, the BC/OC ratio may reflect different human activities and pollution sources in the urban zones than in the rural ones [13].

5. Conclusions

A spatial distribution map of BC and BC/OC revealed hotspots of BC accumulation in Guangzhou, especially in several urban parks with a management action. The amount of BC, the BC/OC ratio, and the EF of BC in the soils were higher in the urban areas, which had a high interference intensity than that in the soils in rural areas, which had a very low interference intensity. Road density had a significant effect on the BC content. Because there are more cars driving in the areas with a high road density, vehicle emissions were the dominant source of BC. Therefore, human activities (anthropogenic wastes, management action, and vehicle emissions) significantly contribute to BC accumulation. In addition, sampling density also strongly affected the accuracy of the geographically weighted regression model. Therefore, in future research, in order to evaluate the effect of urbanization on BC more accurately, more soil samples should be collected and measured to increase the accuracy of the model. Similarly, an appropriate scale is also important for us to evaluate the effects of urbanization intensity on the distribution of BC.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/f13030406/s1, Figure S1: Spatial patterns of coefficients in the multi-scale geographically weighted regression analysis for the proportion of green space in parks (PPG), Figure S2: Spatial patterns of coefficients in the multi-scale geographically weighted regression analysis for population density (PD), Figure S3: Spatial patterns of coefficients in the multi-scale geographically weighted regression analysis for land-use diversity (LUD), Figure S4: Spatial patterns of coefficients in the multi-scale geographically weighted regression analysis for the proportion of industrial land (PI), Figure S5: Spatial patterns of coefficients in the multi-scale geographically weighted regression analysis for the proportion of impervious surfaces (PIS).

Author Contributions: Conceptualization, data collection, data analysis and writing—original draft were performed by M.W. Material preparation was performed by X.L., H.Z., W.W. and M.L. Supervision and writing—review & editing were performed by G.X. All authors read and agreed to the published version of the manuscript.

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