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Interactive comment on “Ten years trends in atmospheric mercury concentrations, meteorological effects and climate variables at Zeppelin, Ny-Ålesund” by T. Berg et al.

T. Berg et al.

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Anonymous review for: T. Berg et al. (2013), Ten years trends in atmospheric mercury concentrations, meteorological effects and climate variables at Zeppelin, Ny-Ålesund, Atmospheric Chemistry and Physics

Discussion General comments Berg and colleagues present a ten-year observational record of GEM and perform statistical trend analysis, as well as correlation analysis with meteorological and climate variables. The authors find no statistically significant trend at Zeppelin, which is an interesting contrast to other high Arctic (Alert) and North-

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ern Hemisphere (Mace Head) sites that do detect a statistically significant decrease. The decreasing trend at Alert and Mace Head and the absence of a trend at Zeppelin presents a conundrum for the Hg community since anthropogenic emissions (and hence the global background of atmospheric Hg) are thought to be increasing globally. Long-term observational records such as the one at Zeppelin are very valuable for diagnosing changes in the Hg cycle. With revision, this work may help shed light on the factors driving change in Northern Hemisphere GEM concentrations. I recommend this work for publication after substantially revising Section 3. Results and discussion, which is overly difficult to read due to lack of clarity. Below are general comments that can be applied to the whole of Section 3 to improve its clarity and impact.

The authors wish to acknowledge the reviewer for his/her detailed and exhaustive constructive comments and suggestions that have greatly improved the manuscript.

Be more quantitative. The authors should provide quantitative information anywhere in the text where it says “correlation” or “relationship”. Qualitative words like “higher”, “lower”, “less”, and “more” are not helpful. Simple quantitative measures like correlation coefficients and percent change would vastly improve the scientific robustness of the discussion. This applies to the Abstract as well.

We have added a little bit more of this information in the text, but find in general a text difficult to read if too much of this information is included in the text. The reader may find this information in the table and some of the figures.

Break up long paragraphs. A number of the paragraphs are very long and contain a number of disparate ideas, which makes the discussion unclear and difficult to read. Break long paragraphs into smaller paragraphs and stick to the rule of one major idea per paragraph.

The text is changed as recommended.

I also suggest paying more attention to the structure of the paragraphs. As a reader,

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you often don't get the main point or major result until several sentences into the paragraph. State the main point or major finding at the very beginning of each new paragraph. This will improve the clarity and impact of your discussion. Provide more concise explanations for observed patterns/trends in your data. The authors present a feature of their data and then attempt to explain this feature, but it frequently comes across as if the authors are just guessing and dumping every possible explanation on the reader. As a result, the discussion is confusing and unconvincing. The authors need to exercise more scrutiny in determining which explanations are plausible and which are not. The discussion would benefit from the authors placing greater emphasis on the explanations they find most compelling, and concisely summarize those explanations which are less plausible (or implausible) and providing justifications for why these explanations should be given less weight.

The text is changed as recommended

Revise Figures 7 and 8. Figures 7 and 8a-g could be more effective than in their present form. Figure 7 only provides qualitative information, which doesn't add any substance to the analysis. Also, the figure caption should explain why April is selected. A more effective figure of sea ice would be to show a time series of total sea ice extent (in km²) over the same period as the GEM data. Does the series of sea ice extent look like the AMDE curve in Figure 6?

Good idea. Fig. 7 (Sea ice maps) is removed and data for the sea ice extent is included in a new Fig 6.

Figures 8a-g are too busy and it's difficult to pull out the useful information. I suggest keeping the color scheme and re-plotting Figures 8a, 8d, 8f, and 8g as scatter plots. Figures 8c-d could be combined into a wind rose showing magnitude and direction. Other plots may be more effective and the authors should use their discretion here, but eight box and whisker plots is not an effective way to present this data and discuss correlations between GEM and meteorological correlations.

The authors find Box and Whisker Plot easy to interpret and show correlations on, but we also see that other people may have another opinion on this. We have chosen to keep them as they are for temperature, wind direction, wind speed, relative humidity and the hour of the day, but have replaced them with other and hopefully better figures for UVB.

Specific comments Title: “Ten years trends” seems grammatically incorrect. I suggest “Ten-year trends”.

Changed as recommended.

Page 2275, line 6: There is little evidence to support a 2-year atmospheric lifetime. More recent estimates (e.g. Corbitt et al. [2011]) are closer to 6 months. Your discussion in Section 3 (Page 2279 lines 6-11) suggest 2 years cannot be the correct atmospheric lifetime, otherwise you wouldn't see such large gradients between the Northern and Southern Hemispheres.

We do agree that recent studies show a shorter atmospheric residence time than 2 years, and hence the text is change to reflect this and we have updated the references. However, the large concentration gradient between the Northern and Southern Hemisphere is not only due to atmospheric lifetime, but also most of the emission sources are located on the Northern Hemisphere and there are strong zonal winds preventing inter-hemispheric mixing. The text now reads:

“Current estimates of the atmospheric residence time of GEM range from months up to 1 year (Hedgecock and Pirrone, 2004; Selin et al., 2007; Corbitt et al., 2011) and is considerably longer than the residence time of GOM and PBM which can be deposited within hours to weeks (Schroeder and Munthe, 1998).”

Page 2275, lines 10-23: Is there a reason why you omitted mentioning AMDEs occurring in non-polar environments (e.g. Dead Sea [Obrist et al., 2011])?

We did not include that AMDEs is occurring in other areas than polar environment since

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this is a publication almost solely covering polar areas. However if the reviewer feel that this would improve the quality of the manuscript, we have no problems including this reference.

Page 25 line 24 to page 2276 line 9: Seems worth mentioning that observed decreases are at odds with emission inventories, which suggest total global anthropogenic emissions are increasing [Pacyna et al., 2010; Streets et al., 2012].

The new updated emission estimates for 1995, 2000, 2005, 2010 (AMAP/UN-ECE) are now included the paper and discussed in the results/discussion.

Page 2276 lines 10-18: This paragraph is difficult to decipher. I'm still not quite sure what the point is. Please revise for clarity.

The paragraph is now revised and reads now : "Intensive measurement campaigns designed to understand the cycling of atmospheric mercury provide considerable information which apply only a snapshot in time. Care should be taken when scaling-up such measurements to apply for longer time periods and over large geographical areas (Pfaffhuber et al., 2012). Long-term measurement programs can provide valuable information towards understanding the processes involved in the cycling of Hg in the polar atmosphere. These observations are also important for addressing potential effects of climate warming on the mercury cycle. Additionally, long-term observations are needed to see possible effects from changes in the global anthropogenic Hg emissions on the deposition of Hg to the Polar Regions (Steffen et al., 2008)"

Page 2278 line 1: Why are you measuring CRS03 hourly dose rate?

We have used CRSO3 dose rates as a proxy for photolysis rates. Unfortunately, photolysis rates could not be estimated because we did not find a full, consistent set of absorption cross sections and quantum yields for all relevant reactions. Furthermore, the conversion of measured irradiances to actinic fluxes remains uncertain and nontrivial (Kylling et al., 2003). Ozone is indicated as an important factor in AMDEs (Lindberg

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et al., 2002), and we assume that the spectral efficiency of light in causing AMDES would resemble that of spectral distribution of ozone absorption cross sections.

* Kylling, A., et al. (2003), Actinic flux determination from measurements of irradiance, *J. Geophys. Res.*, 108(D16), 4506, doi:10.1029/2002JD003236. We have added: "The CRSO3 and UVB dose rates were applied as proxys for photolysis rates".

Page 2278 line 2: Delete "(474 m a.s.l.)". You already said this on the previous page. No need to repeat.

This is the only place in the document the authors feel that adding the altitudes make it easier for the reader to follow the text.

Page 2278 lines 2-3: "...assuming the same ozone and cloud conditions." Is this a good assumption?

This is a perfect assumption if all ozone and clouds are above 475 m.a.s.l. Under pristine Arctic conditions with little local sources of ozone, the tropospheric ozone levels are very low and stable. Most of the ozone is in the stratosphere. Ozone between 25 and 475 m.a.s.l. is small compared to stratospheric ozone. Furthermore, clouds are observed to be above the Zeppelin mountain at 474 masl for most days. Regardless, we correct for the effects of air molecules and aerosols between 25 and 474 meters. In the event that ozone and clouds would occur between 25m (Ny-Ålesund) and 474 m (Zeppelin), our top of mountain UV simulations would be somewhat underestimated. We have added : ":assuming the same ozone and cloud conditions for both measurement elevations at this pristine Arctic location.."

Page 2288 lines 7-10: The authors state that the diurnal pattern of GEM at Zeppelin is driven by "daytime snow surface emissions induced by solar radiation" and then immediately state that UVB and GEM aren't correlated. Isn't this a contradiction?

We agree. We have removed the box and whisker plots for UVB and CRSO3 from the manuscript and made a new figure better showing the relationship between UV and

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emissions. The text now reads: “The role of UV radiation was explored by linear correlations between GEM and UVB dose rates rather than binned box plots because the range of UVB values in March was much smaller than in June. Monthly median GEM values for each month were plotted against the simulated UVB dose rate for that month from 2000 to 2009 where each point represents a given year. These plots and linear regression fits for March and April are shown in Figure 8. The March correlation shows that there is a positive significant correlation between the average UVB dose rate and the monthly median GEM levels ($R^2=0.93$, $p<0.001$). This relationship may be tied to increased snow surface emissions since the higher levels (>1.8 ng m⁻³) are above hemispheric background (1.5-1.7 ng m⁻³). In April, median GEM concentrations were inversely correlated with the average UVB dose rate on an interannual basis ($R^2=0.59$, $p=0.016$). In this plot the years where the UVB dose rates are high include low median GEM which suggests that the oxidation of GEM is photo-initiated. No significant correlations were found in May or June (not shown). The competition between these two effects on the GEM levels (surface emission and atmospheric oxidation) may explain the poor correlations on a finer timescale.”

Page 2278 lines 8-9: “was obtained from the (Fetterer et al., 2012).” Typo? The paragraph now reads: “Monthly sea ice area index and maps for the Northern Hemisphere was obtained from National Sea Ice Data Center.....”

Page 2278 lines 8-11: It’s odd that these two sentences stand alone as individual paragraphs. Would be better woven into paragraphs above. Changed as recommended.

Page 2278 line 11: Surely there is a precedent for defining AMDEs to be below 1.0 ng m⁻³. Cite previous studies that do this (e.g. Cobbett et al. [2007]). Changed as recommended.

Page 2279 lines 2-4: “Month values... from the National Weather Service Climate Prediction Center (Fetterer et al., 2012).” This sentence seems like it belongs in Section 2.2. Changed as recommended.

Page 2279 lines 14-15: “This is the time of the year... mid latitude source regions dominate.” Please provide a citation. Hirdmand et al. (2009) is included as a reference

Page 2279 lines 15-16: “During summer... previously deposited GEM.” Please provide a citation. Hirdmand et al. (2009) is now included as a reference.

Page 2281 lines 14-15: “Emission estimates do not exist for... 2006-2009.” Not quite true. Streets et al. [2011] goes up to 2008 and is distributed by region and sector. The latest UNEP report (Time to Act, http://www.unep.org/PDF/PressReleases/Mercury_TimeToAct_hires.pdf) provides an estimate for 2010. We agree. The text now reads:

“Briefly, most of the available GEM concentration measurements begin after 1995 (Berg et al., 2004; Berg et al., 2008a; Cole and Steffen, 2010) since which total global anthropogenic emissions of Hg have increased a little, although there have been significant changes in emissions from particular source regions during this period. This statement is according to the newest, recalculated and updated emission estimates for 1995, 2000, 2005, 2010 (AMAP/UNEP, 2013) (Pacyna et al., 2010; AMAP/UNEP, 2013). Mercury emissions from East Asia have increased by 50% from 1990 to 2005 whereas emissions from Europe and North America have declined over the same period and estimates for 2010 shows a further increase from East-Asia (AMAP/UNEP, 2013). Overall, these emission trends are smaller compared to those reported from the 1970s and 1980s (Pacyna et al., 2009)..”

Page 2285 line 27: You define PBM in the Introduction but use “PHg” here. Either is fine, but important to be consistent to use the same terminology throughout the paper. Changed as recommended.

Page 2286 lines 18-19: What does it mean for an air mass to have “strong” contact with the ocean?

An air mass has strong contact with the ocean when the air parcel is moving at the

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lowest part of the boundary layer and is affected by the chemistry in the ocean surface layer.

Page 2289 lines 18-19: Be consistent with the capitalization of “sea ice”. Changed as recommended.

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