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Reply on RC1

Xuewu Fu et al.

Author comment on "Isotopic compositions of atmospheric total gaseous mercury in 10 Chinese cities and implications for land surface emissions" by Xuewu Fu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-981-AC1, 2021

Response to Anonymous Referee #1

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

RC1: This manuscript described seasonal variations of Hg isotopes in TGM from 10 Chinese urban sites, and addressed the importance of urban surface emissions to both concentration and isotopic compositions of urban TGM. The provided concentration and isotopic data are precious for enhancing our understanding to the re-emission of Hg legacy in building surfaces or urban soils. The data was archived and presented well. In general, I recommend the publication of this article after the following revisions.

AC1: We greatly appreciate the reviewer for recognizing the merits of this work and for providing the valuable suggestions. We have made revisions following the comments (corrections are marked in blue fonts in the revised manuscript), and the response are shown below.

RC2: L110-112: To my knowledge, the effect of emission and re-emissions of GEM from urban surfaces was frequently neglected because we thought the flux was low in these processes, but not the poor understanding of isotopic signatures of that.

AC2: We agree that the traditional thought of primary anthropogenic Hg emissions is the dominant source of atmospheric Hg in urban areas is one the most important reason to neglect the effect of land surface GEM emissions. We have revised the statement to highlight the anthropogenic emissions in line 111-113 in the revised manuscript, which read: "whereas the effect of emission and re-emission of GEM from urban surfaces was frequently neglected mainly because of the strong primary anthropogenic Hg emissions and poor understanding of emission flux and isotopic signatures of GEM from land surfaces in urban areas".

RC3: L112-115: If we accept anthropogenic emissions as the most important component to urban GEM, which has been proved by many speciation observations in China, the observed d202Hg in TGM should be linked with that in local coal rather than an averaged

value from publications. According to Liu et al. Chemical Geology, 2019, d202Hg in stack emission was similar with feed coal used in CFPPs.

AC3: We read that Liu et al. (2019) estimated the isotopic compositions of total Hg in coal burning flue gas and added this citation in line 87-88 in the revised manuscript. Since isotopic compositions of the total Hg were estimated by this study, we use the fractionation of Hg isotope between GEM and GOM to estimate the GEM isotope signature. In addition to coal fire power plant, many other anthropogenic activities are also important GEM sources in China (Zhang et al., 2015). Therefore, it is needed to use the GEM isotopic signature of all anthropogenic emissions to interpret our observations, which have been estimated by Sun et al. (2016). We agree that isotopic compositions of Hg in feed coal would affect, but currently we do not have this kind of information.

RC4: L213: Did the authors present 204Hg data in this study?

AC4: Yes, some of our samples were measured with δ204Hg signatures, which are presented in the supporting information Table S4. For the rest of the samples, we did not measure δ204Hg signatures because of the limitation of instrumental collector designs.

RC5: L359-367: It's a little arbitrary to evaluate the contribution from urban surface using averaged values from only 3 studies of various surface emissions. The authors should emphasize that.

AC5: Agree. We show that our estimate has large uncertainties as well as their reasons, which is shown in line 371-374 in the revised manuscript, which reads: "We caution that, due to the fact that the isotopic signatures of GEM emitted from many anthropogenic sources and land surfaces in China have not been well constrained, such a preliminary assessment should have large uncertainties".

RC6: L414-416: These study sites are all located in monsoon area in China. How about the effect from monsoon climate? Cities in east part of China, especially in north China plain, are largely controlled by north wind from Siberian in winter. The continental monsoon originates from low human active areas, with low GEM concentration, high d202Hg, and low D199Hg in air parcels. This could also be indicated from Figure 4 in this manuscript. I suggest the influences from monsoon be discussed in this study.

AC6: Yes, the effect of monsoon is discussed in line 421-429 in the revised manuscript, which reads: "Prevailing wind directions during the wintertime and summertime sampling campaigns were similar Jinan, Lanzhou, Zhengzhou, and Shanghai, but were different in other remaining cities (Figure S6). Variations in predominant wind directions would change the relationships between receptor and regional anthropogenic emissions, which could further influence the TGM levels and isotopic compositions in these cities. Given the similarity in wintertime and summertime prevailing wind directions in some cities and consistent summertime lower CO concentrations in most cities, it is postulated that the variations in local anthropogenic emissions and transport of regional anthropogenic emissions were not likely the main cause for the seasonal variations in TGM concentrations and isotopic compositions".

References:

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Zhang, L., Wang, S. X., Wang, L., Wu, Y., Duan, L., Wu, Q. R., Wang, F. Y., Yang, M., Yang, H., Hao, J. M., and Liu, X.: Updated Emission Inventories for Speciated Atmospheric Mercury from Anthropogenic Sources in China, Environmental Science & Technology, 49, 3185-3194, Doi 10.1021/Es504840m, 2015.