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Demonstrating urban pollution using toxic metals of road dust and roadside soil in Chengdu, southwestern China

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Abstract As one of the largest economical hubs in southwestern China, Chengdu is witnessing fast urbanization characterized by rapid urban sprawl, population growth, infrastructural construction, and motorization. However, this rapid urbanization may lead to environmental degradation, placing human health at risk. In this study, toxic metals in road dust and roadside soil are used as proxies to illustrate environmental changes of Chengdu. In August 2009, 133 dust and 132 soil samples were collected from the first, second and third ring roads, along which areas have urbanized for different times. By means of a portable X-ray fluorescence analyzer, concentrations of Pb, Zn and Cu in the samples were determined. The results indicate that the concentrations and contamination levels of Pb, Zn and Cu in dust declined significantly from the first to the third ring roads, paralleling the decreasing trends in traffic and building densities from the first to the third ring roads. However, concentrations of the three elements in roadside soil were relatively stable among the roads. These data may suggest that the metals in road dust can be used as proxies to demonstrate environmental degradation during the urbanization of Chengdu, while concentrations of the metals in roadside soil are affected more by natural factors (e.g., background concentrations, precipitation, and distance to road) than by anthropogenic factors (e.g., traffic and building densities). Furthermore, compared to Pb concentrations measured in the 1990s, Pb concentrations in road dust have been reduced most likely owing to the exclusion of leaded petrol since 2000. Similar situations may be found in many other cities that are experiencing fast urbanization.

Keywords Urban pollution · Toxic metal · Road dust · Roadside soil · Urbanization · Traffic

1 Introduction

Currently half of the Chinese population lives in cities, a proportion that is projected to rise to 60.3 % by 2030 and 72.9 % by 2050 [United Nations Economic and Social Affair (UNESA) 2006]. Thus, urban environments are becoming increasingly important with regard to human health and wellbeing. Although urbanization can offer opportunities for improvements in human health (e.g., better access to health care), it may also result in environmental degradation (Gong et al. 2012), which dominates most of China's large cities nowadays (Chen 2007; Chan and Yao 2008; Gong et al. 2012; Zhao et al. 2012).

Roads play a major role in stimulating social and economic processes (Bai et al. 2009). Concentrations of toxic metals in road dust and roadside soil can be used as useful indicators of urban environmental pollution (Manta et al. 2002) and have been studied in many cities (Fergusson and Ryan 1984; Li et al. 2001; Charlesworth et al. 2003; Al-Khashman 2007; Wei et al. 2010; Apeagyei et al. 2011).

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Previous studies suggest that toxic metal concentrations in road dust and roadside soil are affected by a variety of factors (Bai et al. 2009), including meteorological condition (Othman et al. 1997; Sezgin et al. 2003), traffic density (Nabuloa et al. 2006), vehicle type (Sezgin et al. 2003; Nabuloa et al. 2006), and soil parameters (Viard et al. 2004). In addition, traffic has been identified as the major source of some metals in road dust and an important source of some metals in roadside soil (de Miguel et al. 1997; Al-Khashman 2007; Apeagyei et al. 2011; Luo et al. 2012).

Of the toxic metals indicating environmental pollution, lead (Pb), zinc (Zn) and copper (Cu) pollution in urban dust and soil are usually related to traffic (Apeagyei et al. 2011; Luo et al. 2012). Pb can originate from leaded petrol (de Miguel et al. 1997). Zinc compounds are extensively used in tyre tread (Stigliani and Anderberg 1991) and as antioxidants and detergent/dispersant improvers in lubricating oils (Drew 1975). Oxidation of lubricating oils upon exposure to air at high temperatures could form organic acids and other compounds that are corrosive to metals, releasing Zn and Cu to roads (Luo et al. 2012). The metal pollutants emitted from vehicles can accumulate in dust and soil (Luo et al. 2012). Although the concentrations of Pb, Zn and Cu in dust and soil have been investigated in many cities, none has ever used the metals as indicators to demonstrate environmental changes during urbanization using the space-for-time substitution method.

Using the spatial gradients of Pb, Zn and Cu concentrations in dust and soil, we demonstrate urban environmental degradation during the urbanization of the central city of Chengdu Metropolitan, which is an economic, transportation and educational hub in southwestern China (Fig. 1). In response to the fast motorization and urban expansion, the first, second, third and fourth ring roads have been constructed, and they were opened to public in 1988, 1992, 2002 and 2001, respectively (Zheng 2003; Zhang et al. 2009). The first, second and third ring roads are within the central city and there is no industrial activity within the third ring road. The fourth ring road was a toll highway before 2008, and then it became free for vehicles registered in Chengdu Metropolitan. Therefore, we only measured the concentrations of Pb, Zn and Cu in dust and soil along the first, second and third ring roads in this study. The areas along the first and second ring roads have been urbanized for longer periods, while that along the third ring road have been urbanized for shorter periods. This allows us to understand the relationship between urbanization and metal pollution using the space-for-time substitution method.

The main objective of this study is to understand the correlations of toxic metals in road dust and roadside soil with urban settings (e.g., traffic and building densities). The hypothesis is that Pb, Zn and Cu concentrations in dust and soil decline significantly from the first to the third ring roads.

2 Materials and methods

2.1 Study area

Located in southwestern China, Chengdu is the capital of Sichuan Province (Fig. 1). Of 280 cities in China, in 2007 Chengdu was listed among the top ten cities, which are most favorable for investment. In the recent 20 years, the central city of Chengdu has been experiencing rapid

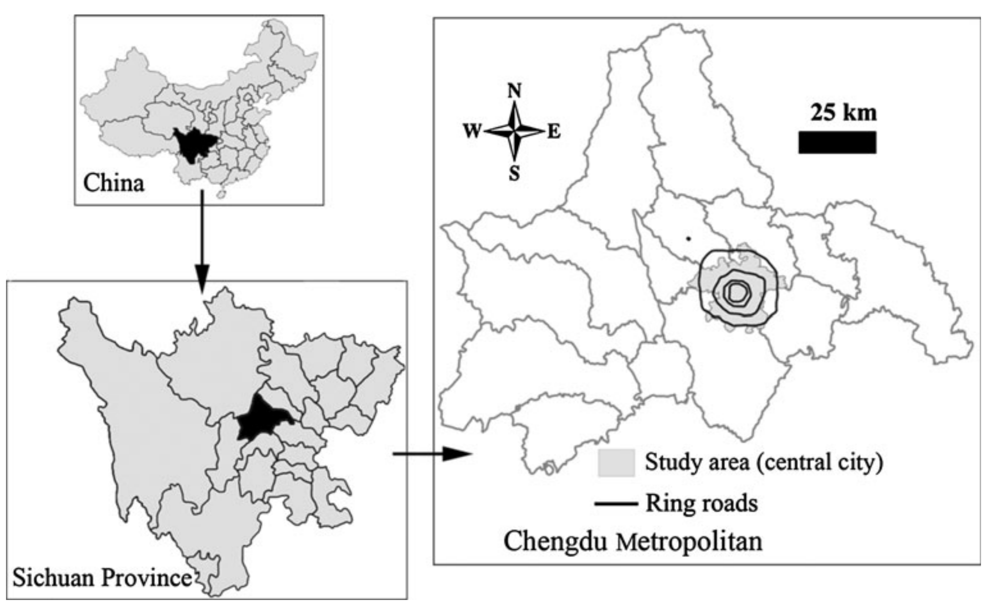

Fig. 1 Locations of Chengdu Metropolitan, study area (the central city of Chengdu Metropolitan) and the ring roads

economic growth and urbanization characterized by urban sprawl, population growth, and motorization (Fig. 2). Built-up area of the central city increased from 129 km^2 in 1995 to 456 km^2 in 2010, GDP of the central city increased from 65 billion Yuan in 2002 to 229 billion Yuan in 2010, and car number of Chengdu Metropolitan increased from 1.48 million in 2005 to 2.23 million in 2012.

2.2 Sample collection and analysis

Dust and soil samples, weighing 500–1,000 g, were collected from 45, 41 and 47 sites along the first, second and third ring roads, respectively, on two sunny days of the wet season (19–20 August 2009) (Fig. 3a); each site was sampled once. Soil samples were collected using wooden spoons from 0–5 cm topsoils in the green belts, while dust samples were collected using plastic brushes from the road surfaces (Fig. 3b). Clean plastic bags were used to preserve the samples.

Prior to metal measurement, the samples were air-dried, grounded by an agate mortar and then sieved through a 2-mm nylon sieve. Concentrations of Pb, Zn and Cu were determined using a portable X-ray fluorescence (XRF) analyzer (Alpha Series 6500, Innov-X systems, Inc, USA). In order to assess the accuracy of the XRF analyzer, reference samples (Montana I Soil, Montana II Soil and San Joaquin Soil from the National Institute of Standards and Technology of US) were measured. The recoveries of Pb, Zn and Cu in the reference samples were between 86 and 95 %.

2.3 Assessment of metal contamination

To assess the contamination level of a given toxic metal in each sample, the contamination index (P_i) was calculated using the following equations (Huang 1987; Bai et al. 2009):

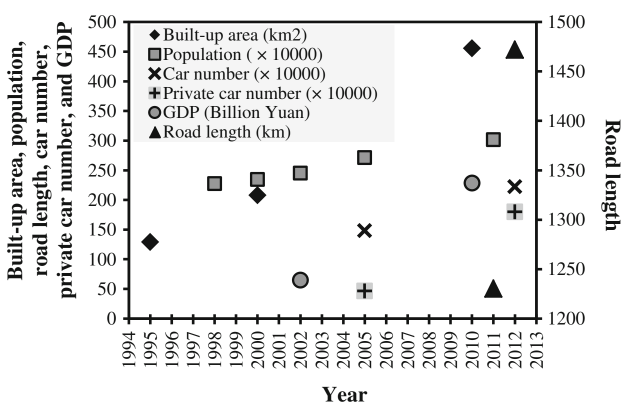

Fig. 2 Increases in the built-up area[a], population[b], road length[b], car number[a], private car number[a] and GDP[b] in Chengdu Metropolitan and the central city. ([a] data of Chengdu Metropolitan; [b] data of the central city of Chengdu Metropolitan)

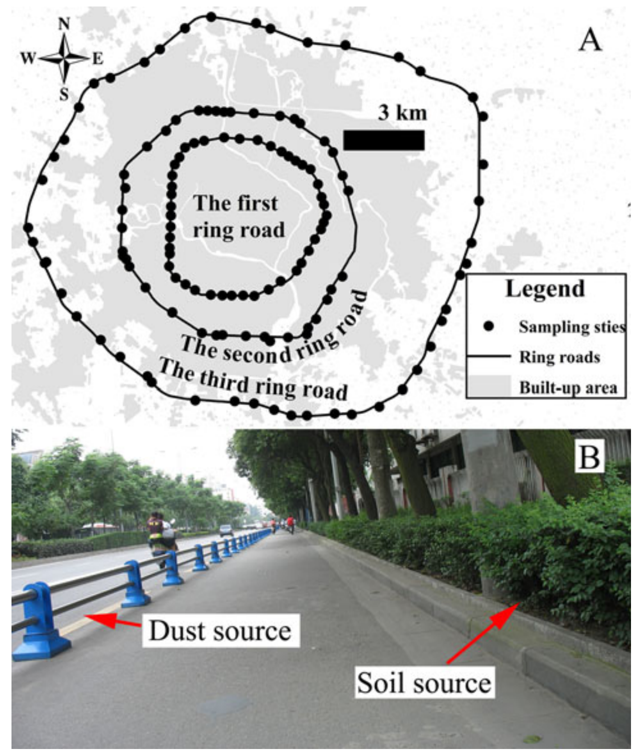

Fig. 3 Locations of the sampling sites (**a**) and sample sources (**b**)

$$P_i = C_i X_a \quad (C_i \le X_a)$$
$$P_i = 1 + (C_i - X_a)/(X_b - X_a) \quad (X_a \le C_i \le X_b)$$
$$P_i = 2 + (C_i - X_b)/(X_c - X_b) \quad (X_b \le C_i \le X_c)$$
$$P_i = 3 + (C_i - X_c)/(X_c - X_b) \quad (C_i > X_c)$$

where C_i is the concentration of a given metal; X_a is the no-polluted threshold value; X_b is the lowly polluted threshold value and X_c is the highly polluted threshold value. Following (Bai et al. 2009; Bai et al. 2011), the X_a, X_b and X_c values are derived from the Chinese Environmental Quality Standard for Soils (GB 15618-1995) [State Environmental Protection Administration of China (SEPAC) 1995] (Table 1).

P_i is described using the following terminologies: $P_i \le 1$ for no contamination; $1 < P_i \le 2$ for low contamination; $2 < P_i \le 3$ for moderate contamination; $P_i > 3$ for high contamination.

2.4 Statistical analyses

Varimax rotated principal component analysis (PCA) was applied to investigate the sources of Pb, Zn and Cu following Shi et al. (2008). The PCA analysis is based on an eigenvalue greater than 1 at the 95 % significance level. Since all the datasets were normally distributed, one-way ANOVA was applied to compare metal concentrations among the ring roads. The statistical analyses were performed by means of SPSS 19.0.

Table 1 The threshold values (X_a, X_b and X_c) for Pb, Zn and Cu by the Chinese Environmental Quality Standard for soils (GB 15618-1995)

	Metal concentrations (mg/kg)		
	Pb	Zn	Cu
X_a	35	100	35
X_b	250	200	50
X_c	500	500	400

3 Results

3.1 Metal concentrations in road dust and roadside soil

The descriptive statistics of metal concentrations in the samples are summarized in Table 2. Mean concentrations of Pb, Zn and Cu in total dust samples were 77 ± 33, 343 ± 149 and 80 ± 30 mg/kg, respectively. Mean concentrations of Pb, Zn and Cu in total soil samples were 55 ± 30, 206 ± 127 and 59 ± 19 mg/kg, respectively. Mean concentrations of each metal in total dust samples greatly exceeded the background values of soil in Chengdu (Shi 1995) and China (China National Environmental Monitoring Center (CNEMC) 1990) (Table 2). Concentrations of Pb, Zn and Cu in total soil samples were at least 2 times greater than their corresponding background values. In general, both dust and soil samples were polluted by Pb, Zn and Cu to certain extents, and dust samples were more polluted by the three elements compared to soil samples (Fig. 4).

3.2 Comparison of metal concentrations among the ring roads

Pb, Zn and Cu concentrations of dust varied remarkably among the three roads (Fig. 4 and Table 2). Along the first,

Table 2 Pb, Zn and Cu concentrations in the soil and dust samples collected from the first, second and third ring roads (mg/kg)

Sites	Pb		Zn		Cu	
	Dust	Soil	Dust	Soil	Dust	Soil
All samples						
N	132	133	132	133	132	133
Range	20–178	17–182	110–846	70–836	30–191	26–120
Median	73	44	315	170	78	53
Mean	77	55	343	206	80	59
SD	33	30	149	127	30	19
The first ring road						
N	45	45	45	45	45	45
Range	41–178	17–123	150–846	85–836	67–154	36–120
Median	92	44	446	170	98	56
Mean	94	57	450	223	99	62
SD	30	29	141	147	19	20
The second ring road						
N	40	41	40	41	40	41
Range	26–168	22–182	160–654	79–636	36–191	26–119
Median	82	42	344	140	78	54
Mean	88	51	365	172	87	58
SD	33	30	125	98	33	19
The third ring road						
N	47	47	47	47	47	47
Range	20–102	20–160	110–417	70–712	30–133	32–102
Median	50	55	218	197	55	52
Mean	53	57	223	219	56	55
SD	19	30	69	126	18	18
Background values of Chengdu[a]		23.1		79.7		28.4
Background values of China[b]		23.5		68.0		20.7

SD standard deviation, *N* number of samples

[a] Shi (1995)

[b] China National Environmental Monitoring Center (CNEMC) (1990)

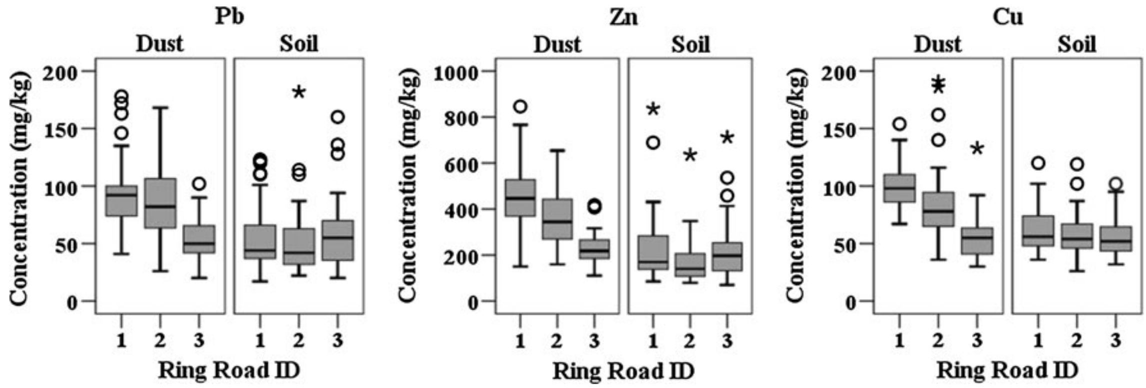

Fig. 4 Concentrations of Pb, Zn and Cu in the samples collected along the first, second and third ring roads (Ring Road ID: *1* the first ring road, *2* the second ring road and *3* the third ring road)

second and third ring roads, Pb concentrations of dust were 94 ± 30, 88 ± 33 and 53 ± 19 mg/kg, respectively; Zn concentrations of dust were 450 ± 141, 365 ± 125 and 223 ± 69 mg/kg, respectively; and Cu concentrations of dust were 99 ± 19, 87 ± 33, 56 ± 18 mg/kg, respectively (Table 2). These values indicate that Pb, Zn and Cu concentrations in dust decreased significantly from the first to the third ring roads ($P < 0.01$; Fig. 4; Table 3), paralleling the time since urbanization.

In contrast, concentrations of Pb, Zn and Cu in soil were relatively stable among the three roads ($P > 0.05$; Fig. 4; Table 3). Along the first, second and third ring roads, Pb concentrations of soil were 57 ± 29, 51 ± 30 and 57 ± 30 mg/kg, respectively; Zn concentrations of soil were 223 ± 147, 172 ± 98 and 219 ± 126 mg/kg, respectively; and Cu concentrations of soil were 62 ± 20, 58 ± 19 and 55 ± 18 mg/kg, respectively (Table 2). Concentrations of Pb, Zn and Cu in soil are independent of time since urbanization.

3.3 Assessment of metal contamination

Contamination levels of dust were highest along the first ring road and lowest along the third ring road (Fig. 5). Most dust samples from the first and second ring roads were moderately polluted by Cu; however, only ~55 % of dust samples from the third ring road had moderate Cu contamination and others had low or no Cu contamination. All the dust samples were lowly or not polluted by Pb, but the third ring road had a higher proportion of dust samples that were with no Pb contamination. Most dust samples from the first and second ring roads were highly or moderately polluted by Zn, while ~40 % of dust samples from

Table 3 Comparison of metal concentrations among the first, second and third ring roads

Elements	Sample sources	The significances of difference
Pb	Dust	0.000*
	Soil	0.539
Zn	Dust	0.000*
	Soil	0.126
Cu	Dust	0.000*
	Soil	0.623

Statistical analysis method: one-way ANOVA

* Difference is significant at the 0.01 level (2-tailed)

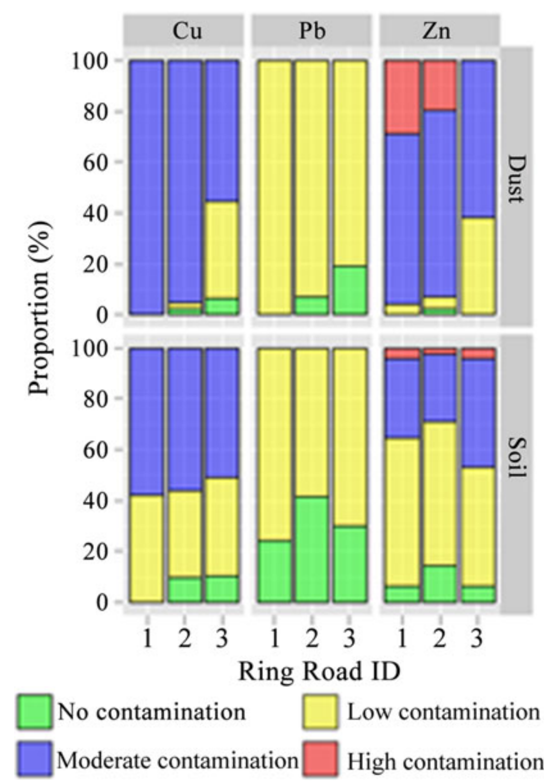

Fig. 5 Proportions of contamination levels of Pb, Zn and Cu in the samples collected along the first, second and third ring roads (Ring Road ID: *1* the first ring road, *2* the second ring road and *3* the third ring road)

the third ring road were lowly polluted by Zn. These suggest that the areas urbanized longer had higher contamination levels of Pb, Zn and Cu in road dust.

Generally, dust and soil samples were more polluted by Zn and Cu than by Pb. Most dust samples were lowly polluted by Pb, but highly or moderately polluted by Cu and Zn. Soil samples were lowly or not contaminated by Pb, but at least 50 % of soil samples were moderately polluted by Cu and at least 30 % of soil samples were highly or moderately polluted by Zn.

3.4 Source identification based on PCA analysis

Pb, Zn and Cu in dust commonly originate from traffic-related materials (e.g., petrol, brake dust, tire tread, yellow paint and exhaust) (Li et al. 2001; Shi et al. 2008), and the dust released from vehicles are regarded as an important anthropogenic source of Pb, Zn and Cu in roadside soil (Cyrys et al. 2003; Gray et al. 2003; Shi et al. 2008). Based on the results of PCA analysis, the metals could be represented by one principal component for dust and soil datasets each, explaining 100 % of the total variance (Table 4). This indicates that Pb, Zn and Cu in dust mainly originated from the same sources, and the main sources of Pb, Zn and Cu in soil were similar. Traffic may be the major source of Pb, Zn and Cu in the dust samples, and an important anthropogenic source of the metals in the soil samples.

4 Discussion

4.1 Correlation of metal contamination with urban settings

The declining trends in Pb, Zn and Cu concentrations of dust from the first to the third ring roads could be explained by the different densities of traffic and buildings along the roads. Because the metal concentrations of soil were insignificantly different among the roads, other anthropogenic and natural factors are explored to explain this similarity.

4.1.1 *Correlation of metal contamination with traffic density*

Although traffic density data are unavailable for the three ring roads, traffic condition maps could illustrate the different traffic densities along the roads to a large extent. In this paper, we only present the traffic conditions of the sampling sites at 9:00 am and 6:00 pm on average Mondays, because the daily and weekly data are unavailable and the rush hours are approximately 8:00–10:00 am and 5:00–7:00 pm on each weekday. As we expected, traffic is more concentrated on the first ring road and less concentrated on the third ring road (Fig. 6), reflecting that ring road with higher traffic density had higher concentrations of Pb, Zn and Cu in dust.

However, concentrations of the metals in soil were relatively stable among the three roads, different from the declining trends in traffic density from the first to third ring roads. This similarity might be associated with the following reasons. First, the sampling sites of soil were covered by plants, which could partly prevent the three elements from depositing on the soil and uptake metals in soil (Princewill-Ogbonna and Ogbonna 2011). Second, since the metal concentrations were lower in soil than in dust (Table 2), bulk soil can buffer metal concentrations of dust. Third, the soil samples were collected on the curbs, which are further separated from the main traffic lanes by a pedestrian or cycling lane. Consequently, the three elements rarely reach curbside soil before they are washed or carried away to the drainage systems. Furthermore, the samples were collected during the wet season, when the

Table 4 Principal component analysis for Pb, Zn and Cu concentrations in the soil and dust samples collected from the first, second and third ring roads

Sample sources	Metals	Factor 1	Communalities
Dust	Pb	0.886	0.795
	Zn	0.932	0.869
	Cu	0.849	0.721
	Cumulative percent (%)	100	
Soil	Pb	0.880	0.774
	Zn	0.826	0.682
	Cu	0.862	0.744
	Cumulative percent (%)	100	

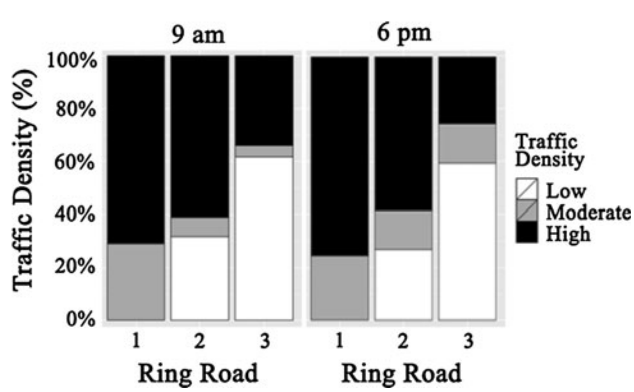

Fig. 6 Traffic densities at the sampling sites along the first, second and third ring roads on two rush hours (9 am and 6 pm) of average Mondays in Chengdu (Ring Road ID: *1* the first ring road, *2* the second ring road and *3* the third ring road)

pollutants could be easily washed away or leaked through the soil.

4.1.2 *Correlation of metal contamination with the "canyon" effect*

The "canyon" effect is that the influence of prevailing winds is not always evident within the environment of tall and dense buildings and, as a result, particles tend to fall-out within the urban roadway "canyon", accelerating toxic metals to accumulate in road dust and roadside soil (Ewen et al. 2009). As presented in Fig. 7, building density is relatively high along the first and second ring roads, while there are more open areas along the third ring road. Therefore, the "canyon" effect would be stronger along the first and second ring roads, leading to the metals from vehicles more easily settle on the first and second ring roads. Areas with higher building density had higher metal concentrations in road dust.

4.2 Urbanization and urban environmental degradation

The urban expansion of Chengdu is closely related to the ring roads (Schneider et al. 2003). Once a ring road was built, the densities of buildings and traffic would gradually grow along the road, increasing metal emissions from vehicles and enhancing the "canyon" effect. Compared to that along the third ring road, the areas along the first and second ring roads have urbanized longer and experience higher traffic density and stronger "canyon" effect. Probably due to these reasons, the concentrations and contamination levels of Pb, Zn and Cu of dust were higher along the first and second ring roads than along the third ring road (Figs. 4, 5). The spatial gradients of Pb, Zn and Cu in road dust reflect environmental degradation during the urbanization of Chengdu.

4.3 Temporal trends in metal contamination

Internal combustion engine accounted for 60 % of total anthropogenic Pb in environment in 1990 (Rayson 1990). To reduce Pb contamination in the environments, the Chinese government has banned the use of leaded petrol since 2000 [General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China (AQSIQ) 1999]. Consequently, the Pb concentrations of road dust in the central city of Chengdu have decreased from about 244 mg/kg (Shi 1995) in early 1990s to 53–94 mg/kg in 2009. Similar reduction in Pb has been observed in other cities, such as Manchester, UK (Anagnostopoulou and Day 2006), Thessaloniki, Greece (Anagnostopoulou and Day 2006; Ewen et al. 2009), Madrid, Spain and Oslo, Norway (de Miguel et al. 1997) as a result of reduced use of leaded petrol.

After the exclusion of leaded petrol, the influence of traffic on road dust can be characterized by Zn and Barium (Ba), and to a lesser extent by Cu and Pb (de Miguel et al. 1997). Probably due to fast motorization, average concentration of Zn in dust along the first ring road has increased from 340 mg/kg in early 1990s to 450 mg/kg in 2009. In addition, in 2009, both soil and dust were more polluted by Zn and Cu than by Pb (Fig. 5). Although the Pb pollution has been reduced, other toxic metals from traffic

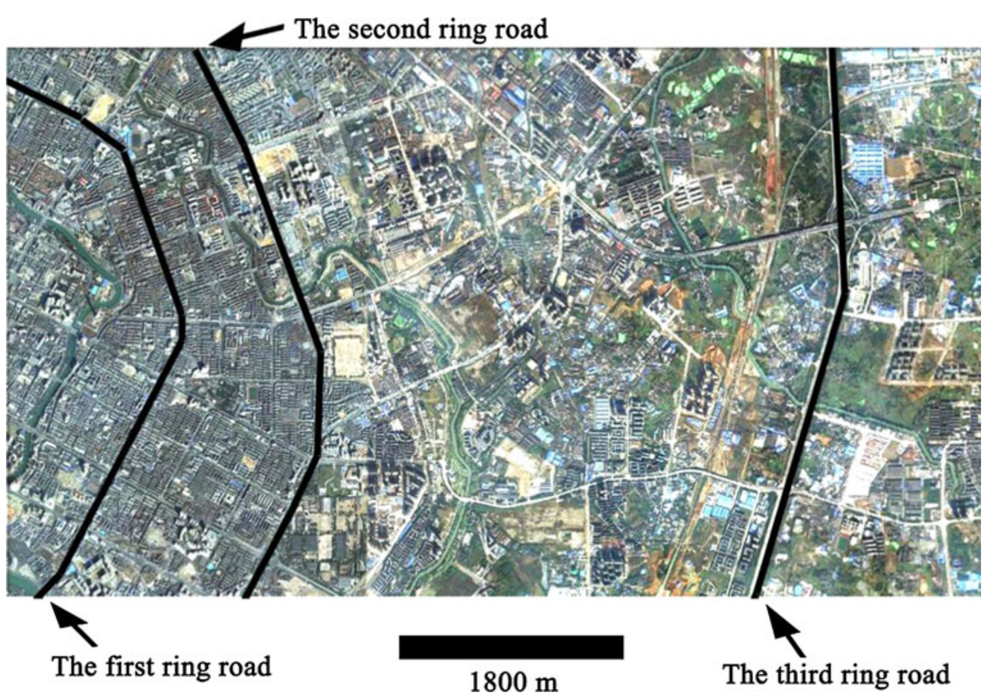

Fig. 7 Building density and open area along the first, second and third ring roads

can also place human health at risk as a result of fast motorization.

5 Conclusions

In this study, Pb, Zn and Cu in road dust and roadside soil are used as proxies to demonstrate environmental degradation during the urbanization of Chengdu. Since areas along the first, second and third ring roads have urbanized for different periods, the space-for-time substitution method was used. Dust and soil samples were collected from the first, second and third ring roads, and Pb, Zn and Cu in the samples were measured. The results suggest that:

(1) Traffic is most likely the major source of Pb, Zn and Cu in road dust, and the "canyon" effect can accelerate the metals released from traffic to accumulate in road dust. Consequently, traffic density, the strength of "canyon" effect, and the concentrations of Pb, Zn and Cu in dust decreased from the first to the third ring roads at the same time, reflecting that the areas urbanized for longer time had higher concentrations of Pb, Zn and Cu in road dust.

(2) Pb concentrations of road dust have been reduced owing to the exclusion of leaded petrol since 2000. However, almost all dust samples and over 50 % of soil samples were still lowly polluted by Pb, suggesting that Pb contamination would still be a problem even though leaded petrol has been banned for 9 years as traffic volume grows and due to the Pb accumulated in dust and soil over years.

(3) Dust and soil were more polluted by Cu and Zn than by Pb. Although controlling Pb pollution has been effective, policies should also be developed to reduce the pollution of other metals.

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