Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2015-842-AC1, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric mercury speciation dynamics at the high-altitude Pic du Midi Observatory, southern France" *by* X. W. Fu et al.

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Received and published: 6 April 2016

RC- Reviewer's Comments; AC – Authors' Comments

RC: This paper evaluates source regions of high GOM and PBM concentration episodes at a high altitude station in westen Europe. The exact vertical distribution of the various mercury species and in particular the oxidation mechanisms of GEM are a major scientific questions in the mercury community. Using backwards trajectories to indentify potential source regions of GOM and PBM, this paper provides a significant contribution to current research questions. The findings are generally in line with recent observations from air-craft campaigns indicating that the free troposphere is a major source of GOM. The paper is well written and the data seems credible. I suggest to

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Discussion paper

publish the paper after minor revisions and clarifications:

AC: We would like to acknowledge the anonymous reviewer for dictating the time to read our original manuscript and provide valuable suggestions. We greatly appreciate all of the comments and have revised the manuscript by addressing all of the comments.

RC: 1) Please clarify how you determined the end points of trajectories (3 hourly endpoints according to the meteo time step?). Why did you choose a fixed period of 7 days for GOM and 10 days for PBM? Did you take into account precipitation events when calculating backward trajectories with HYSPLIT? Concerning GOM and PBM I suggest to stop the backwards trajectories once RH>99% as previously produced/emitted GOM and PBM will probably be scavenged.

AC: The calculation of trajectory endpoints is clarified in line 223-224 on page 8. The total run time of trajectories for GOM and PBM was selected on basis of the atmospheric lifetime of GOM and PBM, which is shown in line 197-198 on page 7. We did not take into account precipitation events when calculating backward trajectories. We agree with the reviewer that humid air mass and precipitation may scavenge GOM and PBM efficiently. In this study, most of the high GOM and PBM events were related to dry air masses from free troposphere and we suggest the effect of precipitation on source identification should be minor. We also introduce the uncertainties of PSCF analysis in line 235-240 on page 8.

RC: 2) Please clarify whether you used any form of ensemble trajectory calculations in HYSPLIT. Already after 2-3 days source regions but especially the altitude can vary significantly. A trajectory could easily originate from the FT in one run and from inside the PBL in a sligthly perturbed model run. (see Figure R1 which depits 27 member ensemble for a single 10 day backward trajectory. Here after 3 days you can see a massive vertical spread of the end points).

AC: We agree with the reviewer that trajectories would have large deviations with time.

In the present study, a 'Normal' type (not Ensemble type) was used to calculate the trajectory.

RC: 3) Please clarify what are the standard conditions for your mercury measurements (e.g. 0 °C, 1024 hPa).

AC: We clarify the standard conditions for mercury measurements in line 128-130 on page 5.

RC: 4) Given your Tekran setup (10 L/min 1h sampling for GOM and PBM) the automatic peak integration should start having problems at concentrations < 12pg/m3. This won't affect the findings of this paper, but in general, I would suggest to check this and possibly apply a correction to small GOM and PBM measurements (compare: Atmos. Meas. Tech. Discuss. doi:10.5194/amt-2015-376, 2016)

AC: We agree with the reviewer that a small Hg load would probably lead to a low bias of GOM and PBM concentrations. In this study, the GOM and PBM concentrations were calculated following the typical method used by previous studies. Also, GOM and PBM concentrations at the PDM Observatory are generally elevated compare to low-altitude sites and may be subjected to minor uncertainties of integration issues. We did not correct small GOM and PBM measurements in the revised manuscript. This also helps to compare with previous observations. Relevant revisions and clarifications are shown in line 123-128 on page 5.

RC: 5) line 21: I would prefer if you gave the concentration range instead of the maximum concentration. (33-98 pg/m3) (91-295 pg/m3) line 40: Higher than 'those' instead of 'that' line 52-53: Please clarify here that PBM is not significantly removed by dry deposition line 61: should read 'which' instead of 'what the' major oxidants line 72-75: You state that CARIBIC found a positive correlation between TGM and O3. However, the publication by Slemr et al. you mention only discusses a correlation between GOM and O3. line 139-140: please rephrase (do not retain half of GOM) line 157,162, and others: I generally suggest to write 'at the PDM observatiory' or 'at PDM' but not 'at

PDM observatory' line 196: Please clarify what exactly you mean by end point and what time step you used (e.g. hourly endpoints?) line 277: ratios instead of ratio. Line 370: relative instead of relatively Supplement Table S3 caption: It should say "high GOM events"

AC: We checked the English writing errors carefully throughout the manuscript, and the errors mentioned above were corrected.

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