

Interactive comment on “Year-round records of bulk and size-segregated aerosol composition in central Antarctica (Concordia site) Part 1: Fractionation of sea-salt particles” by Michel Legrand et al.

Anonymous Referee #3

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The manuscript presents multi-year measurement results of sea salt aerosols and associated ion composition as well as HCl and HNO₃ acid gases in the central Antarctic site. With these data, the authors examine the chloride depletion relative to sodium with respect to freshly emitted sea salt aerosols and the sulfate depletion relative to sodium with respect to the composition of sea water. The seasonal variability of such depletion, the role of acidic sulfur aerosol and nitric acid in the depletion, and the contribution sea-ice and open ocean emissions to the sea salt aerosols load are investigated. The reported data are valuable and such kind of study should be a welcome addition

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to the literature on Antarctic environmental and atmospheric chemistry research. On the other hand, the manuscript appears to be not well written and some discussions are ambiguous without a clear clue to follow. Especially there is a lackness of in-depth analysis based on the fundamental chemistry. In my opinion the quality of this manuscript is not high enough to be published in ACP for its current version. Below are my comments in detail.

What does the authors mean in the term of “acidic sulfur aerosol” (e.g., in P1, L22)? The authors also define nitrate as “acidic compounds” (P6, L26-28). Do they refer to HNO₃ in aerosols? Note that SO₄⁼ and NO₃⁻ should be taken as neutral, instead of acidic, ions in water solution. They calculated the acidic sulfur component as the sum of non-sea-salt sulfate plus MSA after subtracting the amount of ammonium (P6, L27-28). What does the residual mean in acid-base equilibrium chemistry? The authors should have investigated the balance status between cations and anions in measured aerosols before analyzing the chlorine depletion by so called replacement reactions. Figure 7 and related discussions (P7, L10-17) provide some information. However, only when the full set of ions have been taken into account and, if possible, neural compounds (e.g., CaSO₄, Na₂SO₄, (NH₄)₂SO₄, and NaNO₃) diagnosed, the displacement process could be understood clearly. Note that there is a neutralization order for the ions in a solution, e.g., SO₄⁼ > NO₃⁻ for anions and Ca²⁺ > Na⁺ for cations. From Figure 7c, one cannot see whether the displacement had happened or not. Only by investigating NO₃⁻, Na⁺ and other associated ions in the aerosols, the role of HNO₃ in the displacement might be seen. Note that partitioning of HNO₃ between gaseous and aerosol phases depends not only on the acidity of liquid aerosols but also atmospheric temperature.

For Section 3.2 of the manuscript, neither the terms of SO₄, biogenic sulfate, ssSO₄, nssSO₄ and MSA and are well defined, nor their measuring (or calculating) methods clearly introduced. It is difficult for me to follow the discussions as the assumption (e.g., the relationship between biogenic sulfate and MSA in P8, L22-25) has not been

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fully based on fundamental atmospheric chemistry and physics. There is a doubt that the regression results from small particles can be applied to large ones. The authors refer to a companion paper: “Legrand, M., Preunkert, S., Weller, R., Zipf, L., Elsässer, C., Merchel, S., Rugel, G., and Wagenbach, D: Year-round (2006-2015) record of bulk and size-segregated aerosol composition in central Antarctica (Concordia site) Part 1: Sulfur derived aerosol (MSA and sulfate), this issue” (note that both companion papers are labelled as “Part 1”). Considering that sulfate aerosol is a large and important part of this manuscript, I would suggest the paper not to be separated into two parts. Sulfate depletion relative to sodium has been attributed to sea-ice related emissions due to precipitation of mirabilite ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) during freezing of seawater. As both sulfate and sodium will be lost with the deposition of mirabilite, how the depletion of sulfate relative to sodium occurs should be explained quantitatively in detail.

The manuscript needs to be concise and formulas (1), (3), (4), (6), and (7) and associated descriptions can be moved to the Supplement. The authors use the p-TOMCAT model to confirm their conclusion on the source of sea salt aerosols (P10, L11-23). But the model and its simulation results have not been well evaluated and introduced in this manuscript. These discussions (P10, L11-23) provide no more convincing information than the backward trajectory analysis described in the previous paragraph. I do not think that Sect 3.3 should be included in this manuscript as it appears to provide no strong support to explain the observational results of this study.

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