High Enrichment of Heavy Metals in Fine Particulate Matter through Dust Aerosol Generation

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Atmospheric Chemistry and Physics

July 4th, 2023

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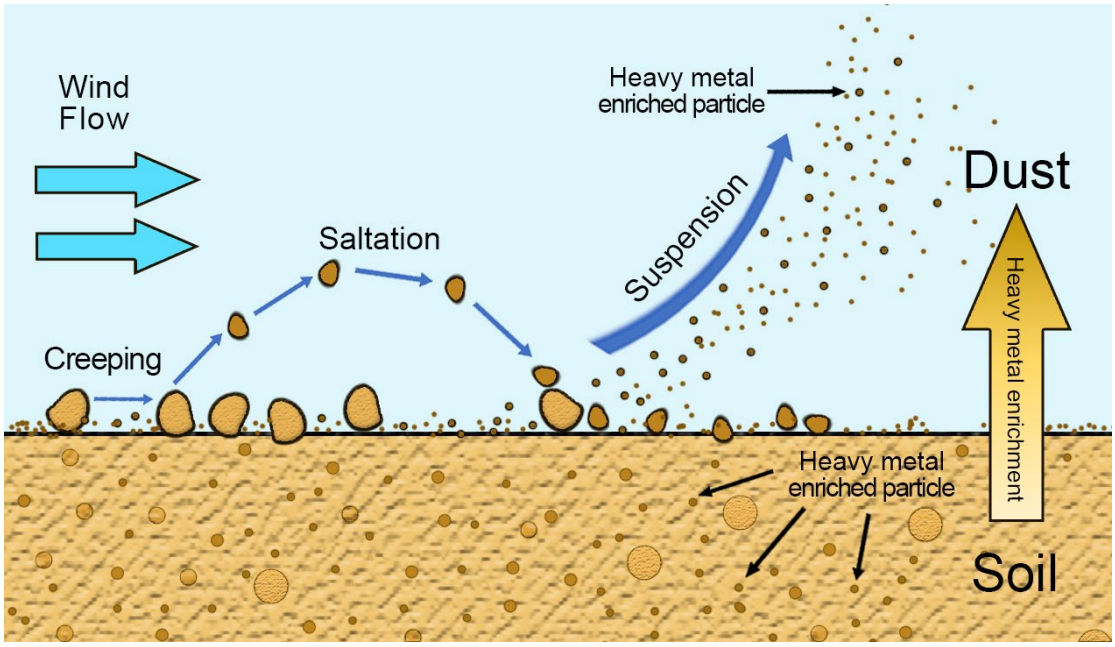
Abstract

Dust is a major source of atmospheric aerosols. Its chemical composition is often assumed to be similar to the parent soil. However, this assumption has not been rigorously verified. Here, we generated dust aerosols from soils to determine if there is particle size-dependent selectivity of heavy metals in the dust generation. Mn, Cd, Pb and other heavy metals were found to be highly enriched in fine ($PM_{2.5}$) dust aerosols, which can be up to ~6.5-fold. To calculate the contributions of dust to atmospheric heavy metals, regional air quality models usually use the dust chemical profiles from the US EPA's SPECIATE database, which does not capture the correct size-dependent selectivity of heavy metals in dust aerosols. Our air quality modeling for China demonstrates that the calculated contribution of fine dust aerosols to atmospheric heavy metals, as well as their cancer risks, could have significant errors without using proper dust profiles.

Graphical Abstract



Short Summary

Dust is a major source of atmospheric aerosols. Its chemical composition is often assumed to be similar to the parent soil. However, this assumption has not been rigorously verified. Dust aerosols are mainly generated by wind erosion, which may have some chemical selectivity. Mn, Cd and Pb were found to be highly enriched in fine ($PM_{2.5}$) dust aerosols. In addition, estimation of heavy metal emission from dust generation by air quality models may have errors without using proper dust profiles.

1 Introduction

The major sources of natural aerosols include mineral dust aerosols produced by wind erosion (Prospero et al., 2002). Dust aerosols are influenced by regional atmospheric circulation, soil characteristics and local weather conditions (Bryant, 2013; Ding et al., 2005; Huebert et al., 2003; Liu et al., 2004; Yang et al., 2008), mainly generated and aerosolized when strong wind passes over soil or sandy areas (Gillette and Goodwin, 1974). Recent studies show mineral dust aerosol accounts for approximately 40% of the mass fraction of natural atmospheric aerosol, with an estimated annual flux of ~2,000 $Tg \cdot yr^{-1}$ (Alfaro, 2008; Griggs and Noguer, 2002; Huneeus et al., 2011; Textor et al., 2006). As the second-largest natural source of atmospheric aerosols in terms of mass flux, dust aerosol has a profound impact on the ecosystem (Middleton et al., 2019), especially the climate (Evan et al., 2014; Kok et al., 2018; Shao et al., 2013). Interactions between dust aerosols and water vapor play a critical role in cloud condensation and ice nucleation processes (Kaufman et al., 2002; Tang et al., 2016). Dust particles can be transported on large scales (Shao and Dong, 2006), and could act as a medium to transport toxic compounds, including heavy metals, which significantly harm human health, particularly the human respiratory system and even cause premature death (Urrutia-Pereira et al., 2021).

Atmospheric studies often assume that the chemical composition of aerosolized dust is similar to the parent soil (Gunawardana et al., 2012; Zhuang et al., 2001). The chemical composition of dust aerosol consists of a key part in source apportionment modeling (Balakrishna and Pervez, 2009; Samiksha et al., 2017; Santos et al., 2017; Ying et al., 2018). A critical approach in source apportionment modeling is the chemical transport model, which predicts the dust aerosol on global

and regional scales based on the prior knowledge of source emission, atmospheric transport, and chemical reaction process. SPECIATE is the EPA's speciation profiles repository of air pollution sources of volatile organic compounds (VOCs) and particulate matter (PM). Therefore, the US EPA's SPECIATE database is an important product to convert total emissions from specific sources into the speciated emissions needed for the chemical transport model. The previous study has combined the US EPA's SPECIATE database and air quality model to predict dust aerosols (Ying et al., 2018), based on the assumption of the chemical composition of dust aerosols is similar to the resuspended soil profiles.

Yet, dust generation and aerosolization are complex processes, which may have some chemical selectivity. Most small dust particles (< 20 μm) are produced either by wind erosion, which leads to soil movements such as creeping, saltation, and suspension (Burezq, 2020) or sandblasting process, which leads soil particles (~75 μm) to be lifted by the wind, move in ballistic trajectories due to the combined effect of aerodynamic force and gravity force (Grini and Zender, 2004; Shao and Raupach, 1993; Shao et al., 1996). The sandblasting efficiency of a soil particle is highly sensitive to its size (Grini and Zender, 2004; Grini et al., 2002). In addition, the chemical composition of soil particles can also vary with particle size. As smaller soil particles are more easily ejected, dust aerosol particles are unlikely to have exactly the same composition as their parent soils (Perlwitz et al., 2015; Wu et al., 2022). Dust deposited samples were the dust samples collected on the road or other surfaces using a brush and plastic tray (Shangguan et al., 2022), while dust aerosol samples were collected by filtering the air. Dust aerosols were produced by the ballistic impacts of wind-driven sand grains (Kok et al., 2023). Indeed, some previous studies do find that in the deposited dust samples (not dust aerosol samples), smaller particles tend to contain higher amounts of heavy metals

93 (Naderizadeh et al., 2016; Parajuli et al., 2016; Becagli et al., 2020). However, the heavy metal
94 profiles for dust aerosols from the US EPA's SPECIATE database seem to have no such enrichment
95 between each particle size, as Table S1 reports profile 41350 as an example. Although these profiles
96 have been widely used in air quality modeling works (Lowenthal et al., 2010; Simon et al., 2010;
97 Ashrafi et al., 2018), they were actually measured in the 1970s and 1980s with the resuspension of
98 soil samples, which placed soil in a glass tube and drew air flow to blow and suspend the soil
99 particles to the air (Miller et al., 1972). This method is not likely to produce realistic dust aerosols,
100 as it does not simulate sandblasting process properly. It is not known whether using such a
101 problematic dust profile could significantly impact air quality model calculations.
102
103 Here we examined the enrichment of heavy metals in the laboratory-generated dust aerosols. A dust
104 aerosol generator that mimics realistic sandblasting and saltation was used to generate dust aerosol
105 from a collection of soil samples (Lafon et al., 2014). The concentrations of heavy metals in soil
106 and dust aerosols were measured by an inductively coupled plasma mass spectrometer (ICP-MS).
107 In this study, some heavy metals, such as Mn, Cd, Zn and Pb, were found to be highly enriched in
108 dust aerosols. Especially, the enrichment factors would be much higher for smaller dust aerosols. In
109 addition, we also utilized a single particle aerosol mass spectrometer (SPAMS) to study heavy metal-
110 containing dust aerosols before, during, and after a dust storm. Regional air quality models usually
111 use problematic dust composition profiles from the US EPA's SPECIATE database. Herein we
112 modeled the contribution of dust aerosol to atmospheric heavy metal loadings, utilizing a range of
113 dust aerosol profiles determined in this laboratory study as well as the SPECIATE profile, to
114 investigate whether using a proper dust profile is critical to air quality modeling and cancer risk

calculations.

2 Materials and methods

2.1 Soil sample collection

Fourteen samples were collected from the top 10 cm of the natural soil profile from various locations in dust source regions and Shanghai, China (Table S2, Fig. S1). S1-S4 were collected from dust sources on the northern slope of Yinshan Mountain in central inner Mongolia and the adjacent areas of the Hunshandake Sandy Land, S5-S12 were collected from dust sources of Hexi Corridor and Alxa Plateau, S13 was collected in Xinjiang Province, in the dust sources of the Taklimakan Desert, and S14 was sampled from Shanghai Yangpu District. Although the soil (S14) collected in Shanghai does not originate from a dust source region, it can still produce dust aerosols in some cases. For example, under dry weather conditions, the soil surface in the Shanghai area could serve as a significant local contributor to the generation of dust aerosols (Liu et al., 2016; Liu et al., 2020). During the prevailing dust storm periods from March to May, Shanghai is primarily influenced by dust originating from the western Inner Mongolia Gobi, deserts in the Tibetan Plateau, and arid deserts in northwest China (Fu et al., 2010; Fu et al., 2014; Sun et al., 2017). Soil texture determination was conducted according to the method outlined in a previous study (Kettler et al., 2001). Soil texture characterization was conducted based on the method outlined in a previous study (Kettler et al., 2001). Soil particle dispersion was achieved by adding hexametaphosphate (HMP) and sodium hydroxide (NaOH) to a soil sample (particle size< 2 mm) and shaking it for 16 hours. The percentage of sand and silt was obtained using a Laser Scattering Particle Size Distribution Analyzer (LA-960). Further details can be found in the SI. As shown in Table S2, they represent

several soil types: S1 was silty loam; S2, S4, S7, S10, S11 and S12 were sand; S3 was sandy loam; S5 and S6 were loam; S8 and S13 were loam sand; S9 and S14 were silty clay loam. Before dust aerosol generation, soil samples were placed in a fume hood and left to dry, without stirring or other treatment, before aerosolization. Fine and coarse dust aerosols ($PM_{2.5}$ and PM_{10}) were produced with a GAMEL dust aerosol generator, which can realistically simulate the sandblasting process. Then, the pH of the soil was measured. Detailed information can be found in Fig. S1 and Table S2.

2.2 Laboratory dust aerosol generation and collection

A laboratory dust generator (GAMEL: "Générateur d'Aérosol Minéral En Laboratoire") (Lafon et al., 2014) was used to produce dust aerosols from the soil samples. The GAMEL dust generator can realistically simulate the sandblasting process. Wind tunnels have the advantage of realistically simulating the generation of dust aerosols. However, conducting this study has certain drawbacks. These include the requirement for a substantial quantity of parent soils and the significant cost associated with eliminating ambient aerosol interference (Alfaro et al., 1997; Lafon et al., 2006; Alfaro, 2008). In GAMEL's dust production system, 10 g of each soil sample was added to a PTFE flask, which was agitated by a shaker simulating the sandblasting process to produce dust aerosols. A constant flow of particle-free air was passed through the dust-generating flask. The optimal generation parameter of the shaker was set at a frequency of 500 cycles/min according to Lafon et al., 2014 with an airflow rate of 8 liter/min controlled by a Mass Flow Controller (MFC, Sevenstar, Beijing Sevenstar Flow Co., LTD). The sample stream was filtered through a cyclone and particles were collected on a 47 mm PVC film held in a metal frame filter holder (Pall Gelman, Port Washington, NY, USA). Dust-$PM_{2.5}$ and dust-PM_{10} were obtained with or without an 8LPM cyclone,

157 respectively. The running time was 1min. To obtain more dust aerosols in different size ranges, size-
158 fractionated particle sampling of dust aerosols was carried out with 10-stage Micro-Orifice Uniform
159 Deposit Impactor (MOUDI 110R; MSP) with size cut points of 10 μm, 5.6 μm, 3.2 μm, 1.8 μm, 1.0
160 μm, and 0.56 μm. Analysis of the size distribution and chemical composition of dust generated by
161 GAMEL and dust generated under natural conditions has shown that the GAMEL generator can
162 produce realistic dust aerosol (Lafon et al., 2014). All the dust aerosol mass collected is shown in
163 Table S3 and S4. The instrument setup is illustrated in Fig. S2.
164

165 **2.3 Analysis of laboratory-generated dust aerosols**

166 The dust aerosol samples collected were weighed with an analytical balance and then put into 25 ml
167 digestion tubes with 6 ml 69% HNO_3 symmetrically. The temperature program of Microwave
168 Digestion (Anton Paar) was as follows: initial temperature of 100 °C held for 5 min, then ramped
169 to 140 °C for 5 min, and finally at 180 °C for 60 min. The whole process was holding 120 min.
170 According to this study (Chang et al., 1984), almost all the heavy metal elements in the natural soil
171 and dust aerosol in concentrated nitric acid were extracted using this experimental procedure. After
172 digestion, the solution was acid-fed at 120 °C for 1.5 h, then deionized water (conductivity 18.25
173 MΩ) was added, the volume was constant with a 25 mL volumetric flask, and then passed through
174 a 0.45 μm membrane. The samples were diluted with 2% HNO_3 by 4 times for further analysis.
175 Three blank PVC film samples were digested using the same method for background control.
176

177 The heavy metal content was determined by inductively coupled plasma mass spectrometer (ICP-

MS; Agilent, 8900). Before analysis, tuning procedures including plasma parameter, ion transmission path, quadrupole mass spectrometer, and detector had been done. During analysis, standard solutions were prepared at concentrations of 0, 1, 2, 5, 10, 20, 50, and 100 μg/L. "In, Bi, and Rn" were used as internal standard elements, and were introduced into the nebulizer by mixing with the sample to be tested and the standard solution in the sampling pipeline by online addition, and the instrument drift and matrix effect were compensated. After each analysis of a sample, 2% dilute nitric acid was used to clean the injection line for 1 min, and then continue to collect the second sample to eliminate the memory effect of the previous sample.

A scanning electron microscope (SEM; Phenom Pro) equipped with an energy-dispersive X-ray detector was used for morphologies of particle examination at the voltage of 10 kV. All the samples (soil, $PM_{2.5}$ and PM_{10}) were on the carbon conductive adhesive, then spray platinum to improve the conductivity. Here, the parent soil of S10 and generated $PM_{2.5}$ and PM_{10} were examined.

Statistical analysis was performed using SPSS Statistics. The correlation analysis was conducted through Spearman's correlation and the significant difference was used with an independent sample T-test.

2.4 Ambient dust aerosol measurements

On May 23rd, 2018 (LT), on-site field measurements were conducted in Shanghai to assess the ambient dust particles. The measurements indicated an average wind speed of 2.2 m/s, which corresponds to a level of floating dust storm with a visibility of up to 10 km. The sampling was located on the sixth floor of the Environmental Science Building in Jiangwan Campus, Fudan University, a typical residential area in a heavily polluted urban area. The chemical composition of

individual ambient particles was measured by single particle aerosol mass spectrometry (SPAMS, Hexin Co., Ltd). Detailed information on SPAMS is available elsewhere (Li et al., 2011). An adaptive resonance theory-based clustering method (ART-2a) was used to classify the mass spectra generated and identify dust/heavy-metal-containing particles (Sullivan et al., 2007). The Hybrid Single-Particle Lagrangian Integrated Trajectory HYSPLIT-4 model developed by the ARL (Air Resources Laboratory) of the NOAA (National Oceanic and Atmospheric Administration), USA, was employed to compute hourly resolved 48 h air mass backward trajectories at 500 m arrival height (Lv et al., 2021; Pongkiatkul and Kim Oanh, 2007).

2.5 Air quality model configuration and application

The source-oriented Community Multiscale Air Quality (CMAQ) model v5.0.1 with an expanded Stratospheric and Air Pollution Research-99 (SAPRC-99) photochemical mechanism was applied to simulate $PM_{2.5}$ levels and track the sources of primary $PM_{2.5}$ ($PPM_{2.5}$) in China during the entire year of 2013 (Guenther et al., 2012; Ying et al., 2018). The simulation domain covered China and its surrounding countries, with a horizontal resolution of 36 × 36 km^2 (127 × 197 grids). Anthropogenic emissions were based on the Multi-resolution Emission Inventory for China (MEIC, v1.3, 0.25° × 0.25°, http://www.meicmodel.org). Biogenic emissions were generated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1 (Guenther et al., 2012). The meteorological inputs for the CMAQ model were calculated by the Weather Research and Forecasting (WRF) model (https://www2.mmm.ucar.edu/wrf/users).

Five major source contributions (windblown dust, residential, transportation, power generation and industrial sources) to $PM_{2.5}$ were investigated based on the inventory-observation-constrained emission factors (Ying et al., 2018). Three control trials were conducted for each heavy metal according to measured soil, dust-$PM_{2.5}$ and the SPECIATE datasets from the four regions (three dust sources and Shanghai). It is worth noting that the emission factors for areas outside these four regions were estimated using Inverse Distance Weight (IDW) spatial interpolation methods. These methods were based on the dataset of emission factors within these four regions, which represent the amount of heavy metal emitted per kilogram of dust (Zhang and Tripathi, 2018). Each heavy metal source concentration from dust aerosol and all four sources were used to quantify the contribution on heavy metal concentrations in the atmospheric dust aerosols, which can be represented in Equation 1:

$$R = \frac{E_1 \times s_1 \times a}{\sum_{i=1}^{5} E_i \times s_i} \qquad \text{Equation 1}$$

Where E_i is the $PPM_{2.5}$ emission from i^{th} source, s_i is the emission factor of the specific heavy metal from i^{th} source, a is a is the concentration of heavy meantal in measured soil, dust-$PM_{2.5}$, and the SPECIATE datasets. E_1, s_1, and a are the values for dust.

In addition, the human health risk of heavy metals was assessed. Three main routes of chemical daily intake (CDI, mg kg^{-1} day^{-1}) of air heavy metals were: (1) direct ingestion of particles or gases existed in the air (CDIing); (2) inhalation of suspended particles through mouth and nose (CDIinh); and (3) daily absorption of heavy metals through skin (CDIdermal) (Luo et al., 2012). To assess the carcinogenic and non-carcinogenic effects of heavy metals, we evaluated these effects in 13 age groups ranging from birth to ≤80 years old. These age groups are as follows: <1, 1 to <2, 2 to <3, 3 to <6, 6 to <11, 11 to <16, 16 to <20, 21 to <31, 31 to <51, 51 to <61, 61 to <71, 71 to <81, and ≥81

years (Gholizadeh et al., 2019b). The variables and values used for estimating human exposure to heavy metals were obtained from the U.S. Environmental Protection Agency (USEPA) and the U.S. Department of Energy (USDoE) (Moya et al., 2011; Doe, 2011). CDIing, CDIinh, and CDIdermal were calculated as:

$$CDI_{ing} = C \times \frac{IRing \times EF \times ED}{BW \times AT} \times 10^{-6} \quad \text{Equation 2}$$

$$CDI_{dermal} = C \times \frac{SA \times AF \times ABS_d \times EF \times ED}{BW \times AT} \times 10^{-6} \quad \text{Equation 3}$$

$$CDI_{inh} = C \times \frac{IRinh \times ET \times EF \times ED}{BW \times AT} \times 10^{-6} \quad \text{Equation 4}$$

Moreover, the total carcinogenic risk (TCR) for each heavy metal were calculated by:

$$carcinogenic\ risk = CDI_{ing,dermal,inh} \times CSF \quad \text{Equation 5}$$

$$TCR = \sum risk = CDI_{ing} \times CSF_{ing} + CDI_{inh} \times IUR + CDI_{dermal} \times CSF_{ing}/ABS_{GI} \quad \text{Equation 6}$$

Here the IRing was Ingestion rate (mg day^{-1}), EF was exposure frequency (day year^{-1}), ED was exposure duration (year), BW was body weight (kg), AT was Averaging time (day), SA was total body skin surface area (m^2), AF was skin adherence factor (mg cm^{-2}), ET was exposure time (hour day^{-1}), ABSd was dermal absorption factor, IRinh inhalation rate (m^3 day^{-1}), ABS_{GI} was gastrointestinal absorption factor, CSF was cancer slope factor. The values of these parameters could be found in the previous study (Gholizadeh et al., 2019a).

3 Results and discussion

3.1 Enrichment of heavy metals in fine dust aerosols

Fig. S3-S4 show the absolute concentrations of heavy metals in dust aerosols and their parent soils. The concentrations of heavy metals in dust-PM_{10} were similar to soil concentrations, which showed a significant correlation between soils and PM_{10} ($p<0.01$) (Fig. S5). While the concentrations of heavy metals in dust-$PM_{2.5}$ were higher than those in soils, especially Mn, Ni, Cu and Zn, showed significant differences ($p<0.001$) (Fig. S6). This trend was consistent across all soil samples. The enrichment factor (EF) of heavy metals in dust aerosols relative to the parent soils was calculated with Equation 8.

$$EF = \frac{C_1/m_1}{C_0/m_0} \qquad \text{Equation 8}$$

Where C_1 is the heavy metal concentration in dust-PM; m_1 is the mass of dust-PM collected on the filter; m_0 is the mass of soil in the ICP-MS sample, and C_0 is the heavy metal concentration of the soil.

Figures 1 and S7 show that many heavy metals were highly enriched in fine dust aerosols ($PM_{2.5}$), i.e., their absolute concentrations were significantly higher in fine dust particles than in the parent soil (Fig. S6). V, Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Ba, Ti, and Pb were all enriched in dust-$PM_{2.5}$ during the process of dust formation. The following trend of heavy metal enrichment was established for dust-$PM_{2.5}$: Cd > Zn > Ba > Cu > Mn > Pb > Ni > Ti > Co > As > Cr > V. Notably, the EFs of Cd were greater than 5 for soil S1, S10 and S11. No other literature has reported the enrichment of Cd or other heavy metals in dust aerosols. However, there is one study showing the

enrichment of water-soluble ions during dust aerosol production from soil (Wu et al., 2022). It reports that the EFs of Ca^{2+} ranged from approximately 5.6 to 223.1, and the EF values of Mg^{2+} were between approximately 2.1 and 90.3 for dust-$PM_{2.5}$ from Sandy soils in the Taklamakan Desert. In this study, it is found that the EF of Cd and other metals falls within the range of EF for these water-soluble ions, consistent with the value reported by Wu et al., (2022). Fig. 1 also illustrates that all heavy metals were more highly enriched in smaller $PM_{2.5}$ dust particles compared to larger PM_{10} dust particles. For example, the Cd's EF reached ~6.4 and ~1.7 for dust-$PM_{2.5}$ and dust PM_{10}, respectively, from soil S1. Most dust-$PM_{2.5}$ should originate from the small colloids in soil, which are defined as soil particles with less than 2 μm in diameter. These soil colloids usually carry large amounts of negative charges, which can help adsorb many cations in soil, including various heavy metal ions (Brady and Weil, 2008). Thus, heavy metals are enriched in small soil aggregates. During the sandblasting process, the smaller soil grains, with higher heavy metal concentrations, are more likely to be ejected and form dust aerosols. The particle size dependence of heavy metal enrichment could have significant ramifications for the health impacts of dust aerosols. The dust aerosol size distribution of dust (Fig. S8) was also measured by an Aerodynamic Particle Sizer (APS, APS Model 3321; TSI Inc.; USA). It is found that the peak of the particle size distribution of dust aerosol was approximately at 2~3 μm. Similarly, the scanning electron microscope (SEM) images of these dust aerosols (generated by S10) also show the presence of a large number of particles with sizes of 2~3 μm. As particle size decreased, the shape of particles changed from flakes to rods, which means a larger surface area (Fig. S9). When examining the impact of soil texture on dust aerosol enrichment, first, notable variations were observed in the EF values from one soil texture, such as sandy soils, specifically S2, S4, S7, S10, S11, and S12. To assess the significance of these

variations, a one-way Analysis of Variance (ANOVA) was conducted using SPSS. In ANOVA, the *p-value* represents the probability of obtaining the observed differences in means (or more extreme differences) by random chance alone, assuming no true difference between the groups. A *p-value* below a predetermined significance level (commonly 0.05) indicates significant differences between the means of the compared groups. Specifically, for sandy soil, analysis results reveal significant variations between these six soils in terms of the EF values for both dust-$PM_{2.5}$ (*p-value*=0.004<0.05) and dust-PM_{10} (*p-value*=0<0.05) (Table S5 and S6). These results indicate that there are significant differences in the EFs of heavy metals within the sandy soil group. Then, the variation between soil types was analyzed. For the six different types of soil samples, the results of ANOVA showed significant differences in the EFs of dust-$PM_{2.5}$ (*p-value*=0<0.05) and dust-PM_{10} (*p-value* =0<0.05) among these soil types (Table S7 and S8). The differences among the six soils from different soil types were greater than those observed among the different soils in the same soil type, indicating a potential role of soil type in affecting EFs, which would require further study to elucidate. Detailed information was found in SI of Texture S3 and Table S5-S10.

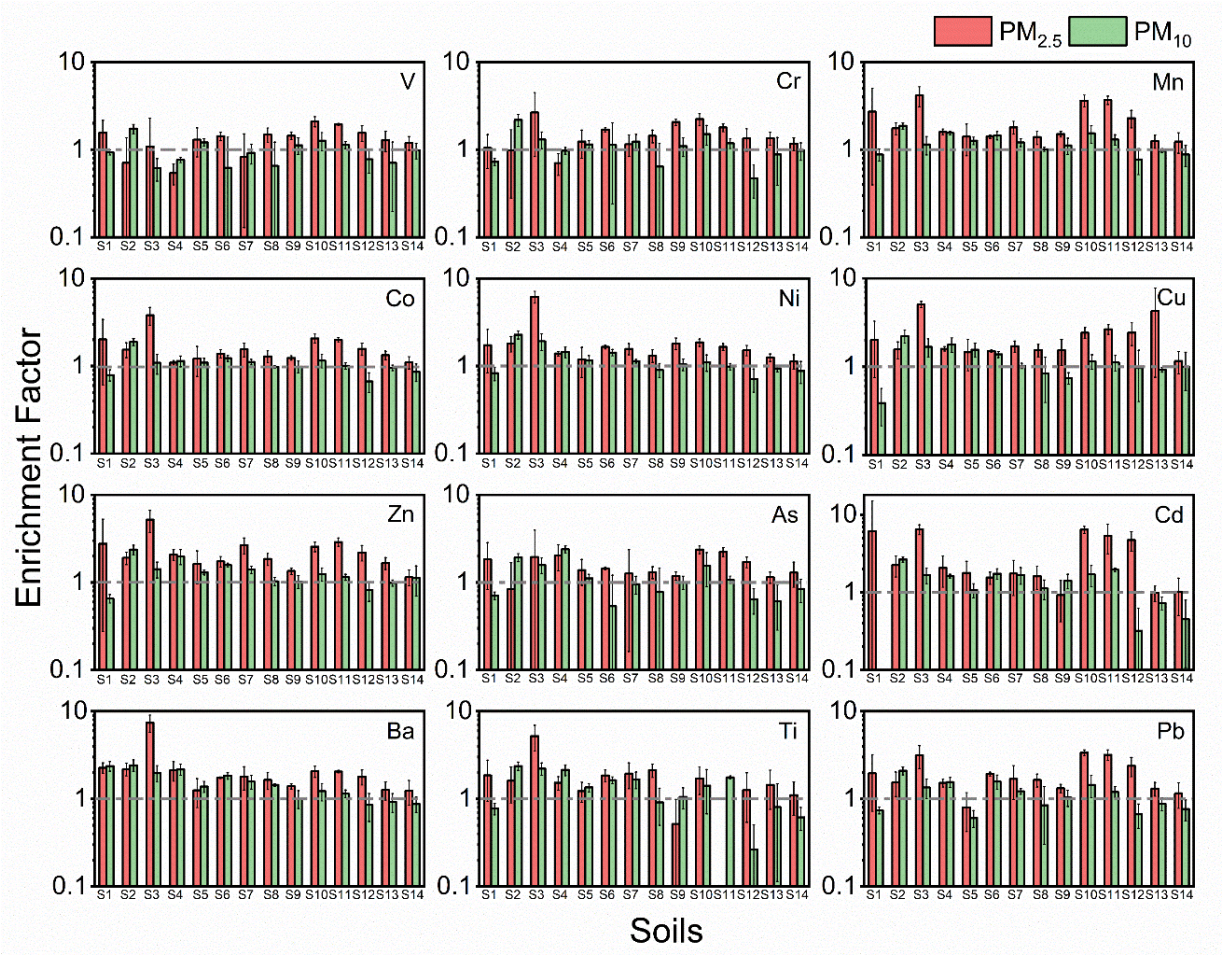


Figure 1. Enrichment Factors of $PM_{2.5}$ and PM_{10}. Enrichment factors of heavy metals in dust aerosols from soil S1-S14; red represents $PM_{2.5}$ and green represents PM_{10}. The grey dotted line represents the EF as 1. The whiskers on the bars represent the standard deviations of triplicates.

To investigate the link between dust particle size and heavy metal EFs in greater detail, a MOUDI impactor was used to collect dust-PM from 0.56 to 10 μm (absolute concentration obtained in Fig. S10). Consistent with the results discussed above, the EFs for some heavy metals, such as Pb, significantly increased with decreasing particle diameter ($r = -1$, $p < 0.01$) (Fig. 2). For the smallest dust particles (0.56~1.0 μm), the EFs for Pb was approximately 83, an order of magnitude greater than the EFs (~3) for the largest dust particles (>10 μm). V, Cr, Co, Mn, Ni, Cu, Zn, As, and Ba show consistent trends, with EFs increasing as the particle size decreases. In detail, V (ranging from

~1.1 to ~18.9), Cr (ranging from ~1.5 to ~23.7), Co (ranging from ~1.7 to ~93.7), Mn (ranging from ~2.3 to ~7.4), Ni (ranging from ~1.6 to ~29.7), Cu (ranging from ~3.3 to ~54.3), Zn (ranging from ~2.3 to ~19.0), As (ranging from ~1.8 to ~112.3), and Ba (ranging from ~1.4 to ~7.0), as the particle size decreases from 10 μm to 0.56 μm. This results demonstrate that some heavy metals are indeed enriched in smaller soil particles, which could be aerosolized during the sandblasting process. The particle size dependence of heavy metal enrichment could have significant ramifications for the health impacts of dust aerosols. In contrast, Cd's EFs remain relatively unchanged with varying particle sizes. On the other hand, Ti exhibits an opposite trend, with EF values decreasing as the particle size decreasing, and the reason for this difference requires further study.

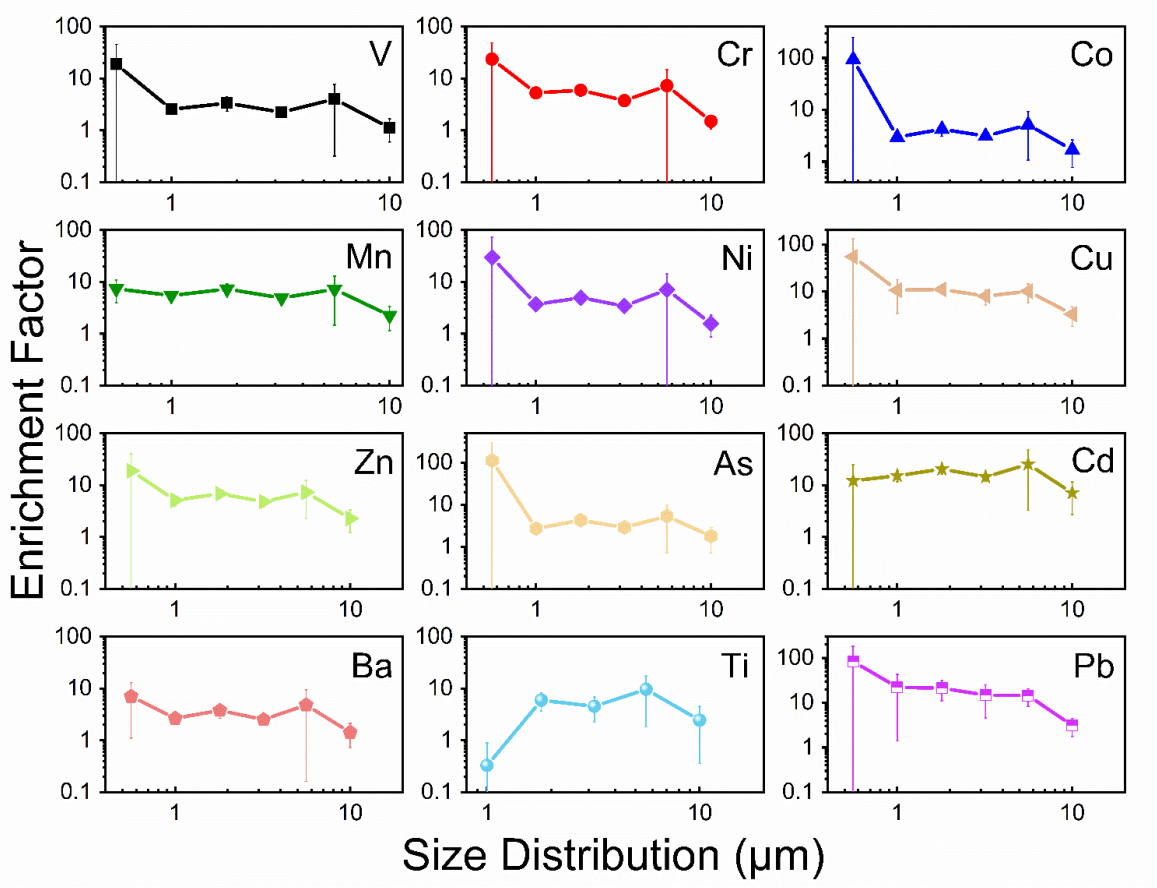


Figure 2. Enrichment factors of heavy metals in dust aerosols with different particle size ranges. The EF data were produced from the Soil S10, with diameters at above10 μm, 5.6-10 μm, 3.2-5.6 μm, 1.8-3.2 μm, 1.0-1.8 μm and 0.56-1.0 μm. The whiskers on the bars represent the standard

deviations of triplicates.

3.2 Modeling of the contributions of dust aerosols to atmospheric heavy metals using the dust profiles from this study and the SPECIATE datasets

It is necessary to know the sources of atmospheric heavy metals to effectively control their emission. Air quality models with emission inventories can estimate the contributions of various sources to atmospheric heavy metals. However, when estimating heavy metal emissions from dust production, some widely used air quality models, such as the CMAQ model, typically use dust profiles from the US EPA's SPECIATE datasets. As discussed in the introduction, this dust profile may be outdated and cannot reflect realistic dust compositions. We used the CMAQ model to assess the potential impact of dust aerosol profile in atmospheric dust aerosol using our measured profile and the profile (No. 41350) from the SPECIATE datasets. The model tracked heavy metals in $PM_{2.5}$ in China for the year 2013 (see Methods) from five major sources: windblown dust, residential, transportation, power generation, and industry.

Figure 3 shows the modeled contributions of the dust source to the Cr and Pb concentrations in $PM_{2.5}$ for China, using the measured soil, dust-$PM_{2.5}$ profiles from this study, as well as the SPECIATE composition profiles (see Methods). In addition, the modeled results for other metals, such as As, Cu, Mn, Ti, and Zn were presented in Fig. S11-15.

For atmospheric Cr, it is clear that the scenario of applying SPECIATE database significantly underestimates the contribution of dust aerosol, with the highest value of ~0.08 $\mu g/m^3$, when

compared to the scenario of applying the measured dust-$PM_{2.5}$ profiles, which had the highest value of ~0.14 $\mu g/m^3$. For Pb, as shown in the right column of Fig. 3, the scenario of applying SPECIATE profile overestimates the contribution of dust aerosol, with the value up to ~0.4 $\mu g/m^3$, when compared to the scenario of applying the measured dust-$PM_{2.5}$ profiles, which had the highest value of ~0.14. Uncertainties associated with the use of SPECIATE have also been identified in previous studies (Ho et al., 2003; Xia et al., 2017). Specifically, the dust $PM_{2.5}$ source profiles obtained from local studies indicated that SPECIATE overestimated the contributions of atmospheric K and Al by approximately 23%, while underestimating the contributions of Ca and Na by 50%. Additionally, the model represents the annual average data for the year 2013. Although there are some field studies conducted in the same year (Wang et al., 2021; Shi et al., 2018), there is no available annual average data for a direct comparison with the model results. These results demonstrate that the modeled heavy metal distribution in the atmosphere is quite sensitive to the input of dust composition profile, strongly suggesting that using a proper dust composition profile is a key in such air quality modeling.

As discussed in the Introduction, many atmospheric studies assume that dust aerosol composition is similar to the composition of its parent soil. Here we also use the soil composition as an input dust profile in the model calculation to see how the modeled results are compared to that using the dust-$PM_{2.5}$ profile. For Cr, an obvious elevation of contribution was found by comparing the map using soil (a) and dust-$PM_{2.5}$ (b) profiles, with the hotspots of contribution (~0.14 $\mu g/m^3$) distributed in northwest China. The region with dust aerosol contribution ranged from 0.02 to 0.08 $\mu g/m^3$ covers most areas in China by using the dust-$PM_{2.5}$ profile. In contrast, the application of

390 the soil profile to the model reveals a significantly reduced area where the modeled Cr concentration
391 from dust aerosols falls within the range of 0.02 to 0.08 $\mu g/m^3$. For Pb, a significant difference is
392 also found. The high contribution areas are also mainly distributed in northwest China for scenarios
393 of applying soil and dust profiles, with the value up to 0.1 $\mu g/m^3$. While the area with low dust
394 aerosol contribution (<0.02 $\mu g/m^3$) shrinks considerably in the scenario of applying soil profile.
395
396 The applied dust enrichment factors to modeled Cr in $PM_{2.5}$ had an even stronger impact on modeled
397 source apportionment (Fig. 3a-3b). The average dust source contribution to the total $PM_{2.5}$ Cr
398 concentration over China was calculated to be 0.03, and 0.05 $\mu g/m^3$in the scenarios of applying
399 soil and dust profiles, respectively. The model results for As, Cu, Mn, Ti and Zn (Fig. S11-S15) also
400 show similar trends, indicating applying realistic enrichment factors to heavy metal concentrations
401 in fine dust aerosols is critical to accurately model the sources of atmospheric heavy metals. These
402 results demonstrate that it is not appropriate to assume dust aerosol composition is equal to soil
403 composition, at least in air quality modeling.

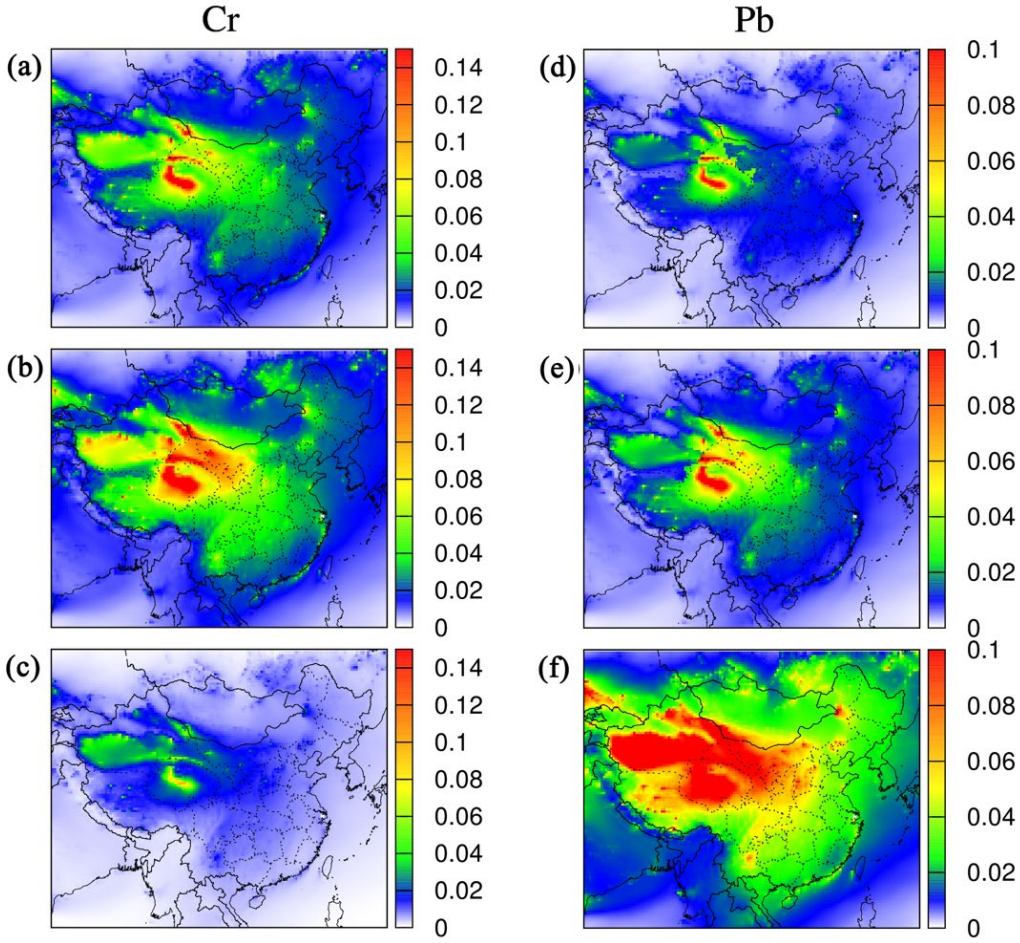


Figure 3. Modeling of the contributions of dust aerosols to atmospheric Cr and Pb concentrations. These results use the dust profiles of measured soil (a, d), dust-$PM_{2.5}$ (b, e), and the SPECIATE datasets (c, f). The unit is $\mu g/m^3$.

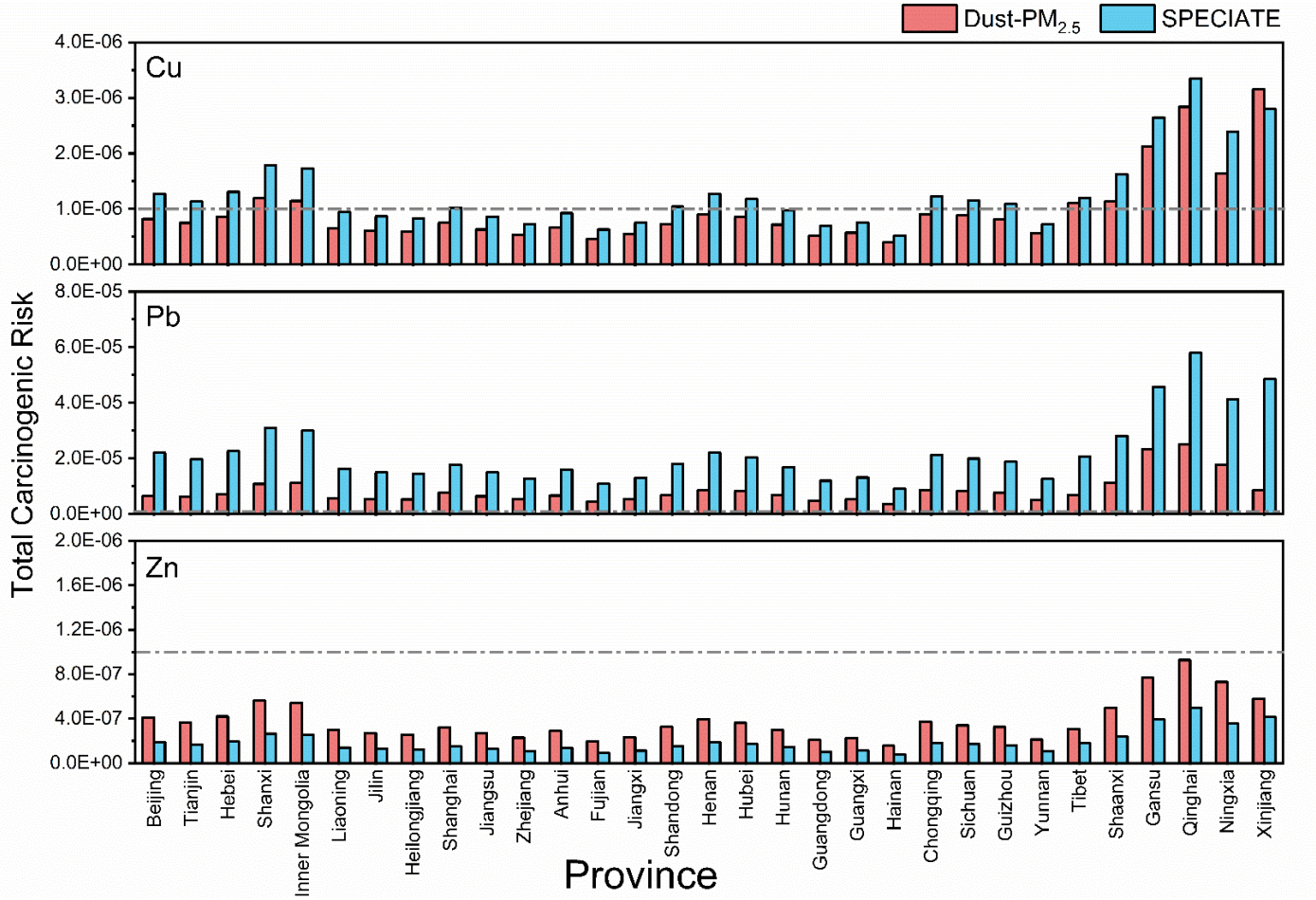
Figure 4 shows the Total Carcinogenic Risk (TCR) of the modeled atmospheric heavy metals (Cu, Pb and Zn) for each province in Mainland, China. The modeled results using the dust-$PM_{2.5}$ and the SPECIATE profiles are compared here. The carcinogenic risks lower than 10^{-6} are considered negligible, and risks above 10^{-4} are not accepted by most international regulatory agencies (Cheng et al., 2015; Epa, 1989; Luo et al., 2012). For Cu, it is evident that using the SPECIATE profile overestimated (the difference range up to ~ 7.5×10^{-7}) the TCR in China compared to using the dust-$PM_{2.5}$ profile, as some regions exceed 10^{-6}, the threshold value. For Pb, although all regions were above 10^{-6}, the TCR using the SPECIATE profile was greatly overestimated (the difference range is

416 ~ 5.5×10^{-6} - 4.0×10^{-5}). The model results for Zn showed that all regions were not above 10^{-6} but

417 significantly underestimated risks using the SPECIATE profile. This indicates that the health risk

418 assessment is also sensitive to dust composition profiles. Using the SPECIATE profile might be

419 problematic for assessing these risks.



420

421 **Figure 4.** Comparison of the total carcinogenic risk (TCR) of the modeled atmospheric heavy metals

422 for each province in Mainland, China between using the dust-$PM_{2.5}$ and SPECIATE profiles. Here,

423 the TCR of Cu, Pb and Zn were calculated. The grey dotted line is 10^{-6}, the threshold value for

424 health concerns.

425

3.3 Field observation before, during and after a dust storm

Our modeling results suggest that dust aerosol could be a major source of multiple heavy metals in $PM_{2.5}$ in China. Therefore, dust storms should significantly increase the concentrations of heavy metals in $PM_{2.5}$. To test this idea, we studied a dust-storm plume, which originated from Mongolia and arrived in Shanghai (Huang et al., 2010) on 23 May 2018 (Fig. S16). Real-time single-particle mass spectra were generated by a single-particle mass spectrometer. Single particle mass spectrometry can offer detailed information on the chemically-resolved mixing state at the single-particle level. According to the similarities of the mass-to-charge ratio and peak intensity of characterized signals, *"Dust"* particles were classified via an adaptive resonance theory-based clustering method (ART-2a, see Method). The number fraction of *Dust* particles was ~4.94% before and after the dust storm and it increased to~9.73% during the dust storm episode (Fig. 5a).

Dust particle mass spectra also contained ion markers indicative of an array of heavy metals (*m/z* 55[Mn^+], 51[V^+], 207[Pb^+], 63[Cu^+], 75[As^+], 91[AsO^+], 52[Cr^+], -84[CrO_2^-], -100[CrO_3^-]) (red sticks in Fig. S17), indicating the existence of heavy metals in the ambient dust aerosols. The time series of Pb-containing and Cr-containing particle number fractions showed similar trends to the *Dust* particles. When the dust storm arrived, both Pb-containing and Cr-containing particle fractions increased as the dust cluster fraction increased. Before and after the dust storm, the percentages of Pb-containing and Cr-containing particles that overlapped with the *Dust* cluster were 41% and 32%, respectively. However, this overlapped ratio increased to 86% and 71% during the dust storm episode. The increase of heavy metal particles in step with the dust particles indicated that the dust particles could be the dominant source of these heavy metal species during this dust storm episode.

We further analyzed the size-resolved number fraction of dust aerosol, Pb-containing, and Cr-containing particles during the dust storm episode (Fig. 5b). The number fraction of *Dust* particles increased with increasing aerodynamic diameter. For particles above 1.0 μm, *Dust* accounted for >12% of the total particles during the storm. However, the Pb-containing and Cr-containing particles made up a larger number fraction of analyzed particles with decreasing particle diameter size (< 1 μm). The number fractions of Pb-containing and Cr-containing particles were 5.7% and 7.9% of all mass spectra for particles from 0.2-0.3 μm. This result was consistent with our laboratory results that there is high heavy metal enrichment in smaller dust particles and our modeling results that dust aerosol is likely a major source of atmospheric Pb and Cr over China.

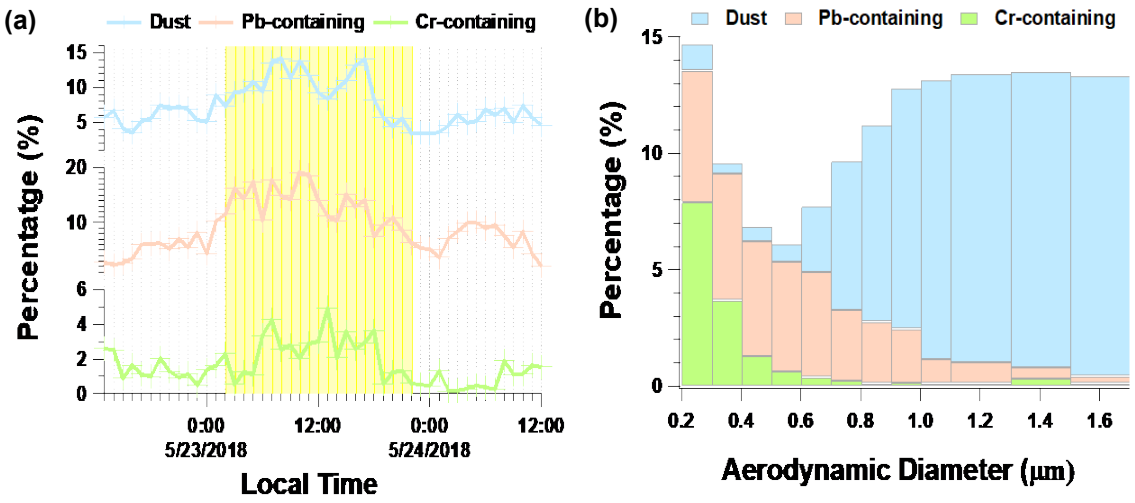


Figure 5. Ambient dust aerosol measurements. (a) Temporal variation of the percentages of dust aerosol, Pb-containing, and Cr-containing particle clusters. The yellow shadow represents the dust storm episode. (b) Size-resolved number fraction of dust aerosol, Pb-containing, and Cr-containing particle clusters.

4 Environmental implications

In this study, many heavy metals were found to be highly enriched in fine ($PM_{2.5}$) dust aerosols compared to their concentrations in the parent soils. We propose that heavy metals tend to be enriched in smaller soil aggregates (Ikegami et al., 2014). During the sandblasting process, the heavy metal enriched smaller soil aggregates are more likely to be ejected and form dust aerosols. This work finds that dust aerosols from different soils may have a range of heavy metal enrichment factors. To study the transfer of heavy metals from soils to the air, it is critical to have a complete set of enrichment factors for each major soil type. There exists a difference among the heavy metal enrichment factors from different soil samples. The variability in the EFs is likely due to differences in soil properties (soil texture and size distribution etc.) which may affect the sandblasting/saltation process. For example, the enrichment factors of heaviest metals for Soil S1, S10 and S11 were higher than other soils. The detailed reason is still unknown and needs further exploration. Moreover, air quality models, including CMAQ models and various CMB models, often use the dust chemical profiles from the US EPA's SPECIATE to calculate the contribution of fine dust aerosols to atmospheric heavy metals, which are outdated and could lead to significant errors in estimating the emission of heavy metals through dust generation. Without using proper dust profiles in estimating heavy metal emissions from dust generation, the contribution of fine dust aerosols to atmospheric heavy metals, and its associated health risks are likely significantly mistaken.

5. Conclusions

Dust generation and aerosolization are complex processes that may have certain chemical selectivity.

Here, we deployed a laboratory generator to produce dust aerosol with a realistic sandblasting process. The concentrations of heavy metals (including V, Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Ba, Ti, and Pb) in soils and fine ($PM_{2.5}$) and coarse (PM_{10}) dust aerosols were measured. With research efforts to elucidate the enrichment process of heavy metal in dust aerosols comparing to their parent soils, our results fill the knowledge gaps of the compositional variation of heavy metal between the parent soils and the generated dust aerosols. Mn, Cd, Pb and other heavy metals were found to be highly enriched in fine ($PM_{2.5}$) dust aerosols, which can be up to ~6.5-fold. These findings were also consistent with our field observation results. In addition, air quality models often use an outdated heavy metal profile for dust aerosols from the US EPA's SPECIATE database, which seems to be lack of enrichment between each particle size. We modeled the impact of the contribution of heavy metals in dust aerosol and their health risks in CMAQ, a widely used air quality model, and determined that atmospheric heavy metal concentrations over China, which drastically changed when we applied different dust profiles, such as the measured soil, dust-$PM_{2.5}$ profiles from this study, as well as the SPECIATE composition profiles. Our air quality modeling for China demonstrates that the calculated contribution of fine dust aerosols to atmospheric heavy metals, as well as their cancer risks, could have significant errors without using proper dust profiles.

Supplement

The supplement related to this article is available online at: http://dx.doi.org/ 0.17632/byg6xk2fg9.1.

Data availability

All data supporting this study and its findings will be available in an online data repository at: http://dx.doi.org/10.17632/wpphf8rd33.1.

Author contributions

X.W. and J.C. conceptualized the work and designed the experiments. H.Z. and S.Z. led the air quality modeling work. Q.G. lead the experimental work of heavy metal enrichment measurements. J.Z. led the field observation. K.Z., Q.W., S.C., S.W., J.H., X.L. and H.C. helped in experimental works. L.Z., L.W., Z.W., X.Y. and H.Z. helped in the experimental design and data analysis. Q.Y. provided the data required for the air quality modeling. All authors contributed to the paper's writing.

Competing interests

The authors declare no competing interests.

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519 # Acknowledgements

520 The authors also thank Xingxing Wang and Xiangcheng Zeng for their help in heavy metal measurement.

521 # Financial support

522 This work was partially supported by the National Natural Science Foundation of China (Nos.
523 92044301, 42077193, 21906024). Comments from Dr. Camille Sultana greatly improved this
524 manuscript.

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