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Contamination characteristics and potential environmental implications of heavy metals in road dusts in typical industrial and agricultural cities, southeastern Hubei Province, Central China

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Abstract

In November 2013, the total concentration of selected heavy metals in 43 urban dust samples, collected from two small-sized cities of industrial E'zhou and agricultural Huanggang, located in the southeastern Hubei province, central China, was detected quantitatively by flame atomic absorption spectrometric (FAAS) for ultimate purpose of pollution monitoring and risk evaluation. Results indicated that the mean concentrations exceeding their respective background values were observed for all the investigated metals, with the exception of Co (13.08mg kg^{-1}) and Fe (38635.02mg kg^{-1}) in Huanggang road dusts, whose average concentrations were close to the background levels. In comparison with the reference data reported from the selected cities worldwide, the urban road dusts were seriously polluted by heavy metals to diverse degrees. The contour distribution maps implied that obviously higher values zones were found between two different types of urban areas, located to both sides of the coastline of Yangtze River. Multivariate statistical analysis revealed that the enriched heavy metals had emanated from the combined effects of both natural sources and anthropogenic sources. Three pollution indices indicated that the riskiest element mainly comprising Cr, Ni, Cu, and Pb appeared to be the major contributors to the urban environmental pollution. Avoiding continuous damage requires, the riskiest metallic contaminants should be paid preferential attention to.

Keywords Urban road dusts · Heavy metals · Contamination characteristics · Environmental risk assessment

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Introduction

Heavy metals, regarded as one of the most important environment pollutants, become an important threat to both ecological environment and public health for lengthened period, because of their toxicity and carcinogenicity (Chen et al. 2014; Dehghani et al. 2017; Lu et al. 2009; Li et al. 2013). As nationwide awareness regarding health protection is markedly improved, environmental contamination by the elevated heavy metal concentrations in urban areas has also attracted considerable attention worldwide (Lu et al. 2010; Wei et al. 2015; Zhu et al. 2013). More noteworthy, the rapid advances of urbanization and industrialization, as well as the population growth, have been taking place on an unprecedented scale in China over the last two decades (Cheng et al. 2017; Lin et al. 2017; Lu et al. 2014). It is generally believed that heavy metals contained in urban dusts are typically influential tracers for the negative consequences of anthropogenic activities

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concentrated in urbanized areas (Dehghani et al. 2017; Men et al. 2018a, b; Qing et al. 2015; Wei et al. 2015; Zhao and Li 2013).

The urban road dusts, acting as the main source and sink of the various anthropogenic contaminants, especially toxic heavy metals, have been widely acknowledged as one of the most pervasive and omnipresent indicators for characterizing urban environmental quality and human health risk (Lu et al. 2009, 2010; Li et al. 2013). Urban road dusts receive the elevated concentrations of heavy metals originating from natural geochemical processes and various anthropogenic activities in urbanized areas, such as vehicle emissions, industrial emissions, atmospheric deposition, and waste incineration (Dehghani et al. 2017; Lu et al. 2009, 2010, 2014; Zhao and Li 2013). Meanwhile, urban road dusts are not only easily transported and deposited at certain circumstances, but also serve as excellent absorbers and carriers of potentially hazardous metallic contaminants due to their large specific surface area, thereby contributing significantly to the polymetallic pollution prevalent in the ambient environment (Keshavarzi et al. 2015; Zhu et al. 2013). Even worse, the elevated levels of heavy metals ubiquitous in urban road dusts can inevitably deteriorate the urban inhabitants' health through resuspension inhalation, hand-mouth ingestion, and dermal contact (Hu et al. 2011; Qing et al. 2015; Li et al. 2013). Additionally, numerous contemporary studies indicate that the most at-risk population in urban regions for metals and metalloid exposure is very young children who are more susceptible to toxic contaminants poisoning than adults (Chen et al. 2014; Hu et al. 2011; Lu et al. 2014). As a consequence, it is critical to conduct on the multi-metal assessment of environmental pollution risks based on the potential ecological risk index and health risk assessment model with explicit consideration of taking protective measures and effective actions.

Hitherto, the existing researches have reported excessively about heavy metal pollution of urban road dusts in many megacities (Bourliva et al. 2016; Keshavarzi et al. 2015; Wang et al. 2016a, b; Yıldırım and Tokalıoğlu 2016). Accordingly, lager amounts of literatures fully reviewed by our research team indicate that the previous studies on heavy metals present in road/street dusts, mainly including concentration levels, distribution patterns, source apportionment, and contamination assessment, have been widely implemented in the scientific community during the last decades (Men et al. 2018a, b; Tang et al. 2017; Wang et al. 2016a, b; Wei et al. 2015; Li et al. 2013). Nevertheless, rare information for small- and medium-sized cities, in particular, both agricultural and industrial cities, is still available.

In response to that, the comprehensive field investigations were launched to highlight source identification and environmental implications for polymetallic pollution in urban road dusts from two cities of industrial E'zhou and agricultural Huanggang, central China. To date, there is a paucity of background metal data obtained for this region regarding urban road dusts. With this background, the straightforward objectives of the present study were to (i) determine the concentrations of selected chromium (Cr), Nickel (Ni), Cobalt (Co), Copper (Cu), Lead (Pb), Zink (Zn), Manganese (Mn), and Iron (Fe) in two distinct sets of dust samples to highlight the impact strength of transportation emissions and industrial operations; (ii) investigate concentration distribution patterns of heavy metals and explore their possible source identification; and (iii) evaluate environmental pollution risks associated with heavy metals. The results would facilitate the local stakeholders and authorities to take effective measures related to metals-mediated risk.

Materials and methods

Description of the studied area

The present environmental investigation was specially conducted in two typical coastal cities of industrial E'zhou (114° 32–115° 05′ E, 30° 00′ –30° 06′ N) and agricultural Huanggang (114° 25′ –116° 8′ E, 29° 45′ –31° 35′ N), located in the southeastern parts of Hubei province, central China, and correspondingly situated to the southern and northern side of the middle and lower reaches of the China Yangtze River basin (Fig. 1). This urbanized areas experiences typical subtropical continental monsoon climate conditions, and its topography is mainly composed by mountains, hills, and plains. In this region, the prevailing wind direction is south . The two studied cities are well known as two important members of the Wuhan Metropolitan Area. To date, the areas have been subjected to rapid and intense industrialization and urbanization in recent decades. Meanwhile, primary, secondary, and tertiary industries areas have come up continually in and around the urban centers. Under these circumstances, the two studied cities are all suffering undoubtedly from the serious atmospheric particulate contamination, highly influenced by increasing population density, the city's fast expansion, and unregulated anthropogenic activities. In particular, the industrial city mostly receives metal contaminants from numerous metalliferous sources, whereas metal pollution sources of the agricultural city are mainly contributed by abundant agronomic practices and limited municipal activities.

Sampling collection and analytical procedures

Forty-three representative samples in total were periodically collected from the whole urban area of E'zhou City and Huanggang City during the dry season in November 2013. At each selected sampling site, an individual composite sample (specific weight of 0.5 kg), composed of at least five sub-dust samples from impervious surface (road, pavement, and building's roof) by gently sweeping an area of about 1 m ×
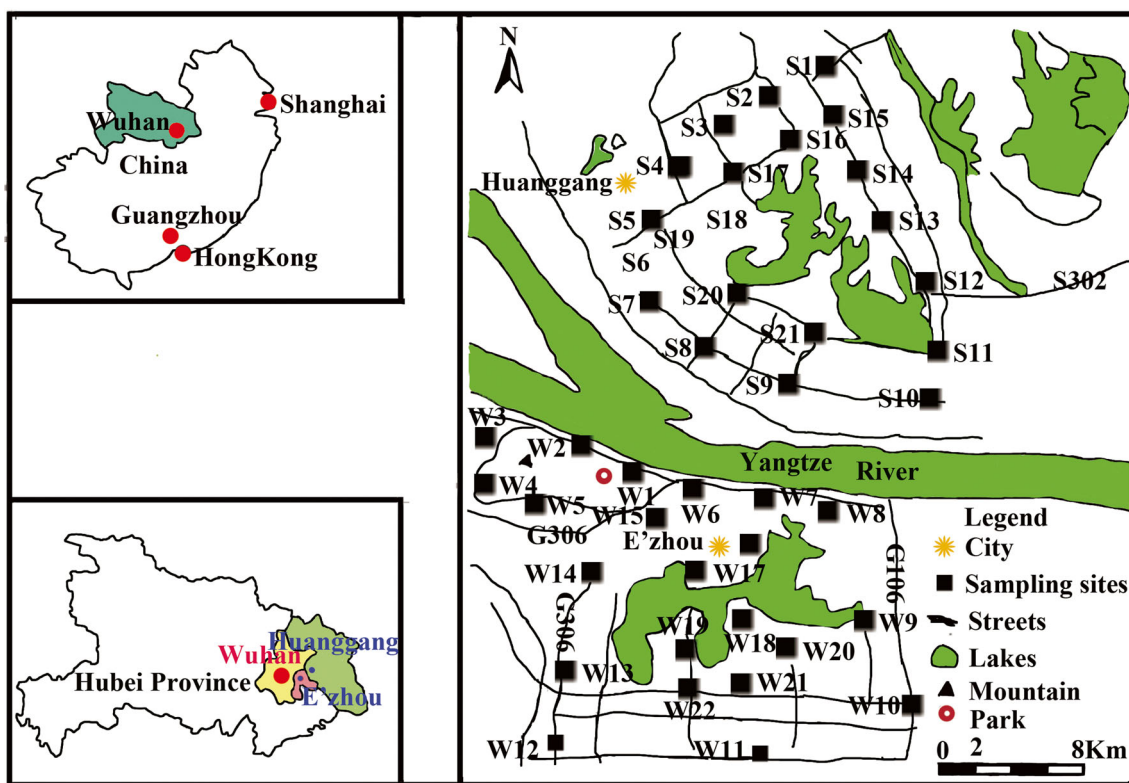
Fig. 1 Geographic location of study area with sampling point locations

1 m using a small brush and a clean plastic dustpan, was collected before daily clean up. Twenty-two dust samples from E'zhou City (sampling sites were coded by S1 to S22) and twenty-one dust samples from Huanggang City (sampling sites were coded by W1 to W21) were collected in this manner to periodically evaluate the accumulation contamination of multiple heavy metals in this study area. The sampling locations, mainly positioned in vicinity of the residential, commercial, and industrial establishments, are depicted in Fig. 1. During the sampling periods, the geographical coordinates for each sampling point were determined using the portable GPS navigator and the surrounding features were correspondingly recorded. All the collected dust samples were stored and labeled in the tightly sealed polyethylene bags and immediately brought back to the laboratory for preparation and analysis.

In the laboratory, all the samples were air-dried naturally at ambient temperature and then passed through a 1.0-mm mesh nylon sieve to remove coarse debris before halving. One half was stored, and the others were finely crushed in mortar-pestle, carefully homogenized , and then gently sieved by a 75-μm nylon mesh for the chemical analysis. After reduction by repeated quartering, the total contents of heavy metals in these resulting samples were further analyzed as outlined below. Of each dust sample, accurately weighed 0.2000 g was transferred into a Teflon crucible on an electrical heating plate and then digested with acid composition of HCl-HNO$_3$-HF-HClO$_4$ in the ratio of 9:3:6:1. After that, the digested solutions were diluted to 50 ml with the addition of 2% (v/v) HNO$_3$ and filtered through using a 0.45-μm membrane filter, and then stored at 4 °C for further analysis. Finally, the total concentrations of typical heavy metals (Cr, Ni, Co, Cu, Pb, Zn, Mn, and Fe) in the residual solutions were quantitatively detected using flame atomic absorption spectrometric (FAAS, Varian AA240, USA).

Quality assurance and quality control (QA/QC)

All the chemicals used in this work were of supra quality and analytical grade reagents and used as received unless otherwise noted. All the container used in analytical procedure was soaked in 10% (v/v) HNO$_3$ overnight, thoroughly flushed with tap water, triply rinsed with deionized water, and then dried in an oven overnight at 40 °C. The procedural blanks, standard samples (GSS-3 and GSS-5, National Research Center for Certified Reference Materials, Beijing, China), and analytical duplicates were conducted in the same batch for the purpose of accuracy and precision of the digestion procedure. Overall, the recovery rates, spiked with standard samples, routinely ranged from 86.5 to 115.2%, indicating the excellent agreement between the measured and the certified values. The analytical precision, expressed as relative standard deviations (RSDs) between duplicate samples, fell within 10% variability for all analyzed elements.

Geochemical factor

Enrichment factor (EF), traditionally employed to quantitatively estimate the pollution levels of anthropogenic heavy metals as beneficial pollutant indicators, was calculated for the targeted element by dividing its ratio to the normalizing element by the same ratio found in the chosen baseline (Buat-Menard and Chesselet 1979; Cheng et al. 2017; Jiang et al. 2017). In general, Fe was served as the conservative element for geochemical normalization since its natural concentration tends to be uniform both regionally and locally (Daskalakis and O'Connor 1995; Jiang et al. 2017; Lin et al. 2017). As originally proposed by Buat-Menard and Chesselet (1979), the EF values can be computed using the following mathematical Eq. (1).

$$EF = \frac{(C_i/C_{Fe})_{Sample}}{(C_i/C_{Fe})_{Background}} \quad (1)$$

where C_{sample} and $C_{background}$ are the concentration of an individual metal in samples and its background values, respectively. Fe_{sample} and $Fe_{background}$ are the determined Fe content in samples and the Fe background value in soils, respectively. On the basis of EF values, contamination categories are classified into five levels as follows (Sutherland 2000): EF < 2, deficiency to minimal enrichment; $2 \leq EF < 5$, moderate enrichment; $5 \leq EF < 20$, significant enrichment; $20 \leq EF < 40$, very high enrichment; and EF > 40, extremely high enrichment.

Environmental risk associated with heavy metal contamination

Potential ecological risk index

The potential ecological risk index method, originally proposed by Hakanson (1980), is widely introduced to not only describe the toxicological effects and accumulative behavior of hazardous multi-metal contaminants in benthic sediments but also comprehensively evaluate their overall potential ecological risk (Hakanson 1980). The index is estimated via the following Eq. (2):

$$RI = \sum E_i^r = \sum T_i^r \times C_i^r = \sum T_i^r \times \frac{C_s^i}{C_b^i} \quad (2)$$

where RI stands for the comprehensive ecological risk index of all risk factors for multiple elements; E_i^r is the monomial potential ecological risk factor of element i; C_i^r is the calculated contamination factor of element i; C_s^i is element i's determined concentration in samples; and C_b^i is element i's reference value in soils. For this study, preindustrial threshold values for heavy metals in background soil of Hubei

Province were adopted as the reference values (CNEMC 1990). T_i^r corresponds to the toxic response factor of individual elements and demonstrated this value for Cr, Ni, Co, Cu, Pb, Zn, and Mn to be 2, 5, 5, 5, 5, 1, and 1, respectively (Hakanson 1980). According to results of Hakanson, the following five categories are used to describe risk levels: $E_i^r < 40$, low risk; $40 \leq E_i^r < 80$, moderate risk; $80 < E_i^r \leq 160$, considerable risk; $160 < E_i^r \leq 320$, high risk; and $E_i^r > 320$, very high risk. Meanwhile, the RI values are correspondingly categorized into four grades: RI < 150, low risk; $150 \leq RI < 300$, moderate risk; $30 \leq RI < 600$, considerable risk; and $RI \geq 600$, very high risk.

Human health risk assessment

Human health risk assessment, which estimates the exposure and tendency of hazardous heavy metals in urban road dusts, is commonly applied to quantify both potentially probabilistic carcinogenic and non-carcinogenic risks caused by polymetallic contaminants to vulnerable population groups via three main exposure pathways (ingestion, dermal contact, and inhalation) (Cheng et al. 2017; Jiang et al. 2017; US EPA 2011; Xu et al. 2017). Human health risk assessment model is recommended by Environmental Protection Agency of United States (US EPA) correlated with four main procedures comprising hazard identification, exposure evaluation, dose-response assessment, and risk characterization (US EPA 1989, 2011).

The average daily doses (ADDs, mg kg^{-1} d^{-1}) for non-carcinogenic metals (Cr, Ni, Co, Cu, Pb, Zn, and Mn), received through the aforementioned exposure pathways, are respectively deliberated by the following Eqs. (3)–(5) (Cheng et al. 2017; US EPA 2011). The lifetime average daily dose (LADD, mg kg^{-1} d^{-1}) for three carcinogenic metals (Cr, Co, and Ni) merely via inhalation exposure pathway is determined in the evaluation of carcinogenic risk and then prescribed by Eq. (6) (Cheng et al. 2017; Lin et al. 2017; US EPA 2011).

$$ADD_{ing} = C \times \frac{lnhR \times EF \times ED}{BW \times AT_{nc}} \times 10^{-6} \quad (3)$$

$$ADD_{dermal} = C \times \frac{SL \times SA \times ABS \times EF \times ED}{BW \times AT_{nc}} \times 10^{-6} \quad (4)$$

$$ADD_{inh} = C \times \frac{lnhR \times EF \times ED}{PEF \times BW \times AT_{nc}} \quad (5)$$

$$LADD = \frac{C \times EF}{PEF \times AT_{ca}}$$
$$\times \left(\frac{lnhR_{child} \times ED_{child}}{BW_{child}} + \frac{lnhR_{adult} \times ED_{adult}}{BW_{adult}} \right) \quad (6)$$

In the case of non-carcinogenic risk assessment, hazard quotients (HQs) are defined as the ratio of each exposure daily

dose (ADD$_{ing}$, ADD$_{derm}$, and ADD$_{inh}$) to the corresponding reference dose (RfD), as described in Eq. (7) (Cheng et al. 2017; Jiang et al. 2017). HQ$_s$ across different exposure pathways to each metal are added and yield an overall estimated value for hazard indexes (HI$_s$) to humans. The total non-carcinogenic risks (HI$_n$) for multiple heavy metals were calculated by addition of the HI$_s$, as expressed in Eq. (8) (Xu et al. 2017). For carcinogenic risk assessment, carcinogenic risk (CR$_s$), the occurrence probability of any type of carcinogenic hazards over lifetime exposure time period for an individuals under specific scenarios, from inhalation exposure pathways for each metal is evaluated by multiplying LADD with the corresponding carcinogenic slope factor (SF, kg d mg^{-1}), as characterized in Eq. 9 (Cheng et al. 2017; Jiang et al. 2017). Similarly, the total carcinogenic risk (CR$_n$) for the studied metals is defined as the sum of individual CR$_s$ across all heavy metals, whose calculation formula is listed in Eq. 9 (Lin et al. 2017; Xu et al. 2017).

$$HI = \sum HQ_s = \sum \left(HQ_{ing} + HQ_{derm} + HQ_{inh} \right)$$
$$= \sum \left(\frac{ADD_{ing}}{RfD_{ing}} + \frac{ADD_{derm}}{RfD_{derm}} + \frac{ADD_{inh}}{RfD_{inh}} \right) \quad (7)$$

Hazard index $(HI)_n = \sum$Hazard index $(HI)_s$
$$= \sum\sum \text{hazard quotient}(HQ_S) \quad (8)$$

Cancer risk $(CR)_n = \sum$Cancer risk $(CR)_s$
$$= \sum LADD_{inh} \times SF_{inh} \quad (9)$$

where ADD$_{ing}$, ADD$_{derm}$, and AAD$_{ing}$ are the daily amount of exposure to metals through ingestion, inhalation, and dermal contact, respectively; C (mg kg^{-1}) is upper limit of the 95% confidence interval for the average (95 %UCL), which is regarded as the conservative estimate of the "reasonable maximum exposure" (US EPA 1992). The threshold values of acceptable or tolerable risk for regulatory purposes are adapted from US EPA (2001). According to Chinese practical situation, the definitions and values of all parameters involved in the above models are preferentially supported by the Supplementary Information (MEEPRC 2014; Xu et al. 2017). Additionally, the RfD and SF values of the investigated eight metals are available in accordance with the data previously reported in these literatures (US Department of Energy 2004; Xu et al. 2017).

Statistical treatment methods of data

The experimental data were processed using Microsoft Excel 2013. Box and whisker plots of element EF values in dusts samples were plotted using Origin Version 2017. Moreover, the statistical analysis of the obtained data, as well as multivariate analysis techniques, was conducted using IBM SPSS

22.0. The interpolation mapping and spatial pattern of heavy metal concentrations were created by Golden Software Surfer 9.0 using the ordinary kriging technique in much effort to distinguish and prioritize potentially contaminated prone areas.

Results and discussion

Total metal concentrations in urban road dusts

The summary statistics results for all investigated heavy metal concentrations in urban road dusts as well as soil background values are presented in Table 1.

Considering this table, the average concentrations of studied heavy metals were markedly higher than those in background soils of Hubei Province, Central China (CNEMC 1990). Similarly, the mean heavy metal concentrations in Huanggang road dusts were comparatively greater than their background levels recorded for Hubei Province, except for Co and Fe (background value 15.40 mg kg^{-1} for Co and 39,100.00mg kg^{-1} for Fe). The significantly broad ranges of multiple element concentrations exhibited anomalous variability with respect to different sampling sites across the whole study area. In addition, the CV values implied that urban road dusts were contaminated by these metals to diverse degrees and further reflected their heterogeneity in distribution. On the basis of mean contents, E'zhou road dusts had accumulated approximately 4.00, 3.13, 3.23, 5.35, 3.44, 2.02, 2.23, and 1.82 times greater amounts of Cr, Ni, Co, Cu, Pb, Zn, Mn, and Fe in sequences than the corresponding mean concentration levels of those metals in the dust samples recorded for Huanggang City. This may confirm the finding that the higher element concentrations had tended to be obviously accumulated in E'zhou road dusts than Huanggang road dusts. It is reasonably inferred that industrial city would have the greater possibility of being highly influenced by two anthropogenic sources, closely tied to frequently vehicular emissions and progressive industrial activities.

A comparison of heavy metal concentrations in this study with those of other cities around the world

A quantitative comparison of the mean concentrations of heavy metals measured in urban road dusts from the studied area with those reported in China and other countries is shown in Table 2.

As summarized in Table 2, the average Ni and Fe concentrations in the present study were exceptionally higher, while the rest metal concentrations in our samples were in the middle range, compared with those reported in other more industrialized and developed cities. The second highest mean concentration of Mn in Huanggan road dusts was lower than the

Table 1 Descriptive statistics of heavy metal concentrations in urban road dusts from study areas and their background values

Cities	Elements	Cr	Ni	Co	Cu	Pb	Zn	Mn	Fe
E'zhou	Mean (mg kg^{-1})	351.54	408.56	42.26	338.27	186.34	353.98	1656.05	70,205.70
	Median (mg kg^{-1})	218.11	250.72	35.67	214.28	167.01	347.95	1614.15	64,339.04
	Minimum (mg kg^{-1})	52.32	85.69	13.60	66.07	40.87	234.51	908.58	26,894.03
	Maximum (mg kg^{-1})	1783.13	2444.58	85.38	2489.24	443.93	481.49	2467.98	154,040.46
	Standard deviation	416.06	500.39	19.60	489.11	93.68	61.73	444.37	35,425.58
	95% UCL[a]	536.01	630.42	50.95	555.13	227.87	381.35	1853.07	85,912.52
	Variation coefficient (%)[b]	118.35	122.48	46.39	144.59	50.27	17.44	26.83	50.46
	Skewness	2.76	3.60	0.54	4.42	1.55	0.12	0.31	1.03
	Kurtosis	7.48	14.12	− 0.24	20.25	3.13	− 0.05	− 0.61	0.51
Huanggang	Mean	87.95	130.42	13.08	63.18	54.24	175.54	742.98	38,635.02
	Median	88.22	136.34	12.93	60.92	51.04	141.43	768.24	37,814.92
	Minimum	37.72	75.45	6.49	32.97	5.72	102.95	560.70	24,565.38
	Maximum	128.83	161.03	20.20	134.53	102.40	378.01	874.88	62,645.41
	Standard deviation	25.46	27.15	3.00	24.08	21.42	68.16	81.46	8914.97
	95% UCL[a]	99.54	142.78	14.44	74.15	63.99	206.56	780.05	42,693.07
	Variation coefficient (%)[b]	28.95	20.82	22.94	38.12	39.49	38.83	10.96	23.07
	Skewness	− 0.35	− 0.63	0.28	1.41	0.32	1.52	− 0.50	0.99
	Kurtosis	− 0.45	− 0.87	1.69	2.51	1.16	2.57	− 0.35	1.45
The background values[c]		86.00	37.30	15.40	30.70	26.70	83.60	712.00	39,100.00

[a] Upper limit of the 95% confidence interval for the mean

[b] The coefficient of variation is calculated using the equation: CV = (Standard deviation/Mean) × 100%

[c] Background values for soils in Hubei Province from the China National Environmental Monitoring Center (CNEMC 1990)

concentration detected in Baoji, Turin, and Tehran, whereas being higher than the concentration measured in the other selected cities around the world. However, in E'zhou road dusts, Mn concentration was far higher than that reported for other metropolitan cities in China and other countries. By integrating data, there was the main conclusion that metallic pollution in both E'zhou and Huanggang road dusts should not be ignored.

Heavy metal enrichment and their contamination levels

The calculated results of EF for selected heavy metals in E'zhou and Huanggang road dusts were demonstrated in Fig. 2a, b.

As shown in Fig. 2, these magnitude results were numerically consistent with the wide ranges of metal concentrations in urban road dusts. The mean EF values of the analyzed elements except for Co and Mn in E'zhou road dusts were higher than 2 (Fig. 2a), which appeared to be a consequence of heavily anthropogenic loading coupled with extrinsic imports of heavy metals. In addition, there were Ni, Cu, and Pb with EF values of 5.48, 6.26, and 4.36, respectively; indicating that E'zhou road dusts was obviously loaded with them. However, in Huanggang road dusts, some EF values obtained

for Cr (1.07), Co (0.89), and Mn (1.09) were lower than 2 (Fig. 2b), suggesting that these non-anthropogenic elements were predominantly contributed by geological weathering sources. Besides this, the EF values obtained for the remaining metals were between 2 and 5, suggesting that their sources were anthropogenic. It is interestingly noted that the EF values for these tested metals from E'zhou City were far greater than those from Huanggang City at each sampling site. This firmly confirms that the pronouncedly higher EF values could be seemingly attributed to the intensive urbanization and industrialization in diverse urban zones.

Implications on risk assessment

Potential ecological risk assessment

Descriptive statistics of the E_i^r values of individual elements and RI values of multiple elements are presented in Table 3. The relative percentage distributions of individual heavy metals at each sampling sites are represented as E_i^r contributing to RI and the results were graphically presented in Fig. 3.

As clearly seen from Table 3, E_i^r and RI values exhibited considerable fluctuations among all the investigated sites from the two studied cities. Among the evaluated elements, the potential ecological risk was strongly associated with Ni,

Table 2 Average heavy metal concentrations found in the study area and other selected cities from the references

Cities	Cr (mg kg^{-1})	Ni (mg kg^{-1})	Co (mg kg^{-1})	Cu (mg kg^{-1})	Pb (mg kg^{-1})	Zn (mg kg^{-1})	Mn (mg kg^{-1})	Fe (mg kg^{-1})	References
E'zhou (China)	351.54	408.56	42.26	338.27	186.34	353.98	1656.05	70,205.70	This study
Huanggan (China)	87.95	130.42	13.08	63.18	54.24	175.54	742.98	38,635.02	This study
Baoji (China)	126.7	48.8	15.9	123.2	433.2	715.3	804.2	NA	(Lu et al. 2010)
Nanjing (China)	126	55.9	10.7	123	103	394	646	34,200	(Hu et al. 2011)
Xining (China)	576.8	22.6	50.0	40.8	53.9	108.9	408.7	NA	(Zhao et al. 2015)
Xi'an (China)	145.00	30.80	30.90	54.70	124.50	286.60	510.50	NA	(Pan et al. 2017)
Beijing (China)	92.10	32.47	NA	83.12	60.88	280.65	553.73	29,774.62	(Men et al. 2018a)
Shiraz (Iran)	67.16	77.52	NA	136.34	115.71	403.46	438.5	20,254.6	(Keshavarzi et al. 2015)
Thessaloniki (Greece)	104.90	89.43	NA	662.30	209.00	452.80	336.40	21,300	(Bourliva et al. 2016)
Turin (Italy)	405	254	NA	128	319	286	857	27,513	(Padoan et al. 2017)
Tehran (Iran)	76.5	57.7	NA	275	213	666	864	NA	(Dehghani et al. 2017)

NA indicates data is not available

Cu, and Pb in all studied sites, exhibiting relatively high pollution levels and assessed as the predominant contributors of the overall ecological risk. More explicitly, the highest E_i^r values in sites E7 from E'zhou City (405.41) and site H7 from Huanggang City (21.91) pointed to relatively high ecological risks for Cu, accounting for 10.19 to 69.76% and 12.20 to 33.50% of RI (Fig. 3), respectively. Inversely, the E_i^r values obtained for Cr, Co, Zn, and Mn posed extremely low ecological risks at all studied sites from the two studied cities with contribution to RI ranging from 0.51 to 12.39% (Fig. 3). Therefore, adequate attention should be paid to the extensive polymetallic contamination, especially Ni, Cu, and Pb. Additionally, the E_i^r values of Co, Zn, and Mn in all studied dust samples and the E_i^r values of the Cr in 95.45%, Pb in 81.82%, Ni in 68.18%, and Cu in 59.09% of dust samples were within 40, respectively, indicating that the concentration

of Co, Zn, and Mn observed in E'zhou City might be occasionally pose the detrimental effects to the living organisms. On the other hand, the calculated values of E_i^r for all studied heavy metals in all dust samples from Huanggang City were below 40 and characterized by a low-risk level, indicating that the urban dusts were possibly unpolluted by these heavy metals. Furthermore, the RI values of each sampling site in the two studied cities varied from 40.27 to 581.14 (mean 173.21) and from 28.38 to 65.41 (mean 47.36) with exceeding 150 detected in 36.36% and 0% of their correspondingly studied locations, respectively.

Taken together, the data comparison of the quantitative evaluation of potential ecological risk further confirmed that the contamination levels of dust samples from E'zhou City with respect to these heavy metal enrichment were more considerably severe than those from Huanggang City. The
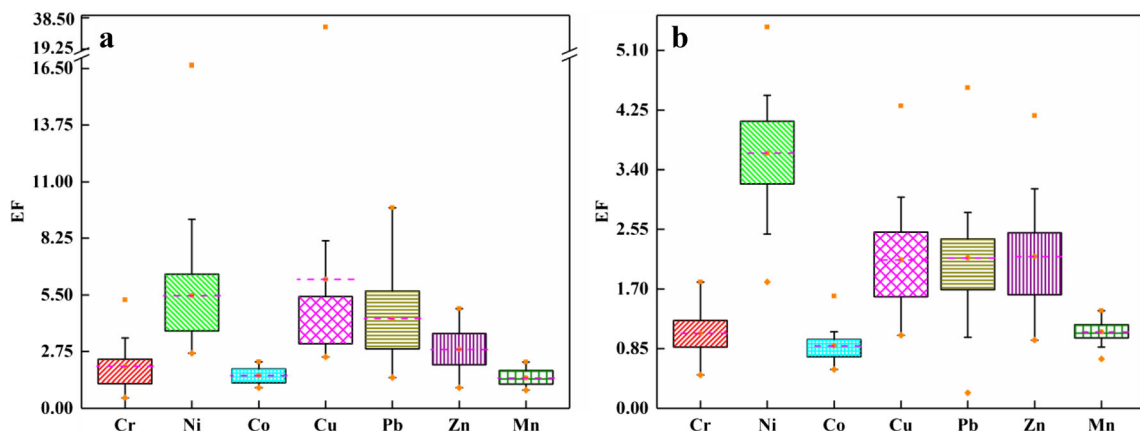

Fig. 2 Boxplot diagrams of the enrichment factor (EF) value for selected heavy metals in urban road dusts from E'zhou (**a**) and Huanggang City (**b**). The red horizontal line in the middle of box represented the mean values; the circles in the bottom and top of box plots represented the minimum and maximum values, respectively; while the ends of the box represented the 25th and 75th percentile values, respectively

Table 3 The calculated E_i^r and RI values, and percent of dust samples included in E_i^r (< 40) and RI (< 150) levels

Cities	Elements	E_i^r (individual metal)							RI (multi-metal)
		Cr	Ni	Co	Cu	Pb	Zn	Mn	
E'zhou	Mean	8.18	54.77	13.72	55.09	34.89	4.23	2.33	173.21
	Minimum	1.22	11.49	4.42	10.76	7.65	2.81	1.28	40.27
	Maximum	41.47	327.69	27.72	405.41	83.13	5.76	3.47	581.14
E_i^r values < 40 or RI < 150 (%)		95.45	68.18	100	59.09	81.82	100	100	68.18
Huanggang	Mean	2.05	17.48	4.25	10.29	10.16	2.1	1.04	47.36
	Minimum	0.88	10.11	2.11	5.37	1.07	1.23	0.79	28.38
	Maximum	3	21.59	6.56	21.91	19.18	4.52	1.23	65.41
E_i^r values < 40 or RI < 150 (%)		100	100	100	100	100	100	100	100

underlying reason for the interesting findings was mainly that as the aforementioned results mentioned above, the concentrations of heavy metals in E'zhou road dusts were appreciably higher as compared to those in Huanggang road dusts.

Human health risk assessment

The evaluation results obtained for non-carcinogenic and carcinogenic health risks involving children and adults separately exposed to heavy metals are listed in Table 4. And the percentage contribution of individual elements to cumulative health risks is illustrated in Fig. 4.

As shown in Table 4, non-carcinogenic risk for children and adults of the reported heavy metals determined in E'zhou City were similarly ordered as $HI_{Cr} > HI_{Pb} > HI_{Mn} > HI_{Ni} > HI_{Cu} > HI_{Co} > HI_{Zn}$, respectively, while HI values recorded in Huanggang City, the overall trend generally followed the sequence of $HI_{Cr} > HI_{Mn} > HI_{Pb} > HI_{Ni} > HI_{Cu} > HI_{Co} > HI_{Zn}$, respectively. As given all the investigated metals

in consideration (Table 4 and Fig. 4), Cr posed the greatest non-carcinogenic risk to local residents, followed by Mn and Pb. Consequently, Cr, Mn, and Pb were identified as the most significant pollutants. Among them, Cr in E'zhou and Huanggang road dusts principally contributed 54.59% and 42.06% for children and 55.86% and 44.26% for adults to HI_n, respectively. The HI values of all the studied metals were lower than 1, ranging from 0.01 to 0.80 for children and 0 to 0.45 for adults, respectively. However, Cr should be of particular concern, and its HI value of 2.44 for adults and 1.43 for children residing in E'zhou City were far more than the safety threshold value (HI = 1), manifested that Cr might pose significant non-carcinogenic health hazard to sensitive population.

By comparison of HI values for children and adults, it could be inferred that children tended to have the relatively higher probability of developing non-carcinogenic risks than adults did, further indicating that they were rather low tolerant to environmental contaminants. This was mostly attributed to
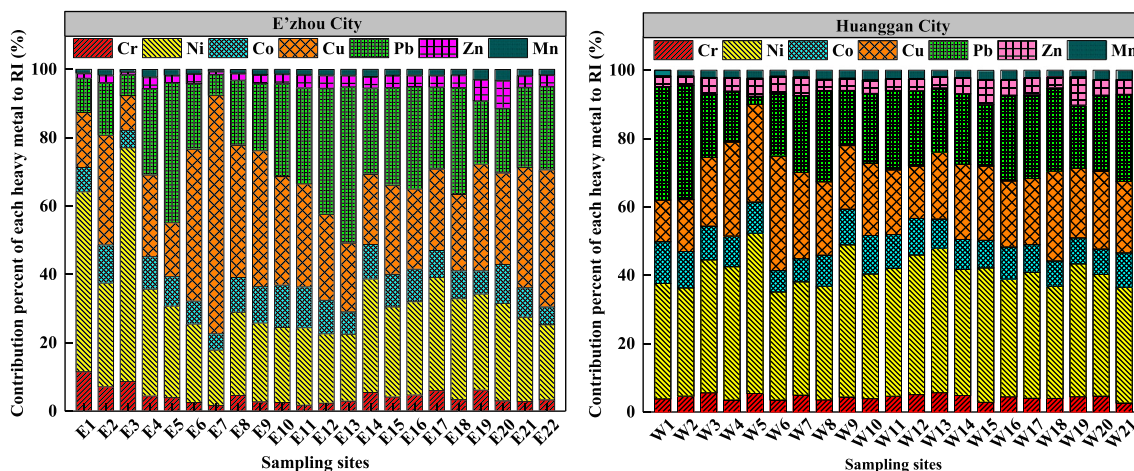

Fig. 3 The relative percentage contribution of E_i^r for each metal to RI for multi-metals at each sampling sites

Table 4 Carcinogenic and non-carcinogenic risks associated with each heavy metal for children and adults

Cities	Population groups	Carcinogenic risk				Non-carcinogenic risk							
		CR_{Cr}	CR_{Ni}	CR_{Co}	CR_n	HI_{Cr}	HI_{Ni}	HI_{Co}	HI_{Cu}	HI_{Pb}	HI_{Zn}	HI_{Mn}	HI_n
E'zhou	Children	2.37×10^{-6}	5.58×10^{-8}	5.26×10^{-8}	2.48×10^{-6}	2.44	0.38	0.04	0.17	0.80	0.02	0.63	4.47
	Adults					1.43	0.22	0.02	0.09	0.45	0.01	0.35	2.56
Huanggang	Children	4.40×10^{-7}	1.26×10^{-8}	1.49×10^{-8}	4.68×10^{-7}	0.45	0.09	0.01	0.02	0.22	0.01	0.27	1.07
	Adults					0.27	0.05	0.01	0.01	0.13	0	0.15	0.61

their pica behavior and hand or finger sucking (Jiang et al. 2017; Li and Ji 2017; Xu et al. 2017). Our observations coincided with the results obtained from other studies (Jiang et al. 2017; Tepanosyan et al. 2017).

Similarly, CR values of the inhalation exposure pathways of the carcinogenic metals derived from E'zhou City decreased in the following order of $CR_{Cr} > CR_{Co} > CR_{Ni}$, whereas, CR values evaluated in Huanggang City appeared as $CR_{Cr} > CR_{Ni} > CR_{Co}$, indicating that during an entire lifetime of sensitive population, the carcinogenic risk mostly resulting from exposure to Cr in the whole study area (Table 4 and Fig. 4). Among the analyzed metals, the CR_n values recorded in the two studied cities were dominated by Cr, accounting for approximately 95.63% and 94.11% of CR_n, respectively. It could be also seen from Fig. 4 that the contribution ratios of CR_s values to CR_n obtained for E'zhou City were highly similar to those for Huanggang City. And the CR value for Cr, detected in E'zhou City ($CR_{Cr} = 2.37 \times 10^{-6}$) stood in the acceptable range of 10^{-6} to 10^{-4}, while in Huanggang City, the individual CR values for Cr, Ni, and Co were all lower than 10^{-6}. Therefore, the carcinogenic risk posed by them was negligible. Interestingly, cumulative carcinogenic risk characterized by CR_n (2.48×10^{-6} in E'zhou City, 4.68×10^{-7} in Huanggang City; Table 4) was found to be within the range of acceptable level (10^{-6} to 10^{-4}), indicating that the carcinogenic elements posed

negligible carcinogenic effects to two groups of population.

In view of their comparatively higher contributing ratios to the health risks, of considerable concern should be Cr and somewhat Pb and Mn. As indicated in the results, it could be also concluded that the non-carcinogenic and carcinogenic health risks in metal-contaminated samples from E'zhou City were obviously higher than those from Huanggang City, further demonstrating that the metallic contamination levels were progressively exacerbated by rapid social-economic development and increasing residents in E'zhou City.

Spatial distribution of heavy metal concentrations in urban road dusts

The distinct variations of element contents in E'zhou and Huanggang road dusts are depicted in Fig. 5a, b.

As observed in Fig. 5a, the distribution features of Cr and Ni displayed remarkably similar patterns, with gradually increased concentrations towards the northern section of E'zhou City along the coastline of Yangtze River in China. Regardless of the concentrations of Co, Mn, and Fe were remarkably different in some of the same areas, an analogous tendency over a large region scale was appreciably observed for Co, Mn, and Fe contents. Those metals were mainly controlled
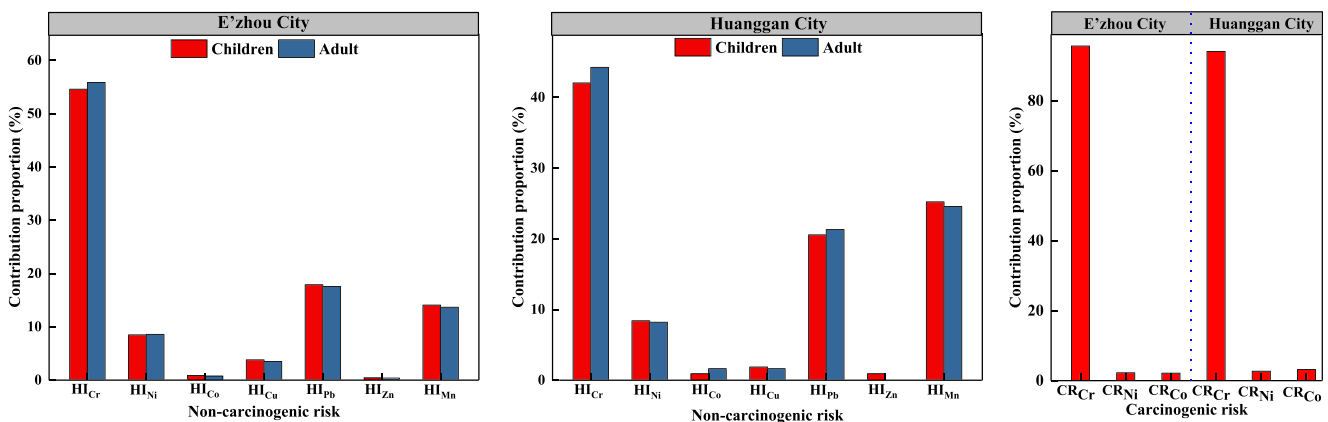

Fig. 4 Percentage contribution of individual heavy metals to HI_n and CR_n in both E'zhou and Huanggang City
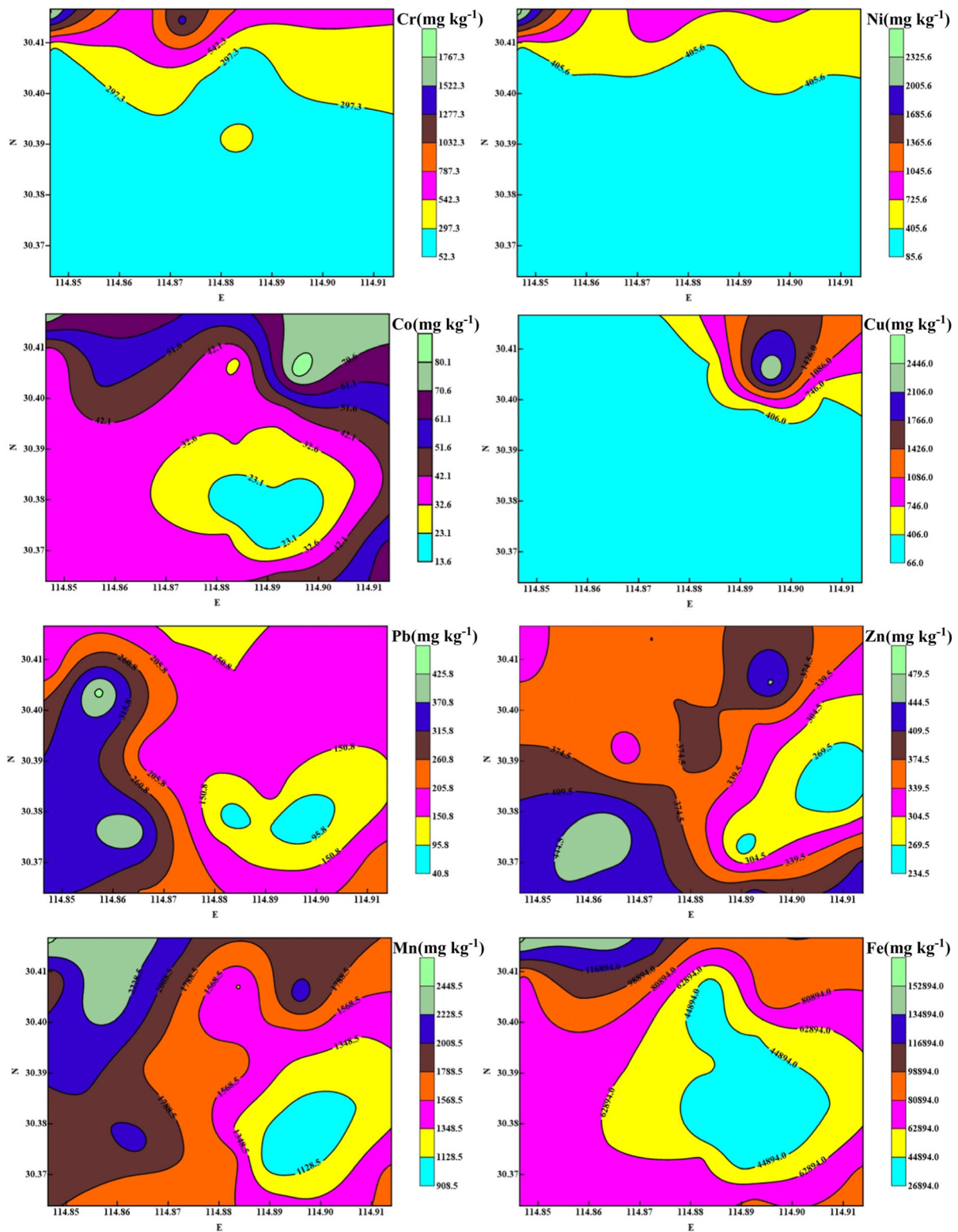
Fig. 5 a The contour maps of selected elements (Cr, Ni, Co, Cu, Pb, Zn, Mn, and Fe) in E'zhou road dusts. b The contour maps of selected elements (Cr, Ni, Co, Cu, Pb, Zn, Mn, and Fe) in Huanggang road dusts

by anthropogenic sources, since their concentrations were far higher their respective soil background values. Moreover, the

preferential distribution for Pb and Zn were also found to be fairly similar and exhibited high contents in the northwestern
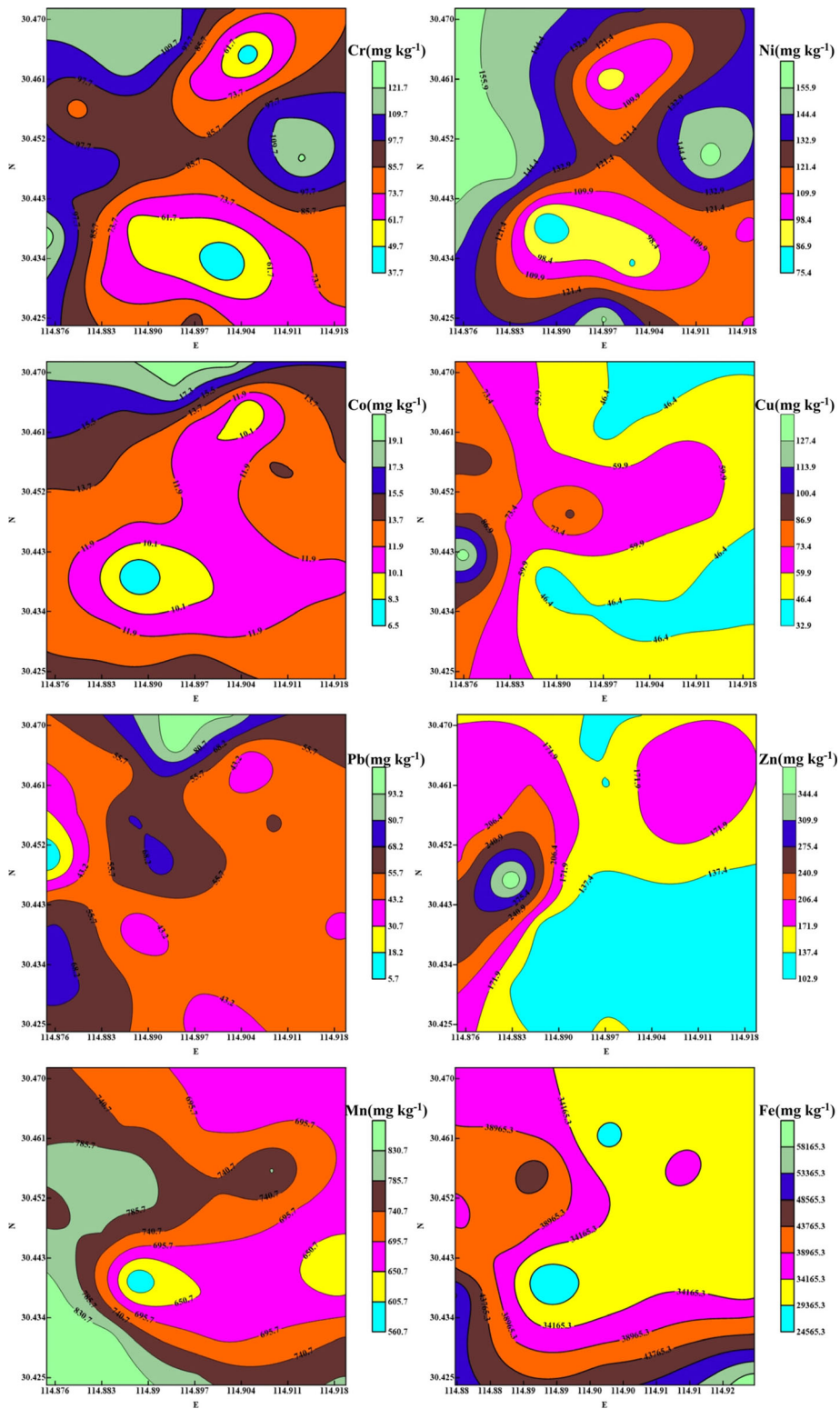
Fig. 5 (continued)

industrial zones and bus stations, suggesting these metals were strongly associated with metallic discharges and vehicles

emissions. However, unlike the remaining metals, the contour map of Cu was quite different and its higher mental

concentrations were preferentially partitioned in the northeastern distal region, compared with those at other most areas.

As depicted in Fig. 5b, these heavy metal enrichment were observable in the northern-western-southern parts of Huanggang City. And their concentrations generally increased from the southeast areas to the other surrounding areas, which was reflective of steel industry emissions from the northwest of E'zhou City along the prevailing winds. The distribution patterns of Cr and Ni are highly similar, and tended to increase from southeast towards northwest of Huanggang City. In addition, the concentrations of Cu, Zn, Mn, and Fe shared similar spatial patterns, with their concentrations decreasing apparently from the eastern towards the western parts. With regard to Co and Pb, the higher value zones were found in the southwestern and northern parts of Huanggang City. These finding suggested that heavy metal contamination in Huanggang road dusts could be possibly subjected to severe major agricultural and partially industrial waste disposal, vehicle emissions, as well as geological weathering.

The distribution patterns of heavy metals in E'zhou and Huanggan road dusts did not well match. It should be noted that obviously higher element concentrations appeared between northwestern E'zhou City and southwestern Huanggang City, located to both sides of the coastline of Yangtze River. The continuous emission sources of steel-smelting activities from E'zhou City were highly responsible for the mentioned heavily polluted zones, with downwind direction considered. The foundation of such deductions came from the results of previous studies (Men et al. 2018a; Ravankhah et al. 2017; Xu et al. 2017). The distribution patterns of these heavy metals could be closely related to various factors, such as combined anthropogenic sources, meteorological characteristics, and geological conditions. As followed from the state above, these inferences further reminded us that urban environmental quality heavily governed by involved natural and extrinsic factors should require serious attention.

Multivariate statistical analysis for pollution sources identification

Pearson's correlation coefficients for heavy metals are presented in Table 5. The Kaiser-Meyer-Olkin (KMO) measure of sampling adequacy was > 0.5, and the significance level of the Barlett's test of sphericity was < 0.01, suggesting that the raw data obtained from the two studied cities was suitable for the multivariate statistical analysis. The results of PCA for heavy metals are listed in Table 5, and the variation diagrams in rotated space are depicted Fig. 6. The resultant dendrograms of HCA analysis are illustrated in Fig. 7.

Geochemical association analysis

The results of Person correlation analysis showed that a statistically positive correlation was found between a group of elements with each other, comprising Cr, Ni, Co, Mn, and Fe in E'zhou road dusts; similarly, the strong correlations were also observed among Pb, Zn, and Mn. In contrast, in Huanggan road dusts, Cr, Ni, Cu, Zn, and Mn concentrations were significantly correlated with each other. Co and Cr, Ni and Pb also showed significant correlation. These findings indicated that similarities in their respective origins.

The correlation matrix also indicated that, the concentration of Cu was correlated negatively with any of the heavy metals in E'zhou road dusts, except for Co, whereas Fe was uncorrelated with the other metals in Huanggan road dusts with the exception of Mn. Moreover, the correlations between the other metal pairs varying from − 0.02 to 0.41were not significant. These findings implied that additional independences in their respective sources.

Source apportionment

From results of the PCA (Table 5), two rotated principal components (PCs) were extracted with eigenvalues great than 1, cumulatively explaining 76.144% for E'zhou City and 67.638% for Huanggang City of the total variance, respectively. In E'zhou road dusts, PC1 accounted for 46.075% of total variation and was influenced primarily by Cr (0.957), Ni (0.940), Co (0.742), Mn (0.710), and Fe (0.879), whereas PC2 explained 30.069% of total variation and was grouped Co (0.522), Pb (0.821) and Zn (0.889), and Mn (0.589) together. Similarly, PC1 (39.945% of the total variance) in Huanggan road dusts contains high load of Cr (0.531), Ni (0.728), Cu (0.822), Zn (0.779), Mn (0.828), and Fe (0.633), whereas PC2 (27.692% of the total variance) was heavily loaded with Cr (0.643), Ni (0.522), Co (0.915), and Pb (0.806). Two element pairs (Co-Mn in E'zhou road dusts and Cr-Ni in Huanggan road dusts) in the two PCs could demonstrate that these elements originated from a mixture of automotive emissions and atmospheric depositions (Lu et al. 2010).

One group of elements comprised Cr, Ni, Mn, and Fe in both E'zhou and Huanggan road dusts, whose average concentrations were far higher or comparable to their respective background levels. Besides this, the integrated results of pollution assessment indicated that, of the four heavy metals, the concentrations of Cr and Ni presented relatively high-potential environmental risk. It is previously reported that Cr and Ni were more related to paints applied in automobile coatings (Trujillogonzález et al. 2016; Bourliva et al. 2016). Therefore, these loading associations were highly characteristic of geochemical and industrial inputs (Lu et al. 2010; Qing et al. 2015).

Table 5 Matrix of correlation and principal component analysis for metal concentrations in E'zhou and Huanggan road dusts

Cities	Elements	Component matrix		Rotated component matrix		Correlation matrix							
		PC1	PC2	PC1	PC2	Cr	Ni	Co	Cu	Pb	Zn	Mn	Fe
E'zhou	Cr	**0.788**	− 0.553	**0.957**	− 0.100	1	0.947**	0.586**	0.09	− 0.05	− 0.03	0.631**	0.765**
	Ni	**0.794**	− 0.507	**0.940**	− 0.057		1	0.603**	0.18	− 0.02	0.02	0.607**	0.727**
	Co	**0.902**	0.095	**0.742**	**0.522**			1	0.587**	0.31	0.41	0.737**	0.849**
	Cu	0.427	0.275	0.239	0.448				1	0.03	0.36	0.28	0.25
	Pb	0.375	**0.731**	− 0.029	**0.821**					1	0.616**	0.541**	0.24
	Zn	0.440	**0.773**	0.008	**0.889**						1	0.485*	0.27
	Mn	**0.907**	0.169	**0.710**	**0.589**							1	0.844**
	Fe	**0.927**	− 0.142	**0.879**	0.328								1
	Eigen values	4.263	1.828	3.686	2.406								
	Cumulative percent (%)	53.290	76.144	46.075	76.144								
Huanggang	Cr	**0.790**	0.268	**0.531**	**0.643**	1	0.817**	0.539*	0.477*	0.34	0.438*	0.40	0.17
	Ni	**0.894**	0.061	**0.728**	**0.522**		1	0.580**	0.632**	0.17	0.561**	0.577**	0.31
	Co	**0.568**	**0.725**	0.102	**0.915**			1	0.10	0.617**	0.00	0.29	0.21
	Cu	**0.748**	− 0.352	**0.822**	0.094				1	0.11	0.578**	0.643**	0.31
	Pb	0.367	**0.721**	− 0.067	**0.806**					1	0.03	0.18	0.04
	Zn	**0.653**	− 0.426	**0.779**	− 0.019						1	0.471*	0.30
	Mn	**0.798**	− 0.284	**0.828**	0.178							1	0.709**
	Fe	**0.549**	− 0.316	**0.633**	0.020								1
	Eigen values	3.804	1.607	3.196	2.215								
	Cumulative percent (%)	47.554	67.638	39.945	67.638								

For this study, by bold are marked values indicated that the factor loadings of PCs above 0.50 were considered for explanation

**Correlation is significant at $P < 0.01$ (two-tailed)

*Correlation is significant at $P < 0.05$ (two-tailed)

A second group of elements consisted of Co and Pb. As widely known, Pb is mainly linked to the secondary industrial combustion processes and vehicle exhausts emissions (Bi et al. 2018; Trujillogonzález et al. 2016). Meanwhile, Co is used as the common additives in the production of carbide alloys, wall paint, and building materials (Tang et al. 2017; Wang et al. 2016a, b). Furthermore, the EF value for Co was between 0.89 and 2.24 in the whole study area, meaning
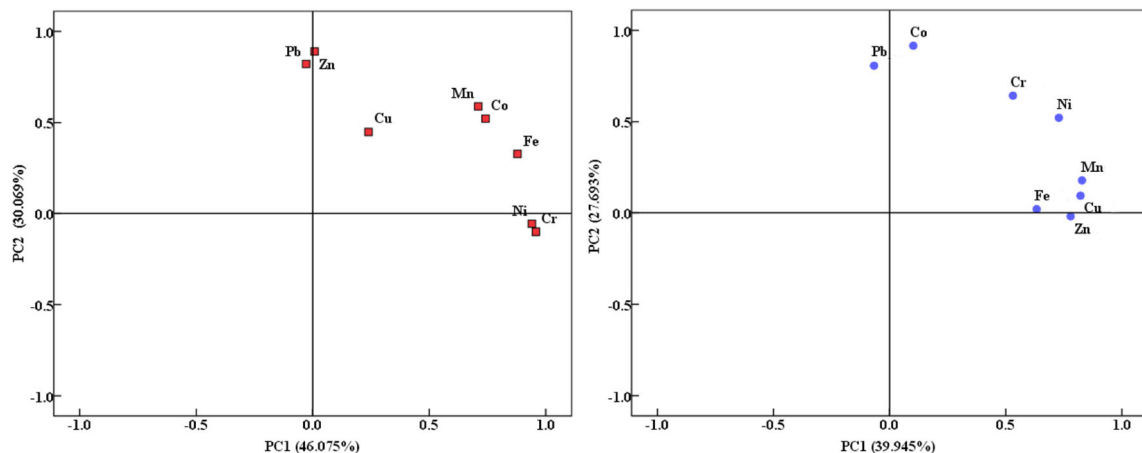

Fig. 6 Loading plots of the PCA for heavy metals in E'zhou (left side of the figure) and Huanggan (right side of the figure) road dusts

extremely low contamination levels. Therefore, this factor could be most probably ascribed to multiple natural and extrinsic inputs.

A third group included Cu, Pb, and Zn. These contaminative elements have been demonstrated to be strongly associated with multiple anthropogenic sources, such as mechanical abrasion of automotive parts, inordinate agrochemicals applications, and metallurgic dusts (Dehghani et al. 2017; Men et al. 2018b; Lü et al. 2010, 2018; Qing et al. 2015; Yıldırım and Tokalıoğlu 2016). Cu, Pb, and Zn were considered to be the typically anthropogenic elements because of their great differentiation in comparison with their respective background values.

According to our present field investigations, the E'zhou road dust samples were heavily contaminated seemingly by the production activities of E'cheng ironworks, E'cheng electric steel mill, and Hubei Steel cold-rolled sheet plant, under the jurisdiction of E'cheng Iron and Steel company. On the other hand, appreciably higher element enrichment were strongly related to long-distance aerial dissemination of soil particles and windblown dusts driven from smelting and processing discharges along the prevailing wind direction. Consequently, large volumes of emissions and improper deposits of municipal wastes were the most likely causes of the elevated metal concentration either in the vicinity of the steel industrial district or the distal parts of the windward areas.

Cluster analysis results

The HCA results given from Fig. 7 deciphered that heavy metals contained in road dusts were categorized into three statistically significant clusters. For E'zhou road dusts, cluster 1 was composed of Cr, Ni, Co, Mn, and Fe; cluster 2 consisted of Pb and Zn; and cluster 3 individually comprised Cu.

However, for Huanggan road dusts, cluster 1 contained Cr, Ni, Cu, and Zn; cluster 2 included Mn and Fe; and cluster 3 comprised Co and Pb. The results from HCA were generally in agreement with those from the discussion above.

Conclusions

In this study, considerably greater concentrations of the reported metals were observable in E'zhou road dusts than those in Huanggang road dusts, providing an information record of the regional anthropogenic impacts.

High concentration variations of heavy metals indicated that their sources were affected by multiple extrinsic factors. Great differences in the distribution trends of heavy metal concentrations could be mostly due to the comprehensive influences of geographical locations, climatic characteristics, and locate point sources in the study area. Multivariate statistical analysis revealed that multiple natural sources and extrinsic inputs exerted profound influences on heavy metal pollution in the urban environment. From an environmental implication perspective, three pollution indices were synchronously introduced. According to the EF values, among the heavy metals considered, Ni, Cu, Pb, and Zn were the most enriched elements in urban road dusts. The E_i^r and RI values of studied metals showed relatively high ecological risk for Cu, Ni, and Pb enriched in both E'zhou and Huanggang road dusts. Additionally, the calculated health risk values suggesting that there was no severe health effects for urban livelihood because the CR and HI values were lower than the threshold safe level. However, HI posed by Cr for vulnerable populations residing in E'zhou City exceeded the maximum acceptable level (1). All comparative data exhibited the finding that the evaluation values reported in E'zhou City were significantly higher than
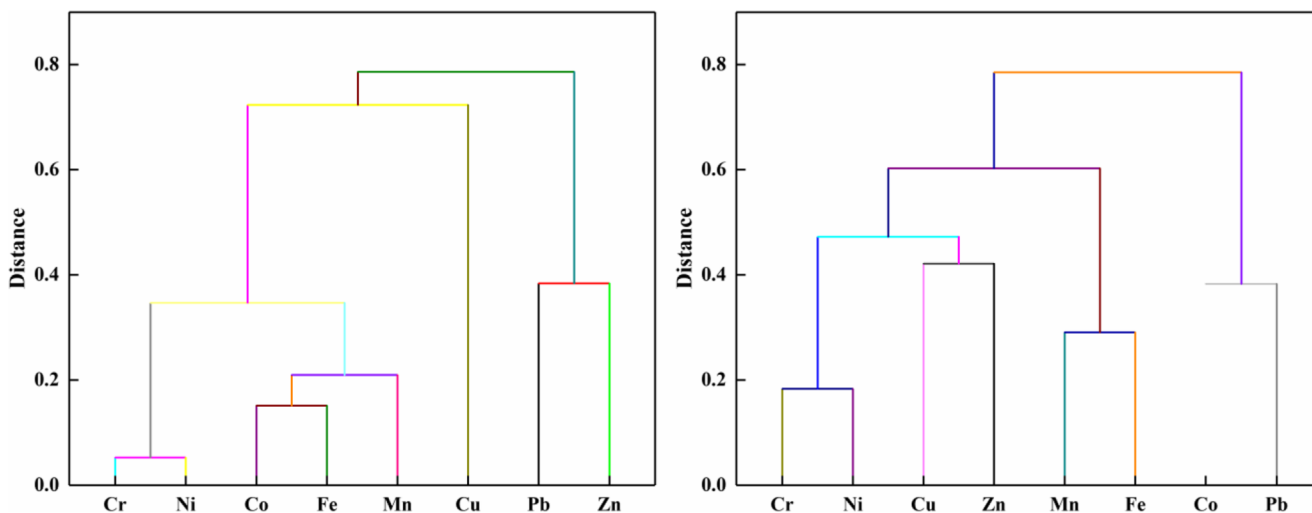

Fig. 7 Dendrogram results of hierarchical cluster analysis for metal concentrations in E'zhou (left side of the figure) and Huanggan (right side of the figure) road dusts

those in Huanggang City. Therefore, urgent proactive actions are of great importance for the future generations to monitor and control polymetallic pollution in urban areas aggravated by human-mediated activities.

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