Source Apportionment and Source-specific Health Risk of Heavy Metals in Urban Road Dust of Jinan City, East China

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Research Article

Keywords: heavy metal, source apportionment, source-specific health risk, bio-availability, urban road dust

Posted Date: November 30th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-1104105/v1

Abstract

For a better regional and source-risk-based control of heavy metals in urban environments, this study provides a source-specific health risk assessment by combining the models of United States Environmental Protection Agency health risk assessment and positive matrix factorization (PMF). The calculated data were optimized by the geochemical speciation of target 10 potentially toxic heavy metals. The results demonstrated that the mean concentrations of most heavy metals in urban dust of Jinan City exceeded their corresponding background values, especially that of cadmium (Cd) and zinc (Zn) exhibiting a mean of 12.9 and 7.84 times those of their backgrounds. Cd, Zn, copper, lead and manganese in road dust existed mainly in extractable forms, exhibiting higher bio-availability. The PMF receptor model determined four sources of heavy metals in urban road dust, namely industrial discharges (41.1%), natural and coal combustion sources (27.8%), traffic emissions (22.8%), and building material and manufacturing sources (8.3%). All the studied heavy metals presented low or negligible non-carcinogenic risk (non-CR) for adults and children, while the lifetime carcinogenic risk (CR) of Cd was in an acceptable level. Regarding source-specific health risks, the highest non-CR was derived from industrial discharges, while CR from traffic emissions, which were mainly associated with the higher content and bio-availability of Pb and Cd in the dust. Moreover, the risk contributions of industrial discharges and traffic emissions were 35.9% and 60.6% for non-CR and CR, respectively, presenting a significant difference with the apportioned source characteristics, thus deep-revealing the potentially source-based risks of heavy metal in urban environment.

1. Introduction

As population-dense and high human activity areas, cities in developing countries are experiencing rapid urbanization and industrialization, which result in severe environmental pollution (Wei et al. 2015; Bi et al. 2018; Men et al. 2018). Various pollutants, such as heavy metals (Men et al. 2018; Zhang et al. 2020), polycyclic aromatic hydrocarbons (PAHs) (Wang et al. 2020), and phthalate esters (Wang et al. 2018b), are discharged and gradually accumulate in media of urban environments (e.g. soil, water, gases/particles, dust, etc.), posing enormous threats to urban ecosystems and resident health.

Urban road dust, a complex matrix of the solid, liquid, and gas phases, is widely distributed in urban areas, and it is closely related to anthropogenic activities (Valotto et al. 2015; Tang et al. 2017; Men et al. 2018). Urban road dust is derived from various sources, primarily including industrial discharges (e.g. fossil fuel combustion, electroplating, printing and dyeing, and metal smelting), traffic emissions (e.g. vehicle exhaust and tire and brake wear), construction activities (e.g. waste and raise dust) (Lu et al. 2009, 2010; Pant and Harrison, 2013; Wang et al. 2018a), and local soil and atmospheric depositions (Pant and Harrison, 2013; De Silva et al. 2016; Rahman et al. 2019). Urban road dust containing toxic pollutants can be re-suspended into the atmosphere, leading to air pollution. Moreover, it can migrate long distances and then be deposited onto surface soil, water, or other media, leading to widespread pollution (Ordóñezc et al. 2003; Ayoubi, 2016). Urban road dust is therefore considered one of the major factors negatively affecting urban environment quality (Shi et al. 2008; Valotto et al. 2015). In addition, the daily ingestion, inhalation, and dermal absorption of dust particles can permanently impact the health of urban residents (Cook et al. 2005). Therefore, urban road dust and its pollutants are important indicators of urban environment quality.

Heavy metals are typical environmental pollutants that have received great attention due to their high toxicity, persistence, bio-accumulation, and non-degradation (Lu et al. 2009; Burges et al. 2015; Men et al. 2018; Rahman et al. 2019; Zhang et al. 2019). Of these, cadmium (Cd), cobalt (Co), nickel (Ni), and chromium (Cr) are carcinogenic, teratogenic, and mutagenic chemicals, and they have been listed as priority pollutants by the United States Environmental Protection Agency (US EPA, 2019). Recently, the occurrence, distribution, bio-availability, source apportionment, and health risks of heavy metals in urban road dust have been extensively researched (Ahmed et al. 2016; Men et al. 2018). However, they are separate, few of which focus on the composite assessments such as health risks considering the bio-availability, source-specific spatial distributions and source-specific health risks of heavy metals. These combinations can provide more comprehensive information on the regional distributions and specific-risks of pollution sources and be helpful in the control and management of heavy metal risks in urban environments. According to current researches, the positive matrix factorization (PMF) receptor model and US EPA health risk assessment model are effective tools for quantifying the source apportionment and multi-exposure human health risks of heavy metals, respectively, in the environment (Wang et al. 2018a; Zhang et al. 2019). Several studies have combined these models and focused PAHs in atmospheric gases/particles (Zhang et al. 2020; Wang et al. 2020), however little attention has been paid to heavy metals in urban road dust. Heidari et al. (2021) and Men et al. (2019) used the two models to assess the source-specific ecological risk of heavy metals in the road dust, but little for the source-specific health risk. Moreover, the bio-availability of heavy metals were generally not considered in the source-specific assessment.

Human health risks associated with heavy metals in urban road dust might be overestimated, as the geochemical speciation of heavy metals is not typically considered (Jayarathne et al. 2018). The geochemical speciation of heavy metals determines their mobility, toxicity, and bio-availability; thus, heavy metals cannot be completely desorbed from dust, and cause fully risks (Jayarathne et al. 2018). The modified sequential extraction procedure of the European Community Bureau of Reference (BCR) divides heavy metal speciation into four types: acid-soluble state, reducible state, oxidizable state, and residual state (Jayarathne et al. 2018; Dong et al. 2020). The acid-soluble state of heavy metals demonstrates the greatest bio-availability. The reducible and oxidizable states exhibit bio-availability levels attributable to their transformation into the acid-soluble state via plant or micro-organism actions. In contrast, the residual state of heavy metals is difficult to extract and exhibits negligible bio-availability (Yıldırım and Tokalıoğlu, 2016; Jayarathne et al. 2018). Therefore, the acid-soluble, reducible, and oxidizable states of heavy metals are considered to be extractable and therefore bio-available. In this study, the bio-availability of heavy metals was applied in the human health risk model to optimize the calculated heavy metal concentrations and obtain the risk results.

Jinan City, the capital of Shandong Province in East China, is experiencing rapid industrialization and urbanization, and the urban environment is suffering serious deterioration as a consequence. Studies have reported mercury and chlorinated paraffin and heavy metals in the atmospheric particulate matters (Li et al. 2017a, 2019) and river sediment (Jiao et al. 2017), respectively, in Jinan City. However, available information on heavy metals in the urban road dust of Jinan City remains limited. To address this issue, this study was conducted with the following objectives: (1) to apportion possible sources and source-specific spatial distributions of heavy metals in the road dust of Jinan City; and (2) to investigate the bio-availability-based health risks and source-specific health risks of heavy metals in urban road dust.

2. Materials And Methods

2.1. Study area

Jinan City (36.40 °N and 117.00 °E), the capital of Shandong Province in East China, is located in the middle-lower basin of the Yellow River. Jinan City is bordered by Mount Tai to the south and Yellow River to the north. The urban area of Jinan City is 8177 km^2, and it contained a population of 7 million in 2018 (JNMBS, 2019). Jinan City has an annual average temperature of 14.8 °C and rainfall of 592.5 mm. Spatially, the industrial area of Jinan City lies to the north, and it contains developed industries and large logistics and storage bases. Commercial and certain residential areas are distributed in the middle-eastern region of the city, with a large portion of the population and numerous vehicles. Other residential, cultural, and educational areas are distributed in the southern mountainous region, a high-tech area is located in the eastern part of the city, and the western region is currently undergoing development. The number of motor vehicles in Jinan City was 2.09 million in 2018 (JNMBS, 2019). In addition, similar to many northern Chinese cities, Jinan City exhibits a long period of coal burning and gas heating during the cold season (Tang et al. 2017).

2.2. Sampling and pre-treatment

The road dust samples were collected from 77 sites arranged along urban major roads that covered the entire second-ring of Jinan City (Fig. 1) during March 2018 on seven sunny days when coal-heating did not occur. Road dust is mainly powdery, and it is concentrated on urban auxiliary roads and the edges of urban main roads. At each sampling site, a composite sample of ~150 g, which was composed of five sub-samples, was collected by cleaning using a polyethylene brush and a tray. In the laboratory, all the collected road dust samples were air-dried for 1 wk, ground, and then passed through a 1-mm nylon sieve to remove debris. Finally, the samples were stored in brown bottles at 4 °C prior to analysis. Metal materials were not used in the sample processing to prevent cross-contamination. Additionally, the latitude and longitude, and surrounding conditions of each sampling site were recorded. The road dust was alkaline with pH values of 7.06–9.64 (mean 8.02), and it predominately contained fine particles of less than 50 µm (> 60%). The electrical conductivity (EC) of dust ranged from 214 to 6230 µS/cm with an average of 1810 µS/cm. The low- and high-frequency susceptibility ranged from 337×10^8 m^3/kg to 1690×10^8 m^3/kg with an average of 608×10^8 m^3/kg, and from 294×10^8 m^3/kg to 1660×10^8 m^3/kg with an average of 561×10^8 m^3/kg, respectively. More detailed road dust physicochemical properties are displayed in Fig. S1 and S2 and Table S1 in the Supplementary Materials.

2.3. Analysis

The total concentration and geochemical speciation of 10 potentially toxic heavy metals (Cd, Cr, copper (Cu), zinc (Zn), lead (Pb), Co, Ni, manganese (Mn), barium (Ba), and vanadium (V)) were analyzed using nitric acid–hydrogen fluoride–perchloric acid (HNO_3–HF–$HClO_4$) digestion, a modified BCR sequential extraction procedure, and inductively coupled plasma atomic emission spectroscopy (ICP–AES, Arcos, Spectro, Germany). The detailed digestion process used to determine the total concentration is described in Text 1 of the Supplementary Materials and in our previous work (Wang et al. 2018a). BCR sequential extraction separated the heavy metals into four geochemical species: acid-soluble (f_1), reducible (f_2), oxidizable (f_3), and residual states (f_4) (Wang et al. 2015, 2018a; Jayarathne et al. 2018; Dong et al. 2020). The specific extraction process is expressed in Text 2 of the Supplementary Materials and in our previous works (Wang et al. 2015; Dong et al. 2020). The bio-availability associated with the speciation of heavy metals in urban road dust can be calculated using Equation 1.

$$F_{bio} = \frac{f_1 + f_2 + f_3}{C} \times 100\% \qquad (1)$$

where F_{bio} is the bio-available geo-chemical fraction (extractable state) of the metal; C is the concentration of the metal (mg/kg), and f_1, f_2, and f_3 are the acid-soluble, reducible, and oxidizable contents of the metal, respectively (mg/kg).

To ensure the accuracy of the experimental results, blanks were used in the experiment as a control, and Chinese soil reference standard substances (i.e. GSS1 and GSD5) were simultaneously analyzed during the total content analysis, and the results exhibited relative errors of 1–14% (Table S2). One-tenth of the road dust samples (n = 8) were repeated, with relative standard deviations of 3–8%. The recovery rates of the speciation analysis were 88–118%. More details regarding the instrumental parameters of ICP–AES are presented in Text 3 in the Supplementary Materials.

2.4. Source apportionment and health risk assessment models

The PMF receptor model can apportion the source composition and contribution of heavy metals in urban road dust (Men et al. 2018; 2019). The PMF model considers the uncertainty of the results to evaluate the quality of the data. Additionally, it provides the fitting results of observed and predicted concentrations of individual species to determine the model reliability. A detailed introduction to the PMF model is available in our previous studies (Zhang et al. 2019, 2020) and is also provided in Text 4 of the Supplementary Materials. In the present study, PMF was applied to determine the source apportionment of heavy metals in the road dust of Jinan City, China.

Human exposure to heavy metals in urban road dust through ingestion, inhalation, and dermal adsorption can have serious health consequences. The health risk assessment model of the US EPA has been widely used to assess the health risks of human exposure to heavy metals. The model exposure dose calculations for carcinogenic (lifetime average daily dose, LADD) and non-carcinogenic (average daily dose, ADD) effects, meanings and values of the assessment parameters (Table S3), reference doses and slope factors of the heavy metals (Table S4), and risk expression (hazard quotient/index, HQ/HI; carcinogenic risk, CR) are described in our previous studies (Wang et al. 2015, 2018b) and are provided in Text 5 of the Supplementary Materials. In this study, the concentrations of heavy metals in the model were modified according to their corresponding bio-availability (F_{bio}).

In addition, the PMF and health risk assessment models were combined to determine the source-specific health risks. In order to obtain the source-specific risk results, the PMF model was first used to obtain a matrix presenting heavy metal data in different factors in the automatically generated source profile; then the heavy metal data of the matrix were modified by their bio-availability (i.e. extractable state); finally the health risk assessment was conducted to obtain a matrix for the source-specific risk results.

2.5. Data processing

Statistical analysis was performed using Microsoft Office 2010 and Statistical Product and Service and Solutions (SPSS) 21.0. Geographic Information Systems (GIS) 10.6 was used to present the spatial distributions of the heavy metals. Dust characteristics and heavy metal compositions were visualized using Origin 9.5C.

3. Results And Discussions

3.1. Concentrations and spatial distributions of heavy metals

As displayed in Table 1 and Fig. S3, the mean concentrations of Co, Cr, Cu, Cd, Mn, Ni, Pb, Zn, V, and Ba in the road dust of Jinan City were 8.25 mg/kg, 114 mg/kg, 87.7 mg/kg, 1.08 mg/kg, 517 mg/kg, 30.3 mg/kg, 85.7 mg/kg, 498 mg/kg, 51.8 mg/kg, and 643 mg/kg, which were 0.61, 1.73, 3.65, 12.9, 0.80, 1.17, 3.32, 7.84, 0.65, and 1.28 times the corresponding Shandong Province soil element background values, respectively (CNEMC, 1990). The mean concentrations of the heavy metals (except for Co, Mn, and V) exceeded their corresponding background values, especially those of Cd and Zn, which were 12.9 and 7.84 times their background values, respectively. In addition, Cr, Cu, Cd, Ni, Pb, Zn, and Ba presented large coefficients of variation (CV), indicating possible anthropogenic sources, while Co, Mn, and V exhibited low CV (< 0.21), implying that they may have been only slightly influenced by anthropogenic activities (Khademi et al. 2019).

The spatial distribution characteristics of heavy metals in urban road dust can reflect their possible sources to a certain degree. As illustrated in Fig. S4, the high concentrations of Zn, Pb, Cd, and Ba were mainly found in the northern part of Jinan City, where many logistics centers and industrial parks, especially those of the coking, chemical manufacturing, and non-ferrous metal smelting industries, are concentrated. This result reflected that these heavy metals in the road dust were mainly associated with traffic emissions and industrial discharges. Additionally, elevated concentrations of Cr and Ni were detected in the northern part of Jinan City and the vicinity of certain coal-combustion plants in the eastern part of Jinan City, indicating that coal combustion and industrial emissions are potential sources of Cr and Ni. Higher concentrations of Cu were mainly distributed in the central, eastern, and northern parts of Jinan City, and they were typically close to construction sites or manufacturing factories, suggesting that Cu in the urban road dust may be derived from metals used in building materials or manufacturing emissions. Higher concentrations of Mn, Co, and V were mainly observed in the northern, southwestern, and eastern areas of Jinan City. However, the distributions exhibited average concentrations lower than those of the background values on average, indicating possible natural sources (local soil). In addition, the concentrations of most heavy metals presented a decreasing trend from north to south.

3.2. Speciation and bio-availability of heavy metals

As illustrated in Fig. S5, Co, Cr, Ni, V, and Ba in the urban road dust were mainly dominated by the residual (f_4); Cu and Pb was controlled by the oxidizable form (f_3), followed by the f_4; Cd and Zn existed mainly in the acid-soluble (f_1) and reducible (f_2) forms; Higher proportion of Mn was observed in the f_4, followed by the f_4 and f_2. According to the classification proposed by Botsou et al. (2016) based on the ratio of f_1 to total concentration, Cd (39.9%) presented high ecological risk to the environment; Zn (27.8%) and Mn (24.3%) exhibited moderate ecological risk; Ni (9.7%), Co (9.3%), Ba (4.5%), Pb (2.7%), Cu (2.5%), and V (1.4%) had low ecological risk; Cr (0.6%) had no ecological risk. The sum of the first three part (f_1 + f_2 + f_3) of heavy metals was considered to be in the extractable state, which represents the bio-availability part. As illustrated in Fig. 2 and S5, the mean proportions of the extractable state of Cd, Zn, Cu, Pb, and Mn were 85.8%, 77.1%, 64.3%, 62.0%, and 51.7%, respectively, indicating bio-availability. For Ba, Co, Ni, V, and Cr, the extractable states were 38.9%, 31.2%, 30.1%, 25.8%, and 23.1%, respectively, suggesting low bio-availability. The proportion order of the bio-availability for heavy metals was Cd > Zn > Cu > Pb > Mn > Ba > Co > Ni > V > Cr.

3.3. Sources apportionment of heavy metals

3.3.1. PMF source apportionment

The PMF receptor model is an effective diagnostic tool for heavy metal sources. Data of 10 species × 77 receptor sites were introduced into the US EPA PMF 5.0 model to analyze the source compositions and contributions of heavy metals in the road dust of Jinan City. Based on the stability and convergence of Q, four factors were determined, with heavy metal signal-to-noise ratios from 2.2 to 4.1, which were defined as strong, and most of the residuals were acceptable. The source compositions and corresponding contributions of heavy metals in the road dust of Jinan City are displayed in Fig. 3.

Factor 1 exhibited a high load of Cd (61.0%) and moderate loads of Zn (31.4%), Pb (28.5%), and Ni (24.4%). Cd, Ni, Pb, and Zn are primarily associated with modern transport modes and are derived from traffic emissions (Zhao and Li, 2013). Zn often originates from the tire wear of motor vehicles and corrosion of galvanized parts (Zhao and Li, 2013). Ni and Cd are commonly applied in the lubrication of automobiles, and they are also released from tire wear (Duan and Tan, 2013). Pb is considered an important traffic fingerprint element, and it is typically released during transport and fuel combustion (Zhu et al. 2013). In this study, Zn and Cd presented the highest concentrations in the traffic-intensive areas of Jinan City, reflecting the influences of lubrication usage, tire wear, and traffic exhaust on heavy metal pollution. Additionally, Zn and Cd exhibited high concentrations in areas far away from significant industrial or traffic areas of Jinan City, which may be due to the migration and sedimentation of dust particulates (Valotto et al. 2015). Therefore, factor 1 can be diagnosed as traffic emissions.

Factor 2 was highly loaded with Ba (70.7%) and Pb (43.9%) and moderately loaded with Cu (30.9%), Cr (27.2%), and Cd (26.7%). Cu, Pb, Ba, and Cd in the road dust exhibited high CV values, indicating strong anthropogenic influences. Ba is an important industrial element that is widely used in the production of paper, textiles, glass, and plastic. Additionally, Ba is often used as a major component of detergents in diesel engines and various internal combustion engines (Chen and Lu, 2018). Pb is an important industrial material that is widely used in various industrial production processes, such as liquid crystal display and electronic product manufacturing (Chen and Lu, 2018; Khademi et al. 2019). In addition, certain metallurgical and industrial wastes contain a considerable amount of Pb. Cd is often used in chemical manufacturing, coking, and smelting, and in the production of pesticides, fertilizers, and medical devices (Khademi et al. 2019). Notably, high concentrations of Cd, Pb, and Ba in the road dust were primarily distributed in the industrially developed areas in the northern part of Jinan City. Therefore, factor 2 represents industrial discharges.

Factor 3 was heavily dominated by Co, Cr, Mn, Ni, and V, with contribution rates of 59.2%, 42.4%, 56.9%, 57.0%, and 60.3%, respectively. Co, Mn, and V exhibited low CV values (< 0.21), and their mean concentrations were lower than those of their corresponding background values. Therefore, Co, V, and Mn can be defined as being derived from natural sources (local soil). The concentrations of Ni in 50.6% of the samples were also lower than its background value, indicating a natural source (Lu et al. 2010). According to JNMBS (2019), coal was widely used as the main fuel in the industrial processes and heating of Jinan City. Half of the Ni and most of the Cr concentrations exceeded their background levels, suggesting possible coal combustion sources (Raja et al. 2014; Wang et al. 2016, 2018a). Therefore, factor 3 is defined as mixed natural and coal combustion sources.

Factor 4 was primarily composed of Cu, which accounted for 65.4% of its total concentration. Cu is a widely used material in manufacturing, industrial production, metallurgical processes, and building materials (Men et al. 2018). Factor 4 can therefore be defined as building material and manufacturing sources. In summary, the PMF receptor model apportioned four sources of heavy metals in the road dust of Jinan City, namely industrial discharges (41.1%), natural and coal combustion sources (27.8%), traffic emissions (22.8%), and building material and manufacturing sources (8.3%).

3.3.2. Source-specific spatial distributions

The source-specific spatial distributions can be determined according to the PMF and spatial interpolation of Geographic Information Systems (GIS). The source-specific spatial distributions of heavy metals in road dust are displayed in Fig. 4. As illustrated in Fig. 4, traffic emissions were primarily distributed in the northwestern, southeastern, and central parts of Jinan City, where logistics centers and transportation hubs were highly concentrated. Sites with lower traffic emissions were located in the northeastern and southwestern residential, cultural, and educational areas. Industrial discharges were mainly distributed in the northern and southern parts of the city, where many industries, including the chemical manufacturing, coking, and non-ferrous metal smelting industries, were distributed. The heavy metals related to traffic and industry showed obvious regional, with a high level approaching to several anthropocentric emission source, and decreasing to the surroundings. This indicated that heavy metal pollution in road dust tends to appear in a form of point and non-point source combination, and associated with the dust migration characteristics. Natural and coal combustion sources were distributed dispersedly. However, they have highest level in the northern and eastern areas of Jinan City, which was primarily related to the higher level of Ni and Cr. In fact, these areas are relatively close to industrial areas and several hot-plants in Jinan City, leading a potential affect by coal combustion. The natural source is mainly affected by the environmental soil (Lu et al. 2010). The building material and manufacturing sources were mainly located in central, eastern, and southwestern urban areas where construction activities were occurring, indicating that the sources of heavy metals in the road dust were significantly affected by the urban layout. The source-specific spatial distribution of heavy metals in the road dust can significantly reflect the spatial variations in heavy metal concentrations and the influences of emission sources on heavy metals (Chen and Lu et al. 2017). The results also support the source diagnostic results of the PMF model.

3.4. Health risks of heavy metals

3.4.1. Health risks based on bio-availability

In the present study, the bio-availability of heavy metals was utilized to optimize the concentration data in the health risk assessment model for more realistic health risk results. The CR and non-CR of heavy metals in the road dust are illustrated in Table 2. The 95% upper confidence limit replaced the maximal value in the health risk assessment. The hazard index (HI) was used to reflect the non-CR of the multi-exposure pathways. For non-CR, the ingestion of dust particles appeared to be the main route for heavy metal exposure to children and adults compared with the other routes, which was consistent with the results of many other studies (Doabi et al. 2018; Men et al. 2018; Rahman et al. 2019). The HI values of the studied heavy metals for both children and adults decreased in the order of Pb > Cr > Mn > Ba > V > Zn > Cu > Cd > Ni > Co, with the highest HI values of Pb for children and adults of 1.18×10^{-1} and 1.52×10^{-2} and lowest values of Co of 9.65×10^{-4} and 1.50×10^{-4}, respectively. The total HI value of all the studied heavy metals was 3.10×10^{-1} for children and 4.13×10^{-2} for adults, indicating low or negligible non-CR. Regarding CR, Co, Cr, Cd, and Ni are considered carcinogens. As displayed in Table 2, the CR values of Co, Cr, and Ni were 1.12×10^{-9}, 5.30×10^{-8}, and 3.63×10^{-10}, respectively, which were much lower than 10^{-6}, indicating low or negligible CR. However, the CR value of Cd was 5.95×10^{-6}, falling in the range of 10^{-6}–10^{-4} and indicating an acceptable CR. Additionally, the CR of Cd was primarily derived from the ingestion route, which was much higher than that of the other pathways. The CR values decreased in the order of Cd > Cr > Co > Ni.

3.4.2 Source-specific health risks

The health risks of heavy metals from different sources in the road dust are displayed in Fig. 5 and Table S6. For the non-CR contribution of each source, industrial discharges exhibited the highest HI values of 8.58×10^{-2} and 1.14×10^{-2} for children and adults, respectively, accounting for 35.9% of the total non-CR and with Pb and Ba as the greatest contributors. Natural and coal combustion sources and traffic emissions exhibited HI values for children and adults of 6.82×10^{-2} and 9.46×10^{-3}, and 5.53×10^{-2} and 7.33×10^{-3}, accounting for 28.5% and 23.2% of the total non-CR, respectively. The building material and manufacturing sources presented the lowest HI values of 2.97×10^{-2} and 3.90×10^{-3} for both children and adults, contributing 12.4% of the total non-CR. Certain toxic metals with higher concentrations, such as Cr, do not present a correspondingly high level of health risk, which is mainly due to their low bio-availability. The total CR values of the carcinogenic metals were 2.73×10^{-6} for traffic emissions (60.6%), 1.22×10^{-6} for industrial discharges (27.0%), 1.92×10^{-8} for natural and coal combustion sources (0.4%), and 5.40×10^{-7} for building material and manufacturing sources (12.0%). Cd was the major contributor to each source. The results indicated that industrial discharges were the primary contributors for the non-CR and were mainly associated with the higher bio-availability and content of Pb in the road dust of Jinan City. In contrast, traffic emissions exhibited the highest CR related to the higher bio-availability and content of Cd. The results of source-specific risks, considering the bio-availability of heavy metals, were different from the source apportionment of heavy metal concentrations, and they can provide a realistic assessment of the impacts on the health of urban residents exposed to heavy metals in road dust.

4. Conclusions

This study evaluated the source-specific spatial distributions and modified health risks of 10 heavy metals using the combined PMF and health risk assessment models, GIS, and bio-availability of heavy metals in urban road dust. The results demonstrated that the heavy metals in the urban road dust were significantly related to the surrounding emissions, primarily industrial discharges and traffic exhausts in the northern part of Jinan City. All the studied heavy metals presented low or negligible non-CR for adults and children, while Cd posed an acceptable CR. Moreover, industrial discharges exhibited the highest non-CR, which was mainly associated with its higher bio-availability and content of Pb, while traffic emissions exhibited the highest CR, which was primarily related to its higher bio-availability and content of Cd. There were significant differences between the apportioned source characteristics and health risks of heavy metals in urban road dust. Thus, utilizing combined PMF and health risk assessment models and considering the bio-availability of heavy metals are useful for assessing the source-specific health risks of heavy metals in urban road dust. The source-specific health risk results imply that greater attention should be paid to the regional effects of pollution sources and heavy metals with high toxicity and bio-availability for the targeted control and management of heavy metals.

Declarations

Acknowledgments

This research was supported by the National Natural Science Foundation of China through Grants 41877516 and 41877517.

Appendices

Supplementary materials

Credit Author statement

Shengwei Zhang: Conceptualization, Methodology, Software, Data curation, Writing-Original draft preparation, Formal analysis, Validation

Ge Ma: Validation, Investigation

Shuzhen Dong: Investigation

Xiang-Zhou Meng: Supervision

Lijun Wang: Conceptualization, Resources, Writing-Review & Editing, Supervision, Project administration, Funding acquisition

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Tables

Table 1 Concentrations of heavy metals in the road dust of Jinan City (mg/kg)

Elements	Min	Max	Mean	SD	CV	RV
Co	6.09	12.7	8.25	1.39	0.17	13.6
Cr	56.5	509	114	58.7	0.51	66
Cu	33.5	412	87.7	52.8	0.60	24
Cd	0.30	6.74	1.08	1.09	1.01	0.084
Mn	362	866	517	110	0.21	644
Ni	16.0	101	30.3	14.1	0.47	25.8
Pb	26.0	413	85.7	66.2	0.77	25.8
Zn	109	1.27×10^5	498	1.43×10^4	2.87	63.5
V	36.4	71.5	51.8	7.06	0.14	80.1
Ba	76.4	2.16×10^3	643	356	0.55	502

SD: standard deviation; CV: coefficient of variation; RV: reference value CNEMC (1990).

Table 2 Non-carcinogenic risk (Non-CR) and lifetime carcinogenic risk (CR) of heavy metals in road dust based on their bio-availability.

Elements	Group	95%UCL	Non-CR				CR			
		(mg/kg)	HQ_{ing}	HQ_{inh}	HQ_{dermal}	HI	CR_{ing}	CR_{inh}	CR_{dermal}	CR
Co	Children	2.67	8.78E-04	8.59E-05	1.26E-06	9.65E-04		1.12E-09		1.12E-09
	Adults		1.12E-04	3.70E-05	2.11E-07	1.50E-04				
Cr	Children	29.4	6.45E-02	1.89E-04	3.71E-03	6.84E-02		5.30E-08		5.30E-08
	Adults		8.26E-03	8.15E-05	6.20E-04	8.96E-03				
Cu	Children	64	1.05E-02	2.94E-07	4.03E-05	1.06E-02				
	Adults		1.35E-03	1.27E-07	6.74E-06	1.35E-03				
Cd	Children	1.14	7.51E-03	2.10E-07	8.64E-04	8.38E-03	5.94E-06	8.82E-14	7.54E-09	5.95E-06
	Adults		9.61E-04	9.05E-08	1.44E-04	1.11E-03				
Mn	Children	280	4.00E-02	3.60E-03	1.15E-03	4.48E-02				
	Adults		5.13E-03	1.55E-03	1.92E-04	6.87E-03				
Ni	Children	10.1	3.31E-03	8.99E-08	1.41E-05	3.33E-03		3.63E-10		3.63E-10
	Adults		4.24E-04	3.88E-08	2.36E-06	4.27E-04				
Pb	Children	62.5	1.17E-01	3.26E-06	9.00E-04	1.18E-01				
	Adults		1.50E-02	1.41E-06	1.50E-04	1.52E-02				
Zn	Children	634	1.39E-02	3.88E-07	7.99E-05	1.40E-02				
	Adults		1.78E-03	1.67E-07	1.33E-05	1.79E-03				
V	Children	13.8	1.29E-02		1.49E-03	1.44E-02				
	Adults		1.66E-03		2.49E-04	1.90E-03				
Ba	Children	281	2.64E-02	3.62E-04	4.34E-04	2.72E-02				
	Adults		3.38E-03	1.56E-04	7.26E-05	3.61E-03				
Total	Children		2.97E-	4.24E-	8.68E-	3.10E-	5.94E-	5.45E-	7.54E-	6.00E-

heavy metals		01	03	03	01	06	08	09	06
	Adults	3.81E-02	1.83E-03	1.45E-03	4.13E-02				

Figures

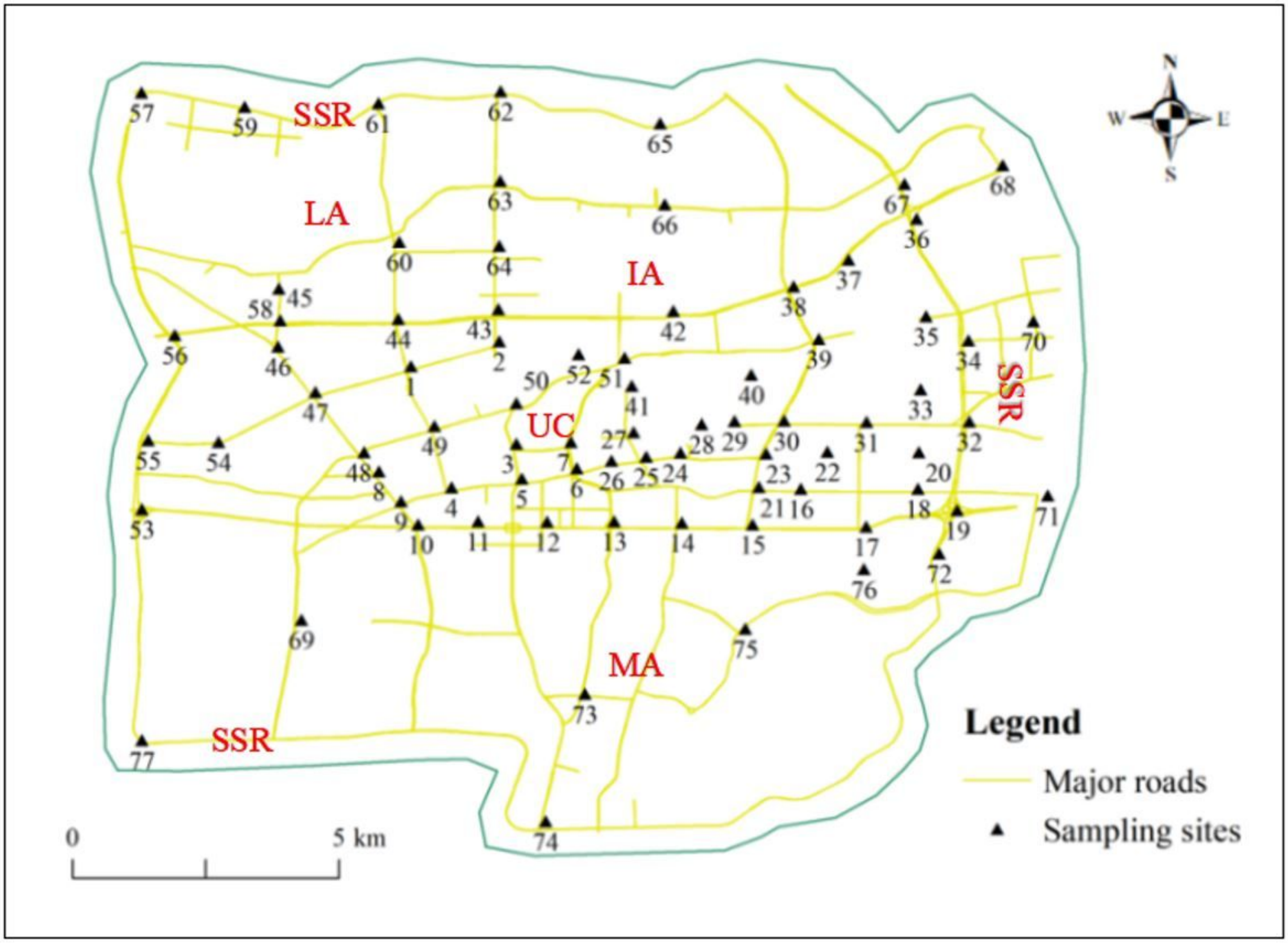

Figure 1

Sampling sites of road dust samples in Jinan City. Logistics area (LA), Industrial area (IA), Urban center (UC), Mountain area (MA), Second-ring road (SRR).

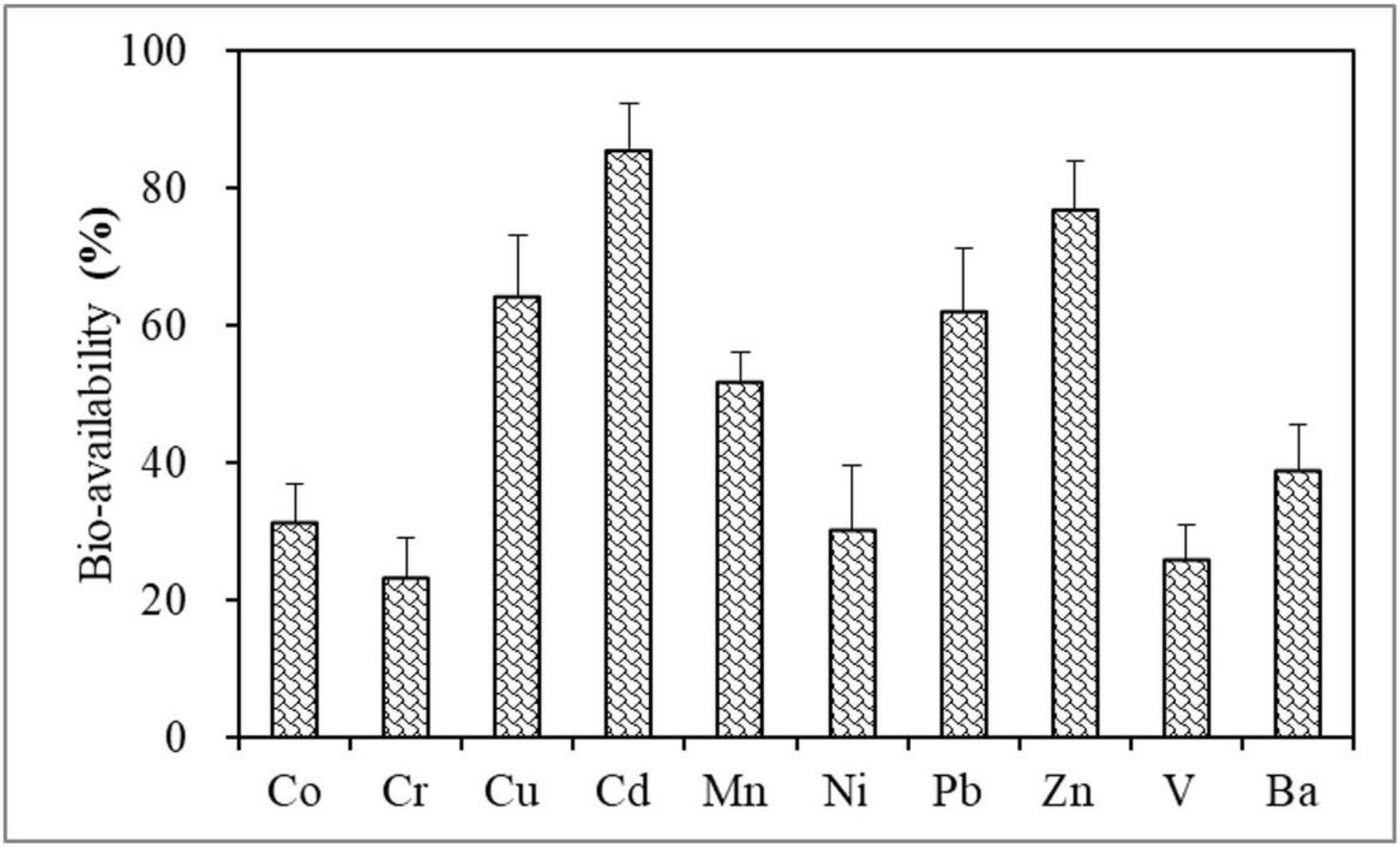

Figure 2

The mean proportion of bio-availability of heavy metals in the road dust of Jinan City.

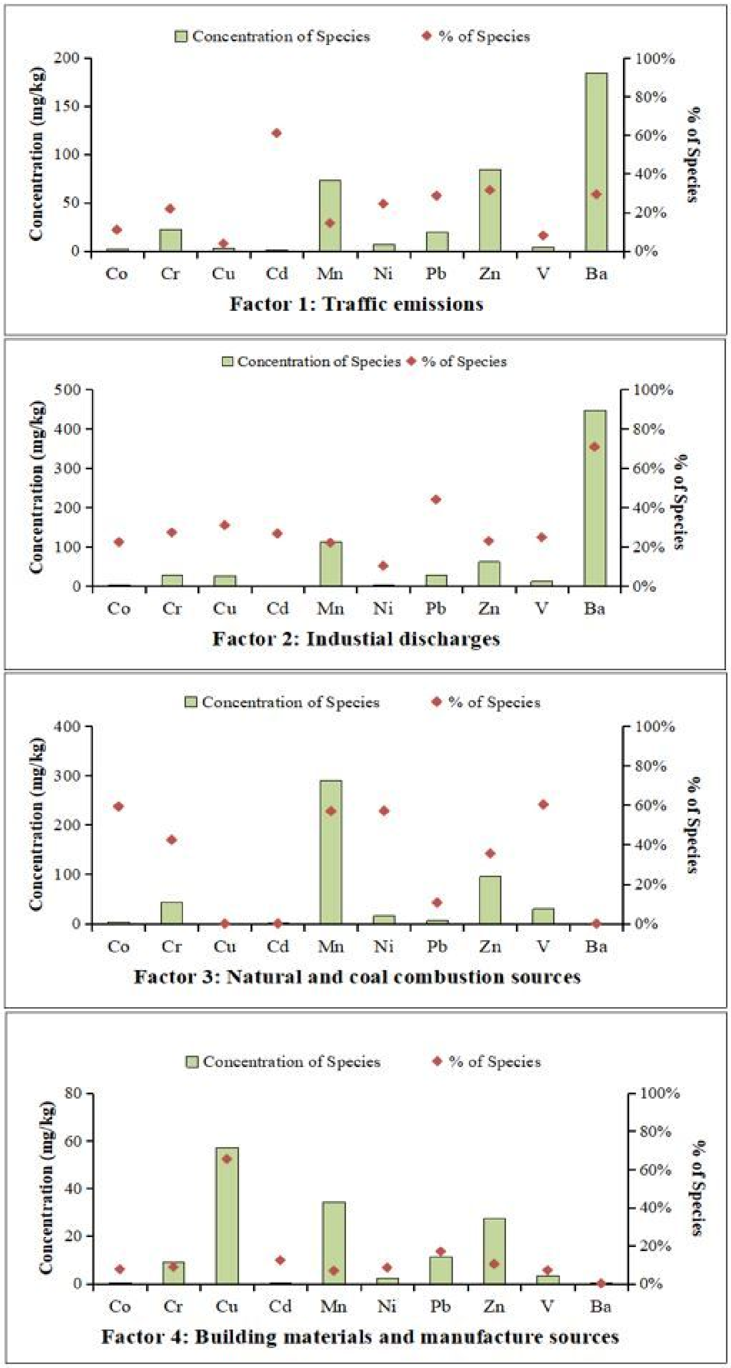

Figure 3

Sources compositions of heavy metals in urban road dust from PMF model.

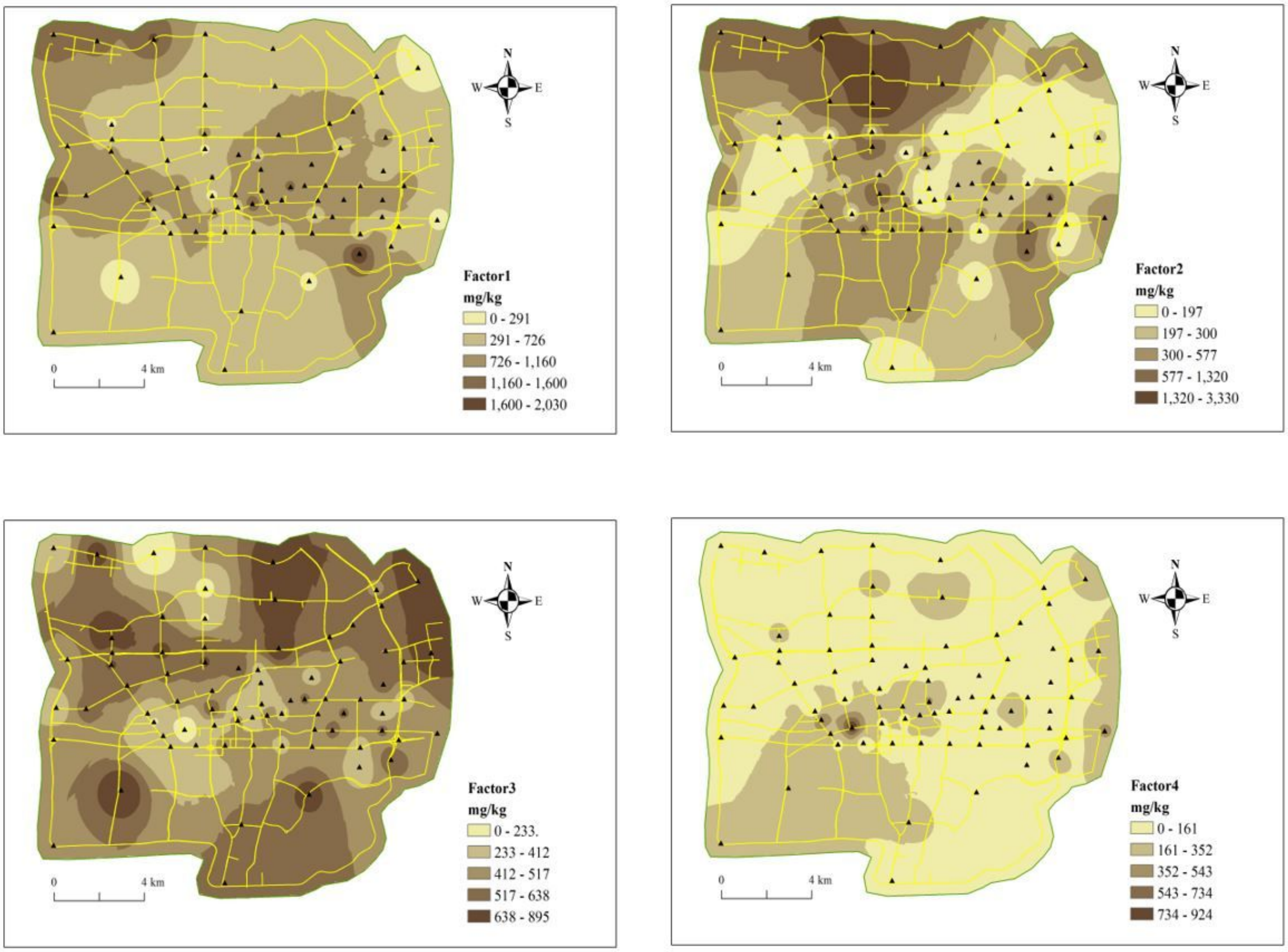

Figure 4

Spatial distribution of sources for heavy metals in the road dust of Jinan City (Factor 1: traffic emissions; Factor 2: industrial discharges; Factor 3: natural and coal combustion sources; and Factor 4: building material and manufacture sources).

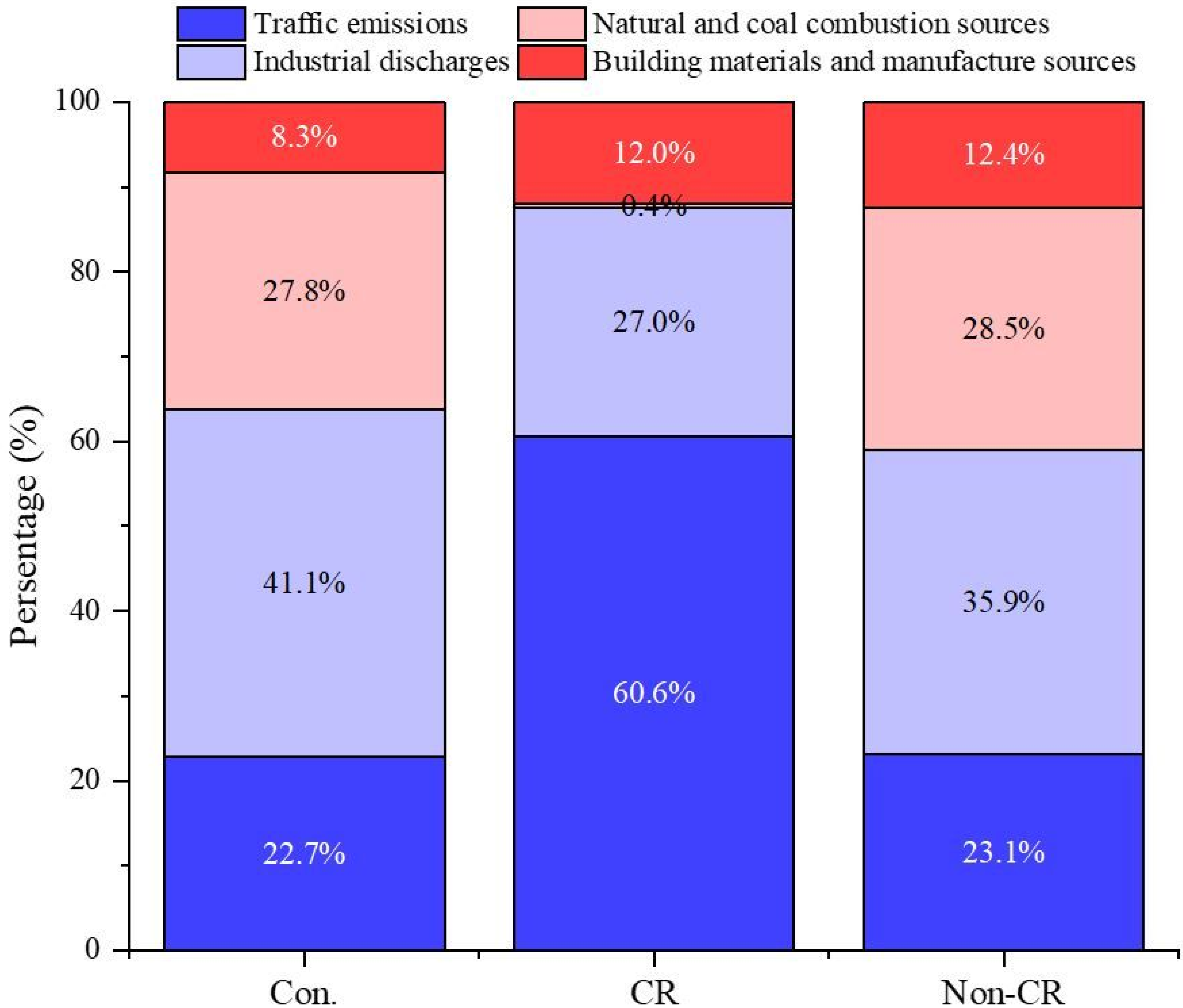

Figure 5

Source contributions of concentration (Con.), carcinogenic risk (CR), and non-carcinogenic risk (Non-CR) of heavy metals in road dust

Supplementary Files

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