Answer to Referee #3

We would like to first thank the reviewer for their comments which have emphasised to us that we needed to explain better some underlying concepts. We hope these additional explanations will answer most of the comments

The manuscript presents multi-year measurement results of seal 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 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.

We note that, in the quick report, our paper was evaluated as "good" for the aspect "scientific quality" and "data presentation".

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) diagnosted, the dis-placement 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 be tween gaseous and aerosol phases depends not only on the acidity of liquid aerosols but also atmospheric temperature.

OK we understand that we made an assumption that readers are familiar with polar atmospheric chemistry and central Antarctic ice core chemistry. So before presenting the calculations of acidic sulfur we remedy that by presenting, as recommended by the reviewer, the ionic balance of aerosol at Concordia. This should indicate very clearly that, apart from the sea salt components, the main cation associated with nitrate and sulfate is H+ :

"An examination of the role over time of chemical species possibly involved in aerosol de-chlorination is reported in Fig. 7, the amount of chloride loss being compared on a molar basis to the main atmospheric acidic components. The mean ionic composition of aerosol collected in spring at Concordia is reported in Table 1. It shows that, apart from sea-salt components, aerosol present at Concordia in spring/summer consists of sulfate (1.2 nEq m^{-3}), nitrate (0.08 nEq m^{-3}), MSA (0.05 nEq m^{-3}), and ammonium (0.18 nEq m^{-3}). Other non-

sea-salt components (nssK$^+$, nssCa^{2+}, and oxalate) remain at the level of 0.01 nEq m^{-3} or less (Table 1). From that, we have calculated the acidic sulfur component as the sum of non-sea-salt sulfate plus MSA after subtracting the amount of ammonium."

Table 1. Mean chemical composition of aerosol collected at Concordia from October to December (2006-2015). Values in bold and in parenthesis refer to the non-sea-salt components of aerosol. All concentrations are expressed in nEq m^{-3}.

Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	$CH_3SO_3^-$	$C_2O_4^{2-}$
0.47	0.18 **(0.18)**	0.02 **(0.01)**	0 .11	0.02 **(<0.01)**	0 .08	0.08 **(0.08)**	1.25 **(1.19)**	0.05 **(0.05)**	0.015 **(0.015)**

Concerning your remark that "From Figure 7c, one cannot see whether the displacement had happened or not.":
Indeed, because only a small fraction of total nitrate is on sea-salt particles and figure 7c shows the gas phase fraction which (independently of sea-salt) increases from spring to summer with the recovery of the photochemistry and NOx emissions from the snowpack.

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.

We think that the readers of Atmospheric Chemistry and Physics are already familiar with most of this terminology, and for example know that SO4 is sulphate. The method of calculation of nssSO4 was already defined on Page 2, line 16. The calculation of ssSO4 was already defined on Page 9, line 6. We think that the wording "biogenic sulphate" would be understood by the readers of ACP. We indeed missed spelling out MSA, and it is now done in the revised version in the introduction: "They also pointed out that, even when examining the chemical composition of particles deposited on the top stages of the impactor, there can be a significant underestimation of the degree of fractionation of sea-salt particles due to a residual presence of biogenic sulfate, as indicated by the presence of MSA (methanesulfonic acid). »
Adding this last sentence, we also clarify what is biogenic sulfate (see below).
Measurements are detailed in section 2, calculations are presented in this section. Please tell us if we need to do more but this seems sufficient for readers with any familiarity with the topic, and the justification for these calculations for those less familiar can easily be found in the cited references.

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 fully based one fundamental atmospheric chemistry and physics. There is a doubt that the regression results from small particles can be applied to large ones.

We do not understand the argument for such a statement that, in any case, is not supported by observations. This point was already discussed in the text and we have included another sentence (mentioning that this has been previously discussed and published by Jourdain et al., 2007): "For the 5 lowest stages (smallest particle sizes) of the impactor, we find that the ratio of nssSO$_4$/MSA is reasonably constant (red dots in Fig 8e (or Fig 9e) falling on a straight line through zero). This then supports the assumption that in winter the size distributions of biogenic sulfate and MSA are the same and that the nssSO$_4$/MSA ratio is constant over the entire size distribution (see also Legrand et al., this *issue). The similarity of the $nssSO_4/MSA$ ratio over the entire size distribution was already pointed out by Jourdain et al. (2007). «*

The authors refers 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").
Thanks for identifying this typographic error.

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.

This suggestion is unrealistic. The topics of the two papers are totally different: part one is dedicated to the origin of sea-salt and its degree of fractionation over Antarctica, part 2 focuses on the understanding of the behaviour of biogenic sulfate and MSA.
Part 1: is 17 pages of text (including references), 1 table, and 13 figures, Part 2: is 22 pages, 5 Tables, and 12 figures. Mixing the two papers (even considering the overlaps in section "sites, sampling and methods", and possible mixing 2 figures together), the resulting paper would be far too long and we feel this is not realistic.

Sulfate depletion relative to sodium has been attributed to sea-ice related emissions due to precipitation of mirabilite ($Na2SO4.10H2O$) during freezing of seawater. As both sulfate and sodium will loss with the deposition of mirabilite, how the depletion of sulfate relative to sodium occurs should be explained quantitatively in detail.

This aspect is introduced and well referenced in the introduction (lines 10 to 21), and it is now a well recognized process. It is obvious that because there is much more sodium than sulfate in sea salt, precipitation of mirabilite removes proportionally more sulfate, leading to a depletion; we do not feel this needs to be explained again.

The manuscript needs to be concise and formulas (1), (3), (4), (6), and (7) and associated descriptions can be moved to the Supplement.

Since theses calculations address two very different processes: equation 1 to 4 refer to calculations for summer (chloride depletion), whereas equations 5 to 7 to sulphate depletion in winter, they have to appear in the corresponding paragraph dedicated to summer and winter, respectively. Mixing them in a supplement would be more confusing, and we do not feel this would help the reader.

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 previous paragraph.

As discussed in these lines, these model simulations were evaluated against observations in the cited previous papers. We feel that these previous works have to be reported here since the present paper clearly assesses (on a far more robust basis) previous observations. The advantage of the p-TOMCAT simulations compared to back trajectories is that they quantitatively assess the source of sea salt aerosol while the back trajectories merely assess the source of air masses.

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.

We disagree. The ice core data can, in no way, here support the atmospheric observations discussed in this study simply because, as clearly mentioned, post depositional effects modified the original atmospheric signal. On the contrary, as clearly introduced in the paper, the atmospheric studies described in this paper are needed to better understand ice core signals. The reviewer 1 clearly highlighted this point : "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."

Also the title of this paragraph is very explicit: "Implications for ice core studies" and not the reverse. From our experience some previous papers also ended with a discussion on "implications for ice core": see for instance the JGR paper "Preunkert, S., Jourdain, B., Legrand, M., Udisti, R., Becagli, S., and Cerri, O.: Seasonality of sulfur species (dimethyl sulfide, sulfate, and methanesulfonate) in Antarctica: Inland versus coastal regions, J. Geophys. Res., 113, D15302, doi:10.1029/2008JD009937, 2008.