Responses to interactive comments

Journal: Atmospheric Chemistry and Physics

Manuscript ID: acp-2022-802

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

We appreciate Referee #1's comments and suggestions to help improve the manuscript. Every comment is addressed, and the detailed responses and related changes are shown below. Our response is in blue and the modifications in the manuscript are in red.

The manuscript entitled "High Enrichment of Heavy Metals in Fine Particulate Matter through Dust Aerosol Generation" examines the relationship between mineral dust aerosols and their parent soils in terms of heavy metal contents, demonstrating enrichment of heavy metals in the fine aerosol particles. The study combines laboratory and numerical modeling experiments and highlights that the heavy metal content may be higher than usually estimated by most models dealing with dust aerosols. Such findings are of great importance for air quality assessment and health implications as it evaluates the sensitivity of air quality models to the size-resolved heavy metal composition established through experimentation. The results are novel and significant and justify the publication of this paper. Nonetheless, I did notice some points that need to be clarified before the manuscript can move forward. Therefore, minor revision has to be done before this manuscript could be accepted for publication in the ACP.

Response: We thank Referee 1 for Referee #1's valuable comments and suggestions. Below are the responses to each specific comment.

Major comments

1. It seems that soil types play a quite important role in affecting the heavy metal

enrichment in dust aerosols. However, it is not clear how soil type was determined. Thus, the authors need to provide detailed information on how the soil type was determined. Please show the details in the Materials and Methods section.

Response:

We have included additional details on how the soil texture was determined in both the revised Materials and Methods section and the Supplementary Information (SI) section.

Changes in manuscript:

"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."

Texture S1. Soil texture characterization

To measure the particle size distribution of the soil, approximately 0.03 to 0.5 g of air-dried soil samples were first passed through a 2 mm sieve. Subsequently, 10 mL of distilled water was added to the soil, and a dispersant was used to adjust the pH based on the soil's alkalinity or acidity. The dispersant consisted of either 1 to 1.5 mL of 0.5 mol/L hexametaphosphate (HMP) or 0.5 mol/L sodium hydroxide (NaOH). The mixture was then left to soak overnight before undergoing ultrasonic vibration for 2 minutes. Finally, the Laser Scattering Particle Size Distribution Analyzer (LA-960) was utilized to measure the soil samples labeled as S1-S14.

2. This paper used GAMEL system for laboratory simulation, which is a small scale dust generator. Please compared and explain the similarity and difference of the GAMEL system and the wind tunnel system.

Response:

The GAMEL system and wind tunnel system share the similarity of being able to realistically simulate dust aerosol generation. However, they differ in several aspects:

Firstly, the wind tunnel typically requires tens of kilograms of soil, which makes it challenging to collect sufficient samples from field sites for conducting tests under various experimental conditions. In contrast, the GAMEL system only requires 10 grams of soil sample.

Secondly, the wind tunnel faces difficulties in preventing contamination from room air due to its large air flow rate. Filtration of such a high air flow would incur significant costs. On the other hand, the GAMEL system operates with an air flow rate of system is 8 LPM (liters per minute), allowing it to be equipped with a small HEPA filter that effectively removes ambient aerosols.

Based on the above considerations, we referenced this literature (Lafon et al., 2014) and used this GAMEL system to simulate the generation of dust aerosols.

Changes in manuscript:

"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)."

3. How is total carcinogenic risk (TCR) calculated is not very clear. For example, what are the 13 age groups. Please provide more details of the calculation.

Response:

We have included additional details on the calculation of TCR (Target Cancer Risk) and referenced the following literature for this purpose (Gholizadeh et al., 2019; Luo et al., 2012; Moya et al., 2011; Doe, 2011).

The 13 age groups are divided as follows (in years): <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. 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) .

Changes in Manuscript:

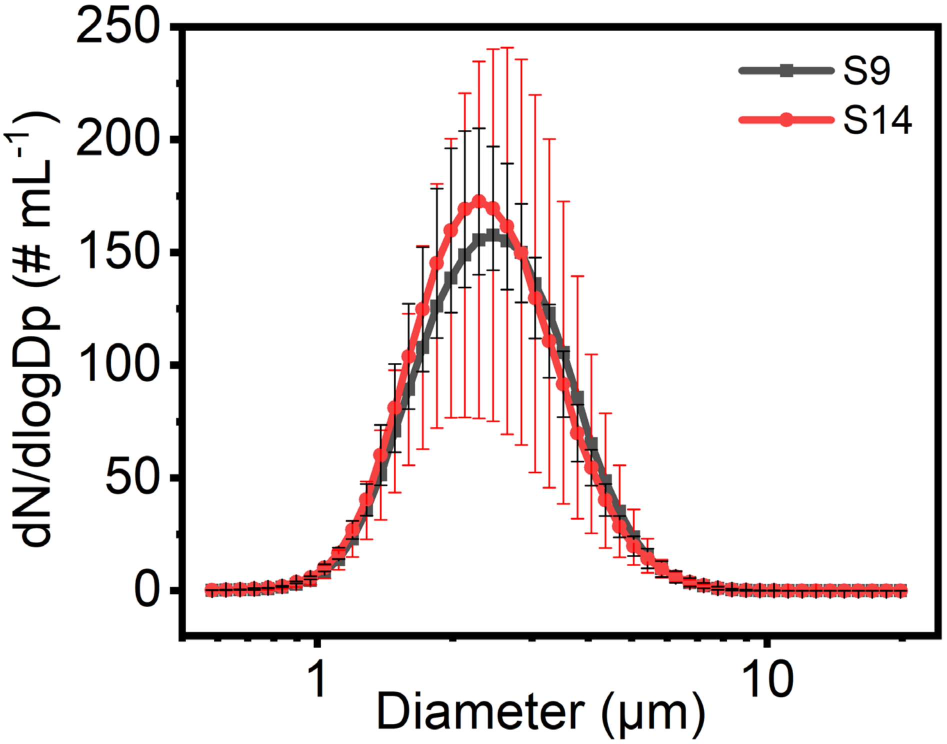
"To assess the carcinogenic and non-carcinogenic effects of heavy metals, we evaluated these effects in 13 age groups ranging from birth to ≤80 years old. These age groups are as follows: <1, 1 to <2, 2 to <3, 3 to <6, 6 to <11, 11 to <16, 16 to <20, 21 to <31, 31 to <51, 51 to <61, 61 to <71, 71 to <81, and ≥81 years (Gholizadeh et al., 2019). 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)."

Minor comments

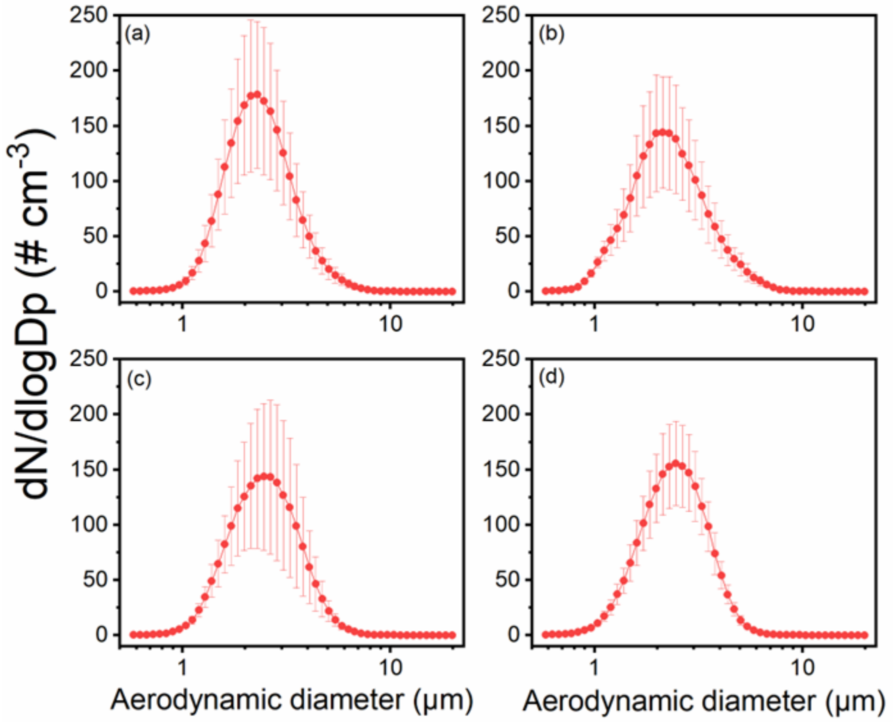
1. Line 137-139: In the GAMEL experiment, the author used an 8 L/min dust aerosol stream to sweep 10 g soil, can it generate dust aerosols stably? Is the particle size distribution related to the wind speed? Please explain.

Response:

The GAMEL generation system has the capability to stably generate dust aerosols. In our study, the experimental duration was set to 1 minute, and the particle size distribution was determined using the Aerodynamic Particle Sizer (APS). In this experiment, the graph below illustrates the observed particle size distribution, with a peak occurring at 2-3 μm. Although this experiment only investigated the particle size distribution generated by S9 and S14, one of our previous studies conducted with the same experimental setup exhibited a similar shape in particle size distributions (Gao et al., 2023). Hence, the GAMEL system can stably generate dust aerosols.



This study: Supplementary Figure S8. Particle size distribution of dust aerosols produced from soil S9 and S14. The size distribution was detected by an Aerodynamic Particle Sizer (APS), which size range is 0.5-20 μm.



Previous study: Enrichment and Transfer of Polycyclic Aromatic Hydrocarbons (PAHs) through Dust Aerosol Generation from Soil to the Air (Gao et al., 2023): The particle size distribution (PSD) of dust particles. All the soil were collected from the top 20 cm of the soil profile in different regions of Hunan province, south China. (a) is Besides the Pedestrian Road, (b) is Under the Street Pipe, (c) is Inside Baling Petrochemical Plant campus 1, and (d) is Inside Baling Petrochemical Plant campus 2.

In terms of the relationship between particle size distribution and wind speed, a study by (Kok, 2011) suggests that the size distribution of dust aerosols released naturally remains unchanged, regardless of any changes in wind speed.

2. Line 153: Please remove the space between "69" and "%".

Response:

Revised accordingly.

Changes in Manuscript:

"…69% HNO_3…".

3. Line 160 and Line 169: Please remove the space between “2” and “%”.

Response:

Revised as suggested.

Changes in Manuscript:

“…2% HNO_3**…**” and “…2% dilute nitric acid…”.

4. Line 163: Besides heavy metal contents, were the mineral elements in dust aerosols detected?

Response:

This study primarily focuses on the enrichment factor of heavy metals, and we did not detect any mineral elements. This limitation arises from the detection range of our chosen method in ICPMS. Routine mineral elements, such as Na, K, S, Mg and Al, fall beyond the detection range of ICPMS.

5. Line 180： What are the types of dust weather? What type of dust weather was observed in 2018?

Response:

The "Dust Storm Weather Grade" is categorized into five levels based on the ground visibility during dust storm weather. These levels include floating dust, sand dust, sandstorm, strong sandstorm, and super strong sandstorm. On May 23rd, 2018 (LT), the observed type of dust weather was floating dust.

Changes in Manuscript:

“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.”

6. Line 193: “… CMAQ model v5.0.1 with an expanded SAPRC-99” Please show the full name for readers who are not familiar with CMAQ and SAPRC-99.

Response:

Corrected.

Changes in Manuscript:

"…Community Multiscale Air Quality (CMAQ) model v5.0.1 with an expanded Stratospheric and Air Pollution Research-99 (SAPRC-99) photochemical mechanism…".

7. Line 209: Please edit as "Inverse Distance Weight (IDW)".

Response:

Revised as suggested.

Changes in Manuscript:

"…Inverse Distance Weight (IDW) …".

8. Line 215: "E1, s1, and a are the values for dust." Please check the font format.

Response:

Corrected.

Changes in Manuscript:

"E_1, s_1, and a are the values for dust."

9. Line 217-222 and Line 233-238: Please check line spacing as it should be consistent with context.

Response:

Changed as suggested.

We thank Referee 1 again for the comments and suggestions!

Reference

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