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Comment on acp-2021-290

Anonymous Referee #2

Referee 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-RC1>, 2021

The authors present one year of measurements of speciated mercury at a mountain station Walliguan (WLG). They interpret the data in terms of diurnal and seasonal variations. Using concentration weighted trajectory approach they identify source regions. Desert dust has been identified a significant source of particle bound mercury (PBM).

The paper is well ordered mostly well written. Unfortunately, the data analysis is rather superficial leading to sometimes questionable conclusions. The major problem is the frequently occurring pollution events combined with a statistical analysis based on averages and their standard deviations which will be substantially influenced by the pollution events. The pollution events are not discussed although their backward trajectories and chemical signatures could provide additional information about the sources. The data are valuable and deserve to be published, after some improvement of their analysis.

General comment: To the best of my knowledge, WLG is a WMO GAW station and many other trace gases and aerosol parameters are being measured there, in addition to Hg and its speciation. I wonder, why only one or two of these in-situ measurements is used for the interpretation of the speciated Hg measurements. Seasonal variation of directly measured dust concentrations would be useful e.g. for the discussion in the section 3.3 and 3.4. The use of these measurement would substantiate the findings and the conclusions of the paper.

General comment: RH is inversely related to AT, and thus essentially redundant to it. Air water content, which can be easily calculated from RH and AT, would be a really independent parameter and thus a preferable one.

General comment: Figure 2 shows numerous pollution events with seasonally varying frequency of their occurrence. Consequently, discussion in terms of averages will blur the differences because of insignificant differences due to large standard deviations. Medians or seasonal and diurnal event frequencies could provide a more transparent insight as would an analysis of event frequencies.

Section 2.2 has a subsection 2.2.1 but no subsection 2.2.2?

Section 2.2.1: GEM detection limit of 0.1 ng m^{-3} is given, but what are the GOM and PBM detection limits? Please provide sampling flow rates and sampling durations for GEM, GOM and PBM. The problem is that with the usual 5 min and 1 l/min for GEM and 2 h with 10 l/min for GOM and PBM not enough mercury is collected for unbiased and precise analysis by Tekran (Ambrose, *Atmos. Meas. Tech.*, 10, 5063-5073, 2017; Slemr et al., *Atmos. Meas. Tech.*, 9, 2291-2302, 2016). The information about sampling intervals and flow rates is thus necessary to assess the accuracy and precision of the presented measurements. Because of the high altitude of WLG it should be also stated whether the concentrations are related to m^3 at standard pressure and temperature.

Section 2.2: Backward trajectories were calculated every 4 h. Presumably GEM, GOM, and PBM were averaged over the same time stamp but this is not mentioned in the text.

Section 3.1: Because of the GEM temporal trends, GEM measured at WLG in 2012 and 2013 should be preferably compared with measurements at other sites made in the same years. Figure 2 shows frequent pollution events which are not mentioned in the GEM discussion. They will drive the averages and standard deviations up, medians would provide a more representative information.

Line 186: ..will be discussed in detail...

Paragraph starting at line 189: The problem with the internal Tekran signal integration mentioned above is another reason for low bias of GOM measured by the KCl denuder. As such it should be mentioned here too.

Section 3.2: Because of the frequent pollution event the discussion here in terms of averages is obscure. A discussion of monthly event frequencies would provide a more transparent insight. E.g. pollution events are much more frequent in the cold season when compared with the warm one.

Paragraph starting at line 232: "... low RH in the cold season was conducive to the formation of GOM and PBM..". Cold season (November – April) is essentially winter, i.e.

GOM and PBM according to this finding are more efficiently produced in winter. This is at odds with observations of wet Hg deposition peaking in summer almost everywhere (e.g. Cole et al., Atmosphere, 5, 635-668, 2014).

Paragraph starting at line 242: The given numbers without the standard deviations and the number of measurements do not allow to judge whether there is a difference between day and night. In addition, because of seasonal GOM and PBM variations the diurnal variations should be investigated separately for different seasons.

Lines 251-255: It is generally very difficult to separate chemistry from transport in diurnal variations without specific tracers because of diurnal PBL dynamics. It is even more complicated at mountain stations with additional upslope and downslope winds, see e.g. Weiss-Penzias et al. (J. Geophys. Res., 111, D24301, doi:10.1029/2006JD007415, 2006). The attribution of diurnal variation to chemistry here is also highly questionable for another reason: With mercury lifetime of 0.5 – 2 yr, mentioned in the introduction, the day/time difference should be nondetectable considering the GOM standard deviations reported here.

Section 3.3: Why is GOM omitted from the discussion?

Paragraph starting at line 332: It is true that gas-particle partitioning is mainly controlled by temperature. At WLG, however, it will be to a large degree controlled also by the available aerosol surface area which is probably orders of magnitude larger in air masses transported from the desert when compared with other air masses. Measured dust concentrations from the GAW monitoring at WLG could provide a better insight in the seasonal variation of PBM/GOM ratio.

Figure 3: What is the meaning of the bars: standard deviations? Monthly medians would provide a more representative seasonal variation, at least for GEM. Alternatively, seasonal variation of pollution event frequencies should be discussed because it determines the monthly averages and their standard deviation.

Figure 8: The caption is confusing: with the ratios at Qomolangma Nam Co, Chinese cities and Chinese remote areas one would expect an additional column d because urban and remote areas are probably different in PBM/GEM and PBM/GOM ratios?

Figure S1: The RH curve without advection should essentially mirror the AT curve, i.e. it should peak at AT minimum and vice versa. The deviation from this idealised relation shows the diurnal change of local transports. Such transports of different air masses prevent the attribution of diurnal variations solely to chemistry.