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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Factors influencing the large-scale distribution of Hg° in the Mexico City area and over the North Pacific" *by* R. Talbot et al.

Anonymous Referee #2

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General comments:

This paper reports data on large-scale distribution of elemental gaseous mercury (GEM) over the North Pacific and Mexico City area based on research flights conducted in spring 2006 (INTX-B campaign). The results on vertical and spatial distributions of GEM along with other atmospheric compounds (O3, CO, C2Cl4, CH4, CO, and CO2) from 13 research flights present an impressive data set and merit publication in Atmospheric Chemistry and Physic Discussions. The paper presents novel data, the scientific methods are clearly outlined, and the scientific questions are clearly within the scope of ACP. I consider the range of altitudes, from the boundary layer to 12 km, a specific strength of this study. The experimental setup and measurement techniques are sound.

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Having said that, there are several points that should be addressed prior to publication. Specifically, I consider section 3 discussing the large-scale distribution as relatively weak and unfocussed. The large-scale spatial distribution is the main objective of the paper and should receive more detailed discussion and conclusions, and should be referenced better to relate patterns to previously reported GEM distributions (e.g., north-south gradient, land-ocean distribution, vertical distribution, modeling studies. Several explanations of observed patterns were difficult to understand or were speculative.

Specific comments:

Section 3. Large-scale distribution While the authors well describe patterns of various atmospheric constituents such as CO, O3, too little discussion actually refers to observed GEM patterns, and some interesting patterns are not discussed at all.

For example, the rather low GEM levels in the boundary layer around Mexico City should be discussed in detail. Although the layer was clearly enhanced in CO, no corresponding enhancement of GEM was observed (page 15540, lines25 to 15541, line5). Why does one of the largest urban areas not show enhanced levels of GEM (in the boundary layer), and why do the two remote Pacific sites actually show higher levels than the urban site? Could the selection of the flight paths have missed polluted air masses? Or does the area simply lack significant GEM emission sources?

However, Figure 4 actually seems to show several occasions with enhanced GEM values in the Mexico City boundary layer, but these events are poorly visible in Figure 7 (showing averaged values) because of generally lower background GEM levels. Could higher levels observed over the Pacific sites be due to ocean sources to the boundary layer? The authors give one possible explanation in section 3.3.(which should be moved into this section). The patterns should be discussed in respect to published literature on north-south distribution patterns, land-sea distribution patterns, altitude gradients, etc.

The authors mention that GEM did not follow the CO altitude trend which indicates

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that it is not being depleted rapidly in the Tropics. The authors should give references why they would expect such depletions in the first place? The backtrajectories show that the upper air masses had a source origin in the Atlantic, probably very different from the boundary layer air mass source origin. Does this not make it very difficult to discuss conversion processes, or a lack thereof, if sources origins of the airmasses are so different?

In the following section, patterns of CO and O3 are described in moving from low to high latitudes, and the authors mention that GEM patterns are subtle and that at high latitudes the steepest decline occurred in the high latitudes. These high troposphere mercury depletion events are discussed in section 3.3. and in detail in another paper (Talbot et al., GRL, in press). The only other point discussed here is an apparent lack of significant marine boundary layer removal of GEM. This explanation was somewhat surprising to me. I am aware of observations reporting increased RGM levels in the marine boundary layer (Sprovieri et al., EST 2003, Laurier et al., JGR 2003, maximum levels of 30.1 and 92.4 pgm-3, respectively). However, is this GEM oxidation not much too low to be detected as an actual depletion in GEM? I am not aware of any study reporting significant GEM depletions due to this transformation in the marine boundary layer, with the exception of recently observed GEM depletions over the Dead Sea (Peleg et al., EST 2007). So why explain a lack of removal if it has not really been observed before?

A further point that merits discussion is the variability in GEM observed with altitude. For example, Honolulu seems to show little concentration variation with altitude (narrow band of concentrations), i.e., a good background site with little industrial pollution? Houston and Anchorage, on the other side, show larger minimum and maximum values. For the Mexico area, specifically at low altitudes, is this likely due to source emissions to a relatively lower background GEM level? For Anchorage, the enhancements might be explained by Asian long-range transport as discussed in detail later in the text. There are, however, enhanced levels observed also at lower altitudes (e.g.,

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around 2km). This could raise important questions in regards to marine boundary layer transport pathways versus free troposphere pathways (e.g., Jaffe et al., 2003 AE, Cooper et al., 2004, JGR). I also suggest to discuss the unusually low values observed in this study (e.g., many values below 50 ppqv observed in Houston and Anchorage flights). It might be worthwhile discussing these in respect to marine boundary layer oxidation, stratospheric influence, and/or arctic mercury depletion events in the high latitudes.

Section 3.1 Correlation with source tracer species This section is well structured and reads well. One notion, though: I am not sure if correlations with other tracers are non-existent in the Mexico data set Figure 8? To me, correlations of the Mexico data do not seem much weaker than in the Honolulu data set (e.g., CO). Can the authors supply r2 and P values to clarify this? I also do not understand the point about the lower CO2 levels around Mexico due to well-aged equatorial air masses? Do the authors mean that these air masses have low CO2 values because the source origin lays over the Atlantic with few (no) combustion sources? In Figure 9 the authors should calculate GEM/CO enhancement ratios (seems to be around 0.33ppqv/ppbv). This might add valuable information since no such enhancement ratios were calculated for the Mexico City area in Fig. 10 due to large scattering.

Section 3.2. Plume GEM-CO relationships The authors might want to discuss measured GEM/CO ratios in more detail. For example, the Anchorage data compares very well to other reported Asian pollution plumes as noted, but what about the Honolulu data? Why are these values almost half of those observed around Anchorage? Are these event hence not attributed to Asian sources, or do they originate from different areas in Asia? Can the authors calculate backtrajectories for some of these plumes to clarify this point? The Mexico City ratio (e.g.,. calculated from Figure 9) could also be mentioned and compared to ratios reported for Center/North American pollution plumes.

Section 3.3. GEM depletion events The authors might want to refer to Figure 4 when

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first mentioning GEM depletions events in the text. Although the results of high troposphsere/low stratosphere GEM depletions are discussed in detail in another paper (Talbot et al., 2007) this information is valuable to explain some patterns. The authors conclude that there was not evidence of a gradient in the GEM mixing ratio at low altitude and refer to Figure 7. I wonder if Figure 7 (showing average GEM levels) is in fact a good way to look if upper-level GEM depletions can mix down to lower areas? Have the authors looked in detail at the low values of 50ppqv in figure 4 and evaluated if those might be due to stratospherically influenced air masses penetrating downward?

Attributions of low Mexico City area GEM levels to urban halogens seems somewhat speculative and should be discussed in section 3. Do the authors have any indication for this process? Are there any correlations of low GEM values with urban tracers to substantiate this?

I also question the statement that a stratospheric influence is not evident in the boundary layer over populated mid-latitude continental areas due to active emission/sink processes obscuring the effect. What is the basis of this statement? Do the authors assume it would be evident without these other sources/sinks? To me, the fact that no stratospheric influence was evident over remote sites either (i.e., Anchorage and Honolulu) indicates that stratospheric influence is not apparent even over unpopulated sites.

The last sentence in the section does not relate to this study.

Section 4. Conclusions The conclusion part does not accurately conclude about the topic of this paper (i.e., large-scale distribution of GEM). The conclusions seem to focus on GEM depletions by halogen chemistry and the possible fate of HgP fate and halogen chemistry, rather than about observed large-scale distribution patterns of GEM over the North-Pacific and Mexico area. I suggest to revise this section.

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