Comments on Legrand et al. ‘Year-round records of bulk and size-segregated aerosol composition in central Antarctica (Concordia site) Part 1: Fractionation of sea-salt particles’

General comments:

This manuscript presents a detail examination of chemical compounds, mainly depletion of sulfate and chlorine relative to sodium with respect to freshly emitted sea salt aerosol, in aerosols that arrived at Concordia in central Antarctica, as well as acidic gases (HCl and HNO_3) in air based on their multiple year-round records of samplings. Both bulk and size-segregated depletion of chloride and sulfate are reported with a major goal of determining the origin of sea salt aerosols reaching the high plateau. To achieve that, the authors have introduced a novel method to remove biogenic sulfate contamination to the sulphates containing in the aerosols. Their approach has been successfully applied to the raw dataset and the conclusion derived indicating rough similar contribution of sea salt aerosols from both sea-ice and open ocean emission during polar winter is well consistent with previous model findings. Together with a back-trajectory result, they further confirm that a larger sulfate to sodium ratio would be observed when air masses had travelled a longer time over sea ice than over open water. Moreover, their novel data seem to show that small sub-micron sized SSA has much larger sulfate depletion that larger ones which is likely to indicate different sized SSA could face quite different preferential production mechanisms as predicted previously.

The second major part of the manuscript is about chlorine depletion in aerosols. Their multi-year data clearly show a maximum depletion occurring in spring which is associating with high level of sea salt amount. The interpretation of the spring peak in chloride depletion has been discussed carefully, in associating with concentrations of nitrate, acidic sulphur in aerosols, as well as nitric acid and HCl in gas phase.

In general, this is a novel and deepening study of aerosol depletions for chlorine and sulphate at a central Antarctica site. It is well written and with huge scientific implications for our understanding of the sea salt ice records taken from inland Antarctica. It deserves a publication in ACP with a minor revision (see my specific comments shown below).

Specific comments:

P12: In terms of the chlorine depletion mechanisms, only two controlling factors (e.g. nitrate and sulphate) are highlighted in the manuscript. Given the fact that the largest chlorine depletion occurs in spring and is associating with highest SSA load, a season seeing the highest BrO in coastal sites of the Antarctic, then the manuscript would benefit if the authors could discuss the potential role of halogen, here mainly bromine in influencing cholrine release from SSA. Particularly, chloride can be liberated via heterogeneous reaction through $HOBr(g)+Cl^- \rightarrow BrCl$ (e.g. see Abbatt et al., 2012). Though at inland sites like Concordia, this process is not important due to the relatively low inorganic bromine species (a few pptv, see their paper of Legrand et al., 2016), however, at coastal sites during bromine explosion events in spring, the boundary layer HOBr concentration could be as high as (or even higher than) HNO_3. Apart from this process, $BrONO_2$ hydrolysis reaction (via heterogeneous reaction on wet surface) has been found dominating nitric acid formation in high latitude. For instance, a global model (pTOMCAT) with a detailed bromine chemistry included clearly showed that when BrONO2 hydrolysis is included, about 60-80% atmospheric NOx in high latitudes will be removed (Fig. 12c of Yang et al., 2005) indicating $BrONO_2$ hydrolysis could efficiently convert NOx to HNO_3 in mid- to high latitude (not in low latitude due to high OH and low BrO there). Does this process play a role, either directly or indirectly, in affecting chlorine depletion? Can bromine be ruled out completely as a role in HNO_3 formation and chlorine depletion? If not, then a discussion should be given.

P24, Figures 6/7: My second concern is again about the chlorine depletion in section 3.1. If the acidic replacement is indeed dominating chlorine release, them it could be possible to see a nearly 1:1 linear scatter relationship between chlorine depleted and $[H^+]$ derived. The $[H^+]$ can be derived from the two major acidic species (nitrate and sulphate without SS_SO_4). Equally, a scatter plot between the ratio of r and ratio of $[H^+]/[Na^+]$ is also indicative to whether the replacement process is working. This examination can be made both for seasonal depletion data shown in figure 7 as well as for size-dependent depletion shown in figure 6. Instead of using $[H^+]$, a sum of $[NO_3]/M_{NO3}+2X[SO_4]/M_{SO4}$ can also be used in the scatter plot. here M_{NO3} and M_{SO4} are molecular weight of NO_3 and SO_4 respectively.

Technical corrections:

P2L32: add ‘mass’ between ‘sodium’ and ‘ratios’

P8L10: remove ‘to’ before ‘due to’

P8L20: ‘Fig.6’ should be ‘Fig. 7’ ?

P8L18: ‘larger’ here should be ‘smaller’?

P9L19: remove the extra ‘and’ in ‘or/and and too high’

P12L3. A reference is needed to ‘high dust levels that neutralized HNO_3 and H_2SO_4’ .

P12L29: change ‘part’ to ‘parts’

In most of figures, change NO3 to NO_3^-, SO4 to $SO_4^=$, Na to Na^+.

P22 figure 4: it would be good to add ‘r’ line to high light the ‘relative depletion factor.

P24 figure 6. Please add a line of total sulphate derived based on MAS (without SS_SO4).

P25 figure 7b: is the number ‘0.64’ in ‘(0.64-3.5 µm)’ actually 0.46? why there is a gap between 0.46 to 0.64 µm ? same question in P6 L25.

P28: what do the two dash lines represent for?

References:

Abbatt, J. P. D., J. L. Thomas, K. Abrahamsson, C. Boxe, A. Granfors, A. E. Jones, M. D. King, A. Saiz-Lopez, P. B. Shepson, J. Sodeau, D. W. Toohey, C. Toubin, R. von Glasow, S. N. Wren, and X.

Yang, Halogen activation via interactions with environmental ice and snow, Atmos. Chem. Phys., 12, 6237-6271, [doi:10.5194/acp-12-6237-2012] 2012.

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Yang, X.; Cox, RA; Warwick, N. J.; Pyle, J. A.; Carver, Glenn D.; O'Connor, FM; Savage, NH; Tropospheric bromine chemistry and its impacts on ozone: A model study. *Journal Of Geophysical Research*, 110 (D23311), doi:10.1029/2005jd006244, 2005.