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Characteristics, source apportionment and health risk assessment of heavy metals in urban road dust of the Pearl River Delta, South China

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ABSTRACT

To investigate the characteristics of heavy metals (As, Cd, Cr, Cu, Pb, Hg, Ni and Zn) in urban road dust from different cities and functional areas in the Pearl River Delta (PRD), South China, a total of 294 dust samples were analyzed. The contamination characteristics and health risk of heavy metals in the dust were assessed, their chemical speciation were distinguished, and their sources were identified by the correlations, cluster and principal component analysis (PCA). The mean concentrations of As (15.89 mg/kg), Cd (1.59 mg/kg), Cr (143.75 mg/kg), Cu (184.42 mg/kg), Pb (114.82 mg/kg), Hg (0.11 mg/kg), Ni (41.53 mg/kg) and Zn (645.94 mg/kg) in urban road dust were in high or moderate levels compare with other previous researches. In this case, the contamination of Cr, Cu, Ni and Zn in the industrial area (IA) and the contamination of Cd and Hg in the commercial area (CA) were significantly higher relative to other functional areas ($P < 0.05$), and the contamination of heavy metals in Foshan City was significantly higher than other cities ($P < 0.01$). The order of mobility of the heavy metals with higher concentration in urban road dust of the Pearl River Delta declined in the following order: Zn, Ni, Cu, Pb and Cr. Statistical analysis result showed the contaminated heavy metals in urban road dust were mainly contributed by industrial activities, traffic activities and building pollution. There were no significant carcinogenic and noncarcinogenic risks for adults, children however showed significant noncarcinogenic effect caused by As and Cr in partial points, albeit with low contamination level of the two metals. The ingestion was a principal pathway for heavy metals via urban road dust to exposure population. More protection measures should be considered to reduce children's exposure to the dust, especially in the CA and IA.

1. Introduction

Urban road dust, a major sink and source of air pollution, has been reflected anthropogenic sources and urban industrial activity and proved an excellent marker to characterize urban environmental quality (Bourliva et al., 2017; Huang et al., 2014b; Liu et al., 2014). With the rapid growth of urbanization and industrialization in the last few decades in China, anthropogenic activities introduce large volumes of contaminants (e.g., heavy metal) into the urban environment, which represent a hidden threat to human health (Acosta et al., 2015; Soltani et al., 2015; Tang et al., 2017; Trujillo-Gonzalez et al., 2016). Previous studies indicated that heavy metals in urban road dust could show noticeable cytotoxicity and detrimental effects on biota even at a low concentration (Bing et al., 2018; Luo et al., 2019). Heavy metals contamination in dust had become serious and ubiquitous in big cities of China (Lu et al., 2010; Wei and Yang, 2010), therefore, identification and evaluation of contaminated areas of early detection becomes an imperative.

Heavy metals such as Cu, Pb, Zn, Ni, Cd and Cr contamination of dust in megacities had been widely reported in recent years (Christoforidis and Stamatis, 2009; Faiz et al., 2009; Lu et al., 2010; Luo et al., 2020, 2011; Wei and Yang, 2010). In urban area, heavy metals in dust are discharged from traffic, industrial and domestic emission, and city construction and demolition activities (Dong et al., 2017; Keshavarzi

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et al., 2015; Li et al., 2013; Liu et al., 2015a; Wang et al., 2016a; Wei et al., 2015). The land uses and human activities are all the important cause leading to heavy metals contamination in urban road dust (Trujillo-Gonzalez et al., 2016; Wei et al., 2015). Thus, the level of dust heavy metals pollution can be influenced by the functional organization of cities (Del Rio-Salas et al., 2012). For example, the higher concentrations of Cu, Cd and Cr in the dust of the commercial area, among the functional areas of Chengdu City, China (Li et al., 2017). In Villavicencio, Colombia, the heavy metals concentrations of dust in commercial area were markedly higher compared with other functional areas (Trujillo-Gonzalez et al., 2016). The higher content of heavy metals was founded in dust samples from the high-density residential areas in Warsaw, Poznan (Lisiak-Zielinska et al., 2021). Therefore, an in-depth look at the distribution and sources of heavy metals in urban road dust are vital to prevent and control the contamination of heavy metal in densely populated cities.

Urban road dust can be a serious harm to human health due to inherent mobility leading to the possibility of exposure, including hand-mouth intake, respiratory inhalation and skin contact, especially for children (Cao et al., 2014, 2015; Man et al., 2010; Zeng et al., 2016; Zhao et al., 2012). However, the total amount of heavy metals cannot completely reflect the environmental hazards due to the peculiarities of migration, bioavailability and biological toxicity (Perez et al., 2008), which have a relationship with the morphology (Luo et al., 2011). Typically, bioavailability of metals in plants, a key-parameters, is used to measure the ecological impact and migration of metals in environment (Guo et al., 2013). Plants absorb and utilize metals, mainly in exchangeable forms of metals, and it is very easy that carbonate-bound form of metals is freed from the soil, leading to high mobility (Zhang et al., 2018). Moreover, the residual form of metals has less migration capacity, activity and toxicity compared to other forms.

The Pearl River Delta (PRD), South China, one of the most dynamic economic zones in the Asia Pacific region, possesses a world-famous manufacturing and service industry base and plays an important role in GDP of Guangdong Province (Wang et al., 2016b). However, the phenomenon like high population density, rapid mobilization, urbanization and industrialization have brought enormous challenges to the ecological environmental development of the Pearl River Delta. In consideration of the adverse impacts created by heavy metals in urban road dust, to explore their pollution level in the Pearl River Delta and pollution source is of great significance. In recent years, a certain number of researches on heavy metal pollution of dust in the Pearl River Delta have been carried out, including air pollution (Dai et al., 2015; He et al., 2018; Li et al., 2018; Mai et al., 2018; Tong et al., 2018), human health risk assessment (Wu et al., 2019; Ye et al., 2021), and source apportionment (Ye et al., 2018). However, there is a lack of a complete system to explore the occurrence, source, chemical speciation, mobility and human health risk for urban road dust in the Pearl River Delta. Moreover, it is the first known study to address their spatial distributions and sources of dust in different functional areas in the Pearl River Delta.

With a view to the economic development level of the Pearl River Delta and the number of permanent residents, the top six cities were selected as the research objects, namely Guangzhou, Shenzhen, Foshan, Dongguan, Huizhou and Zhongshan. More potentially harmful eight metals for urban road dust (Men et al., 2018; Zhang et al., 2019), such as arsenic (As), cadmium (Cd), chromium (Cr), cupper (Cu), lead (Pb), mercury (Hg), nickel (Ni) and zinc (Zn), were analyzed during summer and winter from six cities of the Pearl River Delta, South China. The main objectives are (1) to address the influence of different functional areas on distribution patterns of heavy metals; (2) to evaluate the pollution with heavy metals using the geo-accumulation index (*Igeo*); (3) to distinguish the chemical speciation of heavy metals in urban road dust; (4) to determine possible sources of heavy metals in terms of multivariate statistical analysis, including principal component analysis (PCA), correlations and cluster analysis (Gallego et al., 2002; Yang et al., 2011); (5) to evaluate risks to human health from dust exposures.

2. Materials and methods

2.1. Sample collection

A total of 294 dust samples were collected from six cities in December 2015–June 2016 (Fig. 1), including Guangzhou (GZ, n = 54), Shenzhen (SZ, n = 48), Foshan (FS, n = 48), Dongguan (DG, n = 48), Huizhou (HZ, n = 48), Zhongshan (ZS, n = 48). Each city was divided into residential area (RA), commercial area (CA), industrial area (IA) and transportation area (TA). Among them, 6 samples were collected from IA in GZ and each functional area in SZ, FS, DG, HZ and ZS in summer and winter, respectively, while 7 samples were collected from RA, CA and TA in GZ. The roadway characteristics of sampling sites were listed in Table S1. Pooled samples of about 500 g of dust were collected using a plastic dustpan and a brush, washed with acid and deionized water before each sampling (Pueyo et al., 2008; Wang et al., 2010). All samples were then placed in polyvinyl chloride (PVC) packages and transported to the laboratory.

2.2. Sample extraction

Dust samples were air-dried and then removed coarse impurities (e.g., small stones, refuse and leaves) by a 20 mm nylon sieve. The sieved dust samples were further ground using an agate mortar and pestle and sieved through 200 mesh sieves before chemical assay.

Approximately 0.5 g of each dust sample was digested by CEM MARS-6 microwave digestion apparatus with 2.5 mL HNO_3 and 8 mL HCl at 190 °C, until the solutions ran clear (2.5 mL solution left). The digested samples were cooled and diluted to 50 mL with 1% HNO_3 and then filtered through a 0.45 mm filter. The total As content was measured by atomic fluorescence spectrometry (AFS-930, Beijing Jitian instrument Co., Ltd, China). The total Hg content were measured by a cold vapor atomic absorption spectrometer (CVAAS, Hydra-C, Leeman Labs, USA). The total Cd, Cr, Cu, Pb, Ni and Zn content were measured using inductively coupled plasma-optical emission spectrometry (ICP–OES, Agilent Technologies, 700 Series, Belgium).

The modified BCR procedure was used to determine chemical speciation of heavy metals in urban road dust (Pueyo et al., 2008). The following fractions were determined, including acid soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3), and a residual fraction (F4). The extractants and operationally defined chemical phases (Keshavarzi et al., 2015) are presented in Table S2. The details about the modified BCR procedure can be found in previous studies (Nemati et al., 2009; Tokalioglu and Kartal, 2006; Yıldırım and Tokalıoğlu, 2016). Approximately 1.0 g of each dust sample was applied by the method. The extractants of each step of the BCR scheme were evaporated to near dryness and then completed to 5 mL with 1 M HNO_3, except the third step. The final volume for third step is 6 mL. All samples, including blank, were analyzed in triplicate throughout the analyzes.

2.3. Quality assurance and quality control

In the course of the experiment, in order to completely remove the residual heavy metal ions on the experimental equipment, the Teflon digestion tank and all the glass laboratory equipment were immersed in 20% and 10% nitric acid for more than 24 h, respectively, and then washed with deionized water 3 times.

Two procedural blank, two standard reference materials (GBW-07405 for soils, National Research Center for Standards, China) and at least 10% parallel samples were included with each batch. The procedural blanks for all analytes were below the method detection limit (MDL). The recoveries of the elements ranged from 81.06% to 102.47%. The modified BCR sequential extraction procedure was applied for the BCR-701 certified reference material. The recovery values changed between 86.40% and 102.31%.

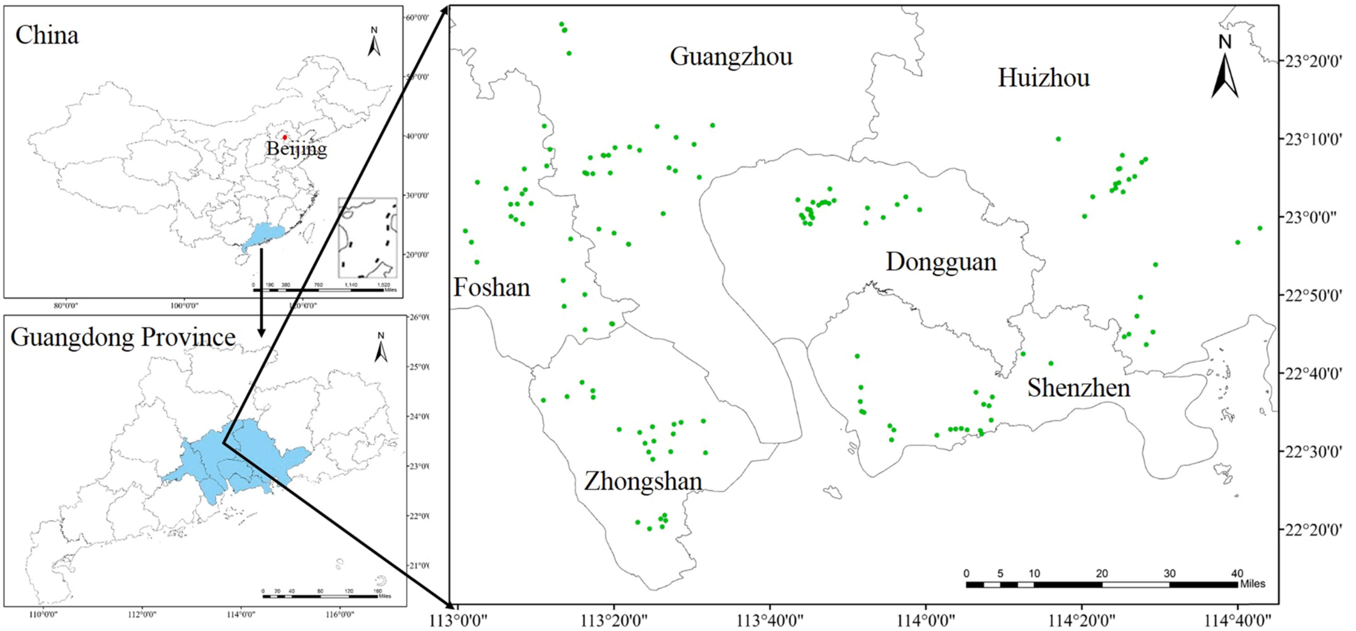

Fig. 1. Map of the urban road dusts sampling sites in the Pearl River Delta, South China.

2.4. Calculations

The geoaccumulation index (I_{geo}) was a quantitative indicator for the level of heavy metal pollution in dust (Wang et al., 2010). The function can be formulated as follows:

$$I_{geo} = Log_2 \frac{C_s^i}{1.5 \times C_n^i}$$

where C_s^i is the heavy metal test content; C_n^i is the background value of heavy metals, which were selected from the soil background value in Guangdong province.

(As 8.9 mg/kg, Cd 0.056 mg/kg, Cr 50.5 mg/kg, Cu 17.0 mg/kg, Pb 36.0 mg/kg, Hg 0.078 mg/kg, Ni 14.4 mg/kg, Zn 47.3 mg/kg) (CNEMC, 1990). The pollution status was classified into 7 grades based on *Igeo* (Pan et al., 2017), listing in Table S3. There are three main pathways of heavy metal intake: ingestion, inhalation and dermal contact. In this study, risk characterization was evaluated for non-carcinogenic and carcinogenic risk, based on those developed by the US EPA (Pan et al., 2018; US EPA, 1989, 2002). The health risk models can be formulated as follows:

$$ADD\text{ing} = C \times \frac{IngR \times EF \times ED}{BW \times AT} \times 10^{-6}$$

$$ADD\text{inh} = C \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT}$$

$$ADD\text{dermal} = C \times \frac{AF \times SA \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6}$$

The model was used to calculate the lifetime average daily doses (LADDs) for Cr, Cd and Ni (Wang et al., 2016a; US EPA, 1996a, 1996b, 2002), and formulated as follows:

$$LADD\text{ing} = \frac{C \times EF}{AT} \times \left(\frac{IngR_{\text{child}} \times ED_{chlid}}{BW_{child}} + \frac{IngR_{adult} \times ED_{adult}}{BW_{adult}} \right) \times 10^{-6}$$

$$LADD\text{inh} = \frac{C \times EF}{AT \times PEF} \times \left(\frac{InhR_{child} \times ED_{child}}{BW_{child}} + \frac{InhR_{adult} \times ED_{adult}}{BW_{adult}} \right)$$

$$LADD\text{dermal} = \frac{C \times EF \times ABS}{AT} \times \left(\frac{AF_{chlid} \times SA_{child} \times ED_{child}}{BW_{child}} + \frac{AF_{adult} \times SA_{adult} \times ED_{adult}}{BW_{adult}} \right) \times 10^{-6}$$

where C is the heavy metal test content. Other parameters are presented in Table S4. The hazard index (HI) and total carcinogenic risk (TR) were calculated as equations (US EPA, 2002):

$$HQ = \frac{ADD\text{ing}/inh/dermal}{RfD}$$

$$HI = \sum HQ_{\text{i}}$$

$$CR = LADDing/inh/dermal \times SF$$

$$TR = \sum CR_{\text{i}}$$

where RfD is the reference dose; SF is the cancer slope factor. Values of parameters were presented in Table S5.

2.5. Statistical analysis

Spearman correlation analysis was performed to elucidate the interrelationships between heavy metals. Cluster analysis and PCA with varimax rotation were run to determine the relationship between the heavy metals and to identify their probable sources. SPSS 18.0, Origin 9.1 and ArcGIS 10.4.1 were used to perform the data treatment, statistical analysis and graphical outputs.

3. Results and discussion

3.1. Distribution characteristics of heavy metal

The heavy metal contents of urban road dust in the Pearl River Delta are presented in Table 1. The arithmetic mean concentrations of As, Cd, Cr, Cu, Pb, Hg, Ni and Zn were 15.89, 1.59, 143.75, 184.42, 114.82, 0.11, 41.53, 645.94 mg/kg, which were 1.8, 28.3, 2.8, 10.8, 3.2, 1.4, 2.9 and 13.7 times the background values. Among the urban road dust, the As, Cd, Cr, Cu, Pb, Hg, Ni and Zn content in 77.4%, 100.0%, 82.2%,

Table 1
The distribution of heavy metals concentrations (mg/kg) in urban road dust in the different functional areas from various cities.

Location		Winter								Summer							
		Pb	Cr	Cd	As	Hg	Cu	Zn	Ni	Pb	Cr	Cd	As	Hg	Cu	Zn	Ni
City																	
Guangzhou	Mean	186.47	122.81	2.16	15.62	0.14	182.20	794.58	40.51	125.09	108.13	1.80	22.93	0.15	140.65	515.31	35.28
	SD	108.39	39.93	1.16	10.17	0.15	98.00	416.94	15.03	75.57	57.21	1.30	7.15	0.14	86.23	236.31	18.52
Shenzhen	Mean	115.09	234.03	1.71	8.37	0.16	218.60	961.20	66.42	150.19	164.60	1.52	15.69	0.10	261.43	893.34	50.67
	SD	78.59	276.49	0.96	5.23	0.15	279.61	1098.54	85.37	122.92	177.77	1.21	6.82	0.11	339.00	724.00	47.07
Foshan	Mean	162.02	255.15	2.17	14.02	0.13	355.23	958.86	66.21	126.47	200.47	1.84	24.32	0.15	312.14	872.80	61.88
	SD	96.05	171.12	1.44	4.98	0.14	432.61	476.75	33.42	84.99	139.84	1.00	8.79	0.12	415.04	708.87	37.98
Dongguan	Mean	101.64	118.16	1.58	11.83	0.11	133.33	713.53	36.24	96.12	106.67	1.34	23.66	0.07	131.32	439.60	31.66
	SD	81.92	86.39	0.94	5.04	0.12	110.14	544.52	23.90	57.72	48.73	0.46	10.74	0.07	81.65	201.72	11.13
Huizhou	Mean	60.45	58.43	0.99	7.99	0.07	56.15	259.33	17.46	72.39	59.65	1.03	14.65	0.07	75.49	296.27	19.66
	SD	27.12	45.03	0.43	5.37	0.12	19.83	225.47	7.87	37.82	21.83	0.38	5.54	0.08	68.79	301.67	11.96
Zhongshan	Mean	92.92	176.30	1.41	11.94	0.08	167.02	508.15	36.79	80.83	126.86	1.42	18.81	0.06	185.13	551.60	37.26
	SD	42.45	197.59	0.75	9.67	0.07	143.42	365.57	15.14	40.29	96.49	0.78	10.20	0.07	219.14	517.43	17.25
Functional area																	
Residential area	Mean	123.39	132.89	1.44	10.84	0.11	119.13	553.50	41.52	104.87	99.02	1.39	19.55	0.10	114.49	563.78	36.18
	SD	86.46	114.58	0.84	5.64	0.10	86.52	400.15	30.00	59.31	66.05	0.97	8.04	0.11	78.35	509.56	23.48
Commercial area	Mean	137.48	143.24	1.91	13.09	0.16	159.81	806.31	41.49	112.20	125.40	1.78	20.69	0.14	143.22	634.83	38.43
	SD	91.56	63.92	0.92	10.68	0.16	82.29	467.52	14.27	55.85	64.32	1.14	10.31	0.13	108.95	454.76	19.23
Industrial area	Mean	109.81	252.13	1.75	12.61	0.09	302.77	918.56	63.54	127.65	196.66	1.52	20.92	0.08	346.22	795.95	53.24
	SD	75.08	270.72	0.95	6.15	0.13	362.15	1006.27	74.12	115.59	181.96	0.81	9.57	0.08	400.09	755.11	44.05
Transportation Area	Mean	111.05	114.38	1.62	10.40	0.10	167.54	531.94	29.02	91.20	94.40	1.30	19.05	0.07	134.33	388.25	30.71
	SD	98.76	136.06	1.44	6.82	0.12	273.30	342.67	17.86	72.15	72.33	0.83	8.48	0.08	209.05	213.71	26.36

100.0%, 95.5%, 45.9%, 89.9% and 100.0% of the dust samples exceeded the background values of local soil, respectively. This indicated the heavy metals of dust samples were most likely from human activities. Concentrations of heavy metals in the Pearl River Delta urban road dusts are compared with data reported for other cities in the world (Table 2). Individually, concentrations of Cr, Cu and Zn in urban road dusts from the Pearl River Delta were higher to those previously measured in Tokat, Mexico and most parts of China, like Beijing, Nanjing, Jiaozuo, Chengdu, Shijiazhuang. The mean concentrations of Cd and Ni in urban road dusts of the Pearl River Delta were comparable to most of the areas listed in Table 2, except for the Cd in dust of Tehran. The concentration of Pb in urban road dust in the Pearl River Delta was higher than those in Beijing, Nanjing, Jiaozuo, Chengdu, but lower than those in Xi'an, Shijiazhuang and all listed foreign countries.

Significant difference was not shown in the contents of Cd, Cr, Cu, Pb, Hg, Ni and Zn during summer and winter in the Pearl River Delta tested by paired sample t-test ($p > 0.05$). However, there were significant differences between eight heavy metal contents in different cities ($p < 0.01$). The concentrations of eight heavy metals showed a trend of decreasing first, then rising and then decreasing again from west to east, with more polluted in Foshan. With the rapid development of economy, wide variety of key industries had occurred in Foshan during the last thirty years, including ceramics, household appliances and furniture manufacturing, which will bring heavy metals polluting (Tan et al., 2016). For example, high levels of heavy metals, such as As, Cd, Cu, Pb and Zn, will be released when the ceramic is fired (Tan et al., 2016; Tian et al., 2010; Zhang et al., 2010).

To further characterize spatial distribution of heavy metals in dust in the Pearl River Delta, heavy metals in dust from different urban land use were explored (Table 1). The concentrations of Cd, Cr, Cu, Hg, Ni and Zn in dust among four functional areas exhibited significant differences ($p < 0.05$), which might be related to the different modes and intensities of human activity in different functional areas (Del Rio-Salas et al., 2012). Among the different functional areas, the hot spots of Cr, Cu, Ni and Zn concentrations in the dust occurred at the IA and most Cd and Hg polluted in the CA. It may originate from the higher vehicular traffic, construction and demolition activities in CA, and industrial emissions in IA (deMiguel et al., 1997; Dong et al., 2017; Fang et al., 2001). The high level contaminations of heavy metals in dust of CA and IA in the Pearl River Delta were consistent with previous studies. Mihankhah et al. (2020) had found the concentration of Pb, Cu and Zn in dust in commercial areas is higher than other functional areas. Meanwhile, the markedly higher concentrations of heavy metals were detected in the commercial area, situated in Villavicencio, Colombia (Trujillo-Gonzalez et al., 2016). On the other hand, the higher Cu, Ni and Cr concentrations in dust were found in industrial areas of Shenyang city, compared to other functional areas (Wang et al., 2021). Spatial variation of heavy metal distribution in dust heavily according to the urban land use. For example, the higher concentration of Cr, Cu and Ni occur at Foshan due to industrial emissions, which was relevant for its types of industries (e.g., appliances and electronics, etc.). The above three kinds of heavy metals were common pollutants in the environment by electronic or metallurgy industries (Yeung et al., 2003).

3.2. Contamination assessment of heavy metals

The mean contamination of heavy metals in the dust based on the *Igeo* showed the order of Cd > Zn > Cu > Pb > Ni > Cr > As > Hg (Fig. 2). According to the classification of *Igeo*, Cd in dust presented heavily to extremely contamination (4.07), Zn and Cu presented moderately to heavily contamination (2.81, 2.31), and Pb, Ni, Cr and As presented uncontaminated to moderately contamination (0.85, 0.64, 0.54, 0.10), while uncontaminated by Hg (− 0.63).

The *Igeo* values for heavy metals indicated the highest pollution of other seven heavy metals were appear in Foshan, except for Pb. The degrees of heavy metals pollution were consistent with the above

Table 2
Heavy metals concentrations (mg/kg) in urban road dusts reported in the referenced literature.

City	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn	Digestion method[a]	Ref.
PRD (China)	15.89	1.59	143.75	184.42	114.82	0.11	41.53	645.94	HNO_3-HCl	This study
Beijing (China)	6	1.1	87	46	54	0.34	34	219	H_2O_2-HF	(Tanner et al., 2008)
Nanjing (China)	13.4	1.1	126	123	103	0.12	55.9	394	HCl-HNO_3-HF-$HClO_4$	(Hu et al., 2011)
Jiaozuo (China)	23.08	1.25	112.07	49.85	55.26	–	51.70	374.30	HCl-HNO_3-HF-$HCLO_4$	(Han et al., 2020)
Chengdu (China)	–	1.66	84.3	100	82.3	–	24.4	296	gastric juice- intestinal juice (synthetic)	(Li et al., 2017)
Xi'an (China)	–	–	145	46.8	124.5	–	30.8	267	–	(Pan et al., 2017)
Shijiazhuang (China)	–	1.86	131.70	91.06	154.78	–	40.99	496.17	HNO_3-HF-HCl	(Cai and Li, 2019)
Kavala (Greece)	17	0.2	196	124	301	0.1	58	272	HNO_3	(Christoforidis and Stamatis, 2009)
Newcastle (UK)	6.4	1	–	132	992	–	26	421	HCl-HNO_3	(Okorie et al., 2012)
Tehran (Iran)	–	10.7	33.5	225.3	257.4	–	34.8	873.2	HNO_3-H_2O_2-HCl	(Saeedi et al., 2012)
Amman (Jordan)	–	1.1	18.3	249.6	976	–	16.3	401	HNO_3	(Jiries, 2003)
Tokat (Turkey)	–	3	30	29	149	–	65	63	HNO_3-HCl-HF	(Kurt-Karakus, 2012)
Mexico (Mexico)		–	51.4	99.7	128.2		36.3	280.7	HNO_3	(Aguilera et al., 2021)

[a] Further details are available in refs.

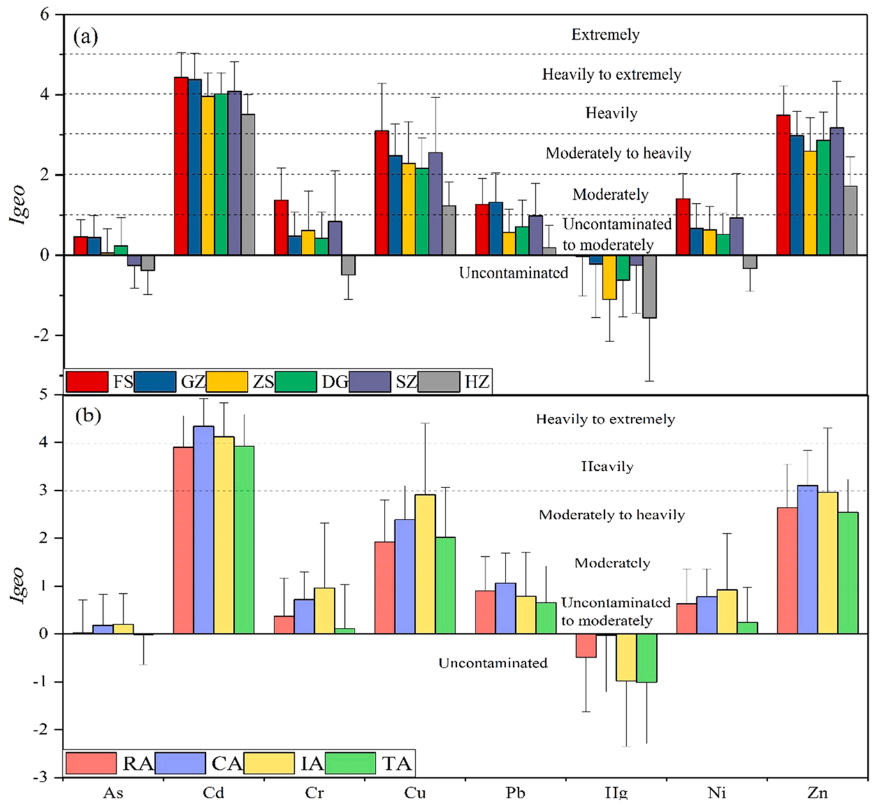

Fig. 2. The geoaccumulation index (*Igeo*) for the eight heavy metals in urban road dusts of in the Pearl River Delta, South China. (a) heavy metals in urban road dust in different cities; (b) heavy metals in urban road dust in different functional areas.

contaminated analysis. This bias seems to be primarily related to its types of industries, for example, Cr and Ni originated from appliances and electronics, and As, Cd, Cu and Zn were released with the ceramic burning (Tan et al., 2016; Tian et al., 2010; Zhang et al., 2010). In addition, the *Igeo* values showed heavy metals contamination in IA and CA was higher than that in others (Fig. 2). These observations agree with the analysis of heavy metal concentrations. The maximum *Igeo* values of As, Cr, Cu and Ni occur in IA, and Cd, Pb and Zn occur in CA. These are all linked in some way to socioeconomic activities (e.g., low-tech auto repair services) that take place in the CA (Trujillo-Gonzalez et al., 2016) and industrial emissions in the IA (Wang et al., 2021). Zn, Pb and Cd are related to tire and brakes wear, lubricants and paints, the socioeconomic activities such as low-tech auto repair services may make the emission of the above heavy metals (Kamani et al., 2015; Martinez and Poleto, 2014).

3.3. Chemical partitioning of heavy metals

Chemical speciation of five metals with higher concentration in urban road dust of the Pearl River Delta determined using a modified BCR procedure are illustrated in Fig. S1. Speciation of Cr and Ni in dust were mostly residual fraction, accounting for 73.11% and 57.87% respectively. The high Cr and Ni concentration in the residual fraction were consistent with that of literature reported (Banerjee, 2003; Passos et al., 2010; Tokalioglu and Kartal, 2006; Tokalioglu et al., 2000). Cu and Pb mainly existed in the form of reducible fraction, while Zn existed in acid soluble fraction. The environmental and health hazards of heavy metals depend on their bioavailability, referring to the traits in which heavy metals cannot be stably presented and are easily released into the environment with environmental conditions changed and thus absorbed by organisms (Luo et al., 2014). The previous researches mainly evaluate the bioavailability of heavy metals based on the content of acid extraction state (Guo et al., 2013; Zhang et al., 2018). Mobility order of heavy metals in the Pearl River Delta was as follows: Zn (40.09%) > Ni (11.88%) > Cu (8.82%) > Pb (5.94%) > Cr (1.52%).

Chemical speciation of heavy metals in urban road dust of the Pearl River Delta were classified based on different cities and functional areas. It indicated the main forms of five heavy metals in six cities and four functional areas were consistent with the analysis of the whole Pearl River Delta.

3.4. Multivariate statistical analysis

3.4.1. Correlations analysis

The Spearman's correlation was employed since the samples were not normally distributed, listing in Table S6. For the four functional areas, positive correlations were observed among Pb, Cr, Cd, Cu, Zn and Ni ($P < 0.01$). The high values of the correlation coefficient between the heavy metals suggest the common sources and mutual dependence (Suresh et al., 2012). Cr and Ni are related to paints used in automobile coatings, while Pb, Cd, Cu and Zn are associated with vehicular emissions and the erosion and wear of tires and alloy (deMiguel et al., 1997; Dong et al., 2017; Pourkhabbaz and Pourkhabbaz, 2012; Yuen et al., 2012), which is in range with earlier studies (Saeedi et al., 2012). The tires and metal parts exposure to high temperature weather can accelerate corrosion processes, which leads metals are released into the urban environment and accumulated in road dust (deMiguel et al., 1997).

3.4.2. Cluster analysis

The variables were standardized by using the z-score, and then Euclidean distances were measured, and Ward's clustering algorithm was used to construct the heat map. The details about the Cluster analysis listed in Table S7. As shown in the hierarchical clustering heat map (Fig. 3), the HCA results indicate five clusters: (1) Cd-Hg-As; (2) Zn; (3) Ni; (4) Cr-Cu; (5) Pb. However, clusters were moderately associated with each other. Cd-Hg-As cluster were well correlated with each other, and joined to the Zn and Ni clusters. Cr-Cu cluster was joined to the Pb cluster. The close association between clusters suggested a common origin. In this case, Cd, Hg and As group was primarily related to erosion and wear of building materials (Fang et al., 2001; Saeedi et al., 2012; Wei and Yang, 2010; Yıldırım and Tokalıoğlu, 2016). Zn and Ni groups mainly derived from industrial activities (Yeung et al., 2003) and corrosion of automobile parts (Li et al., 2001; Lopez et al., 2011; Yuen et al., 2012). Cr, Cu and Pb groups were likely to originate from the contribution of electronic or metallurgy industries (Dhal et al., 2013; Yeung et al., 2003) and using of and paint (Du et al., 2014; Zhang et al., 2020).

3.4.3. Principal component analysis

The method of PCA has been used extensively to determine the sources of heavy metals, based on its ability for classifying multiple metals in the environment with similar behaviors and sources (Han et al., 2020; Luo et al., 2020). PCA revealed the first two principal components (1, 50.7%; 2, 13.8%) accounting for approximately 64.5% of the total variance in the dust (Fig. 4). Factor 1 was dominated by Zn, Cr and Cd with the positive loadings. The higher Zn, Cr and Cd concentrations in the dust were observed in the industrial area, and in Foshan. It was reported the manufacturing bases in Foshan was one of the largest in the world (Tan et al., 2014), and the ceramic plants account for around 30% of total yield of ceramics in world (Tan et al., 2016). The high level of Zn and Cd could be emitted when the ceramic were fired in kiln (Tian et al., 2010; Zhang et al., 2010). Meanwhile, appliances, electronics and non-metallic ore processing were also the key industries in Foshan (Tan et al., 2016). The results of previous studies have shown that Cr and Cd mainly come from electronic or metallurgy industries (Yeung et al., 2003). Thus, it can be inferred that

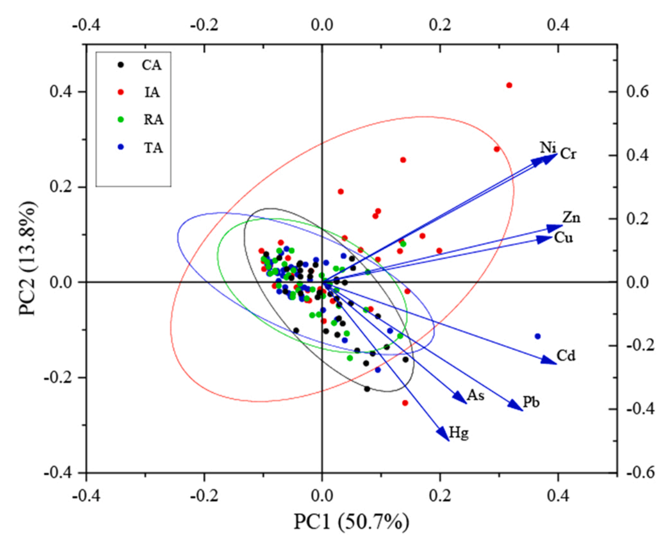

Fig. 4. Plot of PC1 and PC2 from principal component analysis (PCA) of heavy metals as variables for urban road dust of the Pearl River Delta, South China. (Blue arrow means vector resultant of PC1 and PC2).

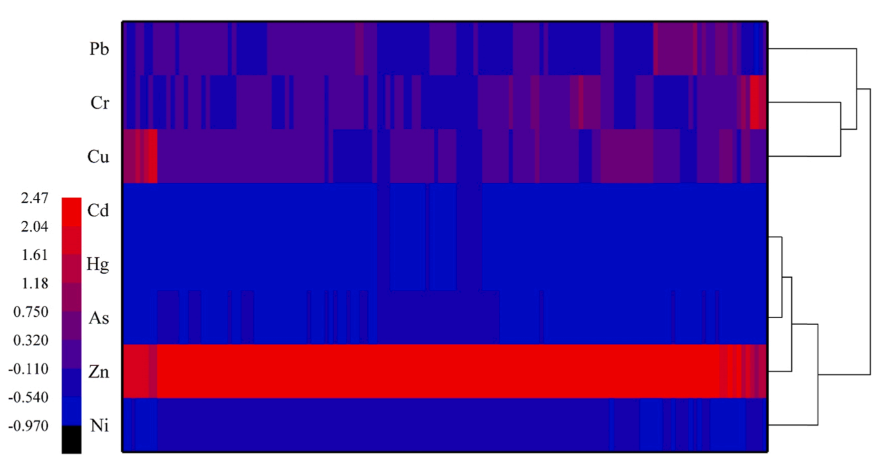

Fig. 3. Heat map of eight metals in urban road dust from the Pearl River Delta, South China.

PC1 is contributed by industrial activities.

The Factor 2 was positively dominated by Hg and Pb and negatively controlled by Cr, indicating the sources of Hg and Pb were different from Cr (Long et al., 2021). The higher Hg and Pb concentrations in the dust were observed in the commercial area, and in Guangzhou. The main source of Hg contamination in environment comes from about ageing of building exterior materials and using mercury-containing batteries and fluorescent lamps (Fang et al., 2001), meanwhile, Pb was mainly associated with vehicular emissions and the erosion and wear of tires (Pourkhabbaz and Pourkhabbaz, 2012; Yuen et al., 2012). Guangzhou is the capital city of Guangdong Province, with intensive human activities and economic output, especially in the CA (Han et al., 2014). Previous studies showed that higher vehicular traffic, construction and demolition activities occurred in CA (deMiguel et al., 1997; Dong et al., 2017; Fang et al., 2001; Mihankhah et al., 2020). Thus, PC2 is contributed by traffic activities and building pollution.

3.5. Health risk assessment of heavy metals

The results of noncarcinogenic risk of heavy metals in dust indicated unlikely adverse health effects for adults in all surveys, but likely adverse health effects for children in partial points of transportation area (Table. S8). It could be explained by the dust was easily suspended in transportation area, which in turn led more exposure frequency for population. The HI values for children were approximately one order of magnitude higher than that of adults, which might originate from pica behavior and hand or finger sucking for children (Li et al., 2015; Wei et al., 2015). The spatial distribution of the non-carcinogenic risks for children are shown in Fig. 5. The non-carcinogenic risk of heavy metals in urban road dust were higher in the west and south of the survey area. In urban road dust, Pb, Cd, As, Hg and Cu have similar spatial risk patterns, which the high HI hot spots are concentrated in the west of the Pearl River Delta (e.g., Foshan and Guangzhou). For Cr, Zn and Ni, in addition to the high HI hot spots in the east, those also exist in south of the Pearl River Delta. However, these high HI for most of heavy metals were below the tolerable limits. Moreover, ingestion was a principal pathway of heavy metals (except for Cd) for exposure population, secondly was dermal contact, followed by inhalation (Fig. S2), which was consistent with previously studies (Huang et al., 2014a; Wang et al., 2014).

The carcinogenic risks for exposure population were all lower than 1×10^{-4} (Table S8), suggesting the individual cancer risk for Cd, Cr and Ni remained the acceptable risk level in the Pearl River Delta. However, several uncertainties and limitations still exist in the present study for risk assessment. The effect factors mainly included (1) some exposure parameters used in this study were derived from the United States Environmental Protection Agency (USEPA) which may not be entirely applicable for the local context; (2) the background soil values were used to replace the lack of background surface dust values; (3) not all carcinogenic metals (such as Fe and Co, etc.) were conclude in this study (Hu et al., 2012; Liu et al., 2015b; Wang et al., 2021). There are still other potentially toxic metals (e.g., V, Mn, Sb, Fe and Co, etc) and organic pollutants (e.g., microplastics, polycyclic aromatic hydrocarbons and polybrominated diphenyl ethers, etc.) in the dust that have not been evaluated (Monira et al., 2022; Wu et al., 2019, 2022). Therefore, more research on other contaminants will be needed in the future.

4. Conclusions

In the Pearl River Delta, the mean concentrations of As, Cd, Cr, Cu, Pb, Hg, Ni and Zn in the dust were exceeded their local background values. The heavy metals concentrations of dust were significantly higher in Foshan than other cities, and in CA and IA than other areas. Cd in dust presented heavily to extremely contamination, Zn and Cu

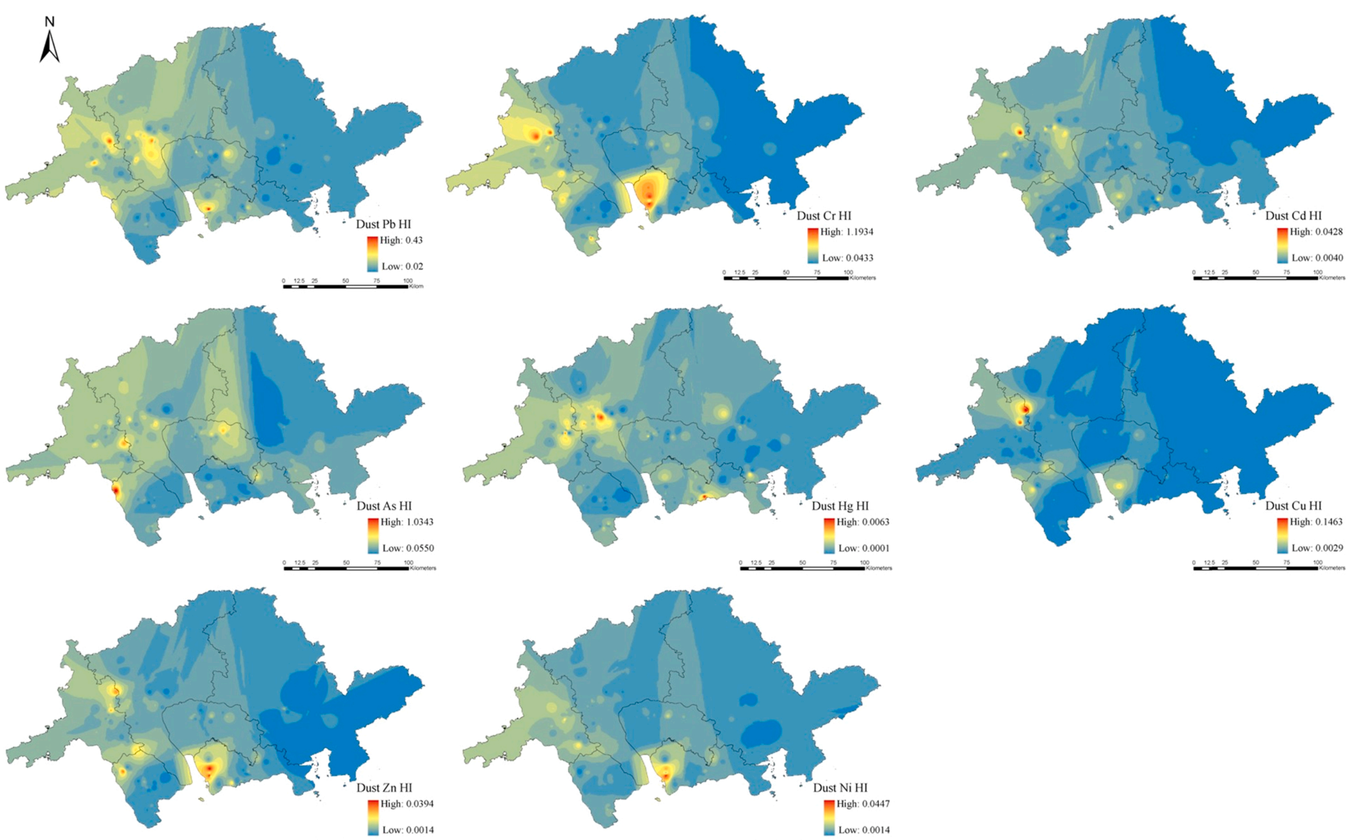

Fig. 5. Spatial pattern of the noncarcinogenic risk from heavy metals for children in urban road dust.

presented moderately to heavily contamination, and Pb, Ni, Cr and As presented uncontaminated to moderately contamination. The order of mobility of the heavy metals in the Pearl River Delta declined in the following order: Zn, Ni, Cu, Pb, and Cr. Industrial activities, traffic activities and building pollution were the main source of heavy metals in urban road dust. The noncarcinogenic risk of heavy metals in urban road dust was unacceptable for children and acceptable for adults. The total cancer risk for Cd, Cr and Ni could be acceptable. Moreover, the present findings have important implications for urban planning and management in the Pearl River Delta.

CRediT authorship contribution statement

Chushan Huang: Writing – original draft, Investigation, Data curation, Formal analysis, Writing – review & editing. **Lijuan Zhang:** Investigation, Writing – review & editing. **Jiuling Meng:** Investigation, Data curation. **Yunjiang Yu:** Writing – review & editing. **Jianying Qi:** Validation, Conceptualization. **Peng Shen:** Conceptualization. **Xin Li:** Investigation. **Ping Ding:** Data curation. **Mianbiao Chen:** Conceptualization, Resources. **Guocheng Hu:** Conceptualization, Methodology, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2022.113490.

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