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## Interactive comment on "Temporal trend and sources of speciated atmospheric mercury at Waliguan GAW station, northwestern China" by X. W. Fu et al.

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Response to reviewer #2

The authors are grateful to the anonymous reviewer for dedicating the time to read our paper and provide comments. We studied these comments carefully and revised the manuscript following of the comments. All of the revisions and corrections were marked with red in the revised manuscript.

General Comments -Multiple grammatical and typos were observed. There are too many to document them all (estimate >30). Please have a proof-reader fix these errors. For example:

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We checked the grammatical errors throughout the manuscript and made corrections to them.

-Consider using TGM, GEM, (Gaseous Elemental Mercury), GOM (Gaseous Oxidized Mercury) and PBM (Particle Bound Mercury) for your nomenclature throughout the document. This is the nomenclature now being used by the GMOS and AMNet monitoring networks.

We replaced all the terms of the RGM and PHg with GOM and PBM in the revised manuscript.

Specific Comments Abstract: -Define first use of WLG as Waliguan

We defined the WLG at the beginning of the abstract

-Line 18: I believe the use of "direct evidence" is overstated. The use of the term "direct evidence" must be supported by a unique chemical signature, for example Hg/CO, aerosol trace metals or organic compound ratios for the source location, in addition to mercury measurements coupled with meteorological models. I recommend changing the sentence starting on line 16 to read "Moreover, we found that Northern India may be a significant source region for WLG during the sampling campaign, and this is the first published evidence suggesting long-range transport of atmospheric Hg from India to the Northeastern Tibetan Plat

We agree the sentence of "direct evidence' is somewhat overestimated, and we revised the sentence as shown in line 15-18 on page 2.

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-Lines 12-16. Recommend that this sentence is clarified, since there is no consensus or clear research results that define the source of mercury re-emission. It cannot be clearly classified as anthropogenic or natural in most cases.

The sentence was revised as shown in line 18-19 on page 3 -Lines 23-24. Fix typo in

references

The references was corrected

Section 1.3

-GEM sampling: Was the filter located at the inlet to the heated line or at the location of the Tekran? It is important to distinguish, since particles and dust can collect in the sample line and create mercury artifacts.

We both had the filter at the inlet of the heated sampling tube and filter at the inlet of Tekran 2537A. This information was shown in line 27-28 on page 5.

What is the abbreviation LT? It is the local time, we replaced the LT with local time in the manuscript

-Sampling bias caused by the use of 25 foot heated line in front of the manual Hg sampling system for RGM and PHg  $\,$ 

We make a state of the artifact by using the 25-ft heated sampling line as shown in line 6-10 on page 6. At the same time, we also speculate that this did not cause significant effect to our observations, and some of the reasons are shown in line 11-16 on page 6. Therefore, we suggested that we should not discuss this artifact in some of the tables and PSCF analysis. For the relatively stable level of GOM and variable PBM and TGM concentrations, we suppose it was mainly resulted from impacts of sources as discussed in the manuscript. Even through there may be a minor artifact of using the heated sampling line, this should not affect the variations of GOM.

-Please list the temperature of the 25 ft heated Telfon tube used for RGM and PHg sampling.

The temperature was shown in line 3 on page 6

- Please list the field blank mean and standard deviation for the denuders.

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They were shown in line 7-10 on page 7

Section 1.4

- There should be a discussion of the limitations for PSCF in this study because of the complex terrain and limited meteorological data in this region. How sensitive is the model to the arbitrarily set criterion for Hg concentrations?

Information of the meteorological data was shown in 23-25 on page 7. The uncertainties of PSCF analysis were listed in 20-28 on page 8.

Section 2.1

- Recommend changing the format of Figure 3 to a bar graph with the width of the bar equal to the sample time. This would more accurately represent the data, since there are large gaps in a single day where no sample was collected.

We changed the format of Figure 3 to better show the variation of GOM and PBM according to sampling time.

Section 2.2

- Since this is a GAW site, it is a little disappointing not to see any comparisons of the mercury concentrations and wind direction with other tracers of pollution, such as CO, carbon soot, fine fraction aerosol or other measured compounds. For example, if continuous CO data is available, does it have the same wind-rose shape as TGM?

Normally, all the data sets of pollutants are not open to the public in China. We tried many pathways but we failed to download these criteria pollutants data.

-There is no discussion or consideration of the effects of upslope/downslope meteorology on the behavior of RGM at this extremely high elevation mountain site? Please discuss the potential impact on the observations at WLG due to the known high concentrations of RGM in the free-troposphere, observed by multiple research groups as referenced in the manuscript (Murphy, Swartzendruber, Faïn) and recent work by Sheu

at the Taiwan high elevation site. In section 2.6, the authors mention that vertical air flow movement reverses during the night, which results in downward transport of the free-troposphere air. If free-troposphere RGM does not influence WLG observations, please explain with respect to the local meteorology for different seasons.

We made a detail discussion on weak relationship between nighttime GOM and downslope flows from free troposphere in line 1-18 on page 16.

- The paragraph starting on page 30065, line 22 should be re-evaluated and adjusted to be less speculative and over-reaching. For example, comparing the seriousness of Hg pollution levels in two countries based on land-area is too simplistic and speculative.

Deletion and revision was made to limit the speculative assessment in line4-6 on page 13.

## Section 2.4

- The analysis in this section relies on and explanation for the RGM results due to its short lifetime and to some extent the limitations of the PSCF model with limited RGM data. Please bring into the discussion the likely RGM sampling artifact mentioned above and the role (or not) of high RGM in the free troposphere as possible explanations for the PSCF results for RGM.

As shown in section 2.2, we opt to minimize the effect of sampling artifact on PSCF discussion. Besides, we did not find relatively higher GOM with trajectories travelled at relatively higher heights (data not shown here), indicating free troposphere had less pronounced effect compared to local and regional sources.

## Section 2.6

-Does Figure 10 include the entire TGM data set? Please clarify.

Yes, we showed this information in line 19 on page 23.

- There appears to be several contradictory lines of reasoning in this section. Implicat-

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ing quiescent nighttime conditions and nearby local settlements for the largest, most rapid jump in TGM concentrations (to their highest average level) just prior to sunrise seems to contradict the notion of long-range transport and the PSCF results. Also, the fact that this site acts opposite to other high elevation sites, as the authors highlight, is difficult to reconcile. The complex local meteorology and potential impacts of local sources (even the site itself) seem more complicated and deserve a more rigorous review and explanation for the reader. The diurnal pattern for TGM does not make sense for a remote site far from sources, considering TGM is a compound with a lifetime of months.

We did not think this section has contradictory discussions. For the peak before sunrise, we listed the first reason of less replenishment of fresh air due to low wind speed, and this may result in accumulation regional atmospheric Hg level resulted from long-range transport and regional sources. We did not have local sources at the sampling site, and the description of the site was shown in sect. 2.1. For the effect of regional emission from several settlements (all are more than 20 km away and should be regarded as regional sources), this source may be an additional contribution to the morning peak. In most areas of China, regional sources inevitable affect atmospheric Hg observations and should be introduced in the discussion even in Tibetan plateau.

- Consider removing the graph and discussion about the diurnal trend for PHg and RGM. Not enough continuous data was generated to make this analysis meaningful.

We think the data sets of PBM and GOM are enough for the diurnal trend evaluation at WLG. We collected more than one month data sets of PBM and GOM.

## Conclusions

- The entire conclusion, especially the last paragraph should reflect all modifications made in the manuscript based on changes requested or recommended by the reviewers.

We made some revisions based on previous discussions.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 30053, 2011.