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RESEARCH ARTICLE

In vitro inhalation/ingestion bioaccessibility, health risks, and source appointment of airborne particle-bound elements trapped in room air conditioner filters

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Abstract

The airborne particle-bound elements (Ca, Fe, Al, Mg, K, Na, Zn, Mn, P, Pb, Cu, Sr, Ti, Ba, Cr, Ni, As, Sb, Cd, Co, and V) trapped in room air conditioners' filters (filter dusts) during recirculating indoor air from different types of rooms were analyzed, and the objectives of this study were to assess the potential sources of those elements and their potential health risks via inhalation/ingestion exposure. Main crustal elements such as Ca, Fe, Al, Mg, and K with an average value of 60.6, 17.9, 11.3, 7.58, and 6.90 mg g^{-1}, respectively, are the preponderant elements, and the mean values of main toxic elements were 2230, 344, 508, 85.7, 71.5, 36.0, 8.02, and 16.9 mg kg^{-1} for Zn, Cu, Pb, Cr, Ni, As, Cd, and Sb, respectively. The enrichment factors indicated the significant enrichment of Cd, Pb, Cr, Cu, Sb, and Zn in the filter dusts. Four potential sources with the contributions of 33.5, 29.1, 22.6, and 14.8%, respectively, were identified by absolute principal component scores-multiple linear regression analysis (APCS-MLR). Enrichment factor and APCS-MLR model reveal the outdoor input of toxic elements. In vitro inhalation and ingestion bioaccessibility of toxic elements showed elemental and in vitro procedure dependence. There are potential carcinogenic risks via ingestion exposure and no non-carcinogenic risks to both children and adults based on bioaccessible contents of toxic elements. This study reveals the potential health risks posed by the particle-bound elements.

Keywords Toxic elements · Enrichment level · Source identification · Bioaccessibility · Health risk · Indoor air quality

Introduction

Humans living in urban areas spend up to 85–90% of their daily life in indoor environments either inside office, school, college, and commercial buildings or inside residential houses (Chen and Zhao 2011). Therefore, the quality of indoor air is an essential determinant of healthy life and people's well-being. Studies show that airborne particles are one of the main contaminants in indoor environment, which have indoor and outdoor source (Ibanez et al. 2010; Praveena et al. 2015; Turner 2011). Cooking and smoking are generally the main indoor particle sources for most office room and residential houses, which have extreme levels of toxic elements, while smoking is banned inside most public office, school, college, and commercial buildings. However, outdoor sources are associated with industrial emissions of nonferrous and ferrous industry and other industries, coal combustion, and vehicle exhausts, which contain higher levels of toxic elements (Duan and Tan 2013). Toxic elements in atmospheric particulate matters (AMPs) can pose acute and chronic health effects to humans via inhalation and ingestion exposure (Cheng et al. 2013; Kim et al. 2015; Wang et al. 2016a). Previous literatures show that the indoor dusts and airborne particles enrich a lot of

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toxic elements (Ibanez et al. 2010; Praveena et al. 2015; Turner 2011). Therefore, long-term exposure to indoor airborne toxic elements via ingestion and inhalation indoors may result in potential health risks posed by the particle-bound toxic elements. It is thus important to reveal sources of the particle-bound toxic elements and to assess their exposing risks to humans in indoor environment.

Risk assessment of contaminants in environmental media has received more and more attention in recent years. Investigations show that the risk assessment based on total contents of toxic elements in environmental media overestimates the potential health risks (Garcia-Santiago et al. 2017; Huang et al. 2018; Kastury et al. 2017). Toxic elements in the ingested media can be dissolved partly or all after contacting with physiological fluids (i.e., bioavailability), which can cause adverse effects to human health. Due to the complexity, expensiveness, being time-consuming, and great variability on intra- and inter-species of experimental animals or human of the evaluation of in vivo bioavailability for toxic elements (Kastury et al. 2017; Zia et al. 2011), in vitro procedures as alternative approaches to measure bioaccessibility (i.e., the fraction that is soluble in the simulated gastrointestinal or lung physiological fluids) are popular in exposure assessment, which are simple, rapid, reproducible, and economic but involve more uncertainties. For example, the common in vitro ingestion bioaccessibility procedures for toxic elements in soils and soil-like materials include the Solubility Bioaccessibility Research Consortium (SBRC), physiologically based extraction test (PBET), in vitro gastrointestinal method (IVG), and Deutsches Institut für Normunge. V. (DIN) (Juhasz et al. 2014). Some in vitro inhalation bioaccessibility procedures such as artificial lysosomal fluid (ALF), simulated lung fluid (SLF), and Gamble's solution have also been developed to investigate the inhalation bioaccessibility of outdoor APM-bound toxic elements (Boisa et al. 2014; Kastury et al. 2017; Mukhtar and Limbeck 2013). In vitro bioaccessibility of toxic metals in household dust has been investigated recently (Huang et al. 2014; Ibanez et al. 2010; Liu et al. 2016; Turner 2011; Turner and Ip 2007). For example, in vitro ingestion bioaccessibility of toxic metals in household dust was investigated by using the PBET procedure (Turner and Ip 2007). The in vitro ingestion bioaccessibility of lead in house dust was analyzed by using SBRC, IVG, PBET, and DIN procedures (Li et al. 2014). The bioaccessible fraction of metals in urban aerosol was determined by using simulated lung fluids (Coufalik et al. 2016), and health risk of trace elements in smoke haze aerosols versus urban aerosols was assessed based on bioaccessible contents by using simulated lung fluids (Huang et al. 2016). But few investigations have been carried out to assess the inhalation bioaccessibility of toxic metals in indoor particles and to identify the potential sources of the particle-bound toxic elements.

Room or window air conditioners (ACs) are widely used in Chinese common resident households and offices, which have many purposes such as cooling, dehumidification, heating, and purification and have become daily comfortable necessities of life (Besis et al. 2014; Olowoyo et al. 2015; Siddique et al. 2011; Zhang et al. 2016). To save energy, room or window air conditioners do not draw fresh air from outside recirculating the indoor air (Besis et al. 2014). The air conditioners' filter the airborne particles in indoor air, which is similar to the filtration of a vacuum cleaner, and filtration can decrease indoor airborne particles (Batterman et al. 2012). Thus, airborne particles trapped in these filters (AC filter dusts) represent the air quality in the indoor environment (Besis et al. 2014; Siddique et al. 2011). Therefore, the dusts collected from the AC filters can provide information on the indoor air quality (Huang et al. 2014; Siddique et al. 2011), which is useful to better understand the occurrence and health implications of toxic metals in indoor environments (Besis et al. 2014; Olowoyo et al. 2015; Siddique et al. 2011).

In the present study, AC filter dusts were collected from different types of rooms in 2014 in Nanjing, China. The objectives of this work were (1) to identify the potential sources by enrichment factor and a receptor model (absolute principal component scores-multiple linear regression analysis, APCS-MLR) based on elemental total contents of Ca, Fe, Al, Mg, K, Na, Zn, Mn, P, Pb, Cu, Sr, Ti, Ba, Cr, Ni, As, Sb, Co, V, and Cd; (2) to compare and evaluate in vitro inhalation and ingestion bioaccessibility of toxic elements by using in vitro bioaccessibility procedures including SBRC, IVG, SLF, ALF and modified Gamble solution (MGS); and (3) to assess the potential carcinogenic and non-carcinogenic risks via inhalation and ingestion exposure based on the bioaccessibility and total contents of toxic elements.

Material and methods

Sample collection

Nanjing (118° 22′ and 119° 14′ E, 31° 14′ and 32° 37′ N) is the capital city of the Jiangsu province, P. R. China, and has an urban permanent population of approximately 6.4 million people in 2014 (NMBS 2014). Nanjing, Hangzhou, and Shanghai are the most important metropolis of the Yangtze River delta, one of the fastest growing areas in China. Previous researches have shown the enrichment of toxic elements in atmospheric particles, urban road dusts, and soils (Ding and Hu 2014; Hu et al. 2011, 2012, 2013; Sun et al. 2014; Wang et al. 2016b).

The filter dust samples in the air conditioners were collected from teachers' office rooms, marketplaces, classrooms, and students' dormitories in different campuses of Nanjing Tech University in 2014. Generally, the samples trapped in the filter

were collected by initially removing the filter screens from the room air conditioners and then brushing with a hairbrush into a white porcelain dish, and filter dusts from two ACs (three ACs in teachers' office rooms of four sampling sites) in a sampling site were combined into one sample of about 1.0~3.0 g. Numbers of the sampling sites were 6 for the teachers' office rooms, 5 for marketplaces, 9 for classrooms, and 10 for the students' dormitories and total 30 samples were collected ($n = 30$). The obtained dust samples were stored in clean labeled polythene bags and brought to the lab for further treatment ($n = 30$).

Pseudo-total contents of toxic elements analyzed using the aqua regia extraction procedure (ISO standard 11466)

Pseudo-total contents of toxic elements (e.g., Zn, Mn, Pb, Cu, Cr, Ni, As, Sb, Cd) and crustal elements (e.g., Ca, Fe, Al, Mg, K, Na, P, Sr, Ti, Ba) in the samples were determined after digestion in *aqua regia* (ISO Standard 11466) (ISO 1995). Briefly, 0.2 g samples were added to the centrifuge tubes and then 2.0 mL nitric acid was added to digest organic matters. When heated until nearly dryness, newly prepared *aqua regia* of 3.0 mL was added and stood for 16 h at room temperature, and then they was heated slowly to about 100 °C under reflux conditions and maintained for 2 h. After cooling to room temperature, they were transferred quantitatively to a 25-mL graduated flask with distilled water. For QA/QC, randomly selected ten samples were performed in duplicate. If the relative deviation of the duplicate sample was over 10%, all the treatments were redone. Reagent blank was performed synchronously.

Extraction of bioaccessible toxic elements using in vitro procedures

In vitro inhalation bioaccessibility of toxic elements was evaluated by using SLF, ALF, and MGS. In vitro ingestion bioaccessibility was evaluated by using SBRC and IVG. Chemical compositions of those extraction procedures can be found in previous studies (listed in the supplementary materials: Tables S1 and S2) (Boisa et al. 2014; Juhasz et al. 2014; Mukhtar and Limbeck 2013; Ng et al. 2015; Wiseman 2015). The environmental labile concentrations were extracted using a dilute acid solution (0.1 mol L^{-1} HCl) (Snape et al. 2004), which is widely used to extract environmental labile concentrations of heavy metals in different environmental media (Snape et al. 2004) and also used as a kind of simulated gut fluid to represent a very simplified approach to simulate the gastric solution (Twining et al. 2005).

Extraction of pulmonary bioaccessible and environmental labile toxic elements using in vitro inhalation bioaccessibility procedures and dilute HCl solution was carried out by accurately weighing about 0.500 g of sample into 50 mL screw-capped centrifuge tubes containing 25 mL of SLF, MGS, ALF, or dilute acid solution. Those centrifuge tubes were placed on an end-over-end shaker within a water bath at 37 °C for 24 h, and then centrifuged at 3500 rpm for 10 min. The supernatant was transferred to a 10-mL centrifuge tube and stored in refrigerator for further analysis.

Extraction of gastrointestinal bioaccessible toxic elements was carried out by accurately weighing about 0.300 g in 30 and 45 mL of stomach phase solution of SBRC and IVG, respectively. Those centrifuge tubes were placed on an end-over-end shaker within a water bath at 37 °C for 2 h, and then centrifuged at 3500 rpm for 10 min. A 5 mL aliquot supernatant, whose pH differed by no more than 0.2 units from the original value, was pipetted into a 10-mL polypropylene centrifuge tube and stored in refrigerator for elemental analyses. Those supernatants were labeled as *stomach phase* (*SP*). Meanwhile, the remaining contents of the centrifuge tubes were resuspended and titrated to pH 7.0 for SBRC and pH 5.5 for IVG, with saturated sodium bicarbonate solution before bile salts and pancreatin (both porcine) were added. Following incubation at 37 °C for 4 h, those centrifuge tubes were centrifuged, and a 5 mL aliquot was transferred to a polypropylene centrifuge tube (as above) labeled as *intestinal phase* (*IP*).

Quality controls for each phase were performed in duplicate. All the treatments were redone if the relative deviation of the duplicate treatment was over 10%. Procedural reagent blank was performed synchronously.

Elemental analysis

The elemental concentrations (Ca, Fe, Al, Mg, K, Na, Zn, Mn, P, Pb, Cu, Sr, Ti, Ba, Cr, Ni, As, Sb, Cd, Co, and V) in the resultant solutions were measured using an inductively coupled plasma optical emission spectrometer (ICP-OES) (Optima 5300DV, Perkin-Elmer), and Cd, As, Sb, Co, Ni, Pb, and V in some resultant solutions of in vitro bioaccessibility procedures were analyzed by using an inductively coupled plasma-mass spectrometer (ICP-MS) (Elan 9000, Perkin-Elmer). Calibration standards were prepared by dilution of a certified multi-element standard solution (SPEX CertiPrep, USA) in the same matrix as the samples (dilute acid or simulated human body fluids). Mixed internal standard (^{103}Rh and ^{209}Bi) was added online through T-junction at a concentration of 0.02 mg L^{-1} to monitor the drift of the ICP-MS signal due to plasma instability and sample matrix effects. The detection limit of elements in dilute nitric acid (5% in *v*/*v*) were 0.01 (Ca), 0.02 (Fe), 0.01 (Al), 0.008 (Mg), 0.01 (K), (Na), (Zn), 0.008 (Mn), 0.02 (P), 0.03 (Pb), 0.006 (Cu), 0.005 (Sr), 0.005 (Ti), 0.009 (Ba), 0.006 (Cr), 0.004 (Ni), 0.02 (As), 0.01 (Sb), 0.001 (Cd), 0.004 (Co), and 0.005 (V) mg L^{-1} for ICP-OES

and 0.041 (Cd), 0.18 (As), 0.15 (Sb), 0.0032 (Co), 0.024 (Ni), 0.032 (Pb), and 0.084 (V) μg L^{-1} for ICP-MS.

Potential health risk from toxic elements

The potential health risks from toxic elements associated with dusts are mainly attributed to chronic risks. Direct inhalation of resuspended AC filter dust particles through mouth and nose and ingestion of deposited particles on food and drinks are the main exposure pathways to indoor residents. According to the risk-based model developed by the United States Environmental Protection Agency (https://www.epa.gov/risk/human-health-risk-assessment), chronic non-carcinogenic and carcinogenic risks are evaluated by the hazard quotient (HQ) and carcinogenic risks (CR) (Eqs. 1–6 in the supplementary information). Carcinogenic risk is the probability of an individual developing any type of cancer from lifetime exposure to carcinogenic hazards and the acceptable or tolerable risk for regulatory purposes is 1×10^{-6}. The acceptable risk for non-carcinogenic risk is 1 for HQ for single element. The hazard index (HI), sum of HQ, is used to assess the accumulative non-carcinogenic risks for multi-elements. An HI below one indicates that there is no significant risk of non-carcinogenic effects; conversely, there is a chance of non-carcinogenic effects occurring, with a probability that tends to increase as the value of HI increases.

Data analysis

The descriptive statistics and mean comparisons of bioaccessible contents among different procedures were performed using SPSS 16.0 for Windows. One-way analysis of variance (ANOVA) was used to determine whether the bioaccessible contents differed among in vitro inhalation bioaccessibility procedures, and post hoc multiple comparisons of means were conducted using the least significance difference (LSD) test. A $p < 0.01$ was taken to indicate statistical significance. Principal component analysis (PCA) is widely used for source apportionment without source profiles. Accordingly, PCA of the elemental data in this study was carried out using SPSS 16.0 for Windows. Varimax with Kaiser normalization rotation was applied to maximize the variances of the factor loadings across variances for each factor.

Results and discussion

Total contents of toxic elements

The descriptive statistics of element contents in the AC filter dusts is summarized in Table 1. Generally, element average contents in the AC filter dusts are the order of Ca (60.6 mg g^{-1}) > Fe (17.9 mg g^{-1}) > Al (11.3 mg g^{-1}) > Mg (7.58 mg g^{-1}) ≈ K (6.90 mg g^{-1}) ≈ Na (6.43 mg g^{-1}) > Zn (2.23 mg g^{-1}) > Mn (0.74 mg g^{-1}) > P (0.99 mg g^{-1}) > Pb (0.88 mg g^{-1}) > Cu (0.34 mg g^{-1}) > Sr (0.27 mg g^{-1}) ≈ Ti (0.24 mg g^{-1} mg g^{-1}) ≈ Ba (0.23 mg g^{-1}) > Cr (85.7 mg kg^{-1}) > Ni (71.5 mg kg^{-1}) > As (36.0 mg kg^{-1}) > Sb (16.9 mg kg^{-1}) > Cd (8.02 mg kg^{-1}) > Co (6.88 mg kg^{-1}) (Table 1). Crustal elements such as Ca, Fe, Al, Mg, and K were the preponderant elements found in the AC filter dusts while toxic elements such as Cr, Ni, As, Sb, and Cd were lower. However, contents of Pb, Cu, Zn, Ni, Cd, and As are higher than their limit values of the grade 2 of the Chinese environmental quality standard for soils (GB 15618–1995), which are set up to protect agricultural production and human health. So the potential human health from those toxic elements via inhalation and ingestion exposure cannot be ignored.

Table 1 Elemental contents in the filter dusts

	Min	Max	Med	Ave	SD	CV
Al (mg/g)	7.78	16.9	11.4	11.3	2.3	20.8
Ba (mg/g)	0.13	0.49	0.21	0.23	0.07	32.8
Mn (mg/g)	0.27	2.12	0.73	0.74	0.30	40.3
Cu (mg/g)	0.11	0.45	0.38	0.34	0.09	25.5
Pb (mg/g)	0.20	0.88	0.50	0.51	0.16	32.4
Zn (mg/g)	0.86	3.1	2.39	2.23	0.53	23.9
Fe (mg/g)	6.53	31.3	17.9	17.9	4.0	22.1
Ti (mg/g)	0.086	0.35	0.24	0.24	0.05	20.3
Sr (mg/g)	0.064	0.47	0.27	0.27	0.07	25.0
Ca (mg/g)	15.1	74.3	64.1	60.6	14.4	23.7
K (mg/g)	1.40	15.9	7.04	6.90	2.42	35.0
Mg (mg/g)	2.77	13.6	7.74	7.58	1.90	25.1
Na (mg/g)	0.98	8.48	6.71	6.43	1.53	23.7
P (mg/g)	0.38	1.27	1.05	0.99	0.19	19.1
Cr (mg/kg)	59.0	186	75.7	85.7	28.8	33.6
V (mg/kg)	27.8	87.3	48.2	48.0	12.3	25.5
Ni (mg/kg)	48.9	152	60.6	71.5	25.3	35.4
As (mg/kg)	14.9	53.1	38.0	36.0	10.7	29.8
Cd (mg/kg)	1.15	21.2	7.58	8.02	3.66	45.6
Co (mg/kg)	2.87	12.1	6.76	6.88	1.58	23.0
Sb (mg/kg)	9.40	29.2	16.4	16.9	4.7	27.5

Min: minimum; Max: maximum; Med: median value; Ave: average value; SD: standard deviation

Source identification by enrichment factors and APCS-MLR

In order to distinguish the anthropogenic inputs in those samples, the enrichment factors (EFs) were calculated using the ratio a certain elemental content (Ci) to a reference element (C_{Fe}) in the studied sample and the reference material $((C_i/C_{Fe})_{sample}/(C_i/C_{Fe})_{reference})$ (Chen et al. 2008). Soil

background values of metallic elements in Nanjing were selected as reference (CNEMC 1990) and the EFs were shown in Fig. 1. Figure 1 shows that Cd exhibits the highest EF values, followed by Zn, Pb, Sb, and Cu. The average EF values for As, Cu, Ni, Pb, and Zn were above 10 (Fig. 1). An EF value greater than 10 is considered as an indicator of a significant anthropogenic input of an element in airborne particles (Chen et al. 2008) while EF < 2 indicates minimal anthropogenic enhancement (Sutherland et al. 2001). Therefore, obvious anthropogenic enhancement of Cd, Cu, Pb, Sb, and Zn was indicated in the filter dusts. These suggest more potential health risks via inhalation and ingestion exposure to toxic elements in the filter dusts.

For PCA, the value of KMO (Kaiser-Meyer-Olkin's test) is 0.760, which meets the limit of 0.600 conventionally held as a critical value. Bartlett's test of sphericity shows that principal component analysis can be applied to the data at the $p < 0.01$ level. Four principal components were identified and they accounted for 83.1% of the total variance in the dataset (Table 2). The first component (factor 1), which explained 35.3% of the total variance with an eigenvalue of 7.42, was most dependent on Cd, Pb, Zn, As, Sb, and Cu (loading coefficient > 0.85) (Table 2). These elements are generally considered as environmental contaminants from wide anthropogenic sources. EF values confirmed the enrichment of Cd, Pb, Zn, and As in the present study (Fig. 1). Pb, Zn, and Cd were representatives of automobile exhaust (Fang et al. 2010). The average As concentration in Chinese coal is 3.18 mg kg^{-1}, and As is one of the characteristic elements of coal combustion in China (Kang et al. 2011). Cu is a marker of automobile brake pads and worn tires (Gietl et al. 2010). These indicate that this component can be attributed to anthropogenic sources mixed with industrial, coal combustion, and traffic emission. The second component (factor 2), which explained 23.7% of the total variance with an eigenvalue of 4.97, appeared to represent natural sources since it was strongly correlated with

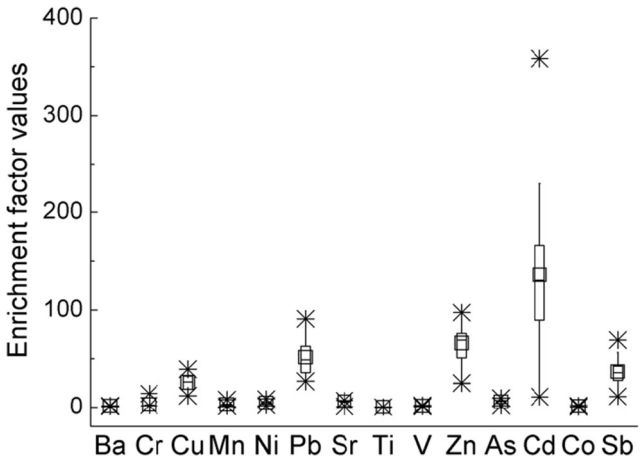

Fig. 1 Values of enrichment factors for the studied elements in the filter dusts

Table 2 Varimax rotated component matrix

	Components				Communalities
	1	2	3	4	
Al	0.35	0.71		0.43	0.83
Fe		0.96			0.95
Ba	−0.67	0.56			0.77
Cr	−0.47	−0.63			0.71
Cu	0.89				0.84
Mn	0.67	0.42		0.42	0.86
Ni				0.78	0.68
Pb	0.94				0.89
Sr	0.57	0.39		0.62	0.87
Ti		0.85			0.82
V		0.80			0.74
Zn	0.91				0.95
As	0.94				0.92
Cd	0.86	0.35			0.87
Co		0.86		0.33	0.91
Ca	0.39		0.59		0.62
K	0.36		0.89		0.96
Mg			0.95		0.96
Na			0.85		0.79
P	0.62		0.48		0.64
Sb	0.91				0.85
Eigenvalues (>1)	7.42	4.97	3.23	1.83	
% of variance	35.3	23.7	15.4	8.70	
Cumulative %	35.3	59.0	74.4	83.1	

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization

Rotation converged in five iterations

Al, Fe, Ba, Ti, V, and Co (Table 2). Cr showed a native correlation with those elements, suggesting different origins (Table 2). Al, Fe, Ba, Ti, and V are generally associated with soil and dust (Fang et al. 2010). This component can be attributed to natural sources of soil and dust. The third component (factor 3) mainly consisted of Na, K, and Na, which explained 15.4% of the total variance with an eigenvalue of 3.23 (Table 2). Ca shows a loading of 0.59 (Table 2). This component appeared to be related to a mixture nature factor. For example, Na is regarded as a marker element of sea salt, sea spray, and marine sources (Fang et al. 2010). Na, Mg, and K are the main elements in the sea salt particles. Moreover, K is also considered to be trace elements of biomass incineration and Ca and Mg are the indicator elements of construction dusts (Fang et al. 2010). This component may be another natural source (sea salt particles and biomass incineration). The fourth component (factor 4) was mainly composed of Ni and Sr and explained 8.70% of the total variance with an eigenvalue of 1.83 (Table 2). Ni is generally considered a fingerprint of oil combustion (Fang

et al. 2010), while coal combustion is also the leading source of atmospheric nickel in Chinese cities (Tian et al. 2012). Sr loading in this factor (0.62) is similar to that in factor 4 (0.57) and Sr also shows higher loading in factor 2 (0.39) (Table 2). Those suggest that Sr have different sources. In a word, this component likely reflects another anthropogenic source (combustion emission of oil and coal). Overall, results of the principal component analysis show that outdoor sources rather than common indoor sources were the preponderant sources of the filter dusts.

For further assessment of source apportionment of filter dusts, the receptor model-APCS-MLR was used to estimate the contributions (%) of the identified four sources mentioned above (Thurston et al. 2011). In this study, enter MLR was applied using standardized values of element contents as dependent variables and absolute factor scores (obtained from PCA) as independent variables. The resultant regression coefficients were then employed to convert the absolute factor scores to produce estimates of each PC source contribution (Harrison et al. 1996). The contributions were 33.5, 29.1, 22.6, and 14.8% for the first, the second, the third, and the fourth component, respectively.

In vitro ingestion bioaccessibility of toxic elements

Due to the difference on the elemental contents and extraction procedures, it is a tough task to compare directly the extractable contents of elements among in vitro procedures. Therefore, bioaccessibility is defined as the soluble fraction of toxic elements in the environmental medium extracted by in vitro procedures (Boisa et al. 2014; Ng et al. 2015). In the present study, in vitro ingestion bioaccessibility of toxic elements is referred to the fraction of an element that can be dissolved in the in vitro ingestion bioaccessibility procedures. The in vitro ingestion bioaccessibility of toxic elements is shown in Table 3. Table 3 shows that the in vitro ingestion bioaccessibility varied greatly among toxic elements. For example, Pb, Mn, and Zn had higher gastric bioaccessibility while Co, Cr, and Sb had lower gastric bioaccessibility for both SBRC and IVG procedures (Table 3). Table 3 also shows that the gastric bioaccessibility of toxic elements by SBRC procedure was significant higher than those by IVG procedure except for Zn and As. So the in vitro ingestion bioaccessibility of toxic elements showed elemental dependence; the in vitro procedures (i.e., SBRC and IVG procedure) had also great influence on the in vitro ingestion bioaccessibility of toxic elements (Table 3). The percentages of Cu, As, Cd, Co, and Sb extracted by dilute HCl were significant higher than their gastric bioaccessibility by both SBRC and IVG procedure (Table S3 and Table 3), suggesting that the bioaccessible fractions resulted mainly from the loose binding of metals to dust particles. Table 3 shows that Mn and Zn showed higher intestinal bioaccessibility, and then V and Ni, while Al, Cr, Co, and Sb showed lower intestinal bioaccessibility for both SBRC and IVG procedure. Table 3 shows that there were no significant differences on intestinal bioaccessibility of toxic elements between SBRC and IVG procedure except for Al, Cu, and Sb. From stomach phase to intestinal phase, bioaccessibility of Al and Pb decreased significantly (Table 3). Therefore, the in vitro ingestion bioaccessibility of toxic elements shows elemental and in vitro procedure dependence.

Table 3 Average values (± standard deviation) of ingestion bioaccessibility of toxic elements in the filter dusts (%)

	SP/SBRC	SP/IVG	IP/SBRC	IP/IVG
Al	30.2 ± 6.1	22.0 ± 4.2	3.27 ± 2.43	13.4 ± 5.0
Ba	35.6 ± 6.6	23.6 ± 5.4	23.6 ± 6.9	23.5 ± 6.3
Mn	62.5 ± 5.5	46.0 ± 7.6	44.8 ± 7.7	56.5 ± 9.9
Cu	31.6 ± 4.5	21.9 ± 3.1	37.7 ± 6.6	21.0 ± 5.5
Pb	60.0 ± 9.3	43.6 ± 10.5	12.0 ± 5.9	21.2 ± 8.2
Zn	59.6 ± 7.4	59.5 ± 8.0	64.5 ± 9.5	56.9 ± 11.5
Cr	13.5 ± 2.7	6.85 ± 2.16	17.7 ± 5.4	10.6 ± 3.3
V	45.5 ± 4.5	28.5 ± 6.2	29.9 ± 6.7	38.4 ± 7.1
Ni	40.3 ± 8.1	27.6 ± 6.1	38.8 ± 8.7	34.5 ± 7.1
As	18.0 ± 4.3	13.2 ± 3.5	14.8 ± 4.6	14.6 ± 4.7
Cd	30.7 ± 6.0	19.4 ± 3.4	27.2 ± 6.3	18.9 ± 4.2
Co	10.1 ± 2.4	5.80 ± 1.14	15.4 ± 2.9	9.03 ± 7.26
Sb	12.4 ± 3.8	8.18 ± 2.89	17.0 ± 4.3	9.28 ± 3.19

SP: stomach phase; IP: intestinal phase

The compositions and pH of in vitro procedures may be the main reasons for the abovementioned differences on the in vitro ingestion bioaccessibility. For example, Table S2 shows that glycine is the only component for the *stomach phase* solution of SBRC procedure while pepsin and NaCl are the main component for the *stomach phase* solution of IVG procedure. Although bile and pancreatin are the main reagents for the *intestinal phase* solution for both SBRC and IVG procedures, their concentrations are different. The pH values are 1.5/7.0 and 1.8/5.5 for the *stomach/intestinal phase* solution of SBRC and IVG procedures (Table S2). The previous investigation confirmed the influence of in vitro assay pH and extractant compositions on As bioaccessibility in contaminated soils (Smith et al. 2014).

In vitro inhalational bioaccessibility of toxic elements

Inhalation is the direct expose route to fine airborne particles entering into the lung. Toxic elements associated with airborne particles may be dissolved in the lung physiological solution and then enter the blood system, which may pose potential health risks. In vitro inhalational bioaccessibility of toxic elements (i.e., solubility of toxic elements in simulated lung physiological fluids) is listed in Table 4. The inhalational bioaccessibility of toxic elements extracted using ALF, SLF,

Table 4 In vitro inhalation bioaccessibility of toxic elements (%)

	Simulated lung fluid		Modified Gamble solution		Artificial lysosomal fluid	
	Range	Ave ± SD	Range	Ave ± SD	Range	Ave ± SD
Al	0.073~0.93	0.28 ± 0.20 [a]	0.031~0.88	0.24 ± 0.21 [a]	0.022~1.15	0.25 ± 0.26 [a]
Ba	0.12~0.63	0.24 ± 0.10 [b]	0.055~0.43	0.17 ± 0.09 [b]	0.13~2.25	0.56 ± 0.45 [a]
Mn	0.55~9.70	4.38 ± 1.74 [a]	0.35~6.19	1.92 ± 1.16 [b]	1.80~18.0	5.53 ± 2.73 [a]
Cu	0.44~72.0	8.95 ± 12.5 [a]	0.66~11.3	3.71 ± 2.59 [b]	0.43~18.9	6.20 ± 4.14 [a]
Pb	0.21~3.46	1.07 ± 0.78 [a]	0.071~3.94	0.46 ± 0.68 [b]	0.19~2.99	0.79 ± 0.64 [a,b]
Zn	0.59~6.59	2.72 ± 1.54 [b]	0.19~5.01	0.89 ± 0.87 [b]	0.66~5.50	2.63 ± 1.12 [a]
Cr	0.48~4.10	2.05 ± 0.87 [a,b]	0.80~3.65	1.72 ± 0.60 [b]	0.79~5.43	2.78 ± 1.30 [a]
V	1.55~7.86	5.40 ± 1.42 [a]	1.41~7.17	3.14 ± 1.21 [b]	1.41~7.17	3.14 ± 1.21 [b]
Ni	3.21~17.6	9.33 ± 3.24 [a]	3.03~22.2	8.98 ± 4.11 [a]	4.22~16.9	9.28 ± 3.13 [a]
As	13.0~47.1	26.9 ± 7.76 [b]	18.8~95.5	42.2 ± 13.5 [a]	8.09~24.7	14.9 ± 4.22 [c]
Cd	0.64~21.6	5.56 ± 4.27 [a]	0.52~19.4	3.75 ± 3.89 [a]	0.79~14.5	3.93 ± 2.98 [a]
Co	3.44~16.9	10.0 ± 3.49 [a]	2.73~12.8	6.23 ± 2.56 [b]	3.45~22.9	11.1 ± 4.18 [a]
Sb	1.29~19.7	8.20 ± 4.12 [a]	0.40~14.5	6.22 ± 3.71[a]	0.44~21.6	8.17 ± 5.28 [a]

Average values with different letters in same row differ significantly by Duncan's test ($p < 0.05$)

and MGS have obvious elemental dependence (Table 4). For example, Table 4 shows that the average value of elemental bioaccessibility was in the order of As > Co > Ni ≈ Cu > Sb > Cd > Mn > Zn ≈ Cr > Pb > Al ≈ Ba for SLF, As > Ni > Co ≈ Sb > Cd ≈ Cu > Mn ≈ Cr > Zn > Pb > Al ≈ Ba for MGS, and As > Co > Ni > Sb > Cu > Mn > Cd > Cr ≈ Zn > Pb > Ba > Al for ALF. As showed the highest bioaccessibility, followed by Co, Ni, Cu and Sb, while Pb, Ba, and Al had the lowest bioaccessibility for the three in vitro procedures (Table 4). The values of inhalational bioaccessibility of toxic elements were generally higher in ALF than those in SLF and MGS (Table 4). Those indicate the influences of the in vitro procedures (i.e., ALF, SLF, and MGS) on the in vitro bioaccessibility of toxic elements. The significant differences were found for most of the studied metals among ALF, SLF, and MGS procedures by Duncan's test ($p < 0.05$) (Table 4).

Table S1 shows that total dissolved solid (TDS) is approximately 30 g L^{-1} for ALF, 10 mg L^{-1} for SLF, and 20 mg L^{-1} for MGS. The main components are NaCl (6.4 g L^{-1}) and $NaHCO_3$ (2.7 g L^{-1}) for SLF and NaCl (6.4 g L^{-1}), NH_4Cl (5.3 g L^{-1}), and $NaHCO_3$ (2.3 g L^{-1}) and NaH_2PO_4 (1.7 g L^{-1}) for MGS (Table S1). The dominant component for ALF is citric acid (22.75 g L^{-1}, about two thirds of the TDS), an organic phase reagent, and then NaOH (6.0 g L^{-1}) and NaCl (3.21 g L^{-1}). The compositions of in vitro procedures used differed greatly. Moreover, the pH values are 4.5, 7.4, and 7.6 for ALF, MGS, and SLF, respectively (Table S1). The pH and chemical compositions of ALF simulate the acidic cellular conditions, which occur following phagocytosis, while MGS and SLF represent the neutral conditions of the interstitial fluid found deep in the human lung. Those may result in the differences on the in vitro bioaccessibility of the studied metals. The in vitro bioaccessibility procedures should be further optimized and validated for the risk-based assessment.

Potential health risk from toxic elements

The non-carcinogenic and carcinogenic risks to residents were evaluated by using the risk-based assessment model advocate by U.S. EPA (Environmental Protection Agency of the United States, https://www.epa.gov/risk/human-health-risk-assessment). According to the classification defined by the IARC (International Agency for Research on Cancer), As and inorganic arsenic compounds, chromium (VI) compounds, nickel compounds, cadmium, and cadmium compounds are class I carcinogenic pollutants, lead compounds (inorganic) are class 2A, and cobalt and cobalt compounds are class 2B. Therefore, their lifetime carcinogenic risks were investigated in the present study. Table 5 shows that values of carcinogenic risks were beyond the acceptable level (1×10^{-6}) for As, Cr, and Pb via ingestion exposure and for Cr via inhalation exposure. Cr toxicity is directly dependent on its valence state (Cr(VI) and Cr(III)); total Cr rather than Cr (III) were determined in this study, which would overestimate the potential risks. Therefore, the potential carcinogenic risks existed based on elemental total contents. The risks based on the in vitro bioaccessibility was also calculated (Tables S4 and S5). Table S4 shows that the carcinogenic risks of toxic elements were within the acceptable level (1×10^{-6}) based on their in vitro inhalation bioaccessible contents extracted by using SLF, ALF, and MGS. Table S5 shows that the carcinogenic risks of As, Cr, and Pb were just beyond the acceptable level (1×10^{-6}) based on their in vitro ingestion bioaccessible contents extracted by using SBRC and IVG procedures.

Table 5 Non-carcinogenic and carcinogenic risks to both children and adults via ingestion and inhalation exposure based on elemental total contents

	HQ				Lifetime CR	
	Ingestion		Inhalation		Ingestion	Inhalation
	Children	Adults	Children	Adults		
As	7.89E−01	8.45E−02	2.03E−03	8.70E−04	4.35E−05	3.05E−08
Cr	1.88E−01	2.01E−02	7.25E−04	3.11E−04	3.45E−05	1.42E−06
Pb					1.56E−05	1.10E−09
Cd	1.05E−01	1.13E−02	6.79E−04	2.91E−04		2.84E−09
Co	1.51E−01	1.62E−02	9.70E−04	4.16E−04		1.22E−08
Ni	2.35E−02	2.52E−03	6.72E−04	2.88E−04		3.66E−09
Al	7.43E−02	7.96E−03	1.91E−03	8.19E−04		
Ba	7.56E−03	8.10E−04	3.89E−04	1.67E−04		
Cu	5.59E−02	5.99E−03				
Mn	2.03E−01	2.17E−02	1.25E−02	5.37E−03		
Sb	2.78E−01	2.98E−02				
V	6.31E−02	6.76E−03				
Zn	4.89E−02	5.24E−03				

Therefore, the carcinogenic risks of toxic elements mainly resulted from the ingestion exposure.

The non-carcinogenic risks from these toxic elements via inhalation and ingestion exposure were also calculated (Table 5). The HQ and HI values for studied toxic elements via inhalation exposure for both children and adults were all lower than the safe level (=1), indicating no non-carcinogenic risks from the inhalation exposure for a single element and accumulative effects of multi-elements (Table 5). Non-carcinogenic risks based on the in vitro bioaccessible contents extracted by using SLF, ALF, and MGS to both children and adults were all lower than those based on total contents (Table S4). Although HQ values for studied toxic elements via ingestion exposure for both children and adults were all lower than the safe level (=1) (Table 5 and Table S5), HI was 1.99 for children and 0.21 for adults, suggesting the accumulative non-carcinogenic risks to children via ingestion exposure. Arsenic (HQ = 0.79) was the main contributor, followed by Sb (0.28), Mn (0.20), Cr (0.19), Co (0.15), and Cd (0.11) (Table 5). HQ values for other elements were lower than 0.1 (Table 5). To adults, HQ values for all toxic elements were lower than 0.1 (Table 5). So there were potential non-carcinogenic risks to children for single element via ingestion exposure. HQ and HI values based on the in vitro bioaccessible contents extracted by SBRC and IVG procedures were all within the safe level (=1) (Table S5).

Conclusions

Crustal and toxic elements in the AC filter dusts were analyzed and significant enrichment of toxic elements such as Cd, Cr, Cu, Ni, Pb, Sb, and Zn in the filter dusts was confirmed by the values of enrichment factors. Four potential sources with the contributions of 33.5, 29.1, 22.6, and 14.8% were identified by APCS-MLR model, revealing the outdoor input of these toxic elements rather than indoor sources. Pb, Mn, and Zn had higher gastric bioaccessibility while Co, Cr, and Sb showed lower gastric bioaccessibility for both SBRC and IVG procedures. Mn and Zn showed higher intestinal bioaccessibility, followed by V and Ni, while Al, Cr, Co, and Sb had lower intestinal bioaccessibility for both SBRC and IVG procedures. Gastric bioaccessibility of toxic elements by SBRC procedure was significantly higher than them by IVG procedure except for Zn and As, while there were no significant differences on intestinal bioaccessibility of toxic elements between SBRC and IVG procedure except for Al, Cu, and Sb. From stomach phase to intestinal phase, bioaccessibility of Al and Sb decreased significantly. The values of inhalational bioaccessibility of toxic elements were generally higher for ALF procedure than those for SLF and MGS procedures. As showed the highest bioaccessibility, followed by Co, Ni, Cu, and Sb, and Pb, Ba, and Al had the lowest bioaccessibility for the three in vitro procedures. Moreover, the environmental labile fractions extracted by dilute HCl were significantly higher than or similar to the gastric/intestinal bioaccessibility, indicating the loose binding of metals to dusts for the bioaccessible fractions. Based on the bioaccessible contents of toxic elements in filter dusts, there were potential carcinogenic risks via ingestion exposure and no non-carcinogenic risks to both children and adults via inhalation and ingestion exposure. Therefore, the outdoor input of toxic elements in indoor airborne particles may result in

potential health risks via ingestion/inhalation exposure to indoor airborne particle-bound elements.

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