

Interactive comment on “Atmospheric Mercury Concentrations observed at ground-based monitoring sites globally distributed in the framework of the GMOS network” by Francesca Sprovieri et al.

Anonymous Referee #3

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Review of the paper ACP2016-466 “Atmospheric Mercury Concentrations observed at ground-based monitoring sites globally distributed in the framework of the GMOS network” by F. Sprovieri et al.

The paper describes the global mercury observation network that has been established in the framework of the EU-FP7 project GMOS in recent years. Observations in the years 2013 and 2014 have been statistically evaluated for the Northern and Southern Hemisphere as well as for the Tropics. The main results are presented and some special features are highlighted.

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The paper is worth to be published in ACP, however it needs some significant revisions with regard to the statistical evaluation. My main concern is the data coverage which is very different among the stations and also from year to year. Many conclusions are based on data sets that cover different times or very different number of observations (e.g. 2013/2014 at Dumont d'Urville). Some results look unreliable, e.g. 2013/2014 differences at Mt. Ailao which are not discussed although the Asian stations are intensively described in section 4.2.2.

In addition, there are some contradictions and redundancies that need to be corrected as well as numerous technical errors. These technical errors should have been corrected by the authors before they submitted their manuscript for review.

All pages and line numbers refer to the pdf version of the manuscript.

Major comments:

page 3, line 21: "... and highlight its potential to support the validation ...". I cannot see where this is explained, shown or highlighted in the paper.

Table 1 contains 27 stations, Figure 1 shows 26 stations and Table 2 includes 23 stations. You should explain why this is the case.

Table 2 and 3 and Fig. 1, 2 and 3: It is necessary that you give more information about the data coverage and how the averaging and statistical evaluation has been performed. For the monthly average: how many days were used to generate this average or is this the average of all observations within the month? For the annual average: Is this the average of all observations (each lasting 300 s) or did you first calculate daily averages and then the annual average? You should always give the number of data points that is behind the values you have in the tables and in Fig. 3. One of the main shortcomings of this paper is that at some stations data coverage is very inhomogeneous (at least that's what I get from Fig. 1) and therefore annual averages might not be comparable between the years 2013 and 2014.

Table 3: The concentrations at MAL are much higher in 2013 compared to 2014. Is there a good reason for this?

page 9, section 4.1: Here, you should give more information about data coverage and consistency, e.g. about the averaging methods, the number of available data points etc. In line 21 you mention that most measurements started at the end of 2011 and your evaluation is for 2013 and 2014. What about 2012?

page 9, line 25: Tables 2 and Tables 3 do not contain all GMOS sites as I mentioned earlier. The stations in Fig. 1 are not consistent with those in Table 1 and Table 2.

page 9, line 26/27: You need to be much clearer with averages, means and medians. What you call “mean concentrations” are station averaged medians. How were the annual medians derived? What is given in Table 3 and what is the basis for the values?

page 11, line 5-8: You say that you fitted a log-normal distribution. Why did you do so and how did you do it? The PDFs in Fig. 4 and Fig. 5 look very much like normal distributions. Can you show that the frequency distribution of the observations has a skewness that differs from zero? The standard t-Test is only applicable to normal distributions, which might be fine if I look at the PDFs. However, you claim that the concentration values are log-normally distributed. In my opinion you test if the means are different on a 99% confidence level. Again, you need to say what the basis for your evaluation and your fit is. Did you simply take all 300s observations from all stations in the individual sub-groups (Northern, Tropics, Southern)? Why do you show the same for the monthly averages?

page 13, Table 5 and page 17, lines 8-11: I think it would make more sense to give a constant p-value (say 0.05, the most commonly used value) and then give the confidence intervals. This could result in the means of MCH and MAL being not significantly different on this level. I wonder why the difference between MCH and MWA should be lower than that between MCH and MAL. This looks wrong in the table.

I also cannot follow the explanation on page 17 (l 8-11), that tries to demonstrate that

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the PDFs of MCH and MWA are significantly different. I do not see why this is to be shown. It is certainly not true on the $p = 0.05$ level.

page 15, lines 31-34: It would be nice if you could elaborate a bit more about what would be needed in order to understand the fate of atmospheric Hg and the reaction kinetics. Certainly global GEM observations are valuable but not enough. This could of course also be done in the conclusions.

page 22, lines 20-22: Shouldn't it be visible in the observations at MAN if there is an influence from regional sources on the mean concentrations? Did you analyse temporally higher resolved data than monthly averages?

page 22, lines 28-31: These statements are very weak. You do not explain how the meteorological conditions influence the observed concentrations. Obviously, GEM concentrations are always influenced by the hemispheric background. The questions is why you do not see a regional impact from the sources that are expected to be present in the area. Can that be answered by the meteorological conditions?

page 23, line 29 - page 24 line 8: These statements about GEO and GEOSS do not fit into the conclusions. GEO is briefly mentioned before on page 4. Some of this text about GEO would fit better there.

page 23/24, Conclusions: To me, the conclusions sound too general. You should say more about what you found out with the analysis presented in the paper and then give an outlook about what can be achieved if the observations are continued or improved.

Minor comments:

page 4, line 7: Where are the guidelines available? Is there a web page where they can be read or downloaded?

Table 1: Put the explanations in the bottom into the caption. Give the "Country" as third column right to "Site". Give units for Lat and Lon. Replace ',' with '.' (e.g. -37.79604).

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page 6, line 34: what exactly are sub ng/m^{-3} levels. This can be much if the concentrations are not higher than 1 ng/m^{-3} .

page 7, line 1: How is it possible that GMOS results have been published in 2010 when the project started in November 2010 as you indicate here? Is there another reference to GMOS intercomparison studies?

Table 2: The percentiles in this Table are also shown in Fig. 3. They could be moved into an appendix. The caption would need more explanations, e.g. what “5th”, “25th”, ... means. Mean and st. dev might then be moved to Table 3.

Table 3: You need to give more explanations in the caption about what is shown. “Monthly based statistics” does not tell much. What is shown? What are the units? Are these mean or median values?

page 10, Fig. 1 and Fig. 2: The details are hard to read because the pictures are too small. Why do you say “some of the ... stations”. This figures contains more stations than Table 1. Some of them, e.g. Iskrba are not used anymore. Why?

page 10, line1: “according to their location”: What is the rule for this? I suppose by latitude from North to South but it is not mentioned.

page 11, Fig. 3: This figure contains the same information as Table 2.

page 11, line 4: I suppose the groups are related to latitude (not longitude).

page 12, Fig. 4: This figure could be smaller. How did you choose the width of the bins for the histograms that represent the observations?

page 12, Table 4: Sometimes you give 3 and sometimes 4 significant digits. What is the reason behind this?

page 13, caption of Table 5: What are “experimental measures”?

page 15, line 6-8: You say that concentrations at EVK are comparable to aircraft ob-

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servations in August 2013 over Europe. The aircraft observations represent just a snapshot. Do you want to argue that GEM concentrations in the free troposphere are always around 1.3 ng/m^3 ? If this is the case you need to present more evidence for this.

page 17, line 31: Is there any proof for higher direct PBM emissions in winter? What are the sources

page 17, line 34 - page 18, line 1: Is there a proven relation between high PM_{2.5} concentrations and high PBM concentrations as you indicate there?

page 18, line 2: How do you know that high GOM values were due to local sources. Is there anything that supports this?

page 19, lines 1/2: How do the observations support the fact that East Asia is the biggest source region the world?

page 19, line 5: Again, I get confused with the median and mean GEM values. You should only use one of those, depending on the frequency distribution of the observations.

page 19, line 19/20: "... anthropogenic emissions from ... can sometimes be observed": Is this shown somewhere or is there a reference for this?

page 19, lines 31/32: "... the lower concentrations ... were associated with ...": Again, you claim something that cannot be verified by the reader through an analysis that you performed. You don't give any reference, either.

page 20, line 7: "..., reported concentrations ...": Please give the reference where they were reported.

page 20, line 23/24: "... exported from ... by strong katabatic winds.": Did you investigate this? How is it proven? Or is it taken from another publication? Then it should be cited, here.

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page 20, line 23: “Observations at DDU also highlighted ... ”: The reader cannot follow this. Where can this be seen?

page 21, lines 1/2: What about Celestun? According to Table 1, this is in the tropics, too.

page 21, line 11: What do you mean with “meteo-climatic conditions”?

page 21, line 12. How close is the thermometer plant?

page 21, line 14: “ India is the third largest hard coal producer ...”: Concerning Hg emissions, it is more important how much is consumed.

page 21, line 23: “... it is necessary for India ...”: I think this necessary at other places in the world, too.

page 21, line 30/31:“SIS site was typically influenced by ...”: Again you claim something that the reader is not able to follow. This leaves the impression that you speculate or that it is published somewhere else and you do not give a reference.

page 22, line 11/12: Please explain how you analysed the influence of different air masses on the concentrations measured at NIK.

page 22, line 34: Here you refer to the year 2012 while all other stations were only evaluated for 2013/2014.

page 22, line 35, page 23, line 1: Again, the reader gets the impression that you analysed the meteorological conditions at the different stations but you do not explain how you did this.

page 23, line 18/19: Over which years has the inter hemispherical gradient been constant? You analyse only two year in this paper. Or do you refer to other investigations?

page 23, line 20: You mention cruises but there is no data from cruises presented here.

Technical corrections:

affiliation 14: Sweden

Write all units correctly, e. g. on page 2, line 24: convert ng m^{-3} into ng m^{-3} . This appears at several places in the entire document.

Improve the way citations are shown, by removing inner brackets when two or more references are given, e.g. page 2, line 23: (Lindberg et al., 2007; Sprovieri et al., 2010b). This appears at several places in the entire document.

Explain TGM earlier in the document. It appears first on page 3, line 32 but is explained later.

Take care about words starting with capitals. E.g. on page 4, line 2, I propose to write “GMOS external partners”. There are several other places where the use of capitals should be re-considered.

Explain PBM2.5 on page 4.

Give more explanations about what is displayed in Table 1 and Table 2 in the respective captions.

The graphs in Fig. 1 and Fig. 2 are too small and therefore hard to read.

page 11, line 4: “latitude” instead of “longitude”.

Fig. 4 and Fig. 5: If one has a title the other should also have one. They could be a bit smaller or combined into one figure with two panels. It is not clear what “raw data” in the caption of Figure 4 really means.

Table 5 looks misplaced, it is first mentioned 5 pages later. The number 0.0.365 is misspelled.

Figure and table captions should end with a full stop.

Abstract, page 1, line 2: understand

page 2, line 30: Wängberg

page 3, line 14: and a QA/QC ...

page 4, line 15: ... at the French site Dumont d'Urville.

page 4, line 29: ... within the GMOS network.

page 6, line 4: This is also in line with a study recently published by Slemr et al. (2015)

...

page 6, line 8: located at the coastline

page 9, line 20: raw data

page 13, Table 5: 0.365. The table is misplaced because it is mentioned much later in the text (page 16).

page 15, line 12/13: misplaced brackets for the references.

page 15, line 15: Which station is STN?

page 17, line 3/4: ... the three distributions come from the same ...

page 17, line 7: Table 5 is on page 13, which is not close enough to the page where it's mentioned.

page 17, line 23: ... during the years 2013 ...

page 17, line 24: I think this has to be GOM (not GEM).

page 18, line 7: ... at the edge of the north-eastern part ...

page 18, line 9: Hg sources

page 20, line 17: in the same period

page 20, line 30: in the Tropics

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