

# Seasonal concentrations, contamination levels, and health risk assessment of arsenic and heavy metals in the suspended particulate matter from an urban household environment in a metropolitan city, Beijing, China

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**Abstract** The levels and health risks of arsenic and heavy metals (As, Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn) in the suspended particulate matter (SPM) collected from an urban household environment in Beijing of China for 12 months were investigated. The mean concentrations of the studied toxic elements were higher and lower than crustal abundance and PM<sub>2.5</sub> in the urban outdoors of Beijing. The concentrations of the studied elements displayed significant seasonality. The highest concentrations of the total elements occurred in winter, followed by autumn, while the lowest concentrations were recorded in summer. Based on the calculated values of enrichment factor (EF) and geoaccumulation index ( $I_{geo}$ ), the levels for As and Cu were heavily contaminated, while those for Cd, Pb, and Zn were extremely contaminated. As and Pb might pose risks to children and adults via ingestion exposure. The accumulative risks of multi-elements resulted from dermal contact and inhalation exposures were not negligible. More

attention should be paid to reducing the non-carcinogenic and carcinogenic health risks posed by the toxic elements bound to urban household SPM particles via ingestion, inhalation, and dermal contact exposure.

**Keywords** Suspended particulate matter (SPM) · Arsenic and heavy metals · Seasonal concentrations · Contamination · Health risk · Urban household

## Introduction

In modern societies, people, especially the vulnerable populations, children and the elderly, spend 80 to 90 % of their time indoors, with most of that time spent at home (Simoni et al. 2003; Katsoyiannis and Bogdal 2012). Thus, the quality of the indoor environment plays an important role in human health (Nastov et al. 2003). Due to complicated and diverse sources, indoor environment pollution, particularly indoor air pollution, is more serious than outdoors (Zhao and Hopke 2006). Indoor suspended particulate matter (SPM), a complex mixture, is the most important source of indoor air pollution. Human exposure to SPM gives rise to adverse health effects. Small particles (PM<sub>10</sub> and smaller) can penetrate into the airways, lungs, and alveolar, causing a number of respiratory and cardiovascular inflammations (Pope et al. 2002; Nawrot et al. 2007; Hsieh et al. 2011; Hung et al. 2012). In addition, a broad range of chemical species, ranging from elemental to organic and inorganic compounds, attached to SPM also have negative

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effects on human health (Fergusson 1990; Dasilva and Williams 1991; Moreno et al. 2007; Gupta et al. 2007). Among the inorganic compounds, the most important are the trace elements which originate from outdoor sources, such as crustal materials, road dust, construction activities, fossil fuel combustion, vehicular traffic, industrial emissions, and indoor sources, such as building and decorating materials, smoking, and cooking (Willers et al. 2005; Ahmed and Ishiga 2006; Shah et al. 2006; Liu et al. 2009). For instance, lead could impact the nervous system and cause impairment of the immune system and insomnia (Canfield et al. 2003; Järup 2003; Bouchard et al. 2007), and arsenic could the gastrointestinal tract, respiratory tract, hematopoietic and nervous systems (Al Rmalli et al. 2005). Because indoor SPM is the most important carrier of trace elements in ambient air (Sternbeck et al. 2002; Wang et al. 2013); therefore, trace elements in indoor SPM are one of the most significant contamination sources to human health.

Over the previous decades, numerous studies have been conducted worldwide to determine the concentrations, origins, distributions, and health risks of trace elements in SPM in the outdoors (Mohanraj et al. 2004; Zereini et al. 2005; Haritash and Kaushik 2007; Fang et al. 2014). Limited attention has been paid to the trace elements in indoor SPM (Farghaly and Ghandour 2005; Satsangi et al. 2014). The household environment (homes) is one of the most common but important types of indoor environments for human life. Most studies of arsenic and heavy metals in the household environment have focused on those attached to household dust (Tong and Lam 2000; Chattopadhyay et al. 2003; Hunt et al. 2012; Yoshinaga et al. 2014). However, higher concentrations of trace elements might have been carried by SPM rather than dust due to the finer size and larger surface per unit mass of SPM (Wang et al. 2006; Hu et al. 2012). Additionally, given the variation of sources with time, concentrations of trace elements might vary on the long-term series (year). Studies on the long-term (year) investigation of trace elements in household SPM are even more limited. Therefore, long-term studies on the trace element characteristics of SPM from the household environment are necessary and urgent.

Beijing, the capital of China, is the center of economy, politics, and culture, representing typical cities experiencing rapid increases in population, traffic, energy usage, urbanization, and industrialization. Air pollution has become a serious problem, and the air quality in Beijing is getting worse in recent years. As a serious

example case, Beijing suffered a continuous fog haze in January 2013 that lasted for 26 days. However, little attention has been paid to metal(loid)s in SPM from the household environment in Beijing. Therefore, a study of the characteristics of trace elements in SPM from urban household environments in Beijing is necessary. For a better understanding of the long-term characteristics and health risks of trace elements in SPM from the urban household environment in Beijing, SPM particles from an urban household environment in Beijing have been collected for 12 months, in this study. The seasonal concentrations, contamination levels, and health risks of arsenic and heavy metals in SPM were analyzed. This work is the first report on the characteristics of metal(loid)s in SPM from the urban household environment in a metropolitan city of China.

In summary, the main goals of this study are to (1) analyze the seasonal variations of arsenic and heavy metal concentrations (As, Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn) in SPM from an urban household environment in Beijing and (2) determine the contamination levels and health risks of arsenic and heavy metals in SPM from the urban household environment.

## Materials and methods

### Study area

Beijing is situated at the northern tip of the roughly triangular North China Plain (39.4° N–41.6° N, 115.7° E–117.4° E, a.s.l. 20–1500 m). The city is surrounded by mountains to its west, north, and northeast. Beijing consists of 16 administrative districts (counties), 6 of which are constituted by the urban areas. The urban areas of Beijing are situated in the south–central part of the municipality. Six concentric ring roads spread out through the city, and the 6th Ring Road passes through several satellite towns. The city has a typical monsoon-influenced climate, characterized by a hot and humid summer and generally cold, windy, and dry winter. The average annual temperature is approximately 11.5 °C, and the average annual precipitation is 500–700 mm. Over the past three decades, Beijing has undergone rapid economic development and urban construction, and the urban population has reached over 22 million with approximately 5 million registered motor vehicles. Fourteen subway routes and 895 bus lines have been built and operated.

The sampling site is located in the Jingshiyuan community of the Chaoyang district and is close to the Olympic Forest Park (Fig. 1). The site is near the Beijing–Tibet (G6) highway, 5th Ring Road, and Metro Line 8 and surrounded by the Kehui and Lincui roads. Generally, every family in this community has a car. The family chosen for sampling lives on the third floor of a 12-story building and consists of three persons, one of which is a smoker. The house is comprised of a kitchen, bathroom, study, living room, and two bedrooms. The most common ventilation modes are opening windows and air conditioning. The locals in this community use natural gas and collective heating system for cooking and warmth.

### Sampling and preparation

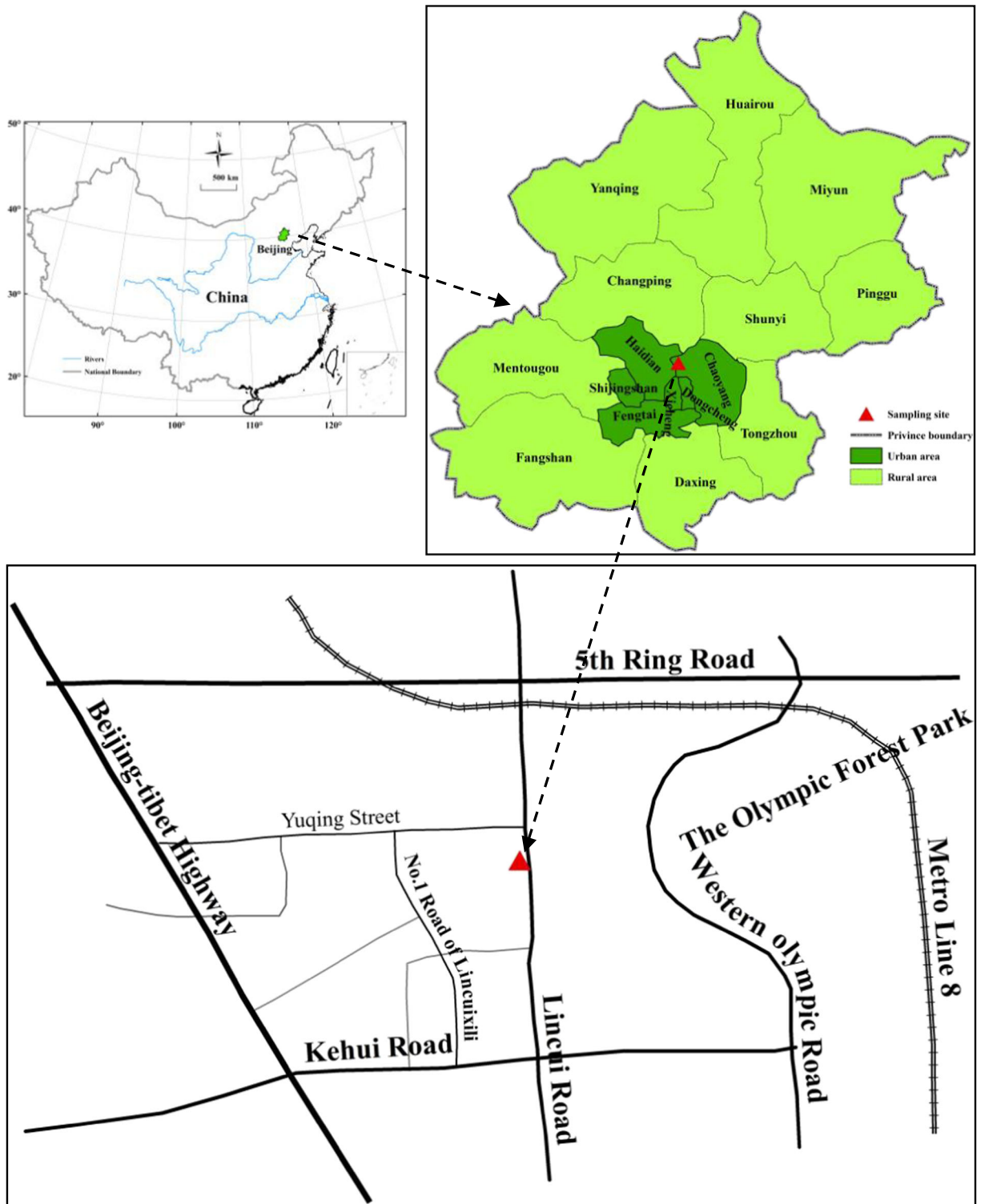
The instrument used for measurements in this study was an air purifier (model TA 240), which was produced by Yuanda Air Products Co., Ltd., China. The air purifier includes a block net, electrostatic particle collector, fan, and control panel. The principle behind the SPM collector of this instrument is that the tungsten wires of the electrostatic precipitator continuously charge the air with high voltage, so that the positively charged SPM can be captured by the negatively charged adsorption plate (Fig. 2). Particulate matter with diameters less than 0.001  $\mu\text{m}$  could be captured by the instrument. The efficiency of particle removal can be up to 99 %. The air volume is adjustable with the maximum of 240  $\text{m}^3 \text{h}^{-1}$ . The measurement instrument was conducted from April 9, 2012 to March 31, 2013 with the air volume of 96  $\text{m}^3 \text{h}^{-1}$ . Each collection month includes 30 to 31 days, except February that includes 28 days. The air purifier was operated 24 h a day, which was in a state of continuous collection of SPM. The air purifier was cleaned thoroughly at the end of every month using distilled water and polyethylene brushes. The SPM were washed out, stored in beakers with a volume of 500 mL, and then dried at 105 °C in an oven (Black 1965). After being dried, the samples that were collected in the same month but in different beakers were mixed together and weighed. The mean weight of sample collected each month was approximately 5.0 g. A total of 12 dry samples were stored in sealed polyethylene bags, labeled, and then transported to a refrigerator (25 °C). Due to the compaction of SPM during drying process, the labeled samples were ground

and then sieved through a 1.0-mm mesh nylon sieve to remove refuse before further analysis.

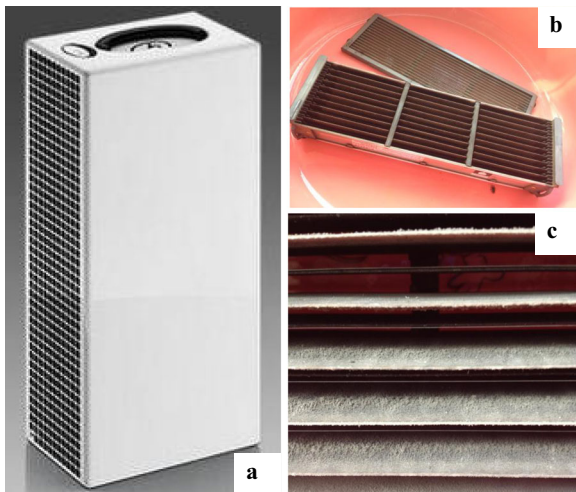
### Analytical methods

#### *Concentrations of arsenic and heavy metals*

The concentrations of arsenic and heavy metals (As, Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn) were measured using an inductively coupled plasma atomic emission spectrometer (ICP-AES, Spectro Arcos Eop, USA) in the Analytical and Testing Center of Beijing Normal University. All chemical treatments were conducted in the ultra-clean laboratory, and all reagents are high purity grade. Before ICP-AES analysis, 0.1 g of dry sample was weighed and dissolved into 10-mL polytetrafluoroethylene vessels. Then, approximately 5 mL of  $\text{HNO}_3$  and 2 mL of HF were added to the vessels. Next, the vessels were introduced to an automatic digestion instrument (ST-60) produced by Beijing Puli Tyco Instruments Co., Ltd. After being closed, the vessels were exposed to a three-stage microwave digestion procedure. The steps of digestion were as follows: (1) heating at 140 °C for 60 min, (2) adding 1 mL  $\text{HClO}_4$  and heating at 160 and 180 °C for 60 and 45 min, respectively, and (3) removing the excess acid. Following digestion, the vessels were cooled to room temperature and then opened, and the contents of the vessels were transferred into volumetric flasks. Those extracts were diluted to 25 mL with deionized water in pre-cleaned polyethylene volumetric flasks and stored at 4 °C prior to analysis. A blank was prepared using the same digestion method (Taner et al. 2013). The limit of detection (LOD) for each element was determined by measuring the signal-to-noise ratio. The LODs for As, Ba, Cd, Cr, Cu, Mn, Ni, Pb, and Zn were 1.2, 0.05, 0.1, 0.3, 0.5, 0.05, 0.3, 1.0, and 0.1  $\mu\text{g L}^{-1}$ . The accuracy of the analysis was assessed by using standard and duplicate samples with relative standard deviation (RSD) and ranges from 0.8 to 1.5 %. The quality assurance and quality control (QA/QC) results show no sign of contamination in the entire analysis. The accuracy of the analytical procedures employed for the analysis of arsenic and heavy metals in SPM was verified using the certified reference material of dark brown soil (GSS-1), and good agreement with the certified values (85.9–107.1 %) was obtained. The QA/QC included the reagent blank and analysis of the standard reference material (GSS-1).



**Fig. 1** The location of study area and sampling site in Beijing



**Fig. 2** Instrument of sampling. **a** The Yuanda air purifier (TA 240); **b** the electrostatic particle collector and block net; **c** the suspended particulate matter attached to the electrostatic particle collector

*Contamination levels of arsenic and heavy metals*

The contamination levels of As, Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn in the SPM samples from the urban household environment were evaluated using the enrichment factor (EF) and geoaccumulation index ( $I_{geo}$ ), which are widely used to assess the contamination levels of dust and soil (Han et al. 2006; Lu et al. 2009a, b, 2014). The EF of an element in the SPM samples was based on the standardization of a measured element against a reference element. A reference element is often characterized by the low occurrence variability, such as the most commonly used elements Al, Fe, Si, and so on (Tasdemir and Kural 2005; Turner and Simmonds 2006; Hao et al. 2007). EF is calculated from the following equation:

$$EF = [C_x/C_{ref}]_{sample} / [C_x/C_{ref}]_{Background} \quad (1)$$

where  $C_x$  is the concentration of the element of interest, and  $C_{ref}$  is the concentration of reference element for normalization. In this study, Al was used as the reference element. Five contamination groups are recognized on the basis of EF: deficiency to minimal enrichment ( $EF \leq 2$ ), moderate enrichment ( $2 < EF \leq 5$ ), significant enrichment ( $5 < EF \leq 20$ ), very high enrichment ( $20 < EF \leq 40$ ), and extremely high enrichment ( $40 < EF$ ) (Han et al. 2006; Lu et al. 2014).

The geoaccumulation index ( $I_{geo}$ ) was introduced by Muller (1969) to assess the contamination level

for a single trace element and is calculated using the following equation:

$$I_{geo} = \log_2 \left[ \frac{C_{mSample}}{1.5 \times C_{mBackground}} \right] \quad (2)$$

where  $C_{mSample}$  is the measured concentration of arsenic and heavy metals in SPM from the urban household environment, and  $C_{mBackground}$  is the geochemical background value. The  $I_{geo}$  for each trace element is calculated and classified into seven groups: non-contaminated ( $I_{geo} \leq 0$ ), non-contaminated to moderately contaminated ( $0 < I_{geo} \leq 1$ ), moderately contaminated ( $1 < I_{geo} \leq 2$ ), moderately to heavily contaminated ( $2 < I_{geo} \leq 3$ ), heavily contaminated ( $3 < I_{geo} \leq 4$ ), heavily to extremely contaminated ( $4 < I_{geo} \leq 5$ ), and extremely contaminated ( $I_{geo} \geq 5$ ) (Muller 1969).

*Health risk assessment of arsenic and heavy metals*

The three major pathways of residential exposure to arsenic and heavy metals in SPM are ingestion, mouth and nose inhalation, and dermal contact (Ferreira-Baptista and De Miguel 2005; Zheng et al. 2010). Exposure was expressed in terms of a daily dose and calculated separately for each element (As, Ba, Cd, Cr, Cu, Mn, Ni, Pb, and Zn) and for each exposure pathway. The exposure dose contacted through each of the three pathways was calculated using a model developed by the following formulas (US EPA 1989; 2011a):

Dose contacted through ingestion of SPM:

$$CDI_{ing} = C_{UCL} \times \frac{\ln gR \times EF \times ED}{BW \times AT} \times CF \quad (3)$$

Dose absorbed through inhalation of SPM:

$$EC_{inh} = C_{UCL} \times \frac{ET \times EF \times ED}{AT_n} \quad (4)$$

Dose contacted through dermal contact of SPM:

$$DAD_{dermal} = C_{UCL} \times \frac{SL \times SA \times ABS \times EF \times ED}{BW \times AT} \times CF \quad (5)$$

where  $CDI_{ing}$ ,  $EC_{inh}$ , and  $DAD_{dermal}$  are the chemical daily intake, exposure concentration, and dermal absorbed dose which were estimated to assess the risks posed by arsenic and heavy metals in the household SPM via ingestion, inhalation, and dermal contact, respectively.  $C_{UCL}$  is the 95 % upper confidence limit

(UCL) on the arithmetic mean concentrations ( $\text{mg kg}^{-1}$ ) of arsenic and heavy metals in SPM.  $\text{IngR}$  is the ingestion rate, estimated in this study to be  $200 \text{ mg day}^{-1}$  for children and  $100 \text{ mg day}^{-1}$  for adults.  $EF$  is the exposure relative frequency, assumed to be  $350 \text{ days year}^{-1}$ .  $ED$  is the exposure duration, assumed to be 6 years for children and 24 years for adults.  $CF$  is the conversion factor,  $10^{-6} \text{ kg mg}^{-1}$ .  $SA$  is the skin area, assumed to be  $2800 \text{ cm}^2$  for children and  $5700 \text{ cm}^2$  for adults.  $SL$  is the skin adherence factor, estimated to be  $0.2 \text{ mg cm}^{-2} \text{ day}^{-1}$  for both children and adults in this study.  $ABS$  is the dimensionless dermal absorption factor that was 0.001 for all elements in this study (Ferreira-Baptista and De Miguel 2005; Man et al. 2010; Li et al. 2013).  $BW$  is the average body weight, assumed to be 15.9 kg for children and 56.8 kg for adults (MEPPRC Ministry of Environmental Protection of the People's Republic of China 2014).  $AT$  is the averaging time, assumed to be  $ED \times 365$  days for non-carcinogens and  $70 \times 365$  days for carcinogens.  $ET$  is the exposure time every day (hours/day), which was 24 h/day in this study.  $AT_n$  is the average time. For non-carcinogens,  $AT_n = ED \times 365 \text{ days} \times 24 \text{ h/day}$ ; for carcinogens,  $AT_n = 70 \text{ years} \times 365 \text{ days/year} \times 24 \text{ h/day}$ . All of the values for  $\text{IngR}$ ,  $EF$ ,  $ED$ ,  $CF$ ,  $SA$ ,  $SL$ ,  $AT$ ,  $ET$ , and  $AT_n$  were found in the User's Guide/Technical Background Document for US EPA Region 9's RSL (Regional Screening Levels) Tables (US EPA 2013).

$C_{UCL}$  (exposure-point upper confident limit content,  $\text{mg kg}^{-1}$ ) is considered to yield an estimate of the "reasonable maximum exposure," which is the upper limit of the 95 % confidence interval for the mean (US EPA 1989; Zheng et al. 2010; Hu et al. 2011; Kurt-Karakus 2012). The 95 % UCL concentration ( $C_{UCL}$ ) has been calculated by the following formula (US EPA 2002; Kurt-Karakus 2012):

$$C_{UCL} = \bar{X} + \left[ Z_{\alpha} + \frac{\beta}{6\sqrt{n}} \times (1 + 2 \times Z_{\alpha}^2) \right] \times \frac{STD}{\sqrt{n}} \quad (6)$$

where  $\bar{X}$  is the arithmetic mean,  $STD$  is the standard deviation, and  $\beta$  is the skewness. For the 95 % confidence level,  $Z_{\alpha}$  is 1.645, and  $n$  represents the number of samples.

After the doses for the three exposure pathways were calculated, the potential non-carcinogenic toxic risk was determined by calculating the hazard quotient (HQ). The carcinogenic risks (CR) were used to assess the

health risks to toxic element exposure by SPM and calculated by the following equations (US EPA 1989):

$$HQ = \frac{CDI_{ing}}{RfDo} = \frac{EC_{inh}}{RfCi \times 1000} = \frac{DAD_{dermal}}{RfDo \times GIABS} \quad (7)$$

$$CR = CDI_{ing} \times SF_o = EC_{inh} \times IUR = DAD_{dermal} \times \left( \frac{SF_o}{GIABS} \right) \quad (8)$$

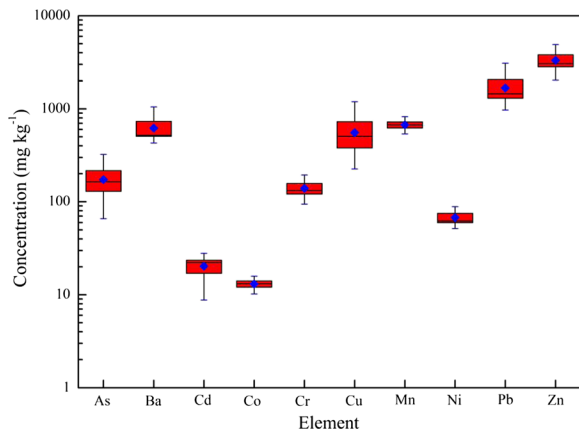
where  $RfDo$  (the reference dose,  $\text{mg kg}^{-1} \text{ day}^{-1}$ ) is an estimate of the maximum permissible risk to the human population through daily exposure during a lifetime,  $RfCi$  is the inhalation reference concentration ( $\text{mg m}^{-3}$ ),  $SF_o$  is the oral slope factor ( $(\text{mg kg}^{-1} \text{ day}^{-1})^{-1}$ ),  $GIABS$  is the gastrointestinal absorption factor, and  $IUR$  is the inhalation unit risk ( $(\text{mg m}^{-3})^{-1}$ ). The  $SF_o$ ,  $RfDo$ ,  $RfCi$ ,  $GIABS$ , and  $IUR$  were cited from the US EPA website (US EPA 2011b (<http://www.epa.gov/region9/superfund/prg/index.html>)). The  $SF_o$  and/or  $IUR$  values for As (inorganic), Cd (diet), Co, Cr (VI), Ni (refinery dust), and Pb (acetate) were found in the screening level (RSL) tables (US EPA 2011c).

The hazard index (HI) is equal to the sum of HQ being used to assess the overall potential for non-carcinogenic effects posed by more than one element. HI values of  $\leq 1$  indicate no significant risk of non-carcinogenic health effects, and HI values  $> 1$  indicate that there is a chance of non-carcinogenic effects occurring, with a probability that tends to increase as the value of HI increases (Kong et al. 2011; US EPA 2011a; Li et al. 2013). The carcinogenic risk is the probability of an individual developing any type of cancer from lifetime exposure to carcinogenic hazards. The tolerable risk for regulatory purposes is in the range of  $10^{-6}$  (1 in 1,000,000) to  $10^{-4}$  (1 in 10,000) (Ferreira-Baptista and De Miguel 2005).

## Results and discussion

### Concentrations of arsenic and heavy metals

The concentrations of arsenic and heavy metals (As, Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn) on a dry weight ( $\text{mg kg}^{-1}$ ) in SPM from an urban household environment in Beijing are presented in Fig. 3. The mean concentrations of the studied elements in the urban household SPM decreased in the following order:  $\text{Zn} > \text{Pb} > \text{Mn} > \text{Ba} > \text{Cu} > \text{As} > \text{Cr} > \text{Ni} > \text{Cd} > \text{Co}$ . The



**Fig. 3** Concentration of the studied toxic elements in the urban household suspended particulate matter in a whole year

arithmetic mean concentration of Zn was 3041.1 mg kg<sup>-1</sup>, while that of Co was 13.1 mg kg<sup>-1</sup>. Compared with the background values of Beijing, the mean concentrations of the elements were all higher than those of corresponding background values, except for Co (CNEMC (China National Environmental Monitoring Center) 1990). The concentrations for As, Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn in the household SPM were 20.1, 1.0, 300.7, 0.8, 2.3, 27.3, 1.3, 1.4, 105.3, and 54.8 times the respective background values. In addition, the mean concentrations for As, Cd, Co, Cr, Ni, and Pb in the SPM of this study were lower than those of urban outdoor PM<sub>2.5</sub> in Beijing during 2009–2010 (Tao et al. 2014). Moreover, compared with outdoor dustfall and road dust in the parks in urban area of Beijing, the mean concentrations for the studied elements in the household SPM were all higher, except for Co and Ba, while the concentrations were lower than those in street dust, except for As, Pb, and Zn (Guo 2008). The lower household concentrations of the studied elements could be accounted by a dilution effect produced by indoor contributions (Hunt et al. 2012). The concentrations of the toxic elements in the household SPM in this study were higher than those in household dust at the same location (Yang et al. 2014), indicating that more elements attached to smaller particles. The levels of the studied elements were also higher than those in urban household dust in other countries, as reported in the literature (Kim et al. 1998; Tong and Lam 2000; Chattopadhyay et al. 2003; Yoshinaga et al. 2014).

The higher concentration was because the sampling location in the urban area is near the Inner Ring Road and highway of Beijing which have high and intensive

volume traffic, and it is also close to commercial and financial center and industries. Most of the toxic elements are derived from traffic, industrial, and incinerator emissions (Adachi and Tainosho 2004; Thorpe and Harrison 2008; Zhu et al. 2008; Duong and Lee 2011). It is worth mentioning that the higher concentration of arsenic in the household SPM might derive from the combustion processes of cooking and smoking (Zeng et al. 2001).

A statistical package (SPSS 19.0) was utilized to identify the correlations among the concentrations of the studied toxic elements in the urban household SPM. The correlation results are listed in Table 1. A strong significant ( $P < 0.01$ ) positive correlation was observed between Ba, Co, Cr, Cu, and Ni. Arsenic was strongly correlated with Cd and Cu. Lead was significantly ( $P < 0.05$ ) correlated with As, Cd, and Cu. No statistically significant correlation was observed for Mn with other elements, except for Zn. The correlation of the above toxic elements in the household SPM indicates the similarity of their sources.

#### Seasonal variation of arsenic and heavy metal concentrations

The total concentrations of the studied elements in the urban household SPM are shown in Fig. 4, and they displayed significant seasonality. Comparatively higher total element concentrations occurred in winter, followed by autumn, while the lowest concentrations were recorded in summer. The total concentrations of the ten studied elements in the household SPM had an average of 6970.0 mg kg<sup>-1</sup> in the studied year. The highest value of the total concentrations occurred in November (9407.1 mg kg<sup>-1</sup>), followed by January (8800.0 mg kg<sup>-1</sup>), while the lowest value presented in July (4804.2 mg kg<sup>-1</sup>). The correlations of the total concentrations of the studied toxic elements in the urban household SPM and the external climate variables were analyzed (Table 2). The results showed that the total concentrations of the studied toxic elements in the urban household SPM were strongly negatively correlated with the monthly average precipitation and temperature ( $P < 0.05$ ). With increasing monthly average precipitation and temperature, the total concentrations of the studied toxic elements in the urban household SPM decreased. The correlations of the total concentrations of the studied toxic elements in the urban household SPM and the monthly average wind speed and relative

**Table 1** Correlation matrix among the studied toxic elements in the urban household suspended particulate matter

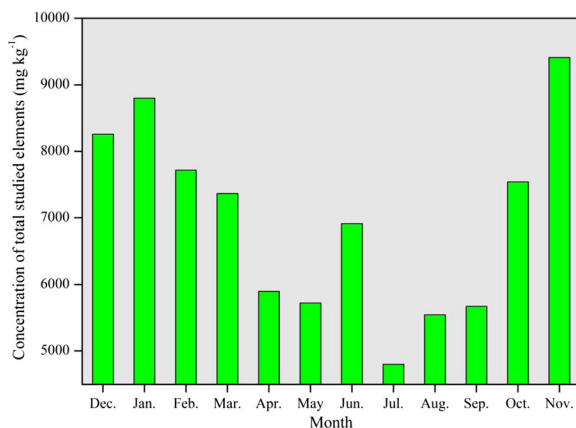
<i>n</i> =12	As	Ba	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn
As	1.00									
Ba	0.22	1.00								
Cd	0.85**	0.15	1.00							
Co	-0.15	0.85**	-0.10	1.00						
Cr	0.20	0.89**	0.15	0.86**	1.00					
Cu	0.73**	0.77**	0.71*	0.44	0.69*	1.00				
Mn	-0.13	0.02	0.16	0.24	-0.10	0.00	1.00			
Ni	0.31	0.84**	0.36	0.82**	0.90**	0.78**	0.18	1.00		
Pb	0.66*	0.41	0.69*	0.28	0.48	0.68*	0.03	0.59*	1.00	
Zn	0.50	-0.04	0.59*	-0.08	-0.10	0.30	0.64*	0.13	0.54	1.00

\*Correlation is significant at 0.05 level (two-tailed); \*\* correlation is significant at 0.01 level (two-tailed)

humidity were not obvious. The highest total concentration in winter was due to the burning of coal or natural gas for urban central heating systems and meteorological conditions outside (such as low temperature and slow air convection). Further, the firecrackers and fireworks set off during the Spring Festival comprised another source for the toxic elements in the household SPM in January. In spring, the wind is strong, speeding up the air convection, and SPM with higher values of the toxic elements in the outdoor atmosphere are blown away, decreasing the outdoor contribution of arsenic and heavy metals in the household SPM. Summer is the rainy season in Beijing. The leaching effect of precipitation in this season is the most plausible explanation for the lowest total concentration and more variability in the studied elements. Additionally, ventilation through

open windows and air conditioning because of the higher temperature is another mode that reduces the household SPM in summer. The higher total element concentration in autumn is due to busier traffic, as autumn is the best time for tourism and travel in Beijing. In addition, the burning of natural gas or coal for heating begins in late autumn (November 15) in Beijing, increasing the contribution of outdoor sources of toxic elements to the household SPM.

The monthly variation of each single element is presented in Fig. 5. The concentrations of all studied toxic elements exhibited the highest concentrations during winter, conforming to the seasonal variations of PM<sub>2.5</sub> in the urban outdoors of Beijing (Yu et al. 2012; Tao et al. 2014), except for Mn and Zn of which the highest concentrations occurred in spring and autumn. The lowest concentrations for As, Cd, Cu, Mn, Pb, and Zn were in summer, while those for Ba, Co, Cr, and Ni were in spring. The maximum for most of the studied elements occurred during November, December, Janu-



**Fig. 4** Monthly variation of the total concentrations of the studied toxic elements in the urban household suspended particulate matter

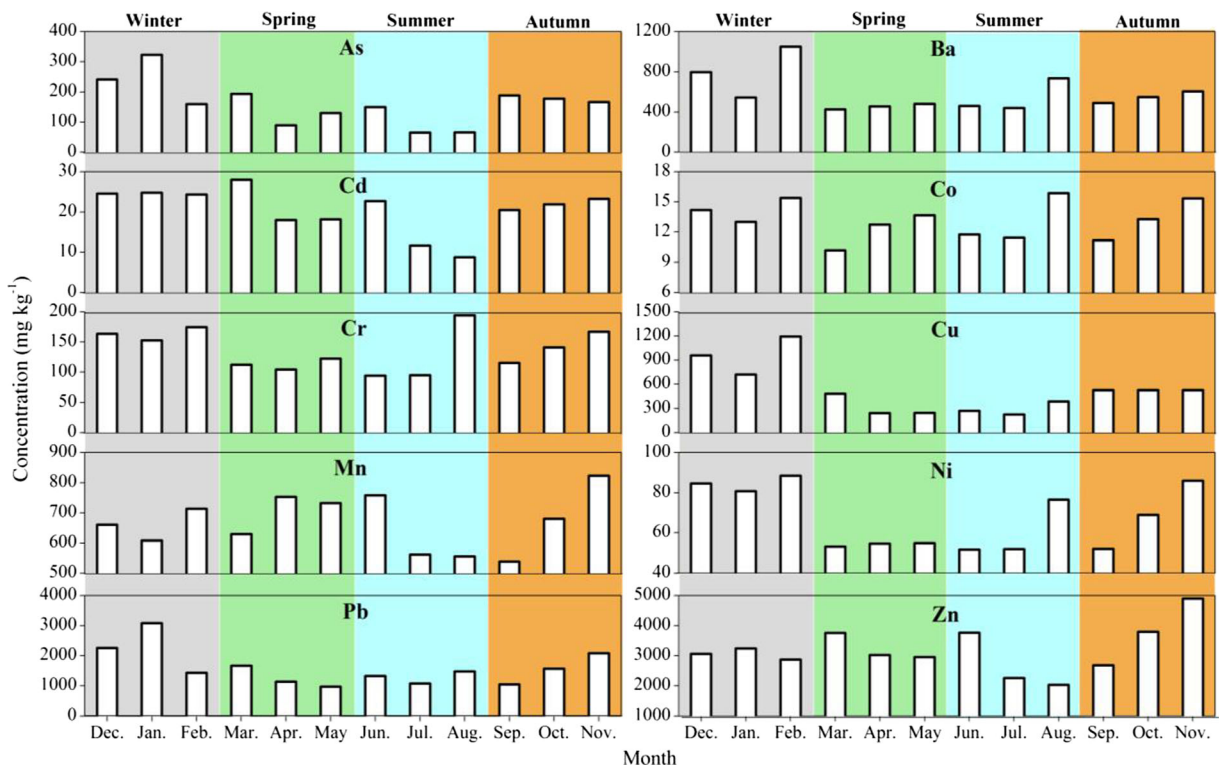
**Table 2** Correlation matrix of the total concentrations of studied toxic elements in the urban household suspended particulate matter and the external climate variables

<i>n</i> =12	TC	P	WS	T	RH
TC	1.00	-0.62*	0.09	-0.90**	-0.28

TC the total concentrations of the studied toxic elements in indoor SPM, P the monthly average precipitation, WS the monthly average wind speed, T the monthly average temperature, RH the monthly average relative humidity

\*Correlation is significant at 0.05 level (two-tailed); \*\*correlation is significant at 0.01 level (two-tailed)





**Fig. 5** Monthly variation of each studied toxic element concentrations in the urban household suspended particulate matter

ary, and February, while the minimum presented during June and July. Most notably, the monthly maximums of Co (15.9 mg kg<sup>-1</sup>) and Cr (194.2 mg kg<sup>-1</sup>) were recorded during August, and the monthly minimums of Ba (429.3 mg kg<sup>-1</sup>) and Co (10.2 mg kg<sup>-1</sup>) were recorded during March. For Mn and Zn in the household SPM, the concentrations in June were relatively higher, leading to a comparatively higher total element concentration in this month. The monthly variations of the individual element levels (Cv) ranged between 0.14 and 0.58. The coefficient of variation for Cu was relatively higher, indicating a higher variation of concentrations for Cu among the different months, while the comparatively lower Cv values for Co and Mn implied lower variations.

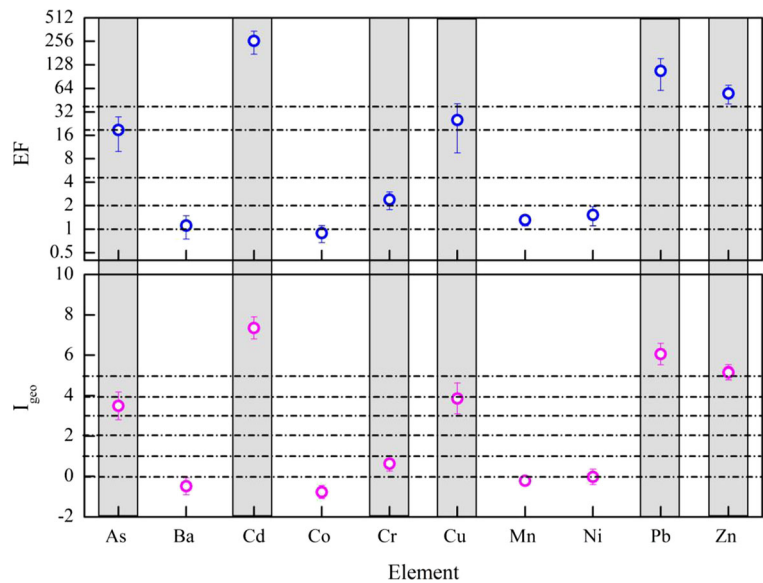
Contamination levels of arsenic and heavy metals

The calculated EF and I<sub>geo</sub> results for arsenic and heavy metals in the urban household SPM are displayed in Fig. 6. The mean EF values decreased in the following order: Cd > Pb > Zn > Cu > As > Cr > Ni > Mn > Ba > Co. According to EF values, Ba, Co, Mn, and Ni in the household SPM (<2) were considered to originate from

natural sources, belonging to the minimal enrichment group. Chromium (2 < EF ≤ 5) was classified as a moderately enriched element, indicating a moderate influence by human activities, while arsenic (18.7) was a significantly enriched element. Copper fell into the very high enrichment group, while Cd, Pb, and Zn were considered part of the extremely high enrichment group, indicating that their sources were predominantly from anthropogenic activities. Although the concentrations of arsenic and heavy metals varied seasonally, the EFs of those elements in the different seasons were generally categorized in the same enrichment group, except for Cu and Zn (Fig. 6).

The mean I<sub>geo</sub> values decreased in a similar order to the mean EF values for the household SPM. All elements measured in this study were considered contaminants at different levels. The mean contamination levels for Ba, Co, Mn, and Ni in the household SPM samples based on the I<sub>geo</sub> values were assessed as non-contaminated (I<sub>geo</sub> < 0), while the value of Cr was determined as non- to moderately contaminated (1 > I<sub>geo</sub> > 0). The I<sub>geo</sub> values indicated that As and Cu belonged to the heavily contaminated group, while the levels for Cd, Pb, and Zn indicated extreme contamination. Therefore, the contamination of the toxic elements in the household SPM should be paid

**Fig. 6** Enrichment factor (EF) and geoaccumulation index ( $I_{geo}$ ) of the studied toxic elements in the urban household suspended particulate matter. The *gray color* points out the contaminated toxic elements



more attention. The  $I_{geo}$  values of those elements over the different seasons generally fell into the same contamination category, except for As and Zn (Fig. 6).

#### Health risk assessment of arsenic and heavy metals

The results of the health risk assessment of arsenic and heavy metals in the urban household SPM are listed in Table 3. Direct exposure to the toxic elements in SPM by the local residents is caused by inhalation, while the indirect exposure pathways include the ingestion of particles deposited on food and drinks and the dermal contact with surfaces of indoor appliances via hand-to-mouth activities (Li et al. 2013). For non-carcinogenic and carcinogenic risks, the highest mean exposure dose (CDI) values of children and adults were for Zn, while the lowest values were for Co in the urban household SPM. The doses of the studied elements in the household SPM via ingestion and dermal contact exposure for children were higher than those for adults, while those via inhalation exposure were at similar levels for both children and adults.

The HQ values of ingestion, inhalation, and dermal contact exposure for the studied toxic elements decreased in the following orders: As > Pb > Cr > Co > Cd > Cu > Zn > Mn > Ba > Ni, Mn > As > Co > Cd > Cr > Ni > Ba, and Cr > Cd > As > Pb > Zn > Ba > Co > Ni > Cu > Mn, respectively (Table 3). The HQ values via ingestion exposure for As (8.12 and 1.14) were higher than the safe level (=1) for both children and adults, while the HQ

value for Pb (6.77) was higher than the safe level for children, indicating potential non-carcinogenic risks to both local children and adults from ingestion exposure to As and to children from ingestion exposure to Pb. The HQ values via inhalation exposure to As, Ba, Cd, Co, Cr, Mn, and Ni for both children and adults were all lower than the safe level, indicating no non-carcinogenic risks from the inhalation exposure of each single element. The HI (sum of HQ for all studied elements) values via inhalation for children and adults (2.55 and 2.55) were higher than the safe level (=1), indicating the accumulative non-carcinogenic risks to both children and adults via inhalation. Arsenic and Mn in the urban household SPM via inhalation should be considered a potential hazardous pollutant found at homes in the urban areas for children and adults. The HQ values via dermal contact exposure to the studied elements for children and adults were also lower than the safe level, implying no non-carcinogenic risks from the dermal contact exposure of each single element. The HI values for children (1.51) were higher than the safe level, and the higher HI values mainly resulted from Cr, indicating the accumulative non-carcinogenic risks to children via dermal contact, while those values for adults (0.86) were below 1, implying that there was no accumulative non-carcinogenic risk to adults via dermal contact.

For children and adults, the carcinogenic risk (CR) levels of As ( $3.13E-04$  and  $1.75E-04$ ) and Pb ( $5.69E-04$  and  $3.18E-04$ ) in the urban household SPM via ingestion exposure were higher than the acceptable level

**Table 3** Carcinogenic and non-carcinogenic risks for each studied toxic element and exposure pathway in the urban household suspended particulate matter

	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child-Ing	Adult-Ing	Child-Inh	Adult-Inh	Child-dermal	Adult-dermal
	$CDI_{ing}$ (mg/kg-day) <sup>-1</sup>	$EC_{inh}$ (µg/m <sup>3</sup> ) <sup>-1</sup>	$DAD_{dermat}$ (mg/kg-day) <sup>-1</sup>						Non-carcinogenic risks-HQ					
As	2.44E-03	1.40E-02	6.82E-05	3.89E-05	8.12E+00	1.14E+00	9.34E-01	9.34E-01	2.27E-01	1.30E-01	2.27E-01	9.34E-01	2.27E-01	1.30E-01
Ba	8.49E-03	4.88E-02	2.38E-04	1.35E-04	4.24E-02	5.94E-03	9.76E-02	9.76E-02	1.70E-02	9.67E-03	1.70E-02	9.76E-02	1.70E-02	9.67E-03
Cd	2.74E-04	1.57E-03	7.66E-06	4.36E-06	2.74E-01	3.83E-02	1.57E-01	1.57E-01	3.06E-01	1.75E-01	3.06E-01	1.57E-01	3.06E-01	1.75E-01
Co	1.69E-04	9.74E-04	4.74E-06	2.70E-06	5.64E-01	7.90E-02	1.62E-01	1.62E-01	1.58E-02	9.00E-03	1.58E-02	1.62E-01	1.58E-02	9.00E-03
Cr	1.85E-03	1.06E-02	5.18E-05	2.95E-05	6.16E-01	8.63E-02	1.06E-01	1.06E-01	6.90E-01	3.93E-01	6.90E-01	1.06E-01	6.90E-01	3.93E-01
Cu	8.46E-03	4.86E-02	2.37E-04	1.35E-04	2.11E-01	2.96E-02	2.11E-01	2.96E-02	5.92E-03	3.37E-03	5.92E-03	2.11E-01	5.92E-03	3.37E-03
Mn	8.58E-03	4.93E-02	2.40E-04	1.37E-04	6.13E-02	8.58E-03	9.87E-01	9.87E-01	1.72E-03	9.78E-04	1.72E-03	9.87E-01	1.72E-03	9.78E-04
Ni	9.00E-04	5.18E-03	2.52E-05	1.44E-05	1.80E-02	2.52E-03	1.04E-01	1.04E-01	1.26E-02	7.18E-03	1.26E-02	1.04E-01	1.26E-02	7.18E-03
Pb	2.37E-02	1.36E-01	6.64E-04	3.78E-04	6.77E+00	9.48E-01	6.77E+00	9.48E-01	1.90E-01	1.08E-01	1.90E-01	6.77E+00	1.90E-01	1.08E-01
Zn	4.36E-02	2.50E-01	1.22E-03	6.95E-04	1.45E-01	2.03E-02	1.45E-01	2.03E-02	4.07E-02	2.32E-02	4.07E-02	1.45E-01	4.07E-02	2.32E-02
Sum	9.84E-02	5.66E-01	2.76E-03	1.57E-03	1.68E+01	2.35E+00	2.55E+00	2.55E+00	1.51E+00	8.59E-01	1.51E+00	2.55E+00	1.51E+00	8.59E-01
	$CDI_{ing}$ (mg/kg-day) <sup>-1</sup>	$EC_{inh}$ (µg/m <sup>3</sup> ) <sup>-1</sup>	$DAD_{dermat}$ (mg/kg-day) <sup>-1</sup>		Carcinogenic risks (CR)									
As	2.09E-04	1.20E-03	5.85E-06	4.66E-05	3.13E-04	1.75E-04	5.16E-06	5.16E-06	8.77E-06	2.00E-05	8.77E-06	5.16E-06	8.77E-06	2.00E-05
Cd	2.34E-05	1.35E-04	6.56E-07	5.24E-06	2.43E-07	9.71E-07	2.43E-07	9.71E-07	3.00E-06	2.02E-04	3.00E-06	2.43E-07	3.00E-06	2.02E-04
Co	1.45E-05	8.12E-06	4.06E-07	3.24E-06	7.92E-05	4.44E-05	7.66E-05	7.66E-05	8.88E-05	2.02E-04	8.88E-05	7.66E-05	8.88E-05	2.02E-04
Cr	1.58E-04	9.11E-04	4.44E-06	3.54E-05	1.07E-07	4.26E-07	1.07E-07	4.26E-07	1.59E-05	3.63E-05	1.59E-05	1.07E-07	1.59E-05	3.63E-05
Ni	7.72E-05	4.44E-04	2.16E-06	1.72E-05	5.69E-04	3.18E-04	9.35E-07	9.35E-07	1.13E-04	2.59E-04	1.13E-04	9.35E-07	1.13E-04	2.59E-04
Pb	2.03E-03	1.17E-02	5.69E-05	4.54E-04	9.61E-04	5.38E-04	8.38E-05	8.38E-05	3.35E-04	2.59E-04	3.35E-04	8.38E-05	3.35E-04	2.59E-04
Sum	8.44E-03	4.72E-03	4.85E-02	1.88E-03										

$CDI_{ing}$ ,  $EC_{inh}$ , and  $DAD_{dermat}$  (mg kg<sup>-1</sup> day<sup>-1</sup> or µg m<sup>-3</sup>) are the exposure dose through ingestion, inhalation, and dermal contact, respectively. Ing and Inh represent ingestion and inhalation. HQ and CR represent hazard quotient and carcinogenic risks

( $1 \times 10^{-4}$ ), indicating that the carcinogenic risk to children and adults posed by As and Pb via ingestion is not negligible in the urban household SPM of Beijing. The CR levels of Cr for children and adults via ingestion exposure were lower than  $1 \times 10^{-4}$  and can be acceptable. However, the carcinogenic risk value of Cr in the household SPM for children via ingestion was  $7.92E-05$ , slightly lower than  $1 \times 10^{-4}$ , implying a potential carcinogenic risk to children. For inhalation exposure, the carcinogenic risks of As, Cd, Co, Cr, and Ni in the household SPM samples were less than  $1 \times 10^{-4}$  for children, and the accumulative risks of the studied elements were also within the acceptable level, indicating that the carcinogenic risk posed by the studied toxic elements to children via inhalation is acceptable. However, the carcinogenic risk value of Cr in the household SPM for children was  $7.66E-05$ , slightly below the acceptable level, implying a potential carcinogenic risk to children. The carcinogenic risk posed by Cr ( $3.06E-04$ ) was slightly higher than  $1 \times 10^{-4}$  for adults, indicating that the carcinogenic risk posed by Cr to adults via inhalation should not be ignored. The carcinogenic risks for children posed by As, Cr, and Pb in the household SPM via dermal contact exposure were below the acceptable level, but the accumulative risks of those elements were higher than the acceptable level, indicating accumulative carcinogenic risks to children by those toxic elements via dermal contact exposure. The carcinogenic risk level for adults by Cr ( $2.02E-04$ ) in the household SPM via dermal contact was slightly higher than the acceptable level, indicating that the carcinogenic risk to adults posed by Cr via dermal contact should be paid more attention.

Even though the assessments of the non-carcinogenic and carcinogenic risks are uncertain because the health risks are evaluated based on a number of assumptions and estimates including the types of parameters ( $ED$ ,  $EF$ ,  $RfD_o$ ,  $RfC_i$ ,  $IUR$ , and so on), our assessment still offers a valuable evaluation of the health risks associated with the exposure of As and heavy metals bound to household particles. The results could alert policymakers that the potential carcinogenic and non-carcinogenic risks posed by the toxic elements in household SPM at the present levels should not be ignored and some measures should be taken to control household SPM contamination for residential health concerns.

This study only focused on As and heavy metals in the SPM experienced by one family in an urban area of Beijing. But the studied family is located at the typical

urban area of Beijing, in which the value of these measurement results could represent the overall levels of arsenic and heavy metals in particulate matter in residences in Beijing. However, more families in different urban areas can be investigated in future studies. Moreover, the distributions of the toxic elements in household SPM by the different particle-size fractions could be considered. Further, the health risks of the toxic elements in household SPM in different particle-size fractions should also be determined.

## Conclusions

In this work, the seasonal concentrations, contamination levels, and health risks of arsenic and heavy metals (Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn) in the suspended particulate matter from an urban household environment in Beijing, China, have been investigated. The mean concentrations of As, Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn in the studied urban household SPM were 160.4, 545.0, 21.9, 13.0, 127.8, 485.0, 660.5, 60.1, 1428.4, and 3057.4  $\text{mg kg}^{-1}$ , respectively. Compared with the background values and  $\text{PM}_{2.5}$  in the urban outdoors of Beijing, the mean concentrations of the studied elements in the household SPM were higher and lower, respectively. The concentrations of the studied elements in the household SPM displayed significant seasonality. Comparatively higher total element concentrations occurred in winter, followed by autumn, while the lowest concentrations were recorded in summer.

The calculated enrichment factor (EF) and geoaccumulation index ( $I_{\text{geo}}$ ) values of the studied elements indicated the order of EF and  $I_{\text{geo}}$ :  $\text{Cd} > \text{Pb} > \text{Zn} > \text{Cu} > \text{As} > \text{Cr} > \text{Ni} > \text{Mn} > \text{Ba} > \text{Co}$ . Based on the EF and  $I_{\text{geo}}$  values, Ba, Co, Mn, and Ni in the household SPM were assessed as non-contaminated, whereas Cr was determined to have no to moderate contamination. The levels for As and Cu in the household SPM were heavily contaminated, while Cd, Pb, and Zn were extremely contaminated.

For the non-carcinogenic and carcinogenic risks to children and adults, the highest mean exposure dose values were for Zn, while the lowest values were for Co. The non-carcinogenic risks to children mainly resulted from As and Pb via ingestion exposure, and the potential non-carcinogenic risks were from As and Mn via inhalation exposure and Cr via dermal contact. For adults, the non-carcinogenic risk was caused by As via

ingestion, a potential non-carcinogenic risk was from As and Mn via inhalation exposure, and there were no non-carcinogenic risks from the studied elements via dermal contact. For children, the carcinogenic risks were from As and Pb in the urban household SPM via ingestion exposure, the integrated (accumulative) carcinogenic risks of the studied elements via dermal contact were above the acceptable level, and the integrated carcinogenic risks of the toxic elements via inhalation exposure were within the acceptable level. The carcinogenic risks to adults were from As and Pb via ingestion exposure and Cr via inhalation and dermal contact exposure. Therefore, the carcinogenic and non-carcinogenic risks to children and adults from the toxic elements in household SPM of urban area in Beijing via ingestion, inhalation, and dermal contact must be considered.

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