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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Annual cycle of Antarctic baseline aerosol: controlled by photooxidation-limited aerosol formation" *by* M. Fiebig et al.

M. Fiebig et al.

markus.fiebig@nilu.no

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We thank Reviewer #1 for a very competent review, especially with respect to the very specific and helpful comments that did improve the manuscript. We respond to the comments in an itemized fashion for clarity:

1. "Focussing on the most crucial discovery i.e. the predominance of photochemistry over source strength, there are some issues which I feel deserve some more discussion. First, the assumption that source strength (e.g. sea-air exchange of precursor gases) is of negligible

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importance for the baseline aerosol budget in Antarctica relies largely on FLEXPART results. These model calculations demonstrate that air masses (20-days backward plumes) virtually never resided within the lowest 100 m above sea surface. The case in point is that apart from inherent uncertainties of dispersion model calculation itself, the marine boundary layer is typically well mixed up to several hundred meters. Hence I am not really convinced of this reasoning. In this regard an additional FLEXPART analysis comparable to that shown in Fig. 5 but now showing 20-day backward plumes were particles reside above the boundary layer in the free troposphere / lower stratosphere (i.e. about 1500 m or so above ground) would be advisable to support this chain of argumentation."

We took this comment of Reviewer #1 rather seriously and revised Figure 5 presenting the concerned FLEXPART analysis completely. The Figure now uses the same criterion for distinguishing baseline from non-baseline air as the remainder of the article. Furthermore, the footprint maps of the FLEXPART backward plume calculations for Troll station now cover the whole Southern hemisphere, and 2 panels contrast the footprint maps (relative to the mean footprint) of non-baseline and baseline cases. Whereas the non-baseline footprint covers the whole Southern Ocean, but seems to exclude the Central Antarctic continent, the baseline footprint is concentrated on Central Antarctica (where the baseline air descends) and the Souther Ocean mid-latitudes and the ITCZ (where the the baseline air ascends). The present version of Figure 5 thus supports the point made in the article about the origin of the Antarctic baseline air much better than the previous version. The discussion of Figure 5 in section 5 was adapted accordingly.

We didn't adapt the definition of the footprint to include all altitudes where the marine boundary layer can be located as proposed by the reviewer. The boundary layer magnitude is rather variable, and including the whole range where it

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may reside will necessarily include free troposheric air, which would make the analysis less significant. Since the marine boundary layer is normally well-mixed, including only the lowermost model layer in the footprint will represent all cases where the backward plume enters the marine boundary layer.

2. "A second point concerns the fact that the results are restricted to baseline aerosol data. In order to judge the relevance of the main conclusion (photooxidation control) for the aerosol budget of Antarctica on the whole, the percentage of baseline aerosol (particle number and volume concentration) related to the total budget should be assessed."

The main conclusion that aerosol production is controlled by photooxidation concerns only and is limited to Antarctic baseline air. Quantifying the contribution of this process to the total Antarctic aerosol budget would go beyond the scope of this article, especially since this assessment would be highly dependent on location and distance from the Antarctic shoreline.

3. "Page 23065, line 21: Why using here a 20th percentile and not the 2.5 times of the running 5th percentile introduced in chapter 2.2?"

This question was rather warranted. The previous version of Figure 5 was based on a very early version of the analysis, and has now been replaced by a version that uses the same criterion for baseline air as defined in section 2.2 (see above).

4. "Page 23066, line 15-23, impact of NOx emissions: To my knowledge NOx emission from the snow surface leads to ozone production (and not destruction), at least on the Antarctic plateau (see e.g. Crawford et al.: Evidence for photochemical production of ozone at South Pole surface, Geophys. Res. Lett., 28, 3641-3644, 2001)."

Thanks to the reviewer for pointing this out. The annual cycle of ozone in low-NO_x environments such as Central Antarctica is due to photochemical production of

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hydrogen peroxide in summer, with subsequent destruction of ozone (Ayers et al., 1992). The text has been changed accordingly, and the quotation changed from Helmig et al. (2007) to Ayers et al. (1992).

5. "Page 23077,the correct reference is: Yang, M. Y. M., Vay, S.A., Stohl, A., Choi, Y., Diskin, G. S., Sachse, G. W., and Blake D. R: Chemical composition of tropospheric air masses encountered during high altitude flights (11.5 km) during the 2009 fall Operation Ice Bridge field campaign, J. Geophys. Res., 117, D17306,doi:10.1029/2012JD017858, 2012."

   Corrected.

6. "Page 23066, line 4: cyanobacteria"

   Corrected.

7. "Page 23070, line 24: boundary"

   Corrected.

References

Ayers, G. P., Penkett, S. A., Gillett, R. W., Bandy, B., Galbally, I. E., Meyer, C. P., Elsworth, C. M., Bentley, S. T., Forgan, B. W., 1992. Evidence for photochemical control of ozone concentrations in unpolluted marine air. nature 360, 446 – 449. http://www.nature.com/nature/journal/v360/n6403/abs/360446a0.html

Helmig, D., Oltmans, S. J., Carlson, D., Lamarque, J.-F., Jones, A., Labuschagne, C., Anlauf, K., Hayden, K., 2007. A review of surface ozone in the polar regions. Atmos. Env. 41 (24), 5138 – 5161.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 23057, 2013.