Reply to interactive comment on "Atmospheric mercury concentration and chemical speciation at a rural site in Beijing, China: implication of mercury emission sources" by L. Zhang et al.

To comments from Anonymous Referee #1:

This paper conducted one-full year's continuous measurements of speciated atmospheric mercury concentrations at a rural site in North China plain, which is an important anthropogenic source region of mercury in China and has not been well studied regarding the atmospheric mercury. I think the dataset presented in the study will help the scientists better understand the mercury distributions, sources, and transport of atmospheric mercury in China. This study also made some interesting discussions on the relationships of atmospheric mercury and criteria pollutants, and stories of the intercept of the trend line as well as the RGM/Ozone ratio are quite new to me. I think this manuscript could be published in the journal of ACP after the following comments are addressed.

Reply: We thank the reviewer for supporting the publication of our manuscript. We address all of the reviewer's comments below. The original comments are in black and our responses are in blue.

One general comment is that more discussions regarding the anthropogenic sources of the different atmospheric mercury species are needed. As I learnt from the manuscript, anthropogenic sources were an important factor regulating the distributions of mercury species. GEM, GOM, and PBM showed quite different seasonal trends in the study area, and this may imply the three mercury species may have distinct anthropogenic sources. I would like to encourage the authors to make some detailed discussions on the elevated GEM, GOM, and PBM events. They can also compare the ratios of GOM/GEM and PBM/GEM with the published speciation of mercury compounds released from typical anthropogenic sources in China.

Reply: We thank the reviewer for this valuable comment. The ratios of RGM/GEM and PBM/GEM have been added to Table 2 and comparison with existing literatures has been added to the Section 3.1 in the revised manuscript. Please see Line 202-206 on Page 8:

"The PBM/GEM ratio at Miyun site is higher than most of the monitoring sites in China except Guiyang urban site, while the RGM/GEM ratio at this site is lower than most of the sites in China except Mt. Gongga site (see Table 2). This is possibly due to the heavy PM pollution in North China."

We agree with the reviewer that the discussion on the elevated GEM, RGM and PBM events is very important. We have discussed these events in Section 3.1, 3.2, 3.3 and 5.1. There are several extreme peaks in the observation of the three mercury species. Most of the GEM and PBM peaks match well with the highest API values in Beijing urban area, indicating the influence of heavy pollution episodes in Beijing. The heavy pollution episodes in autumn were longer and heavier than those in other seasons due

to the disadvantageous diffusion condition. This is also the reason why the RGM concentration peak for autumn is higher than that for summer. HYSPLIT modeling was performed for three heavy pollution episodes respectively in spring, summer and autumn. Both the HYSPLIT results and the RGM/O_3 ratios suggest that the heavy pollution episode in autumn is more impacted by the local sources.

Specific comments: Sect. 2.2: please add some relevant information of the field maintenance of the speciated mercury system. How often did you change your denuders, RPF, and impactor plate? The method or reference related to the preparation of denuders should be also addressed.

Reply: Information on the field maintenance of the Tekran system has been added. Please see Line 136-139 on Page 6.

"The impactor plate was changed every two weeks. The quartz filter was changed once a month. The denuder was recoated every two weeks following the procedure developed by Landis et al. (2002)."

Line 8 on page 12182: please clarify the method for the calculation of detection limit, or add reference here.

Reply: The method detection limit (MDL) was provided by Tekran company. The MDL for 2537B is <0.1ng/m^3. The sampling time of GEM is 5 min, and the sampling time of RGM and PBM is 60 min. The sampling flow rate of GEM is 1 L/min, and the sampling flow rate of RGM and PBM is 10 L/min. Therefore, the MDL for RGM and PBM should be 120 times lower than that for GEM, which is about 0.5 pg/m^3. The detail can be found in the study of Landis et al. (2002). Please see Line 119 and 126 on Page 5.

Line 6 on page 12183: Is the ending height of 500 m referred to sea level height or elevation above surface ground. Does the start time mean local time or UTC time?

Reply: The ending height referred to 500m above surface ground, and the start time means UTC time. These two points have been clarified in the revised manuscript. Please see Line 154-155 on Page 6.

Line 14 on page 12183: please specify the criterions of GEM, GOM, and PBM in the PSCF simulations.

Reply: In this study, only GEM is used for PSCF simulations since its lifetime in the atmosphere is relative long. The mean GEM concentration is used as the criterions. This point has been clarified in the revised manuscript. Please see Line 160 on Page 7.

Line 21 on page 12185: the dominant wind here is inconsistent with Figure 5D, please check it.

Reply: The dominant wind direction has been checked and modified in the revised manuscript. Please see Line 218-220 on Page 9.

"The dominant wind direction for spring, summer and autumn was southwest, while

the wind was mainly from north and northwest in winter."

Sect. 3.2: the distinct season trends in GEM, GOM, and PBM are very interesting. The authors declare that some of the pollution episodes worked here. Are there some difference in the dominant wind direction and long-range atmospheric transport among the four seasons?

Reply: The seasonal variation of GEM is mainly affected by the long-range transport. Based on the PSCF results, in winter and autumn, most of the mercury comes from the remote west and north area, including the Loess Plateau and Outer Mongolia. On the contrary, the major sources of mercury in spring and summer are located in the south and east area to Beijing, including Shandong, Hebei, Henan, Anhui and Jiangsu. This is in line with the wind directions. The wind was mainly from north and northwest in winter which is different from other seasons. Please see Line 217-220 on Page 9 and Line 397-400 on Page 15.

Line 12-16 on page 12188: the contribution of natural sources to the GEM/CO ratio should be discussed.

Reply: The contribution of natural sources is mainly reflected by the intercept of the trend line for the GEM/CO correlation. Please see Line 308-314 on Page 12.

Line 22-23 on page 12188: IF the pollution episodes dominated the decreased intercept in autumn, these episodes may have relatively higher GEM/CO ratios. Can you speculate a little bit of the major sources for these episodes?

Reply: These episodes did have relatively higher GEM/CO ratios. The major source could be coal-fired power plants, because the Hg/CO ratio for power plants (25.2) is larger than that for industrial and residential boilers (2.9 and 0.4). In autumn, the influence of coal-fired power plants was enhanced due to the disadvantageous diffusion condition.

Figure 3 on page 12203: please add the mean concentrations of GEM, GOM, and PBM.

Reply: The mean concentrations of GEM, RGM, and PBM have been added to Fig. 3.

Figure 4 on page 12204: why there is a significant difference in PBM concentrations between 23:00 and 0:00?

Reply: There is a peak of PBM in the early morning from 0:00 to 2:00, resulting in the significant difference between 22:00 and 0:00. This peak is probably caused by atmospheric stratification during nighttime against laminar fluxes during daytime, driven by wind secular periodicity. Please see Line 244-246 on Page 9.