

## **Responses to the Reviewers' Comments**

### **Unexpectedly high concentrations of atmospheric mercury species in Lhasa, the largest city on the Tibetan Plateau**

Dear editor and reviewer,

We greatly appreciate the useful comments and suggestions from the editor and reviewers. We think the novelty and importance of this study have been acknowledged by the reviewers. We have revised the manuscript thoroughly based on the reviewers' comments. Detailed point by point responses are provided below. All the revisions have been highlighted in blue in the revised manuscript. We hope the revised manuscript could meet the standard of ACP. Thanks again for your consideration.

#### **Anonymous Referee #2**

This article entitled 'Unexpectedly high concentrations of atmospheric mercury species in Lhasa, the largest city on the Tibetan Plateau' by Lin et al. analyzes the monitoring concentrations and source analysis of atmospheric mercury in important cities on the Tibetan Plateau. According to the local meteorological conditions, the author divided the monitoring period into two periods. Indian Summer Monsoon and Westerly Circulation. The authors analyzed the unusual phases of high atmospheric mercury concentration events and attempted to analyze the causes, and also analyzing the diurnal variation of atmospheric mercury in Lhasa. This article reveals the changes of urban atmospheric mercury in clean background areas, indicating the impact of urban anthropogenic activities and wind speed on regional atmospheric mercury concentrations. The result makes sense, and the output of the study will be useful for future studies on global mercury cycling and modeling. The manuscript is generally well organized and written. After revising the questions listed below, the study is accepted for publication.

#### **Response:**

Thanks for your detailed comments and suggestions. We have reviewed the paper in detail according to the reviewer's comments and made corresponding modifications accordingly. Please see the revised manuscript. All the revisions have been highlighted in blue. Detailed responses to your comments are provided as follows.

#### **Specific comments**

##### **Comment #1**

In line 175, the author only calculated the backward trajectory of GEM in the article, why did the author not consider analyzing the trajectory of GOM and PBM.

##### **Response #1**

Thanks for the comment. In this manuscript, we carried out trajectory analysis for GEM in Lhasa. Considering the complex topography of the Tibetan Plateau and the fact that most of the trajectories would pass through dramatic elevation rise when entering Tibetan Plateau, where the subsidence of GOM/PBM is very complex, we think that backward trajectory simulations of GOM and PBM in

Lhasa may introduce considerable errors and uncertainties.

**Comment #2**

Line 226, the author should show the timing of ISM and WEC in Figure 2.

**Response #2**

Thanks for the suggestion. We have added time markers to Figure 2.

**Comment #3**

Line 240, Table 1, what is the purpose of dividing WEC into WEC1 and WEC2?

**Response #3**

Thanks for the question. To better understand the changes of atmospheric Hg concentrations in different periods, the WEC period was further divided into WEC1 and WEC2 periods. We have added some explanations in Section 2.1. **‘To better understand the changes of atmospheric Hg concentrations in different periods, the WEC period was further divided into WEC1 (October 1 to December 30, 2016) and WEC2 (January 1 to February 2, 2017) periods.’**

**Comment #4**

Line 229-231, is it possible that the change in the relationship between the concentration of GOM and PBM is related to the change in the gas-solid distribution ratio of atmospheric mercury?

**Response #4**

Thanks for the comment. We agree with the reviewers that temperature, humidity, and particulate matter composition may affect the partitioning of Hg(II) between GOM and PBM. During the period transiting from ISM to WEC, the climate and atmospheric composition of Lhasa changed significantly, which is likely to affect the partition ratio of GOM and PBM. We have added some relevant discussions in the paper. **‘GOM and PBM may undergo mutual transformation in the atmosphere, which may be related to temperature, humidity, and atmospheric composition (Rutter and Schauer, 2007; Rutter et al., 2008), and thus the concentration distributions of GOM and PBM may also be related to the changes of local climate and atmospheric composition from S-ISM to WEC periods.’**

**Comment #5**

Line 268& 440, the author claims that strong winds may reduce the concentration of atmospheric mercury in the city. I wonder how the atmospheric mercury concentration in Lhasa compares with the background area (such as Nam Co) under the condition of strong winds?

**Response #5**

Thanks for the comment. We counted the GEM concentration data at the period with wind speed greater than  $4 \text{ m s}^{-1}$ , and the GEM concentration was  $1.31 \pm 0.93 \text{ ng m}^{-3}$ , similar to that of Nam Co at  $1.33 \pm 0.24 \text{ ng m}^{-3}$ . We have added the relevant comparison in Section 3.4. **‘At wind speed greater than  $4 \text{ m s}^{-1}$ , the average GEM concentration in Lhasa was  $1.31 \pm 0.93 \text{ ng m}^{-3}$ , which is similar**

to the average concentration of Nam Co ( $1.33 \pm 0.24 \text{ ng m}^{-3}$ ) in the Tibetan hinterland.'

**Comment #6**

Line 297, how did the authors confirm that the high mercury concentration events occurred randomly?

**Response #6**

Thanks for the question. The observed high Hg concentration events occurred at different time intervals, in different forms of concentration change curves, and the values of high Hg concentrations were also different, so we concluded that the high Hg concentration events occurred randomly.

**Comment #7**

Line 323 Table 3, Please note that colored values in tables are not allowed in ACP, please consider replacing them with italic or bold lettering.

**Response #7**

Thanks for the suggestions. Revisions have been made accordingly.

**Comment #8**

Line 396, does the author think that the changes in atmospheric mercury in Lhasa can be extended to more cities?

**Response #8**

Thanks for the suggestions. Based on the analysis in this paper, we believe that the effects of anthropogenic source emissions of atmospheric Hg on urban atmospheric Hg concentrations are widespread and may occur in other cities as well. However, the ability of wind fields to remove urban Hg pollutants depends on the strength and source of the wind, and only clean wind sources and sufficiently strong and persistent wind fields can remove urban Hg pollution. This hypothesis needs to be confirmed by monitoring atmospheric Hg concentrations in other cities and surrounding areas.

**References**

Rutter, A. P. and Schauer, J. J.: The impact of aerosol composition on the particle to gas partitioning of reactive mercury, *Environmental science & technology*, 41, 3934-3939, 2007.

Rutter, A. P., Schauer, J. J., Lough, G. C., Snyder, D. C., Kolb, C. J., Von Klooster, S., Rudolf, T., Manolopoulos, H., and Olson, M. L.: A comparison of speciated atmospheric mercury at an urban center and an upwind rural location, *Journal of Environmental Monitoring*, 10, 102-108, 2008.