"Multi-year record of atmospheric mercury at Dumont d'Urville, East Antarctic coast: continental outflow and oceanic influences" by H. Angot et al.

Response to referee comments by Referee #2.

We provide below a point-by-point reply to the comments (points raised by the referee in bold, changes made in the manuscript in red).

1. General comments

This manuscript describes novel measurements of Hg(0) and surface snow samples from a coastal Antarctic location. It is not the first paper presenting a multi-year data from on atmospheric mercury from Antarctica as the other anonymous reviewer states; however, it is the first dataset from the east Antarctic coast and reveals a different annual pattern of atmospheric mercury as compared to previously published Hg measurement from other Antarctic coastal stations. This difference makes the manuscript interesting and a valuable addition to Antarctic atmospheric mercury measurements. I agree with the other reviewer that the manuscript is well written and presented, and is of high quality. The discussion of results, and theories presented are supported by the measurements. I recommend publication of this manuscript after addressing some minor issues as outlined below.

2. Specific comments

- Line 92: Estimated MDL sound very unscientific. I would suggest changing it to "According to the instrument manual, MDL is…" The reference Tekran, 2001 is missing in the reference list.

We agree. This has been changed in the revised manuscript: "According to the instrument manual, the detection limit is 0.10 ng m^{-3}"

The reference Tekran 2011 is in the reference list:

Tekran: Tekran 2537 mercury monitor detection limit. Summary of known estimates, Tekran Instruments Corp., Toronto, ON, Canada., 2011.

- Line 102: You state using internal standards as a part of the QA/QC of the Hg-tot measurements with the Tek2600. This is to my knowledge not common and has to be explained. Or do you mean external standards?

These questions have been addressed in the revised manuscript: "The instrument was calibrated with the NIST SRM-3133 mercury standard. Quality assurance and quality control included the analysis of analytical blanks, replicates, and internal standards (Reference Waters for mercury: HG102-2 at 22 ng/L from Environment Canada)."

- Line 105-110: I do not understand the logic of this paragraph, in particular the last sentence. You refer to surface snow samples collected at McMurdo as being representative for your surface snow samples collected very different locations and even year. This has to be explained a little more in detail. See also my comment on surface snow further down.

This paragraph aims at discussing the (in)homogeneity of surface snow samples. We do not say that surface snow samples collected at McMurdo are representative for our surface snow samples collected during the traverse between Concordia station and Dumont d'Urville in 2009. We, however, highlight the fact that surface snow samples collected at McMurdo at various locations and on different days give very similar results. This indicates that the spatial and temporal representativeness of surface snow samples collected in Antarctica can be fairly good. This experiment carried out at McMurdo is the only one, to the best of our knowledge, dealing with the representativeness of surface snow samples. This is the reason why we are referring to it.

This paragraph has been modified in the revised manuscript:

"Surface snow samples collected during traverses may have limited spatial and temporal representativeness given the variability of chemical species deposition onto the snow surface, and the occurrence of either fresh snowfall or blowing snow. The (in)homogeneity of surface snow samples was investigated at MM by Brooks et al. (2008). ~~However, the daily Hg_{tot} concentration of surface (upper 3-5 cm) snow samples collected at different snow patches at MM averaged 67 ± 21 ng L (n=14) (Brooks et al., 2008), indicating that the spatial and temporal representativeness of surface snow samples can be satisfactory.~~ Surface (3-5 cm) snow samples were collected daily (n = 14) at different snow patches. Hg_{tot} concentrations averaged 67 ± 21 ng L^{-1}. This result indicates that the spatial and temporal representativeness of surface snow samples collected in Antarctica can be satisfactory and gives us confidence that spatial differences in Hg_{tot} concentrations reported in section 3.2.2 are not due to samples inhomogeneity."

- Line 190-193: The fact that NM, HA and TR are not significantly impacted by air from the Antarctic plateau does not explain why the Hg concentrations at NM and TR are stable in winter. This statement should be changed.

This has been changed in the revised manuscript:

"Conversely, several studies showed that stations such as NM and HA are not significantly impacted by air masses originating from the Antarctic plateau (Helmig et al., 2007; Legrand et al., 2016b), consistently explaining why Hg(0) concentrations ~~remained rather stable~~ did not decrease at NM and TR throughout winter (Ebinghaus et al., 2002; Pfaffhuber et al., 2012)".

- Section 3.1.2: The section deals with AMDEs and that they are not frequently occurring at DDU caused by weak Br-chemistry at DDU. This is a probable cause however, what about the katabatic winds? Are they also dominant in spring? If so I would assume they to some extent can explain the lack of AMDE-observations at DDU.

Yes, you are right. This has been added in Section 3.1.2:

"Additionally, air masses originating from the Antarctic plateau prevailed (62 ± 23 %, Fig. 4) in spring at DDU according to the HYSPLIT model simulations. This can also explain, to some extent, the lack of AMDE-observations at DDU".

- Line 272-277: You explain about the summertime diurnal cycle of Hg(0) as emissions from snow covered soil. How thick is the snowpack in summer at DDU? Figure 7 and 9

show that the diurnal cycle is quite large, so I am just wondering how it can be possible that the penguin excreta on the soil is the source of this diurnal variation. This means that Hg emitted from the soil has to penetrate the snowpack. What about other possibilities, have you considered meteorological phenomena such as boundary layer height? The penguin excreta explanation is funny but I do not see it as a plausible cause.

As mentioned in the conclusion, several processes may contribute to this diurnal cycle, including a local chemical exchange at the air/snow interface in the presence of elevated levels of Hg(II) species in ambient air, and emissions from ornithogenic soils present at the site. We do not believe that penguin excreta can be held responsible alone of the diurnal cycle. However we cannot rule out that penguin excreta do have an influence on the Hg(0) concentrations in summer.

- Section 3.2.2, in particular the paragraphs dealing with surface snow samples. Figure 10 shows a quite nice snow concentration gradient between the two stations. However, I think you should mention that the snow samples are from 2009 and the atmospheric measurements between 2012 and 2015. Papers about surface snow in the Arctic have repeatedly shown how inhomogeneous the snow is and that the deposition of Hg onto surface snow quickly is being re-emitted to the atmosphere. Why should the Antarctic be any different? You state Cl in the snow complexes HgII and prevents re-emission, did you do any anion analysis on your snow samples to back up your Hg snow measurements? Your snow samples are very much higher than the transect studies you compare with, even your non-coastal snow samples. Any thought on why you observe such big differences? Your coastal snow samples are compared to Brooks et al 2008 from MM, and yours are also higher than these. Any thoughts on why?

- The fact that snow samples are from 2009 is mentioned in section 2.2.2. However, this has been added in section 3.2.2 of the revised manuscript:

"The Hg_{tot} concentration of snow samples collected in summer 2009 between DC and DDU".

- The comment regarding snow samples inhomogeneity has been addressed above (see comment Line 105-110).

- We did not do any anion analysis on the surface snow samples. However, Legrand et al. (2016a) reported a large gradient of sea-salt concentrations in bulk aerosols between Dumont d'Urville and Concordia station.

- As mentioned in the manuscript, our non-coastal snow samples give results in good agreement with Hg_{tot} concentrations reported by Angot et al. (2016) at Concordia station in summer. The fact that our concentrations are higher than concentrations reported by Brooks et al. (2008) at McMurdo might be due to the advection of inland air masses enriched in Hg(II) species. As explained in the manuscript, Dumont d'Urville is most of the time influenced by inland air masses (enriched in Hg(II) species) due to strong katabatic winds.

- Line 369-371: You state that during summers of Hg measurements there were a significant unusual amount of sea ice. Was it more or less than normal?

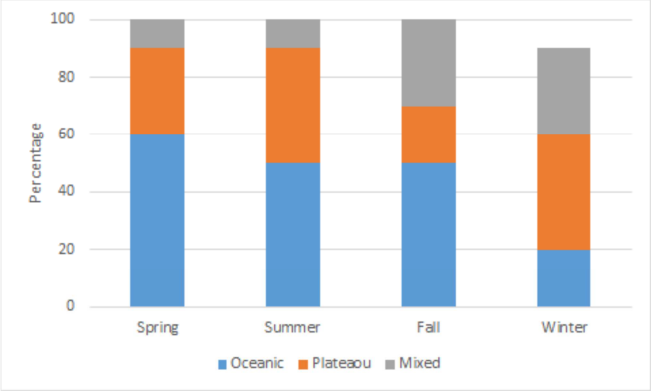
Indeed. This has been clarified in the revised manuscript:

“It should be noted that during summers 2011/2012, 2012/2013, and 2013/2014, areas of open waters were observed but with a significant unusual large amount of sea ice”.

- Line 388-391: This paragraph seems a bit out of context as it is located here. Should it go with sections 3.2.1 dealing with the diurnal cycle of Hg(0)? Is it even expected that oceanic emissions are follow a daily cycle?

We agree that this paragraph is a bit out of context here. It has been removed in the revised manuscript.

- Figure 4: This figure is an attempt to visualize the air mass origin, and I find this figure very busy and it is difficult to get any useful information. Have you considered plotting the percentages as bars instead, such as the exemplified figure below?



Thank you for this suggestion. This figure has been changed in the revised manuscript:

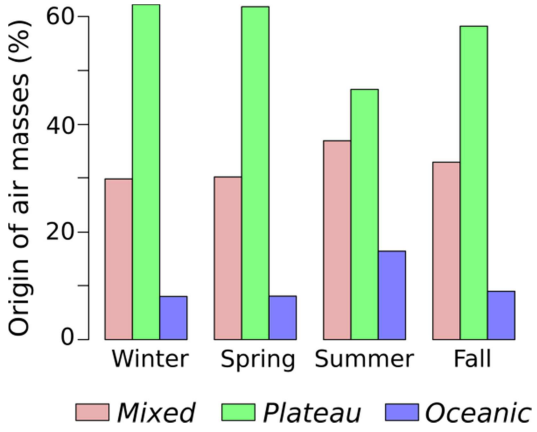


Figure 4: Mean percentage (%) of continental/oceanic mixed air masses (pink), and of air masses originating from the Antarctic plateau (green) or the ocean (blue) according to the HYSPLIT model simulations in winter (May to August), spring (September-October), summer (November to February), and fall (March-April).

3. Technical corrections

- Line 32: Consider replacing “since” with “and”.

Done.

- Line 38: This is a very oddly constructed sentence, not grammatically wrong, just odd. Consider revising it.
This sentence has been changed in the revised manuscript: "Hg(0) is the most abundant form of mercury ~~– a toxic element –~~ in the atmosphere."

- Line 143: Consider removing "a" from ": : : impacted by a local pollution"

Done.

References

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