Atmos. Chem. Phys. Discuss., 11, C11827–C11829, 2011 www.atmos-chem-phys-discuss.net/11/C11827/2011/

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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.

Anonymous Referee #1

Received and published: 14 November 2011

The manuscript reports one year of speciated mercury (GEM, RGM and PHg) measurement at a monitoring site that is relatively elevated and potentially located in the free troposphere. This is a useful addition to the existing mercury data reported in an important source region (China) of global anthropogenic mercury emissions. The manuscript is well organized. Although there are occasional editorial issues (e.g., grammar and selection of words) in the text that need revisions, they do not substantially impair the readability. The data quality is excellent and I do not have question on the research group's capability in mercury measurements and interpretation of the observational data. However, since the most important source-receptor conclusions in this study are based on the PSCF modeling results, I do feel that there is a need for the

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authors to clarify regarding how the backward trajectory analysis was performed and what the uncertainty level is. I recommend the manuscript be accepted after addressing the following points:

- 1. Page 2, line 16 in Abstract. PSCF model is a conditional probability model and the PSCF results must not be regarded as "direct evidence" of the claimed long-range transport. Additional evidence (such as other chemical signature) is needed for that claim.
- 2. Figure 2. Please show the time series plots of the measured PHg and RGM as well.
- 3. Section 2.3. This is a section that needs substantial clarification. The reliability of backward trajectory calculation is highly dependent on the meteorological data and there is essentially no mention of what meteorological analysis was used for the backward trajectory calculation. Particularly important aspects such as the spatial (vertical and horizontal) and temporal resolution of the met data, as well as the spatial coverage of the meteorological domain, must be stated clearly. The measurement of GEM has a 10-minute temporal resolution and that of RGM/PHg has a few hours of temporal resolution. How was the selection of the "elevated" observations aligned with the temporal scale of the meteorological data? How many trajectories (for GEM, RGM and PHg) respectively were calculated? Redundant calculation of backward trajectories can result in large false signal during PSCF modeling. Also, what is the uncertainly of the trajectory calculation? This can be assessed by performing forward and backward trajectory calculations originated from the source and receptor locations and examining if the starting and end points of the trajectories are closely co-located. This allows quantitative evaluation of the reliability of the trajectory analysis. Finally, depending on the arrival height of the backward trajectories, there may be significant number of trajectories being "grounded" before arriving at the receptor site, were those trajectory endpoints excluded for the PSCF modeling?
- 4. Section 3.3. Page 11, Lines 5-11. This passage sounds very speculative and does

not seem to have any scientific basis. Higher emission cannot be directly translated into a high level of pollution. In addition, there is no relationship between the level of pollution and the proximity to coast. Consider deleting this passage. In Figure 5, I was somewhat surprised that the "mean" trajectory for each trajectory cluster can extend so far away from the receptor location. Can the authors provide the subplots showing the six clusters of trajectories?

- 5. Section 3.4. I was also surprised that the very weak signal of PHg near New Delhi, India can be picked up by the PSCF modeling using 48-hour backward trajectories and wonder if the false signal introduced by potentially redundant trajectory calculation might have caused this. Can the authors provide the number of trajectories contributing to the high PSCF values and a description of the transport pathway associated with these trajectories?
- 6. From Figure 1, there are obviously some extreme events (say, TGM > 6 ng/m^3) observed during the study period. The backward trajectories associated with these events (and similarly, those associated with extreme events of PHg and RGM) may yield important insights on the transport pathways of pollution events. Some discussion regarding this will be very helpful.

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

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