

Article

Contamination Identification of Trace Metals in Roadway Dust of a Typical Mountainous County in the Three Gorges Reservoir Region, China, and its Relationships with Socio-Economic Factors

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Abstract: Trace metal contamination in urban road dust has attracted global concern due to its potential risk to the urban environment and human health. Compared to big cities, relative studies in counties and small towns have not been well quantified. This research identified the trace metal contamination characteristics and possible sources in the road dust of a typical mountainous county and a town in the Three Gorges Reservoir region, southwest China, and their associations with major regional socio-economic factors. The trace metal concentrations were determined, and the contamination levels were assessed. Concentrations of Zn, Pb, and Cu were relatively high in both locations, and a significant accumulation of them was confirmed by the geo-accumulation method. Multivariate analysis and geographic information system (GIS) mapping were combined to explore the sources of trace metals in the investigated area. Anthropogenic activities predominantly affected the contamination levels of Zn, Pb, Cu, and Co, and traffic emission, agricultural activities, and fossil fuel combustion were their main sources. The significant accumulation of Zn should attract special concern for its wide use in industrial and agricultural activities. Population and vehicle density were the main factors that controlled the trace metal contamination levels in the roadway dust. Rapid urbanizing promoted trace metal accumulation in counties and towns. Therefore, it is urgent to make appropriate strategies for trace metal pollution mitigation in the process of urbanization.

Keywords: trace metal; road dust; source identification; contamination evaluation; socio-economic factors

1. Introduction

Road dust is a valuable indicator of urban environment quality, which has attracted global concern in recent years due to its negative environmental impacts [1–8]. Trace metals in urban roadway dust were considered a priority for their high toxicity, long-distance transport, and bioaccumulation properties [8,9]. Trace metal pollution in urban areas has been widely investigated across the world [10–14]. Cadmium (Cd), lead (Pb), chromium (Cr), copper (Cu), zinc (Zn), nickel (Ni), and arsenic (As) were reported as ubiquitous pollutants in urban roadway dust [1–9,15–17]. Road dust might effortlessly enter the human body via ingestion, dermal contact, and inhalation because of small particle size and inherent mobility in windy weather, which poses a potentially high health risk to human beings [15–23]. According to Wei and Yang [24], approximately 65% of all Chinese cities presented a high trace metal pollution level in urban roadway dust, indicating significant effects

of anthropogenic activities. In the last two decades, investigations of trace metal accumulation in roadway dust have been extensively conducted in China [1–6,14–28]. Concentration, spatio-temporal distribution, environmental risk, and source of trace metals in roadway dust have been extensively studied [25–29]. However, these studies were mostly carried out in large cities [15], and limited information could be found about counties or small towns.

China has experienced rapid industrialization and urbanization in recent decades. By the end of 2018, there were 2851 counties and 39,945 towns in China, and the average growth rate of urban residence was 3.4 % in the last two decades (2001–2018) [30]. In this process, continuously increasing population density and anthropogenic activities will inevitably place great stress on the local environment. Except for traffic and industrial emission, the misuse of chemical fertilizers and pesticides might also be a potential source of trace metals in counties and small towns [31]. Furthermore, long-distance transport by atmospheric dust might be another source of some trace metals in roadway dust [32]. The mobility and various sources of road dust make it difficult for us to fully understand the mechanism of the accumulation characteristics and fates of trace metals in the urban environment, especially in counties and small towns. Meanwhile, the quality of public service and medical care in these areas is far behind those in big cities. Therefore, the prevention and control for trace metal contamination in counties and small towns are great challenges for the government of China.

Chongqing is the only municipality in southwestern China. The urbanization rate of Chongqing was 35.6% in 2000, and it became 65.5% by the end of 2018 [30]. During the rapid process of urbanization, intense anthropogenic activities, such as industrial, agricultural, commercial, and traffic activities, greatly affected the local environment [33]. Moderate pollution of trace metals was observed in the Chongqing surface environment [34–36]. The massive use of fossil fuels in industrial production and the rapidly increasing number of motor vehicles were suggested to be predominant sources of trace metals in Chongqing [37]. Furthermore, a higher health risk resulted from trace metals in road dust was observed in southeast China, Chongqing included [1,37]. Wushan is a typical mountainous county in the Three Gorges Reservoir Region of China, which is located in the northeast of Chongqing. Previous studies reported that the soils in a town of Wushan were polluted by cadmium, which might cause adverse risk to the local residents [38,39]. Nonetheless, the metal pollution status of road dust in this area is limited. This research characterized the spatial variation, assessed the accumulation degree, and determined potential sources of the target trace metals in road dust. Furthermore, the associations of trace metal accumulation levels and the major regional socio-economic factors were analyzed. The findings of this research will enhance our understanding of the trace metal accumulation in counties and towns during the rapid urbanization process.

2. Materials and Methods

2.1. Study Area and Sample Collection

The study area is situated in the northeast of Chongqing Municipality (105°11'~110°11' E, 28°10'~32°13' N), which is surrounded by two rivers and four mountains (Figure 1). Wushan county (109°33'~110°11' E, 30°45'~23°28' N) is known as the “East Gate of Chongqing” and is located in the central region of the Three Gorges Reservoir Region, which has several key enterprises, including thermal power plants, building-material chemical plants, cement plants, and cigarette factories. The sampling campaign was conducted from July to August in 2017. A total of 80 road dusts were sampled in Wushan county, and 10 samples were collected in Duping town. Detailed information was provided in the supplementary information (SI).

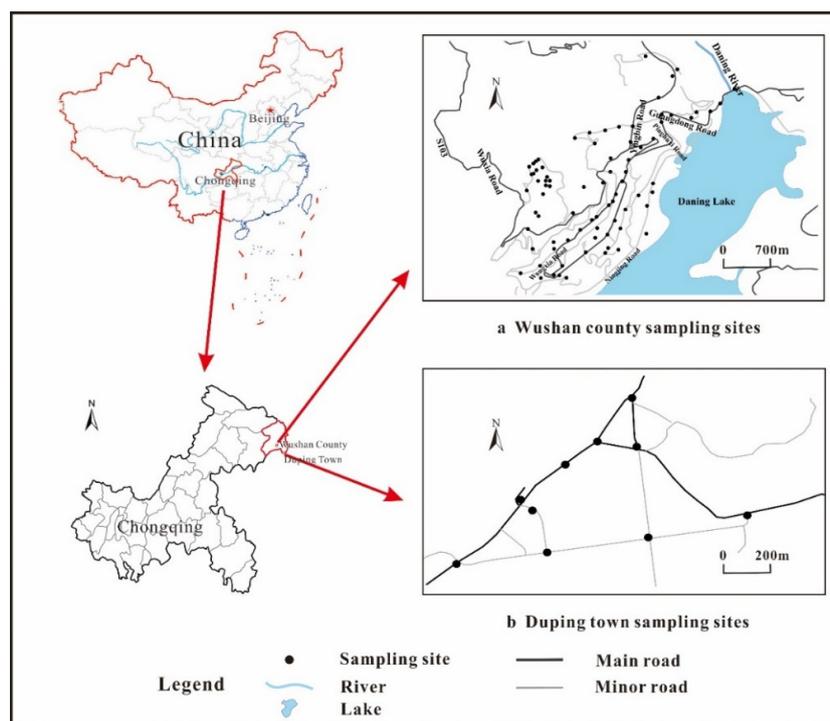


Figure 1. The study area and sampling sites in Wushan county and Duping town, Chongqing, China ($n = 90$).

2.2. Analytical Procedures and Quality Assurance

All samples were pretreated according to our previous method [5]. Briefly, the dust samples were put through the procedure of air-drying, slightly crushing, and screening with a 200-mesh sieve. Eight trace metals (Co, Cu, Zn, Cr, Ni, Mn, Pb, V) were detected with X-ray fluorescence spectrometry (XRF, Bruker S8, Germany). Method detection limits (MDLs) were 1.6, 1.2, 2.0, 3.0, 1.5, 10.0, 2.0, and 4.0 mg/kg for Co, Cu, Zn, Cr, Ni, Mn, Pb, and V, respectively. The milled road-dust sample (4.0 g) and boric acid (2.0 g) were weighed out accurately and placed in the mold, then pressed into a 32-mm diameter pellet under 30-ton pressure for metal determination. The method accuracy was evaluated by analyzing the reference materials (GSD12 and GSS1, Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences). Replicate analytical errors of the target elements were in the range of -8.6 % to 9.3 %. The average relative standard deviation (RSD) of two replicates of 18 dust samples was between 2.6% and 7.9%. More details were provided in the SI.

2.3. Contamination Evaluation

The geo-accumulation index (I_{geo}) was often applied to identify the influence of anthropogenic activities on the hypergene environment and estimate the accumulation levels of trace metals [16,17]. I_{geo} is computed as the equation (1):

$$I_{geo} = \log_2 \left[\frac{C_n}{1.5 \times B_n} \right] \quad (1)$$

where C_n is the content of trace metals in the road dust, B_n is the baseline value of the trace metals, and the soil background values in Chongqing [40] were used as B_n values in this study [5]. The seven pollution levels are classified in Table S1.

2.4. Statistics and Data Processing

Statistical analysis of trace metals in dust was conducted using SPSS 25.0. Multivariate statistical analysis combined with GIS mapping was introduced to conduct the source analysis of the trace metals by using JMP 13.0 and ArcGIS 10.2, respectively. Redundancy analysis (RDA) was carried out using Canoco 5 to quantify the impact that environmental variables might make on trace metal accumulation. Tukey-Kramer HSD test was conducted to examine the significant differences at a significance level of 0.05 [41].

3. Results and Discussion

3.1. Contents of Trace Metals in Roadway Dust

Statistical results of the analyzed trace metals in roadway dust for both locations are listed in Table 1. For samples in Wushan, the median concentrations were 57.3, 49.4, 33.7, 22.4, 427.6, 31.9, 62.1, and 98.0 mg/kg for Cr, Co, Cu, Ni, Mn, Pb, V, and Zn, respectively. For Duping, the median concentrations of the eight trace metals were 72.0, 24.8, 23.9, 28.0, 1020.0, 27.3, 84.1, and 121.6 mg/kg. Although trace metal contents in this study were lower than those in big cities in China, according to the results of Wei and Yang [24], several trace metals already presented a certain degree of accumulation in both locations. Contents of Zn and Co in the road dust in both locations were generally elevated in comparison with the soil background value. In addition, Co and Pb in Wushan and Mn in Duping were relatively higher.

Table 1. Descriptive statistics of trace metal concentrations in the road dust from Wushan and Duping (mg/kg).

Elements		Co	Cr	Cu	Mn	Ni	Pb	V	Zn
Wushan	Minimum	17.5	29.4	14.1	295.8	13.9	15.6	42.1	39.9
	Maximum	127.6	150.7	160.2	1278.1	81.5	118.5	120.1	461.4
	Mean	53.2	61.2	37.0	464.5	24.4	38.4	66.6	120.7
	Median	49.4	57.3	33.7	427.6	22.4	31.9	62.1	98.0
	SD	25.3	18.4	19.4	164.1	8.5	22.4	15.1	78.9
	CV (%)	47	30	52	35	35	58	23	65
	Kurtosis	−0.33	6.21	20.01	13.72	24.93	2.94	2.31	4.69
	Skewness	0.60	1.77	3.62	3.37	4.10	1.78	1.53	2.05
	Reference value [40]	17.6	79.0	31.1	657	32.6	30.9	96.0	86.5
Duping	Minimum	20.8	45.3	14.3	374.7	21.5	19.3	54.1	49.0
	Maximum	59.0	84.9	44.2	1194.4	35.7	129.5	101.6	369.5
	Mean	31.4	70.0	26.0	851.7	26.8	41.0	80.6	134.7
	Median	24.8	72.0	23.9	1020.0	28.0	27.3	84.1	121.6
	SD	14.8	12.6	9.5	348.1	4.4	36.0	16.4	92.3
	CV (%)	47	18	36	41	16	88	20	69
	Kurtosis	0.68	0.08	−0.15	−2.04	0.50	4.05	−1.28	5.08
	Skewness	1.49	−0.83	0.76	−0.42	0.62	2.14	−0.38	2.04
	Reference value [40]	17.6	79.0	31.1	657	32.6	30.9	96.0	86.5

SD: standard deviation; CV (%): coefficient of variance.

Furthermore, the results of the Tukey-Kramer HSD test suggested a significant difference for the contents of Mn, Co, and V in Wushan and Duping, and insignificant difference for other trace metals ($\alpha = 0.05$) (Figure S1). Cobalt concentration in Wushan was significantly higher than that in Duping, which might be due to the thermal plant in the east of Wushan. Contrarily, Mn and V in Wushan were significantly lower than those in Duping. Apparently, this result seemed to contradict with the common conclusion that the trace metal pollution level generally reduces from the urban to suburban [12]. But the recent research confirmed that recently introduced metals in automotive technology were higher in younger roads, which might be relative to the higher concentration of Mn in the street of Duping than that in Wushan. Vehicular speed, road age, and traffic density might be the influencing factors that led to the difference in Wushan and Duping [42]. In addition, this result might

also be due to the differences among the sample amounts or potential sources in the surrounding area, which should be further explored.

The result of this study was compared with the data reported for cities in China and other countries, Chongqing included (Table S2). The trace metal concentrations were generally at a moderate level. Specifically, the trace metal contents in Chongqing were generally lower than in the megacities of Beijing [43] and Shanghai [44], and they were comparable to the provincial capital cities, such as Xi'an [5], Lanzhou [45], and Wuhan [46]. These results reflected that the high concentrations of trace metals were closely related to the larger population [47]. Similarly, trace metal contents in Wushan and Duping were lower than those in the urban area of Chongqing [48], especially Cu and Pb. The result indicated that population density could have an obvious impact on the spatial distribution of trace metals.

As presented in Table 1, a wide range of concentrations for each metal was observed as shown by the high values of variation coefficients (CV%). Generally, a CV% \leq 20 % indicates low variability, a CV% range of 21–50% indicates moderate variability, and a CV% range of 51–100% suggests high variability [5]. For Wushan, Pb, Cu, and Zn presented high variability (CV% > 50%), while the other trace metals showed moderate variability (CV% > 21%). For Duping, only Pb and Zn showed high variability (88% and 69%). Cobalt, Cu, and Mn showed moderate variability, while the other trace metals showed low variability. Therefore, Pb and Zn in both locations presented spatial heterogeneity, which might indicate the input of anthropogenic sources as observed in metropolises [49].

3.2. Trace Metal Pollution Levels in Roadway Dust

To quantitatively assess the trace metal accumulation level, the I_{geo} values were calculated for each metal (Figure 2). The mean values of I_{geo} of the trace metals were generally lower than 0, indicating no contamination of these elements at present. Only the mean I_{geo} value of Co in Wushan county and those of Co and Pb in Duping were slightly higher than 0, which suggested no contamination to moderate contamination [50].

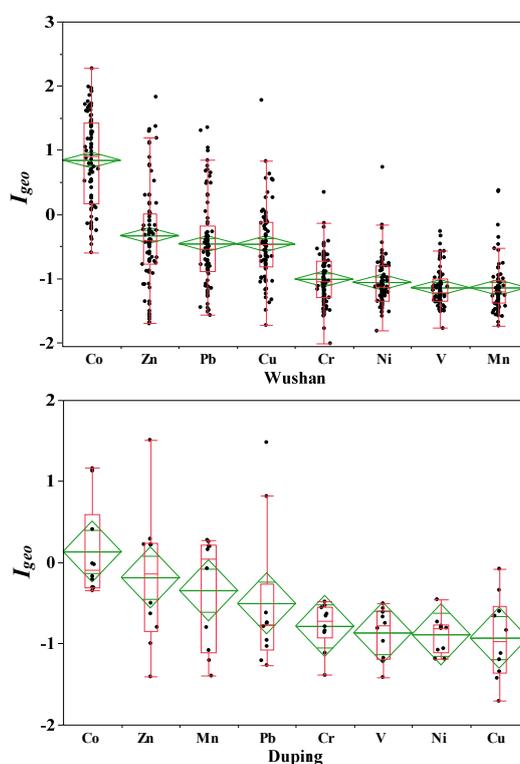


Figure 2. Geo-accumulation index (I_{geo}) of trace metals in the road-dust samples of Wushan and Duping, respectively.

But it should be noticed that many individual values of I_{geo} were higher than 0, which might indicate some degree of trace metal contamination in the sampling sites. Specifically, the I_{geo} of Cu, Co, Pb, and Zn in Wushan and Co, Pb, and Zn in Duping were mostly higher than 1, which indicated moderate contamination of these metals in corresponding sampling sites. Therefore, the above metals were the main potential pollutants in both locations. Compared to those in Chongqing, the accumulation levels of these elements in Wushan and Duping were generally lower [34,35]. This result further confirmed that trace metal contents in road dust might increase along with city growth and its dynamics [51].

3.3. Multivariate Statistical Analysis and Source Identification

3.3.1. Correlation Analysis between Trace Metals

Spearman's correlation coefficients are illustrated in Tables S3 and S4 to reveal the interrelationships among trace metals, Al and Fe. For Wushan, moderate positive correlations existed in the following trace metal pairs: Mn-Cr, Cu-Cr, Pb-Cr, Ni-Cr, V-Cr, Zn-Cr, Cu-Pb, Cu-Ni, Cu-Zn, Mn-V, Mn-Ni, Zn-Ni, V-Ni, and Zn-Pb. Since significant correlations of trace metals suggested a similar origin of them [52], three groups of the analyzed trace metals were identified. Of the eight metals, significant correlations were found between Cr, Ni, Mn, and V, and they all significantly correlated to Fe and Al. This result indicated similar geochemical behaviors of these metals and natural sources of them. Copper, zinc, and lead also showed a significant correlation between each other, but they presented weak or no correlations to Al and Fe. Therefore, the main source of them was from anthropogenic activities [2]. Only Co was not correlated to any of the other elements, which suggested a different source of this element. For Duping, similar results were found. Significant positive correlations were observed between Cr-V, Cu-Pb, Mn-Ni, Mn-V, and Ni-V. Similarly, Cr, Ni, Mn, and V were indicated to primarily originate from natural sources. Copper and Pb were significantly correlated to each other and had similar sources. Zinc and Co were not correlated with any of the other metals, and they might have different sources.

3.3.2. Principal Component Analysis (PCA)

Principal component analysis (PCA) was conducted with the data from Wushan to provide an estimation of the amount of variance explained by a particular number of components (Table S5). The results of PCA exhibited that four principal components (PCs) were obtained to interpret 82.46% of the total variance. PC 1 was predominately composed of Cr, Mn, and V, which explained 26.64% of the total variance. PC 2 explained 25.74% of the data variance, which was heavily weighted by Cu, Pb, and Zn. PC 3 and PC 4 accounted for 16.61% and 13.47% of the total variance, and the main element was Ni and Co, respectively.

An intuitive representation of the result was illustrated by a rotated component loadings plot (Figure 3). The vectors representing the contents of Cu, Cr, Pb, Mn, Zn, and V were close to 1, indicating a good representation of these elements by PC 1 and PC 2. By analogy, the direction and length of the vectors of Ni and Co indicated that they were well represented by PC 3 and PC 4, respectively. Interestingly, the loading characteristics of Ni and Co were visibly different. The vector of Ni directed towards PC 2 to some degree, but that of Co only directed towards PC 4.

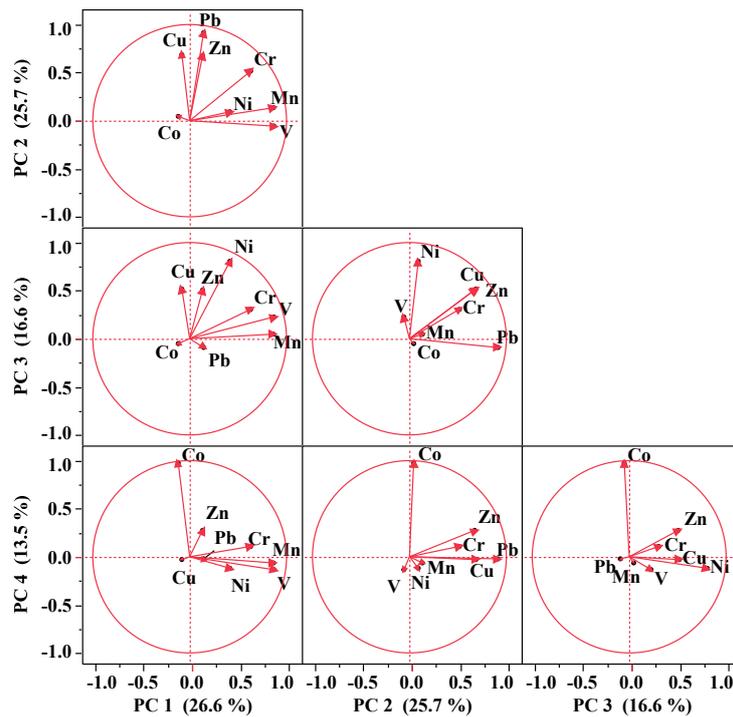


Figure 3. Rotated component loadings plot of the trace metals in road dust from Wushan. PC: principal component.

3.3.3. Source Identification

Trace metals are considered to be markers of some typical sources [53]. The results of correlation analysis and PCA combined, three groups were identified for the eight trace metals studied in Wushan. To validate the correlation between trace metals and environmental factors, RDA was conducted and similar results were obtained (Figure 4). Several environmental variables of road dust, such as magnetic susceptibilities (i.e., low-frequency (LF), high-frequency (HF), frequency-dependent (FD)) and total organic carbon (TOC), were included. Both magnetic susceptibilities and TOC were helpful indicators of the anthropogenic sources of trace metals [54].

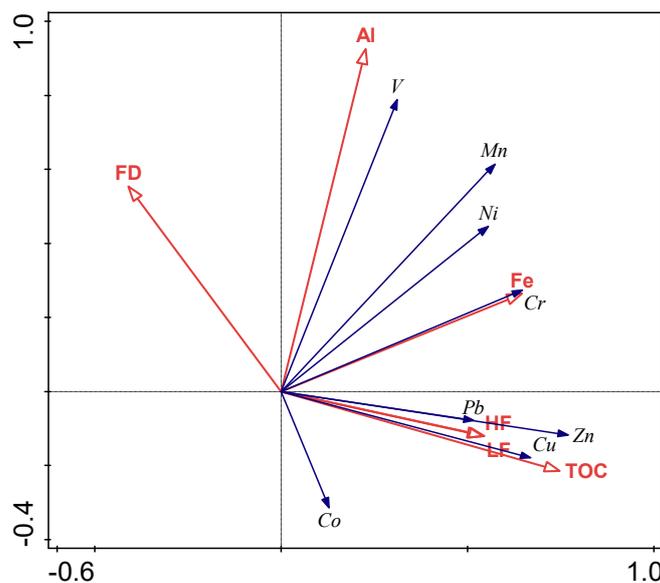


Figure 4. Redundancy analysis of trace metals and physicochemical indexes in road dust. FD: frequency-dependent; HF: high-frequency; LF: low-frequency; TOC: total organic carbon.

The first group included Cr, Mn, Ni, and V, and their close relationships to the conservative elements, i.e., Al and Fe, indicated that they mainly originated from natural sources [55]. This result was consistent with that of the I_{geo} , which indicated no obvious contamination of the above metals in the study area.

The second group included Cu, Pb, and Zn, and their close relationships to LF, HF, and TOC suggested that they mainly came from anthropogenic sources [54]. It was generally acknowledged that traffic emission was the dominant source of Pb, Cu, and Zn in roadway dust [1,52,55]. Except for traffic emission, fertilizers and agrochemicals applied in agricultural activities were also suggested to be one source of trace metals in road dust [56,57]. In our study, TOC was measured and significant correlations were observed between TOC and Cu ($R = 0.49$, $\alpha = 0.05$) and TOC and Zn ($R = 0.72$, $\alpha = 0.05$). Since TOC represented the organic content in urban dust, the value of TOC was closely related to agricultural activities in the study area. Continuous and heavy application of inorganic fertilizer, pesticides, and soil amendments would cause the metal accumulations in road dust [58]. Thus, traffic emission and agriculture activities were identified as the main sources of Pb, Cu, and Zn in roadway dust.

Of the three metals in this group, Zn presented a high level of accumulation in most areas of Wushan. According to Liu et al. [29], Zn might become the most dominant metal in road dust due to its wide utilization in the components of vehicles. In addition, Zn might also originate from agricultural fertilizers and livestock manure [59]. Considering the phase-out of leaded gasoline, a historical trend of an obvious decrease has been observed for Pb in recent decades [6,60]. Therefore, more attention should be gradually shifted to Zn.

Cobalt was separated from the other trace metals, which might originate from the large consumption amounts of coal and fossil fuel in China [61]. Since the amount of trace metals from the atmospheric deposition in this area was high, the thermal power plant in the east of Wushan might affect the distribution pattern of Co by air pollutant migration [7,62].

As for Duping, similar results were observed. Chromium, Mn, Ni, and V presented significant correlation to Al and Fe, which indicated the natural source of them. Copper and Pb were significantly correlated to each other, which might originate from traffic emission. Different from Wushan, Zinc presented no significant correlation to other metals. As discussed above, Zn might have a mixed source, which includes traffic emission and agriculture activities [28,63].

3.4. Spatial Distribution Characteristics of Trace Metals

Considering the non-normal distribution of trace metal contents, Gauss Kriging interpolation method was applied to explore the spatial distribution characteristics of trace metals in Wushan. As shown in Figure 5, low concentrations and spatial heterogeneity of Mn, Ni, Cr, and V further confirmed the natural origins of these elements. Some high metal concentrations were observed in the southwest corner of Wushan, and this might be due to the scattering point pollution sources related to steel product processing [64].

Copper, Pb, and Zn presented similar spatial distribution characteristics, which suggested that except for industrial land use, residential land use was also characterized as having a high accumulation of Cu, Pb, and Zn [65]. Meanwhile, most of the heavily polluted points scattered along the diagonal of Wushan, especially for Pb and Zn. This diagonal was consistent with the two main road directions, which confirmed that these metals were closely associated with traffic emissions [66].

As discussed above, Co was the most contaminated metal in this study. It could be observed in Figure 5 that Co concentrations in most areas in Wushan were higher than the background value and the concentrations increased obviously from northeast to southwest. The wide distribution of Co might be the result of the wide use of biomass fuels by local residents [67]. While the increasing tendency was due to the thermal power plant in the east of the county and the prevailing wind direction in this area.

In conclusion, the results of GIS mapping combined with the application of multivariate statistical analysis indicated that trace metal contamination in Wushan and Duping were not as serious as that in

big cities at present. However, the accumulation and potential environmental risks of trace metals should not be neglected. Trace metals in road dust exhibited complex spatial variations and patterns in counties and small towns impacted by the rapid industrialization and urbanization [68].

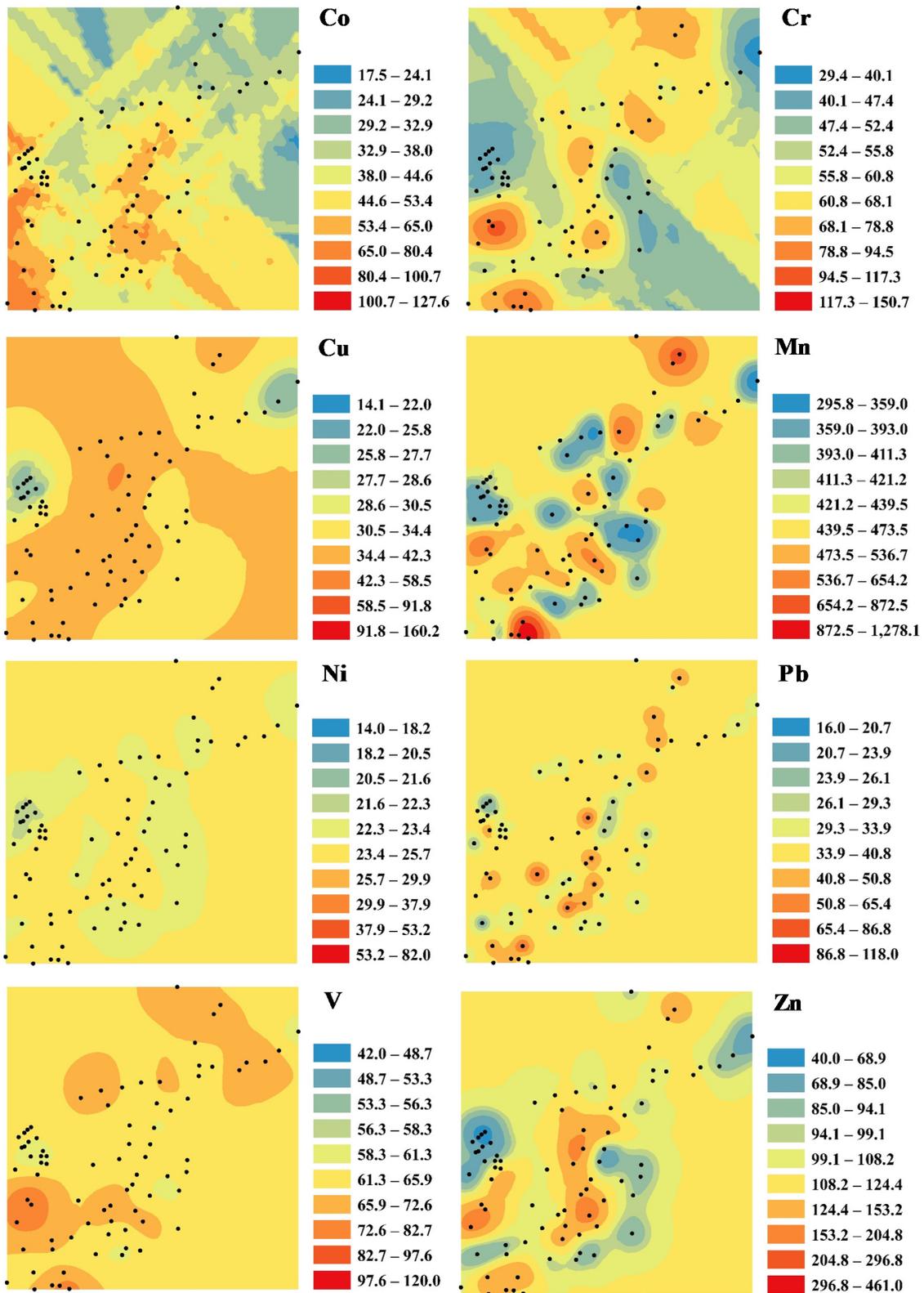


Figure 5. Spatial distribution characteristics of the trace metals in road dust.

3.5. Relationships between Trace Metal Contamination and Socio-Economic Factors

Socio-economic factors such as the population size, population density, built-up area, industrial structure, gross domestic product (GDP), and the number of automobile vehicles were commonly related to the environmental contamination levels [42,69]. In reference to previous research and data availability, the following factors of the Chongqing region were selected: (1) per capita GDP; (2) population density; (3) number of vehicles per 100 persons; and (4) number of vehicles per square kilometers [52,69]. Data for the above factors were obtained from the data library of Chongqing statistical yearbooks and regional economic statistics yearbooks in corresponding years. The average I_{geo} values were applied to represent the average levels of trace metal contamination.

Trace metal contamination was related to the socio-economic factors (Figure 6). Specifically, significant linear relationships were observed between the average I_{geo} values and vehicle density ($R^2 = 0.50$) and population density ($R^2 = 0.41$). A weak positive linear relationship was also observed with the number of vehicles per 100 persons ($R^2 = 0.21$). However, the relationship between trace metal contamination and per capita GDP was nonlinear. Therefore, population and vehicle density were key factors affecting the pollution level of trace metals. An elevated population density leads to more energy consumption and pollutant emissions. Similarly, increasing amounts of vehicles would lead to the translocation and accumulation of trace metals in the roadway dust. Since population and vehicle numbers will increase greatly in counties and towns in the process of rapid urbanization, a large number of counties and towns tend to face increasing risks of trace metal contamination.

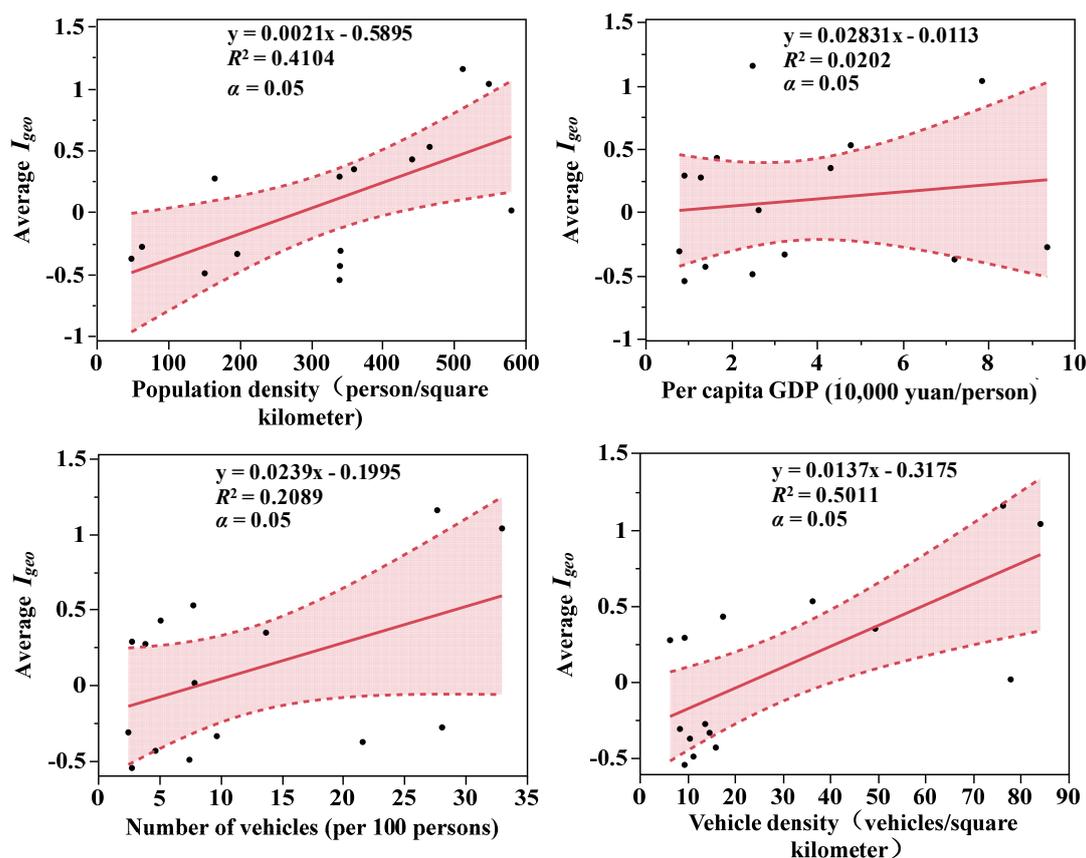


Figure 6. Relationships between the average I_{geo} of trace metals and socio-economic factors; the solid line on each scatterplot displays the linear fitting curve of the relationship; the shadow represents the 95% confidence interval.

Compared to the classical Environmental Kuznets Curve (EKC) hypothesis with inverted U-shaped fitting curves [69], a linear relationship was observed between trace metal pollution and population and

vehicle density in this study. Limited by data availability, the analysis in this study only concentrated on Chongqing, which might not present the overall trend between the socio-economic factors and trace metal pollution in China. The result observed in this study might only present one stage of the U-shaped fitting curves in the EKC hypothesis. In future work, ground-based monitoring datasets should be used to quantify the spatial pattern in a fairly long period and understand the relationships between trace metal accumulation and socio-economic variables. Therefore, the control point during the urbanization process might be revealed which were related to the differences in urbanization gradients.

4. Conclusions

This research investigated the trace metal accumulation in roadway dust in a typical mountainous county in the Three Gorges Reservoir Region, Southeast China. Moderate contamination was suggested by I_{geo} in specific sites in both locations. Specifically, more concern should be focused on the high anthropogenic accumulation of Pb, Co, Cu, and Zn. The analysis of multivariate statistical analysis and GIS mapping identified a natural source for Cr, Ni, V, and Mn, and an anthropogenic origin for Co, Pb, Zn, and Cu. It was also revealed that Co distribution was controlled by coal combustion and Pb, Cu, and Zn by traffic emission. Meanwhile, agriculture activities, especially the wide application of fertilizers and pesticides, should pay more attention to the control and prevention of trace metal contamination. The associations between trace metal contamination and socio-economic factors reflected that rapid urbanization might increase the pollution risk of trace metals in counties and towns.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2071-1050/12/14/5624/s1>. Figure S1: Comparison of trace metal concentrations between Wushan and Duping. Table S1: Pollution categories on the basis of I_{geo} values. Table S2: trace metal contents in the road dust of this study and other cities. Table S3: Spearman's correlation coefficients among trace metals, Al and Fe, in road dust from Wushan. Table S4: Spearman's correlation coefficients among trace metals, Al and Fe, in road dust from Duping. Table S5: Rotated component matrix of principal components analysis of trace metals in road dust.

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