Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., author comment AC1
https://doi.org/10.5194/acp-2021-290-AC1, 2021

Reply on RC2

Hui Zhang et al.

Author comment on "Speciated atmospheric mercury at the Waliguan Global Atmosphere Watch station in the northeastern Tibetan Plateau: implication of dust-related sources for particulate bound mercury" by Hui Zhang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-290-AC1, 2021

Response to Referee #1

RC- Reviewer's Comments; **AC** – Authors' Response Comments

RC1: This paper reports a full-year continuous measurement of speciated atmospheric mercury at Waliguan Baseline Observatory, which is a valuable addition to our understanding of atmospheric mercury cycle especially as this site is an important background site for many contaminants. The data quality reported here is quite high and the related analysis is also appropriate and deep. The paper is generally well written with high-quality figures, and the discussions about PBM and dust levels are also interesting and novel. I suggest publishing this paper after a minor revision.

AC1: We appreciate Prof. Zhang for reading our manuscript and providing constructive comments. We have studied these comments carefully and made corrections accordingly, which have been marked in blue in the revised manuscript. The response to these comments are presented below.

Specific comments:

RC2: Line 26: Can GEM also be adsorbed by aerosols?

AC2: Yes, GEM can be adsorbed by atmospheric aerosols to form PBM, and the gas-partitioning equilibria is expected to be controlled by Henry's law (Ariya et al., 2015).

RC3: Line 280: Not clear what each point in Figure 6a represents, a group of 0.5x0.5 grid

cells? How do you aggregate them? Which emission inventory do you use?

AC3: We have specified these information in the caption of Figure 6 in the revised manuscript.

Yes, each point in Figure 6a represents a group of CWT values (or anthropogenic emission) in 0.5×0.5 grid cells. As shown in the caption of Figure 6, gridded anthropogenic emissions in all grid cells (0.5°×0.5°) in the study domain were ranked starting from the lowest value to the highest value, and then the gridded anthropogenic emissions were divided into 20 groups with equal number of grid cells, starting from the lowest to the highest. The average anthropogenic emissions as well as the matched GEM and PBM CWT values in these 20 groups were then calculated to conduct the relationship analysis.

Anthropogenic emission data are from AMAP/UNEP, 2018.

RC4: Line 285-301: The observed high PBM concentrations are caused by the high dust load? I suggest reporting the dust concentration levels at Waliguan as well, and a comparison with urban sites. Also, the instrument only measures PBM minus than 2.5 micron, but the dust is probably mainly in the coarse mode. How to reconcile this discrepancy?

AC4: The suggestion of further analysis between PBM concentrations and dust concentrations are appreciated. Unfortunately, we are not able to collect the dust concentrations datasets at WLG. Absorbing Aerosol Index (AAI) is one of the most useful space-borne data sets, which dominantly presents the levels of desert dust and biomass burning related aerosols (biomass burning could be neglected in the Tibetan Plateau, more detail please see Figure S5). Therefore, we deem that the use of AAI is suitable to investigate the effect of dust related sources on PBM in the study area.

For the difference regarding the aerodynamic diameter between PBM and dust aerosols, previous studies observed that the $PM_{2.5}$ concentration in the dust is significantly positively correlated with the concentrations of TSP and PM_{10} concentration (Fadi A. Al-Jallad et al., 2017; Hemraj Bhattarai et al., 2021). Therefore, the total concentrations of dust aerosols, which is highly linked with the $PM_{2.5}$ concentrations within the dust aerosols well, could be useful to investigate the impact of dust sources on PBM at WLG.

RC5: Line 322-325: It's also important to specify the size range of these dust emission estimates.

AC5: good point, the ranges are added in line 345 and 347 in the revised manuscript, which reads: "*total particulate bound mercury (Hg-TSP) emissions from desert dust related sources were roughly estimated to be 606 ± 298 (range from 165 to 1650) Mg* yr^{-1} *globally and 80 ± 40 (range from 33 to 151) Mg* yr^{-1} *in China (Table S3)*."

RC6: Line 325-330: I would also refrain from suggesting an "emission flux" from the suspended dust particles, as the Hg on particles may be from adsorbing from the ambient atmosphere during transport after release.

AC6: we understand reviewer's concerns. Actually, our estimate represents the sum of

PBM emitted from primary desert sources and PBM formation during subsequent atmospheric processes (e.g., adsorption of gaseous Hg). Currently, we are not able to calculate the exact values of these two potential sources. In the revised manuscript, referred these two sources as to "desert dust related sources".

RC7: Line 332-346: Both temperature and particulate matter (PM) load influence the PBM/GOM ratio. Have you checked the relationship between temperature and the PBM/PM/GOM? I guess if you normalize the PBM/GOM ratio by the particulate matter load, you would get a more consistent results with other Tibetan sites.

AC7: we agree with the reviewer the gas-particle portioning of gaseous Hg could be controlled by air temperature and the concentrations of particulate matter. The point that we would like to show here is that the elevated PBM concentrations in winter and early spring at WLG was not likely caused by the enhanced gas-particle portioning of GOM under their low air temperature, and we have revised this part in line 364-366 in the revised manuscript.

Reference used in this response letter

- AMAP/UNEP, 2013. Geospatially Distributed Mercury Emissions Dataset 2010v1.
- Ariya, P.A., Amyot, M., Dastoor, A., Deeds, D., Feinberg, A., Kos, G., Poulain, A., Ryjkov, A., Semeniuk, K., Subir, M., Toyota, K. (2015) Mercury Physicochemical and Biogeochemical Transformation in the Atmosphere and at Atmospheric Interfaces: A Review and Future Directions. Chemical Reviews 115, 3760-3802.
- Fadi A. Al-Jallad, Clarence C. Rodrigues, Hamda A. Al-Thani: Ambient Levels of TSP, PM10, PM2.5 and Particle Number Concentration in Al Samha, UAE, Journal of Environmental Protection, 8, 1002-1017, 2017.
- Hemraj Bhattarai, Lekhendra Tripathee, Shichang Kang, Pengfei Chen, Chhatra Mani Sharma, Kirpa Ram, Junming Guo, Maheswar Rupakheti: Nitrogenous and carbonaceous aerosols in PM2.5 and TSP during pre-monsoon: Characteristics and sources in the highly polluted mountain valley, journal of environmental sciences 115, 10–24, 2021.

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2021-290/acp-2021-290-AC1-supplement.pdf