Seasonal Source Identification And Source-Specific Health Risk Assessment of Pollutants On Road Dust In Tianjin

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Seasonal source identification and source-specific health risk assessment of pollutants on road dust in Tianjin

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Abstract

Human exposure to metals on road dust might have potential health risks through touching, ingesting, and inhaling. There were limited studies to link seasonal emission sources to health risks from metals on road dust. In this study, metals on road dust from different functional areas were seasonally monitored. The pollutant sources in study city varied slightly with the seasons, but the major pollutant source in the particular study site were significantly affected by the seasons. By combining the source apportionment model (PMF), line sources model and health risk models (HI: Hazard index and ILCR: Incremental Lifetime Carcinogenic risk), industrial and construction activity was identified as the crucial source of both the pollutants on road dust (29% - 47%), and the HI for adults (27% - 45%) and children (41% - 50%) in different seasons. The traffic non-exhaust emission dominated in the carcinogenic risks for children in spring (45%) and summer (36%). Factors such as seasons, particle size, metal bioavailability, human exposure time, and exposure area were all taken into consideration to avoid overestimating or underestimating health risks. The carcinogenic risks for children (1.6 E-06) and adults (2.8 E-06) exposed to Cr both exceed the minimum threshold (10^{-6}). Measured metals mainly posed hazard to human health through ingestion route. Pb and Mn, Fe and Mn were the main harmful elements that induced non-carcinogenic risks for adults and children, respectively. Effectively identifying the source-specific health risks in different seasons will help in the formulation of adaptive strategies to diminish the potential risks.

Keywords

Road dust, Heavy metals, Source apportionment, Risk assessment, Seasonal source-specific risks

Introduction

Pollutants on road dust can be measured as a comprehensive indicator of urban pollution level (Dietrich et al., 2019; Skrbic et al., 2019; Wahab et al., 2020; Živančev et al., 2019). Metals are prevalent on the road surface and build up on the road dust from multiple pollution sources by natural or human activities. The accumulation of road dust itself also serve as a notable pollutant source (Tian et al., 2019). Road dust can be transported into the drainage systems by runoff, and the associated pollutants could reach receiving water body and adversely affect the water ecosystem (Gavrić et al., 2019); it could also be resuspended into the atmosphere by wind or vehicle movement, affecting air quality and threatening human health (Pateraki et al., 2019). The source analysis of the atmospheric particles in Tianjin reported by the former Ministry of Environmental Protection indicated that road dust was the vital source accounting for 30% of the $PM_{2.5}$ (particles with an aerodynamic equivalent diameter of 2.5 microns or less) (Zhao et al., 2019). The key to reducing the adverse effects of road dust is to quantitatively identify and evaluate its pollution sources and risks (Men et al., 2020). Many studies have emphasized that the pollution sources might vary in different seasons, but there is a lack of detailed supporting data (Liu et al., 2015; Men et al., 2020).

Source apportionment was extracted by the receptor model, and the source contribution to the contaminants on road dust was quantified mathematically based on the source composition or fingerprint. Many studies have applied the receptor model, such as multiple linear regression (PCA-MLR) (Zhang et al., 2019b), principal

component analysis (PCA) (Asheim et al., 2019), factor analysis (FA) (Bzdusek et al., 2004), positive matrix factorization (PMF) (Men et al., 2019), and forward stepwise regression (FSWR) (Liu et al., 2018) to evaluate and identify the possible sources of target pollutants in air (Wang et al., 2018), soil (Wang et al., 2019b) and water (Salim et al., 2019; Škrbić et al., 2018). This has accumulated a lot of knowledge on the source tracking of pollutants such as polycyclic aromatic hydrocarbons (Skrbic et al., 2019) and metals (Wang et al., 2019b). Obviously, different models have different advantages and disadvantages (Huang et al., 2018c). In order to quantitatively analyze the contribution proportion of pollution sources on metals and human health risks, the PMF model was selected in this study (Men et al., 2019). This model considers the uncertainty of sampling and data analysis, and could quantify the source contribution to meet the research goals.

The metals, such as Fe, Mn, Pb, Zn, Cr and Cu, are ubiquitous on road dust and have potential adverse effects on human health, including cell injury, inflammation or heart diseases (Huang et al., 2018b; Men et al., 2020). The Hazard Index (HI) model was selected to assess the potential non-carcinogenic risk of human exposure to metals (Wahab et al., 2020). Adults and children exposed to the road dust made pollutants easier to enter the human body through three pathways: ingestion (exposure of mouth and digestive tract), inhalation and dermal contact (Skrbic et al., 2019). Cr and Pb were not the essential elements for living and showed higher toxic to animals, plants and human beings (Wahab et al., 2020). The Incremental Lifetime Carcinogenic risk (ILCR) model was used to assess the carcinogenic risks of Cr and Pb to human health which

has been widely applied in many studies (Liu et al., 2015). However, in previous studies, a common practice in calculating the daily exposure dose (ADD) of road dust through the inhalation route was to classify it as soil using a particulate emission factor (PEF) (Hou et al., 2019). This might underestimate the amount of inhalation or ingestion, because it ignored the impact of particle size and severe traffic disruption. Comprehensive and mature models have been developed in air pollution simulation, including NORTRIP, and AERMOD (Tian et al., 2019). Although these methods were valuable, they require a large number of parameters like road condition and climate with large calculation costs. This study was the first attempt to combine the road dust emission model AP-42 (EPA, 2001b) with linear model, representing the pollutant diffusion, to obtain a relatively accurate estimate of suspended road dust. Furthermore, season was often overlooked in the risk assessment. According to the Exposure Factors Handbook of Chinese Population (Ministry of Ecology and Environment, 2013), the exposure time and exposure area of people varied in different seasons，which would affect the accuracy of the risk assessment.

Accurate source control is the key to mitigating the health risks of metal exposure. Affected by the characteristics of the functional areas and seasons, this study emphasized that the seasonal sources of pollutants in each functional area might be significantly distinctive. In addition, many studies have focused on the source apportionment of pollutants on road dust, but ignored the crucial sources leading to health risks (Men et al., 2020). Due to the different toxic reaction factors of metals, the major sources of metals on road dust might not generate the major risks to human health

(Liu et al., 2015). And the crucial sources of health risks might also be affected by the seasons. The combination of the PMF model and the risk assessment model could achieve accurate and effective evaluation of the source apportionment of health risks.

In order to mitigate hazardous pollutants on road dust effectively, the objectives of this work contained four items: 1) investigate the pollution characteristics of metals on road dust in different functional areas and seasons; 2) identify the vital pollution sources of measured metals on road dust in different seasons; 3) evaluate the carcinogenic and non-carcinogenic risks of metals on road dust for different human groups (adult and children) through three pathways in different seasons and functional areas; 4) distinguish the crucial sources of health risks. This study on the source–specific risk assessment of metals on road dust could provide a theoretical basis and guidance in controlling the potential risks to human health.

1. Materials and Methods

1.1 Sampling site

Tianjin is a typical metropolis with an area of 11966.45 km^2 and a population more than 15.6 million, and it is a largest port city in northern China. There were few studies on the metals on road dust and its potential risks to human health in Tianjin recently (Živančev et al., 2019). Hexi District, as one of the administrative districts in the city center, was selected in this research. Tianjin has a typical warm and semi-humid continental monsoon climate with distinct seasons: hot and rainy summer, and cold and dry winter. The relatively large wind speed appears in the spring and winter, and the average wind speed is about 2 $m \cdot s^{-1}$ throughout the year (Wang et al., 2019a). Each

functional area exhibited different regional characteristics. Four typical areas, traffic areas (JT), commercial areas (SY), residential areas (JM), and central commercial streets (BJ) (with more intensive business activities, people and traffic volume than JM), were selected in this study due to the significant differences in the population and traffic volume as shown in Figure S1 and Table S1. There were large construction activities near the traffic area during study period.

1.2 Sampling and measurement

In this work, three sampling spots were set at each study area for each sampling event. The detailed sampling process and pretreatment methods were performed according to our previous work as shown in SI (Wang et al., 2020). The survey duration included four quarters, started from December 2016 to November 2017, and the detailed sampling date was displayed in Table S1. Thereout, an average of 15 samples was taken each quarter at each site, and a total of 240 samples were obtained in four study areas. The pretreated samples were further sieved into three groups of particles with size <75 μm, 75-150 μm, and 150-500 μm. The particles with different sizes might pose threat to human health in different pathway. Many studies have found that different size particles have various behavioral characteristics (Gbeddy et al., 2018; Wang et al., 2019a; Wijesiri et al., 2016). Fine particles (<75 μm) were more likely to be suspended into the air and endangered human health through inhalation (Deletic and Orr, 2005; Tomašević et al., 2005). While coarse particles might be more inclined to cause health risks through dermal contact or ingestion (Zhao et al., 2016). Therefore, metals in different size particles (Table S2) were evaluated for risks through three

pathways.

Metals (Fe, Mn, Pb, Zn, Cr, Cu) in size-fractioned particles were analyzed using the microwave acid extraction with Agilent-7800 ICP-MS instrument system. The nutrients (NH_3-N, NO_3 -N, TOC and TP) in the samples were extracted and measured according to the China standard methods (HJ 535-2009, GB/T 32737-2016, HJ 632-2011) (SI). NH_3-N, NO_3-N, TOC and TP, as nutrients, were considered to obtain more accurate source apportionment results. The analysis process of elements conformed to the quality assurance and quality control (QA/QC) procedures and the detailed information was shown in SI.

1.3 Positive matrix factorization (PMF) model

PMF 5.0 was selected for source apportionment in this study (Paatero and Tapper, 1994). The positive matrix factorization (PMF) model employs non-negative constraints to quantify the contribution of potential sources to the samples, and considers the error estimation. It overcomes the occurrence of unexplained factors such as non-negative factors that are prone to appear in other receptor models such as principal component analysis (PCA). PMF analyzes source factors by a weighted least-squares fit method (Zhang et al., 2019b). Positive matrix factorization analysis comes with bootstrap uncertainty analysis, which is the best choice for quantitative analysis of pollutant sources.

PMF is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into two matrices: factor contributions (G) and factor profiles (F). These factor profiles need to be interpreted by researchers to identify the source types that

may be contributing to the samples using measured source profile information, and emissions or discharge inventories. As input files, data set can be viewed as a data matrix X of *i* by *j* dimensions, in which *i* number of samples and *j* chemical species were measured, with uncertainties *u*. The goal of receptor models is to solve the chemical mass balance (CMB) between measured species concentrations and source profiles, as shown in Eq. (1), with number of factors *p*, the species profile *f* of each source, and the amount of mass *g* contributed by each factor to each individual sample:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad \text{Eq. (1)}$$

where e_{ij} is the residual for each sample/species.

The residual error matrix *eij* is calculated by the minimum value of the objective function *Q* in Eq. (2):

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{u_{ij}}\right)^2 \quad \text{Eq. (2)}$$

where u_{ij} is the data uncertainty calculated using a fixed fraction of the species-specific method detection limit (*MDL*) (Men et al., 2020; Wang et al., 2019b).

Q is a critical parameter for PMF with two versions of *Q*, *Q(true)* and *Q(robust)*, providing the assessment of model stability. To run the model, it requires the proper species concentration and uncertainty files. The uncertainty was calculated using the following Eqs. (3-4):

If concentration < *MDL*, $u_{ij} = \frac{5}{6} \times MDL$ Eq. (3)

Otherwise, $u_{ij} = \sqrt{(Error\ \ Fraction \times concentration)^2 + (0.5 \times MDL)^2}$ Eq. (4)

Where concentration is the content of species, *MDL* and Error Fraction (relative standard deviation) are shown in Table S3.

The signal-to-noise ratio (S/N) of all elements was >1.0. The values of scaled residuals for all the elements were between−3 and +3, and were normally distributed. The model *Q* (robust) and *Q* (true) values were fitted and compared, and the difference was less than 3% which meant the model run stably and the data fit well. The standard error (*SE*) between the observed and predicted values was less than 0.1. Through Bootstrapping (BS) and Displacement (DISP) error estimation, the uncertainty of model could be explained and accepted, and the number of factors was also appropriate.

1.4 Human exposure and health risk assessment

Based on the US Environmental Protection Agency health risk handbook (EPA, 2001a), the exposure assessment model was used to assess the potential risks of toxic elements on road dust to human health. According to the model, people exposed to metals on road dust could occur through three main pathways: direct ingestion (ADD_{ing}), dermal absorption (ADD_{der}) and inhalation (ADD_{inh}) (Rahman et al., 2019). The average daily dose received by adults and children was evaluated by formula calculation (Zhao et al., 2014). The parameters in the numerical expressions of cancer and non-cancer risk assessments for humans was selected from the US Environmental Protection Agency health risk handbook (EPA, 2001a; 2004; 2009a) and the Exposure factors handbook of Chinese Population (Ministry of Ecology and Environment, 2013). The potential health effects of metals were described as non-carcinogenic and carcinogenic risks according to the exposure assessment. The non-carcinogenic risks of

six metals to human health were evaluated by the Hazard Index (HI) and Hazard Quotient (HQ) (Bourliva et al., 2016; Zheng et al., 2010). According to the Agents Classified by the International Agency for Research on Center (IARC) Monographs, Cr and Pb were carcinogenic hazards to humans and the associated carcinogenic risk were assessed by Incremental Lifetime Carcinogenic risk (ILCR).

The particle emission factors from soil in the risk assessment model were replaced by the emission of road fugitive dust to avoid underestimating potential health risks (Tian et al., 2019). The AP-42 model (U.S. EPA, 2001) and the Line Sources model were used to evaluate the particle emissions from road surface in different functional areas, and the detailed model was displayed in the SI. The relative bioavailability (RBA) of metals, which represented the bioavailability fraction of the metal content (%), was also essential to avoid overestimating health risks (Table S4) (Hou et al., 2019). Due to the limitations of experimental conditions, the ratio of metal bioavailability was referred to previous research results in Tianjin (Hou et al., 2019; Hu et al., 2011; Yu et al., 2014). In addition, the exposure time and body exposure area of people in different seasons were also considered into the model in this study.

1.4.1 Exposure dose

The daily intake dose (ADD) of each metal through three pathways was calculated by using Eqs (5-7).

$$ADD_{ing} = C \times \frac{IngR \cdot ET \cdot EF \cdot ED}{BW \cdot AT_{non}} \times 10^{-6} \quad \text{Eq. (5)}$$

$$ADD_{inh} = C \times \frac{InhR \cdot ET \cdot EF \cdot ED \cdot RED}{BW \cdot AT_{non}} \times 10^{-9} \quad \text{Eq. (6)}$$

240 $$ADD_{der} = C \times \frac{AF_d \cdot SA \cdot ABS \cdot ET \cdot EF \cdot ED}{BW \cdot AT_{non}} \times$$

241 10^{-6} Eq. (7)

242 Where, ADD is the average daily dose through three routes (mg/(kg·day)). *C* in
243 Eqs. (5-7) is the metal concentrations on road dust in mg/kg. *IngR* is the ingestion rate
244 of road dust in mg·day^{-1}. *InhR* is the inhalation rate in m^3·day^{-1}. *ET* is the exposure time
245 of day (h/d). *EF* is the exposure frequency in day/year. *ED* is the exposure duration,
246 years. *RED* is the exposure dose of road dust suspended from the road surface in ug/m^3.
247 *SA* is the exposed skin area in cm^2. AF_d is the skin adherence factor in mg·cm^{-2}·day^{-1}.
248 *BW* is the body weight of human and child in kg. *AT* is the average exposure time in
249 hours (h). *ABS* is the dermal absorption factor (unitless). The parameters are shown in
250 Table S4a.

251 1.4.2 Hazard index (HI)

252 HQ and HI values can be calculated by the following equations (8-12) from EPA
253 guidance documentation to assess the health risks with a threshold pollutant (EPA):

254 $$HQ_{ing} = \frac{ADD_{ing} \cdot \quad RBA}{RfD_{ing}} \qquad \text{Eq. (8)}$$

255 $$HQ_{inh} = \frac{ADD_{inh} \cdot RBA}{RfD_{inh}} \qquad \text{Eq. (9)}$$

256 $$HQ_{der} = \frac{ADD_{der} \cdot RBA}{RfD_{der}} \qquad \text{Eq. (10)}$$

257 $$HI = \sum_{1}^{i} HQ \qquad \text{Eq. (11)}$$

258 $$HIt = HIx \qquad \text{Eq. (12)}$$

259 Where, i is the exposure pathway. *x* is the relevant metal species. *RfD* is the
260 reference permissible daily intake dose in mg/(kg·d) for ingestion and dermal contact

or mg/m^{-3} for inhalation. Relative bioavailability (*RBA*) of the metals was shown in Table S4b. The HQ calculated for each exposure route was added to obtain HI to assess the potential non-carcinogenic risk effects of specific metal. HIt was used to estimate non-carcinogenic risks for multiple metals (Zhang et al., 2019a). HI value less than 1 suggests no significant non-carcinogenic risk.

1.4.3 Incremental Lifetime Carcinogenic risk (ILCR)

Pb and Cr, as non-threshold contaminants, were assessed by the Incremental Lifetime Cancer Risk (ILCR) for the carcinogenic risk (CR) effect. It was used to measure the probability of carcinogenic risk during a 70-year lifetime continuous metal exposure. It was calculated using Eqs. (13-15):

$$LADD_{inh} = \frac{C \cdot ET \cdot EF \cdot RED}{AT_{car}} \times \left(\frac{InhR \cdot ED}{BW}\right)_{child} \quad \text{Eq. (13)}$$

$$LADD_{inh} = \frac{C \cdot ET \cdot EF \cdot RED}{AT_{car}} \times \left(\frac{Ingh \cdot ED}{BW}\right)_{adult} \quad \text{Eq. (14)}$$

$$ILCR = LADD \times RBA \times SF \quad \text{Eq. (15)}$$

Where, *ILCR* is the incremental lifetime cancer risk. *LADD* is the lifetime average daily dose exposed by the inhalation pathways (μg/m^3). *ET* is the exposure time (h/d). *SF* is the cancer slope factor. Carcinogenic risks in the range of 1.0×10^{-6} to 1.0×10^{-4} were considered non-negligible and should be controlled through risk management decisions (Tian et al., 2019).

1.4.4 Source contribution for risk assessment

According to the source apportionment resulted from the PMF model, the new

combination of PMF - ILCR was used to assess the source contribution to human health risks, which was calculated as follows:

$$S[ILCR]_{jk} = S[LADD]_{jk} \cdot SF \quad \text{Eq. (16)}$$

$$S[HQ]_{jk} = \frac{S[ADD]_{jk} \cdot \quad RBA}{RfD} \quad \text{Eq. (17)}$$

$$SC_{jk} = C_j \cdot S_K \quad \text{Eq. (18)}$$

Where, $S[ILCR]_{jk}$ and $S[HQ]_{jk}$ is the risk exposed in the *jth* metal and contributed by the *kth* pollution source for carcinogenic and non-carcinogenic effects, respectively. $S[LADD]_{jk}$ and $S[ADD]_{jk}$ is the average daily dose of the *jth* metal contributed by the *kth* pollution source (mg/kg/d). *C* in Eqs. (5-7) and Eqs. (13-15) presenting metal concentration is replaced by SC_{jk}. C_j is the *jth* metal concentration in road dust (mg/kg). S_k is the contribution proportion of the *kth* source to the metal concentration (%). *SF* is the cancer slope factor, and *RBA* is the relative bioavailability of metal which shown in Table S4b.

1.5 Statistical analysis

The figures and data analysis in this study were completed in the GraphPad Prism 8 software. The concentration difference of metals on road dust between different seasons is expressed by coefficient of variation (CV) and the equation is as follows:

$$CV = \frac{sd}{mean} \times 100\%$$

2. Results and discussion

2.1 Concentration characteristics of heavy metals on road dust

Figure 1 displays the seasonal concentration distribution of metals in functional areas. The concentrations of Fe and Mn in both the traffic area (JT) and the residential

area (JM) were relatively higher except in spring. While the concentrations of Pb, Zn, and Cr were prominent in the commercial area (SY) and the central commercial street (BJ). The concentration of Cu in different sites was most affected by the season, with the highest concentration in commercial area (SY) in spring and autumn, but the highest concentration in residential area (JM) in summer and the highest concentration in the central commercial street (BJ) in winter. Obviously, the metal concentrations were significantly affected by the ambient environment of different functional areas (Huang et al., 2018a). A pronounced seasonal variation was also observed in metal concentration distributions. The concentrations of most metals were lower in spring, but higher in other seasons. Many studies have confirmed the seasonal differences in metal concentration (Men et al., 2018). The concentrations of Fe, Mn, Pb and Cu varied greatly among functional areas in summer with a large Coefficient of Variation (CV), while the concentrations of Zn and Cr differed significantly among functional areas in autumn (Table S5). This might be related to the extensive human activities in summer and autumn, like construction activities and industrial activities (Skrbic et al., 2019).

Compared with metals Zn, Cr, Cu, and Pb, metals Fe and Mn were found to have the highest concentrations on road dust. The average concentrations of Fe and Zn in most areas were more than five times of the soil background value in Tianjin (Table S6), and the concentrations of Pb and Cu on road dust were more than twice (CNEMC). While the concentrations of Mn and Cr in most areas were lower than the background values. The concentration of Mn was similar to that in other cities of China, but it was higher than that in London and lower than that in Honolulu (Robertson et al., 2003;

Sutherland and Tolosa, 2000). Compared with the metal concentrations studied in other cities in China or other countries (Table S6), the concentration of Fe in this study area was significantly lower than that in Beijing (Men et al., 2020), but it was close to the findings in Ottawa Canada and Queensland Australia (Gunawardana et al., 2014; Rasmussen et al., 2001). The concentration of Pb in study areas was similar to Birmingham UK (Charlesworth et al., 2003), but significantly lower than that in other cities such as New York US, Tokyo Japan and Bhopal India, and Chinese cities such as Shanghai and Xi'an (Banerjee, 2003; Fergusson and Ryan, 1984; Shi et al., 2008; Wijaya et al., 2012; Yongming et al., 2006). The concentration of Zn in study areas was comparable to the survey results of most cities in the world. The concentrations of Cu and Cr were lower than those in the cities of China, but higher than those in Ottawa Canada (Rasmussen et al., 2001). This might affect by factors such as the geological conditions, economic development and intensity of human activity in different cities，which need further research (Hou et al., 2019).

2.2 Seasonal and regional source identification

A receptor model (PMF) has been widely used to identify and quantify the main sources of elements on road particles. However, the contribution of the sources to the elements which might affected by the seasons, was not well studied in previous literature (Men et al., 2020). Five major factors were identified for the elements on road dust in different seasons, including atmosphere deposition, fertilizer, domestic waste, industrial and construction activity, and traffic non-exhaust emission. The contribution ratios of factors to different elements on the road dust were calculated by Eq. 1.

The contributions of factors to the elements on road dust in different seasons are shown in Table 1. The first factor was characterized by a high contribution of NO_3-N which does not depend on the seasons. As an important component in the air, nitrogen oxides might come from the burning of fossil fuels, or the natural behaviors in the atmospheric environment (thunderstorms, photochemical reactions, etc.) (Giroux et al., 1997; Lin et al., 2015). In recent years, the increasing nitrate in atmospheric particles could accumulate on the road surface as sediment (Feng et al., 2021). Therefore, factor 1 was identified as atmosphere deposition. The second factor was dominated by NH_3-N (48.6%) in spring, while dominated by TP (53.2% - 61.3%) in summer, autumn and winter. There are many green belts near or on the urban roads, and chemical fertilizer can provide the necessary nitrogen and phosphorus elements for the trees (Salim et al., 2019). The seasonal differences in the contributions of nitrogen and phosphorus to the factor could be explained by the different types of fertilizer. Therefore, factor 2 was identified as fertilizer. The third factor was contributed by total organic carbon (TOC). Considering the characteristics of the study area, there are many commercial activities in the commercial area, and daily life in the residential area, which will generate a lot of domestic waste. Previous studies have found that the runoff from the residential area has significant concentrations of organic carbon (Rhee et al., 2012; Salim et al., 2019). Therefore, the factor 3 was identified as domestic waste. The fourth factor was mainly characterized by Fe (41.6% - 50%) and Mn (42.4% - 50%) in four seasons. Even this factor was also partly contributed by Pb, Cr and Cu in spring, Cr in summer, Cu and Zn in autumn, Cu, Zn and Cr in winter. Fe, Mn and Cu were often used in industrial

activities such as electronic industry, mechatronics and construction (Men et al., 2020). There are many industrial parks near the study area, thus the fine particles could release into the environment by the wind and accumulate on the road surface. The fourth factor was identified as industrial and construction activity. The fifth factor was dominated by Pb (18% - 46.2%), Zn (26.9% - 51.4%) and Cr (29.6% - 44.1%). Cu could partly contribute to the factor in spring and winter. Restricted use of leaded gasoline and diesel has significantly reduced the content of Pb in vehicle exhaust emissions (Hong et al., 2020). Pb, Zn, Cr and Cu were widely contained in brakes, tires, and pavements (Huang et al., 2018b; Men et al., 2020). Therefore, the fifth factor was regarded as traffic non-exhaust emission. The main sources (five factors) of measured elements in road dust in study city were not affected by the seasons. However, the relative contribution percent of each element to the factors varied significantly with seasons. Therefore, research on the risk assessments of pollutant sources in specific season is more necessary.

Affected by different human activities, the source contribution percent in different study sites is also fluctuant with seasons (Table S7). Due to the relatively higher green area in residential areas (JM), fertilizer was a major source in different seasons. In addition, atmospheric deposition was also an important source in summer and autumn, while domestic waste and industrial activities were important sources in spring and winter, respectively. For the traffic area (JT), construction activity was the major source in spring, summer and autumn. This might be due to the large number of construction activities beside the road during the study period. This source feature was consistent with the characteristics of construction activities, which activities would decrease in

393 winter. Besides, domestic waste became the main source in winter. What was
394 interesting was that traffic non-exhaust emission is not an important source, even
395 though the traffic volume in the traffic area was very large. It was presumed that express
396 way had no traffic lights on the road, which reduced the abrasion of brakes, tires, and
397 pavement. Research indicated that traffic congestion could increase vehicle emissions
398 and reduce the air quality (Zhang and Batterman, 2013). Furthermore, the higher
399 contribution of construction activities to the measured elements on road dust made the
400 contribution of traffic non-exhaust emissions insignificant. Compared with the central
401 commercial street (BJ), commercial area (SY) had reduced business activities and
402 traffic volume. However, the vehicles were constantly braking at intersections due to
403 the narrow roads. As a result, non-exhaust emissions from traffic became the main
404 source in commercial area (SY) in different seasons. In addition, fertilizer and domestic
405 waste were also important sources of elements in autumn. Traffic non-exhaust emission
406 was the main source of central commercial street (BJ) in winter. Due to more
407 commercial activities and human flow, domestic waste became the main source in
408 spring and summer. The fertilizer and atmospheric deposition were also the main
409 sources in spring and autumn, respectively.

410 From the perspective of overall measured elements on road dust, the pollutant
411 sources in this study city did not vary with the seasons (Figure S2). However, the major
412 pollutant source in the particular study area were significantly affected by the seasons.
413 Seasons might indirectly affect the pollution sources of road dust by directly affecting
414 the economy activities, human activities, environmental policy, or other factors, which

need to be further studied.

2.3 Seasonal non-carcinogenic risk health assessment

The health risks of road dust were evaluated by summarizing the risks of different groups of people exposed to metals on road dust in various functional areas and seasons. Table S8 shows the Hazard Quotient (HQ) and Hazard Index (HI) for adults and children exposed to six metals on road dust through three pathways. The potential health risks of metals to adult in different seasons and functional areas are presented in Figs. 2-3. Figs. S3-S4 focus on the potential non-carcinogenic risk to children. HI <1 was considered to be free of non-carcinogenic risks. The results showed that the non-carcinogenic risk for adults caused by a single metal through any exposure route or the exposure to six metals was less than 1. It was confirmed that the target metals would not pose a significant threat to the adult health. And the metals on road dust mainly effected adult health through the ingestion route (Figure 2). Especially for metals Pb and Mn, the HI values were 2.6 E-03 and 1.3 E-03 respectively. Although the evaluation results were lower than the risk threshold, it still needed to pay more attention to these two metals which often used in the construction activity and vehicle fabrication. Many studies have implied that Mn was an important hazardous element and could cause a higher non-cancer risk (Dietrich et al., 2019; Rahman et al., 2019; Tian et al., 2019). A high concentration of Mn was determined in many cities in China (Sutherland and Tolosa, 2000; Yang et al., 2010; Yongming et al., 2006), and it was also true in this study. It is worth noting that the health risks induced by Mn need advanced investigation based on the doses and concentration levels (Alves et al., 2017). Then, the

metals that caused the health risks were in the order of Zn>Fe> Cr > Cu. Excessive intake of Fe might also have negative impacts on human health, although Fe was always an essential element to human body. There have been many reports on the relationship between excess iron and heart disease (Ghafourian et al., 2020; Wood, 2004).

In addition, the potential non-carcinogenic risks to children was found to be higher than that of adults. Children might be more vulnerable than adults for ingestion rate, inhalation exposure pose and dermal exposure. The exposure route through ingestion was also the main way for children to face the health risks. The non-cancer risks for children caused by exposure to any single metal or the exposure to six metals on road dust were all below the risk threshold. However, it was worth noting that high non-cancer risks exposed to six metals were undoubtedly a serious potential peril to children health. Fe was the main metal causing non-cancer risk, followed by Mn and Zn, and the next lower level was Pb, Cu and Cr. Obviously, compared with the high non-cancer risk to children exposed in Kuala Lumpur (Wahab et al., 2020), Hubei Province (Zhang et al., 2019a) and Gary (Dietrich et al., 2019) areas, the health risks to children exposed in this study area were relatively low. Moreover, the main exposure route for children in this study area was ingestion which was same as the investigation in Dhaka (Rahman et al., 2019) and Queensland (Jayarathne et al., 2018) areas, while differed from that in Serbia (Skrbic et al., 2019) which were mainly through dermal contact. It was worth noting that the Hazard Quotient of metals for human was related to the number of the investigated metal species and the above discussion on the non-cancer risks was based on six target metals.

459 The non-carcinogenic risks for adults (Figure 2) and children (Figure S3) varied
460 significantly with seasons, and the variation pattern was basically similar. For most
461 metals, road dust in the summer posed more serious potential health risks through three
462 exposure pathways. The potential health risks in spring were notably reduced. However,
463 the risk characteristics of Cr exposure were not the same, which exhibited the highest
464 health risks in winter and the smallest risks in spring. The relatively higher metal
465 concentration on road dust in summer than in spring might be one of the important
466 causes for serious potential health risks presented in summer in this study. Another
467 important factor was probably due to the increased exposure time and exposure area of
468 human skin during the summer. Non-carcinogenic risk assessments caused by dermal
469 contact also proved that the increase in exposure area of human skin in the summer
470 greatly raised the potential health risks (Table S3a and Figure 2). There were few
471 studies on seasonal non-cancer risk assessment exposed to metals on road dust (Men et
472 al., 2020; Skrbic et al., 2019).
473 In view of the differences in population, traffic flow and human activities between
474 the functional areas, the mass of build-up particles on the road, the metal concentration
475 and the emission factors of road dust were different (Liu et al., 2019; Patton et al., 2017).
476 Except for Fe and Mn, metals on road dust in commercial areas mainly caused the
477 highest potential health risks through ingestion. The health risks caused by the ingestion
478 of metals Fe and Mn on road dust in traffic area were highest. According to the source
479 analysis, the industrial and construction activity was the main source of Fe and Mn on
480 road dust. There were always construction activities near the traffic area, which might

be the vital reason for the higher potential risks in traffic area. In addition, all metals in the traffic area displayed the highest potential non-cancer risk through direct inhalation route, which was mainly related to the large amount of suspended road dust caused by the dense traffic. Fe, Mn and Zn in the traffic area, Pb and Cr in the commercial area, and Cu in the residential area all posed threat to human health through dermal contact. Due to the low traffic volume in residential area, the potential health risks exposed to various metals through direct inhalation were relatively low. Timely road sweeping carried out in central commercial street and thus the accumulated particle load was less. Therefore, the health risks caused by dermal contact in central commercial street were low, even if the traffic volume was heavy.

2.4 Seasonal carcinogenic risk health assessment

Cr and Pb were still widely used in industry and daily life, and they were considered as carcinogenic elements for the Incremental Lifetime Cancer Risk (ILCR) of adults and children (Table S9). ILCR between 10^{-6} and 10^{-4} was determined to be an acceptable range for carcinogenic risk but could not be ignored. Exposure to metal Pb on road dust through the direct inhalation route did not pose a significant cancer risk for both adults and children. However, the cancer risk caused by metal Cr on road dust exceeded the minimum threshold (10^{-6}) for adults and children in a probability larger than 5%. Most studies have shown that Cr was within the cancer threshold (Jayarathne et al., 2018; Rahman et al., 2019). These evaluation results were enough to attract people's attention and take measures.

Adults and children exposed to Pb in road dust had relatively high potential cancer

503 risks in summer and autumn, while exposure to Cr posed comparatively high potential
504 cancer risks in winter (Figure 4). Undoubtedly, the traffic areas with a lot of road dust
505 would bring high potential cancer risks to adults and children through the direct
506 inhalation exposure route. This was followed by the central commercial area, which
507 had a relatively higher traffic volume. Identifying metals that made principal
508 contributions to human health risks in different seasons and functional areas was
509 essential to effectively recognize source control targets and improve risk control
510 efficiency.

511 **2.5 Critical sources accounting for human health risk**

512 Particular metals were believed to have special risk factors for health risks.
513 Pollutant sources that contributed a lot to the metals on road dust might not necessarily
514 have a large impact on health risks. Figure 5 depicted the contribution proportion of
515 metal sources to Hazard Index (HI) and Incremental Lifetime Cancer Risk (ILCR) for
516 adults and children in different seasons. Even if the concentration proportion of each
517 source fluctuated significantly due to the seasons, industrial and construction activity
518 was the main source for the metals on road dust (29% - 47%) and the HI of adults (27%
519 - 45%) and children (41% - 50%) in different seasons. During the study period, there
520 were a lot of construction activities near the study area. Therefore, the potential risks
521 from the construction activities to the surrounding human health could not be ignored
522 (Khademi et al., 2019; Liu et al., 2014). It is worthwhile for the government and people
523 to take active measures to reduce the risks. In addition, traffic non-exhaust emission in
524 spring and domestic waste in summer were also major sources of pollution that pose

potential health risks.

Unlike the hazard index, elements with carcinogenic risks were more likely to get attention. And it was obviously that the main pollution sources for the carcinogenic risks were different from the Hazard Index (Figure 5). The main source that posed carcinogenic risks to adult's health in spring and summer was traffic non-exhaust emission. The industrial and construction activity in winter and domestic waste in autumn were important sources for the potential carcinogenic risks. Traffic non-exhaust emission and industrial and construction activity also made significant contributions to the potential carcinogenic risks in winter and spring, respectively. Affected by the height of child, the exposure way and amount of metals for children might be different from those for adults (Huang et al., 2018a). In addition, Pb and Cr posed different potential carcinogenic risks for children and adults. However, the main pollution sources that pose carcinogenic risks to children was similar to that of adults in different seasons.

It was worth noting that the analysis of pollution sources in this study was only based on the assessment for the carcinogenic risks of Pb and Cr. If the carcinogenic risks of more metals were considered, there might be a little error in the contribution of pollution sources. Considering the Hazard Index and Carcinogenic Risks, industrial and construction activity and traffic non-exhaust were the critical contribution sources. Identifying specific contribution sources for risk assessment is of great significance for effectively controlling risk sources and reducing potential risk.

3. Conclusion

This study comprehensively evaluated the concentration characteristics, source apportionment and health risks of metals on road dust. The metal exposure mainly posed a hazard to human health through ingestion route. Pb and Mn, Fe and Mn were the main harmful elements that caused non-cancer risks for adults and children, respectively. The cancer risks for adults and children exposed to Cr exceeded the minimum threshold. Higher cancer risks for adults and children exposed to Pb were shown in summer and autumn, while shown in winter exposed to Cr. Five major concentration factors were identified for the elements on road dust in different seasons based on the PMF model, including atmosphere deposition, fertilizer, domestic waste, industrial and construction activity, and traffic non-exhaust emission. More importantly, the major contribution sources for metals on road dust and health risks for adults and children in different seasons might be inconsistent. By combining the PMF model and health risk models, industrial and construction activity was identified as the critical source of both the pollutants on road dust (29% - 47%) and the HI for adults (27% - 45%) and children (41% - 50%) in different seasons. However, traffic non-exhaust emission dominated in the carcinogenic risks for children and adults in spring (45%) and summer (36%). Different source control regulations should be taken in different seasons. It is essential for effectively identifying source control targets and consequently improving risk control efficiency. Considering the impacts of the amount of metals on risk assessment, more metals will be investigated in the future research.

DECLARATIONS

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

The authors declare that all the datasets generated during and/or analysed during the current study are available in the article and its SI Appendix.

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

Jingshu Wang: put forward ideas, performed the experiments and wrote the initial draft.

Jinhui Jeanne Huang: provided study materials, reagents and materials, supervised research activity, and revised draft.

Catherine Mulligan: revised draft.

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Table 1. Relative contributions of the measured elements on road dust to the factors (1-5) in different seasons.

	Spring (%)					Summer (%)					Autumn (%)					Winter (%)				
	F1	F2	F3	F4	F5	F1	F2	F3	F4	F5	F1	F2	F3	F4	F5	F1	F2	F3	F4	F5
Fe	6.7	8.1	18.4	41.6	25.3	14.5	3.1	34.8	47.7	0	29.8	17.3	8	44.5	0	18.7	8.4	2.9	50	20
Pb	0	10.4	6.4	37	46.2	16.2	0	36.2	14.4	33.3	19.8	24.6	20	17.5	18	13.1	4.5	3.8	39.1	39.5
Cu	9.6	0	27.7	31.2	31.4	22.1	5.7	36.6	21.9	13.8	17.6	14.7	16.8	40.4	10.5	14.4	3.6	0	49.9	31.8
Zn	5.9	14.8	10	26	43.4	16.5	10	37.3	9.3	26.9	0	0	8	40.5	51.4	16.6	1.3	0	46	36.1
Cr	3.9	4.6	8.5	39	44.1	7.8	17.2	0	36.5	38.5	11	27.4	37.4	24.2	0	21.1	0	3.8	45.5	29.6
Mn	0	12.5	21.1	49.2	17.2	13.2	6.8	28.7	43.2	8.1	32.4	20.1	0	42.4	5.2	22.3	9	2.2	50	16.5
TOC	15.7	2.4	53.1	9.4	19.4	0	10	74	0	16	0	41.8	57.8	0	0	0	1	44.5	23.5	31
NH_3-N	22.6	48.6	11.3	0	17.6	13.9	28.3	21.9	19.4	16.4	24.7	36.3	16.3	15.7	7.1	22.9	0	0	59	18.1
NO_3-N	65.9	1.1	0	33.1	0	60.2	31.4	8.4	0	0	71.1	0	21.3	0	7.3	74.2	2.8	23	0	0
TP	11.2	7.9	26.6	26.2	28.1	0	61.3	20.9	0	17.1	22.6	53.2	0	0	24.1	11.1	61.3	0	0	27.6

Figure captions

Figure 1. Histogram of metal concentrations on road dust in different seasons and functional areas (including metal concentrations for different size particles). JM, JT, SY and BJ represent residential area, traffic area, commercial area and central commercial street, respectively.

Figure 2. Contribution of metals to non-carcinogenic risks of adult through three exposure pathways in different seasons.

Figure 3. Contribution of metals to non-carcinogenic risks of adult through three exposure pathways in different functional areas.

Figure 4. Carcinogenic elements display potential health risks to adults and children in different seasons and functional areas.

Figure 5. Source contributions of metals to Hazard Index (HI) and Incremental Lifetime Cancer Risk (ILCR) for adults and children in different seasons. a) HI for child; b) HI for adult; c) LICR for child; d) LICR for adult.

Figures

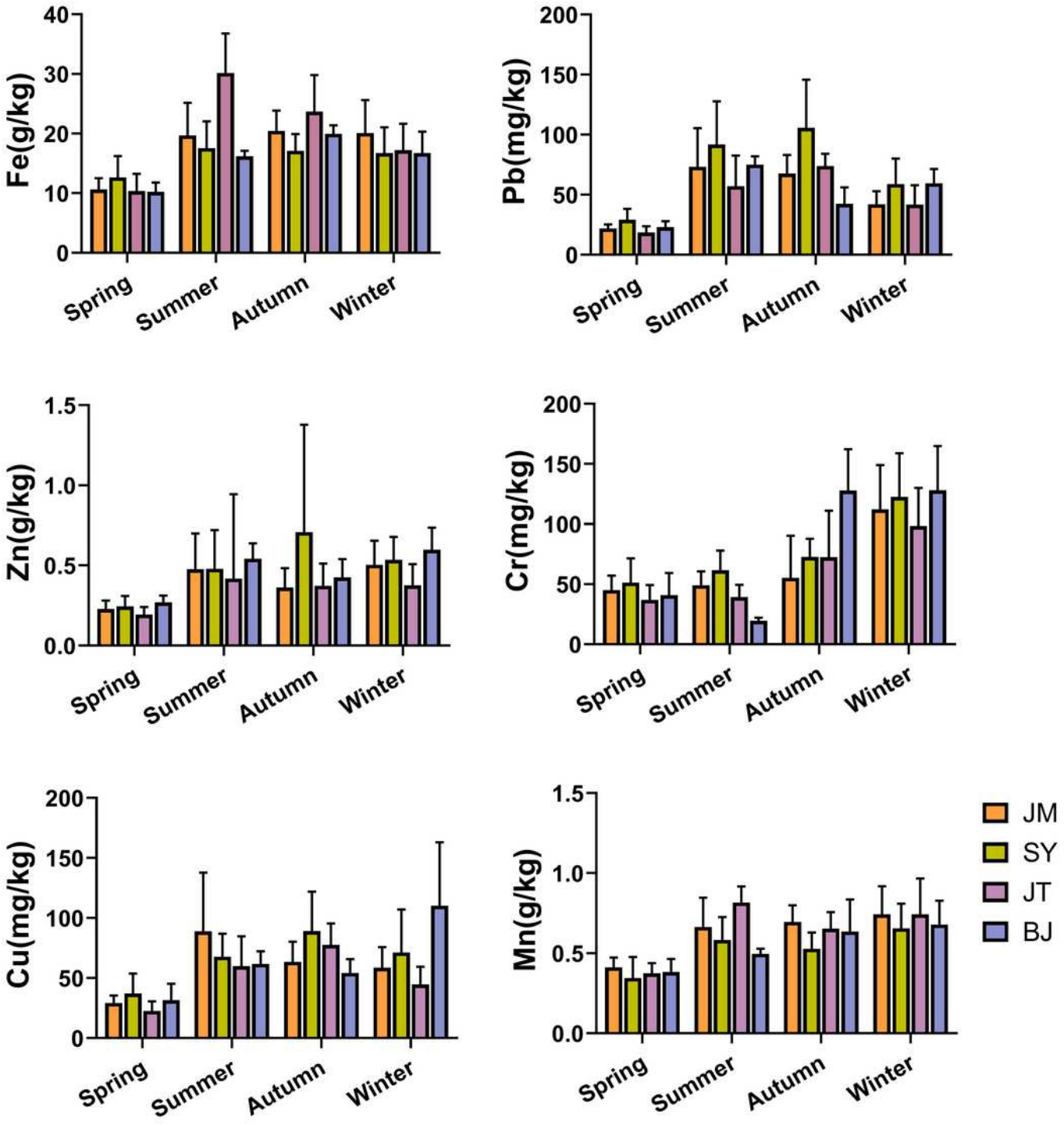

Figure 1

Histogram of metal concentrations on road dust in different seasons and functional areas (including metal concentrations for different size particles). JM, JT, SY and BJ represent residential area, traffic area, commercial area and central commercial street, respectively.

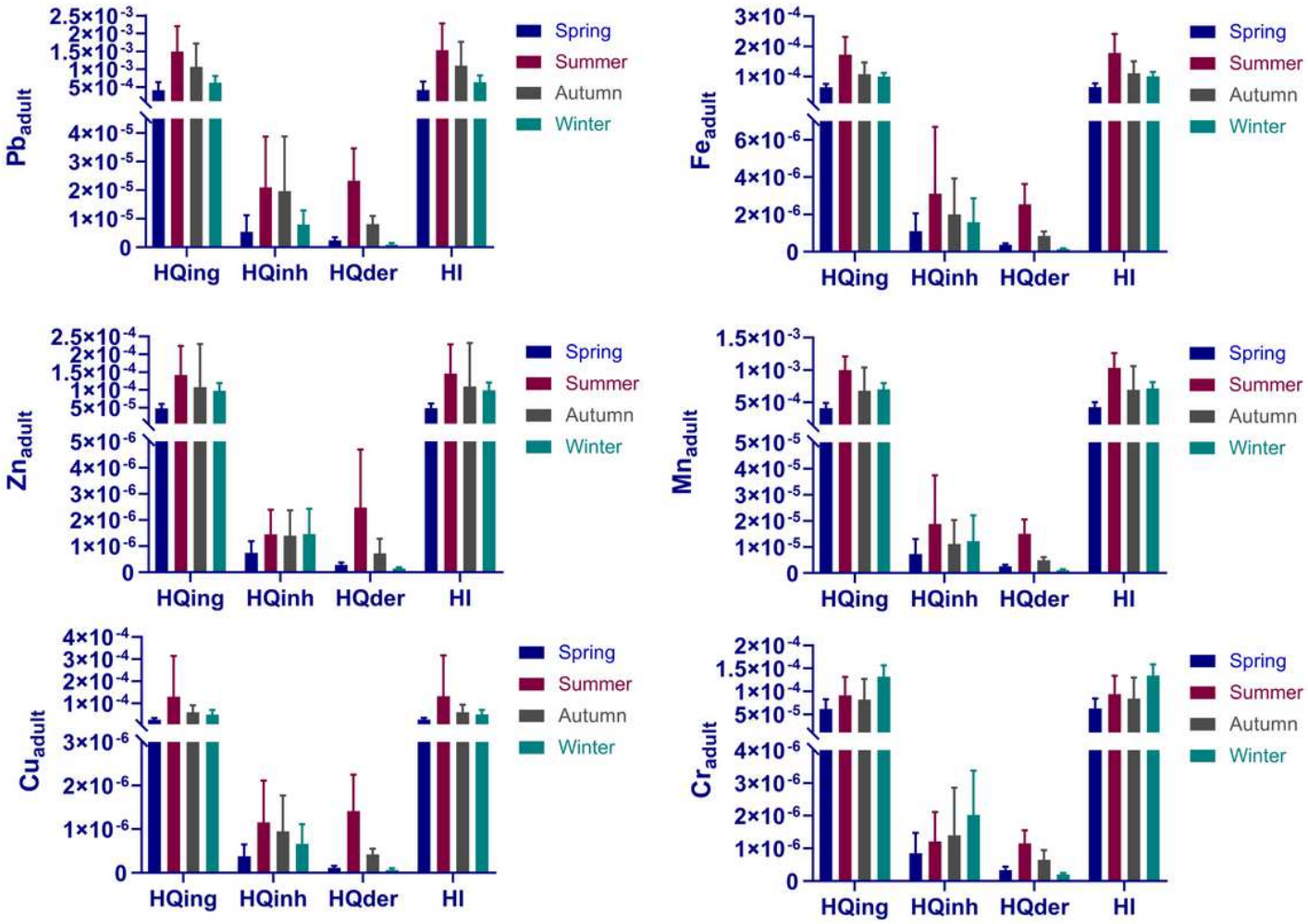

Figure 2

Contribution of metals to non-carcinogenic risks of adult through three exposure pathways in different seasons.

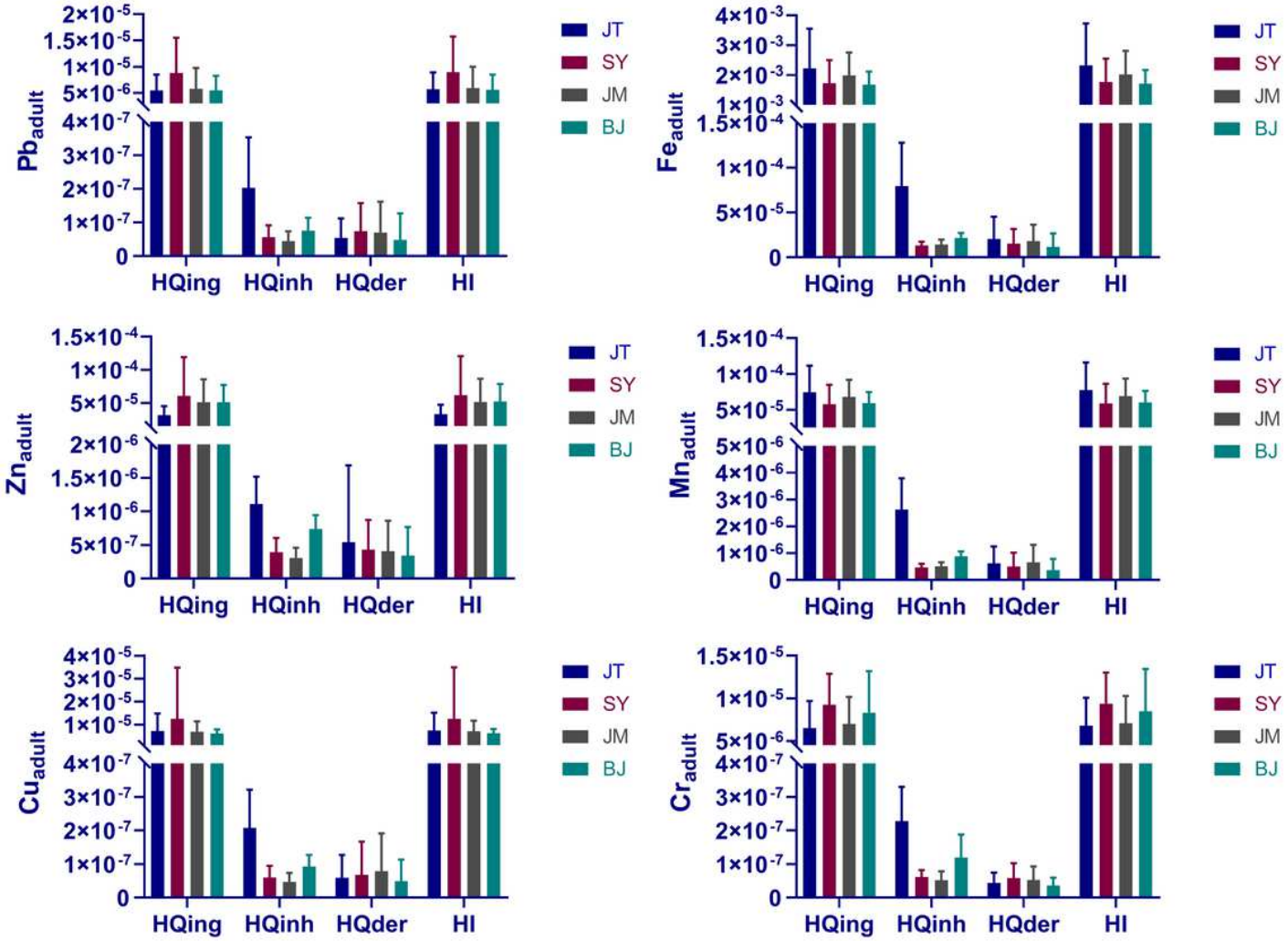

Figure 3

Contribution of metals to non-carcinogenic risks of adult through three exposure pathways in different functional areas.

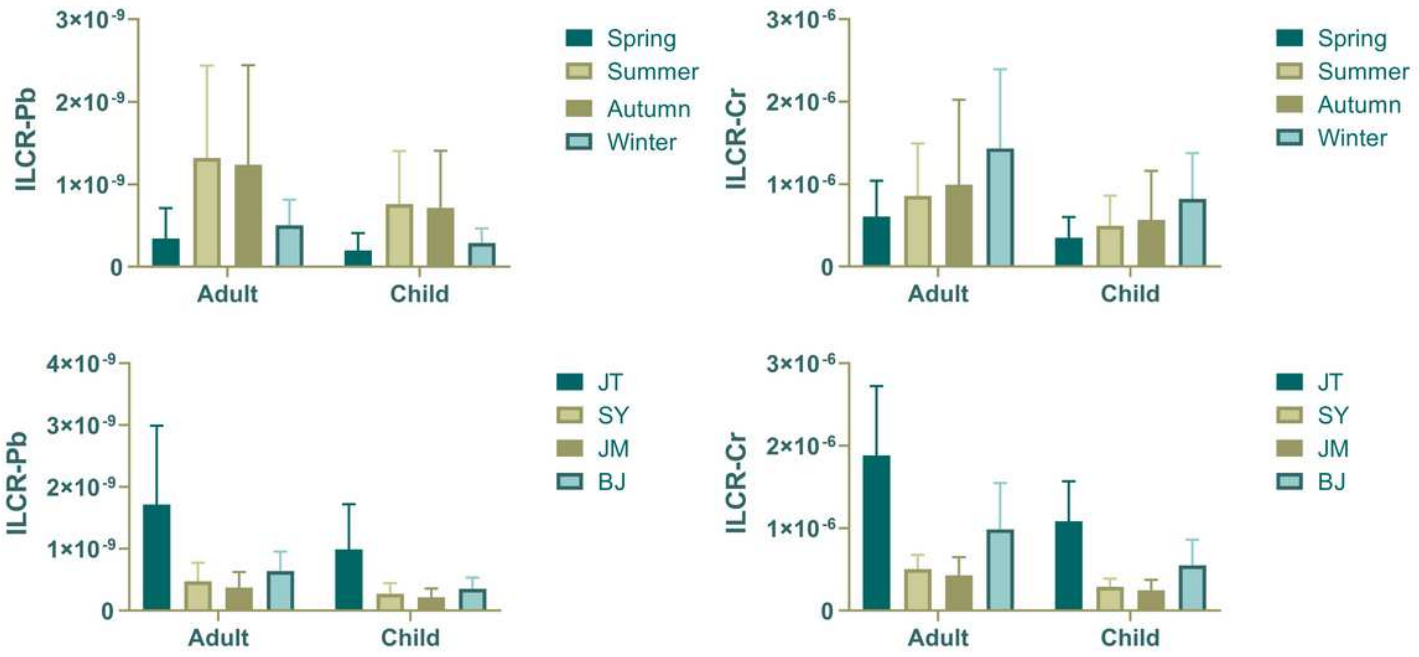

Figure 4

Carcinogenic elements display potential health risks to adults and children in different seasons and functional areas.

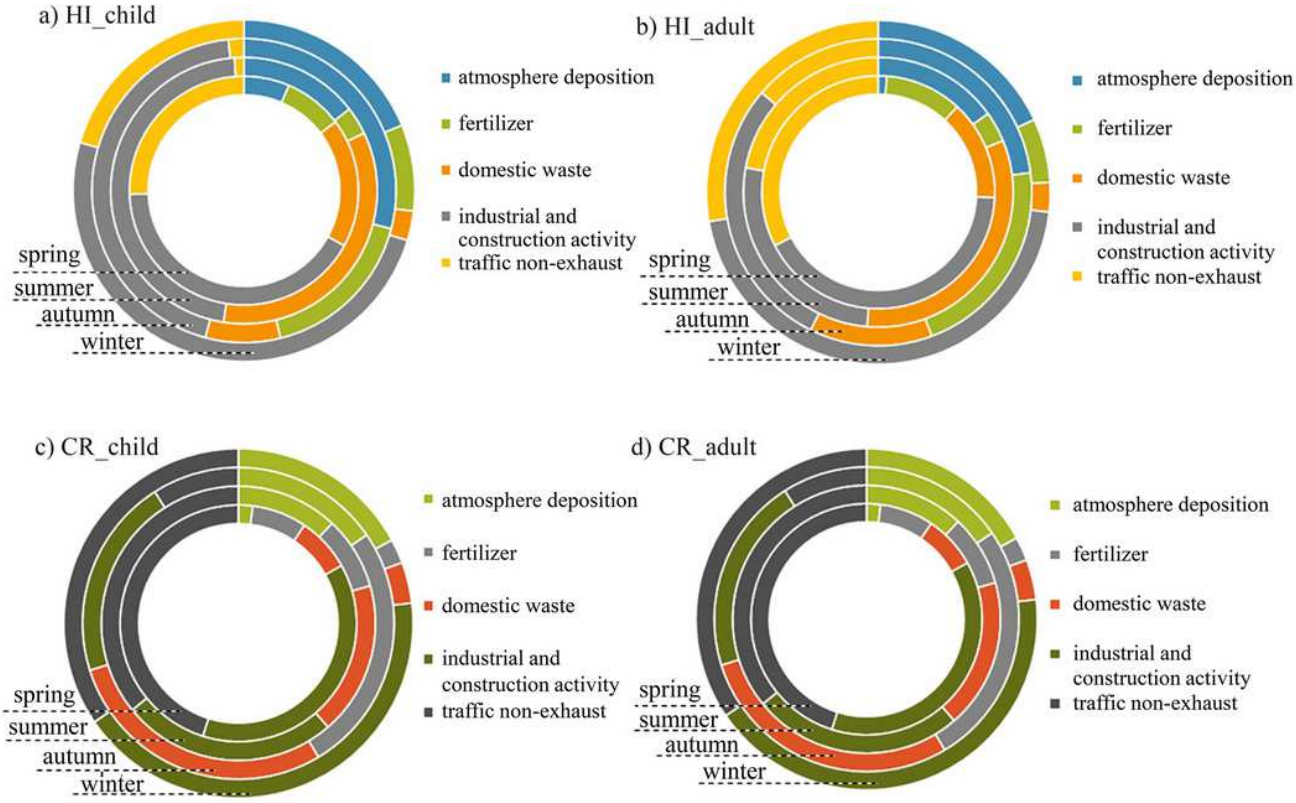

Figure 5

Source contributions of metals to Hazard Index (HI) and Incremental Lifetime Cancer Risk (ILCR) for adults and children in different seasons. a) HI for child; b) HI for adult; c) LICR for child; d) LICR for adult.

Supplementary Files

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- SI.docx