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Atmospheric Chemistry and Physics Discussions

Interactive comment on "High levels of reactive gaseous mercury observed at a high elevation research laboratory in the Rocky Mountains" *by* X. Faïn et al.

Anonymous Referee #1

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General:

This manuscript presents an interesting data set for speciated atmospheric mercury at a site in the Rocky Mountains. The authors state that previous research at the site suggests little influence from boundary layer air. I think the authors need to dig into the data deeper and demonstrate (or not) this point. I think, as stated in my specific comments, that there is reason to believe that the high RGM may be related to boundary layer air reaching up to the site. I think one way to resolve this would be to collect canister samples for analysis of biogenic hydrocarbons. This would clearly indicate the presence of boundary layer or free tropospheric air at the site. I am mystified as to how

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these elevated RGM events can persist for up to nearly one week in time. However, the scale on Figure 1 does not allow a detailed look as to the timing of the see-sawed shape of the RGM during the events. What does the time series of ozone and CO look like? This could offer some clues as to what is going on in this data set. The authors need to take a very careful look at the data, and sort it out instead of just plotting all the data together. Separation of the peaks and valley data might yield answers to what is driving the trends at the sampling site. What I am thinking here is what the authors labeled as individual events, may actually be a series of days where boundary layer air reaches up to the site causing multiple sharp peaks in RGM (see below). These aspects of the data set should be explored to try and gain a better understanding of the processes influencing the site.

There are papers documenting the extremely high levels of Hg(0) and RGM in the southwest that are related to mining operations (Gustin group papers). The mining operations could add some CO but little ozone. This elevated RGM might be carried in very dry air from the arid mining areas. Terrain-forced flows could potentially transport these emissions to the sampling site. I think this source must be considered. Again, the biogenic hydrocarbons might be key indicators in this analysis.

The word “height” is used in the text repeatedly, sometimes multiple times in one sentence. It would seem better to use “altitude” since your analysis concerns the middle and upper troposphere. The times reported in this manuscript appear that they are local (not UTC), and as such this should be stated explicitly.

From p. 16651 onward, the text contains a lot of speculation, and much of the information might fit better in the Introduction section. The text goes on and on, but does not say much in explanation of the data set of interest. I think more detailed analysis and a re-working of the last few sections of the manuscript would improve it greatly. This is an interesting data set, but the authors have not conducted an in-depth analysis of it. I believe that there is much more to be learned from the data than the hand-waving explanations offered in the current version. I encourage the authors to do this.

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Specific:

Abstract – The authors have used both CO and carbon monoxide in this section. Also, ozone is written out throughout the text, but the chemical symbol is used for CO and mercury. There should be consistency in the usage of these terms.

p. 15643, lines 1-4 – nobody really knows, in my opinion, if Hg-particulate is divalent or not.

p. 15643, last few lines - In fact, there are two papers that discuss Hg-particulate measurements from aircraft.

Murphy, D. M., D. S. Thomson, and M. J. Mahoney (1998), In situ measurements of organics, meteoritic material, mercury, and other elements in aerosols at 5 to 19 kilometers, Science, 282, 1664– 1669.

Murphy, D. M., P. K. Hudson, D. S. Thomson, P. J. Sheridan, and J. C. Wilson (2006), Observations of mercury-containing aerosols, Environ. Sci. Technol., 40, 3163–3167. The PALMS instrument used by Murphy is an in situ measurement, so the text here and previously requires some re-wording.

p.15646, first sentence, last paragraph – did you re-calibrate the instrument every 26 hours, or just verify the overall calibration?

p. 15666, Figure 1 – It would be better if the relative humidity was converted to specific humidity. I believe you have all the meteorological data to do this. This would provide a real indication if the air with enhanced RGM consistently had the same characteristics and potential altitudinal source.

p. 15667, Figure 2 – I would advise that either symbols or colors be added to make it easier for the reader to distinguish between the variables plotted in this figure. The afternoon peak in aerosols suggests boundary layer air is reaching the site – 3000 cm-3 is a typical boundary layer number density, but only in pollution would it reach this high in the free troposphere. The close tracking of aerosols, RGM, and temperature

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suggest to me that this is indeed caused by boundary layer air reaching up to the sampling location during the day. Again, specific humidity would be the preferred over relative humidity here.

p. 15668, Figure 3 – I'm a bit confused about ozone. In my initial screening of this paper ozone mixing ratios spanned up to about 250 ppbv. In this version it tops out at about 60 ppbv. There was no mention of this in the text. What's going on here? In my earlier interpretation I thought that there might have been an issue with the CO calibration. Now the ozone values look more like clean boundary layer mixing ratios. They seem low for free tropospheric air. In this version the ozone seems too low for the moderately high CO. The CO seems too high for free tropospheric air masses.

I would sort the data with elevated RGM and plot it versus ozone. I would do the same with CO. A little further analysis is warranted here in my opinion. There is obviously no correlation with aerosols, so I would omit this panel and just say so in the text. I would also plot elevated RGM versus specific humidity.

p. 15651, Section 3.2 – I honestly don't seem the point of the discussion in this section. In general, there has not been budget closure found in studies of Hg(0), RGM, and Hg-particulate. This area of mercury research needs significant work, and much of the text here seems superfluous.

p. 15652, Figure 5 – Just because these 10 day trajectories show the general flow was from the over the North Pacific, doesn't rule out very local terrain-forced flow in the mountains. This would never be picked up by the data used in the trajectory calculations.

p. 15653, second paragraph – Why would you expect to find a correlation between RGM and ozone? This doesn't make any sense to me – omit?

p. 15652, Section 3.3 – Much of the information in this section is more suited for the Introduction as its general information rather than directly related to the data reported

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here.

p. 15654 – As I suggested in my initial screening of this manuscript, I think that the authors should look at the PV over the North Pacific in the vicinity of the trajectories. This will tell if stratospheric air possible influenced air masses advected to the sampling location. The authors need to explore all possible explanations for their data set.

p. 15655, paragraph 2 – I doubt that halogens released from sea salt have an influence on air masses in the middle and upper troposphere (based on your trajectories). This discussion seems highly speculative. In the middle of the North Pacific boundary layer, the air masses are in general quite clean, indicating little heterogeneous chemistry and subsequent halogen release.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 15641, 2009.