Anonymous Referee #4

The authors address a pressing topic; sea salt, its size distribution in aerosols and co-located gas phase measurements over annual cycles. The interpretation of sea salt records in Antarctica is relevant to sea ice distribution patterns recorded in glacial records. This manuscript contributes significantly to this discussion, demonstrating that approximately half the sea salt aerosol load comes from open ocean versus sea ice. The relationship of sea salt, aerosol acidification and the production of HCl and HNO3 is also examined by considering denuder measurements of acid gases and their effect on aerosol characteristics. The authors argued that HCl emission from surface snow or differences in the lifetime of HCl versus sea salt transported from the coast drove differences observed between denuder and aerosol measurements. These results are novel and intriguing. This paper provides constraints to some long-standing questions regarding the source of anomalies in the ratio of Na to Cl in snow, whether and by how much Cl in aerosols is depleted in the continental Antarctic and in ice and the factors driving it. These points are all strong and convincing. Arguments about sea salt and biogenic sulfate however, are less well-rounded: likely this is because detail is provided in a separate manuscript dealing with sulfate and sulfate to sodium ratios. The manuscript is strong, but some sections, particularly with respect to sulfate and sulfate to sodium ratios need additional attention prior to publication.

Portions of this manuscript associated with interpretation of sulfate needs additional description (that perhaps can be found in the accompanying paper on sulfur-derived aerosol) to be fully understandable. Page 8 line 24 refers to quantifying biogenic sulfate using MSA and the relationship between MSA and sulfate on particles with a diameter <0.46 microns. This assumes that all non sea salt sulfate in aerosols <0.46 microns in diameter is biogenic. Is this because anthropogenic sulfate is negligible and volcanic sulfate is absent in the Antarctic interior? A previous study in the Arctic demonstrated that MSA in for aerosols <0.49 microns correlated with the presence of fine particles rather than with biogenic sulfate (Rempillo et al., 2010). In that study the surface of <0.49 micron aerosol, rather than biogenic sulfate, were the dominant factor affecting MSA formation. The authors need to demonstrate why anthropogenic and volcanic sulfate are not factors in MSA formation at this continental Antarctic site. Line 23 on page 8 ".. we therefore have corrected the concentrations of sulfate present on the upper stages of the impactor for the (nss rather than biogenic contribution)".

Thank you for this comment: Yes, at these high southern latitudes, anthropogenic and volcanic sources can be neglected with respect to DMS oxidation. Such a statement was drawn from model approaches (Gondwe et al., 2003), sulfur isotopic measurements (Patris et al., 2000) and examination of MSA to nssSO4 (Minikin et al., 1998). Furthermore, indeed this point is discussed in the accompanying paper. It is concluded that, even for the low winter levels of nssSO4 (6 ng m^{-3}), its presence is mainly attributed to long-range transport of marine biogenic emissions from mid-latitude, the non-biogenic sulfate level remaining limited to 1 ng m^{-3}. But we have considered your comment and in section 3.2., we first refer to sea-salt sulphate and non-sea-salt sulphate modes, then we refer to the accompanying paper arguing that we can assume that non-sea-salt sulfate present in winter there mainly originates from marine biogenic emissions, and finally use the wording "biogenic sulfate".

Gondwe, M., Krol, M., Gieskes, W., Klaassen, W., and de Baar, H.: The contribution of ocean-leaving DMS to the global atmospheric burdens of DMS, MSA, SO_2, and NSS SO_4, Global Biogeochem. Cycles, 17(2), 1056, doi:10.1029/2002GB001937, 2003.

Minikin, A., Legrand, M., Hall, J., Wagenbach, D., Kleefeld, C., Wolff, E., Pasteur, E. C., and Ducroz, F.: Sulfur-containing spacies (sulfate and methanesulfonate) in coastal Antarctic aerosol and precipitation, J. Geophys. Res., 103, 10,975-10,990, 1998.

Patris, N., Delmas, R. J., and Jouzel, J.: Isotopic signatures of sulfur in shallow Antarctic ice cores, J. Geophys. Res., 105(D6), 7071-7078, doi:10.1029/1999JD900974.

On page 10 how were the values of 40% and 60% of travel time within the boundary layer determined? This is not evident from the examples of air mass back trajectories provided in Figure 11. Is there a reference or data to support a boundary layer of ~600 m for open ocean and over sea-ice in the Antarctic at this time of year? The discussion suggests a quantified analysis that is not shared with the reader which makes this section less convincing.

You are right, our wording concerning 600 m was confusing. Also Figure 11 only shows the location of the trajectories and not the altitude. The altitude of the trajectories is provided by trajectories calculations but not shown here. So we reworded the paragraph as follows:

"The temporal variability of sea-salt aerosol reaching Dome C, its level and composition with respect to the sulfate relative to sodium fractionation, were examined in the light of air masses reaching the site, as shown by calculated 10-day backward trajectories. The model was run every 6 h in backward mode for three different altitudes (0, 250, and 500 m above ground level, agl) for time periods corresponding to impactor run sampling. It is seen that when air masses arriving at Dome C have spent more than two days (up to 4 days) over oceanic sectors their mean sodium concentrations reach 47 ± 21 ng m^{-3} against 12 ± 5 ng m^{-3} when air masses have spent less than 0.5 days over oceans. Some typical examples of 10-day backward trajectories corresponding to strong, mid and weak sulfate relative to sodium fractionations are reported in Fig. 11. Backward air mass trajectory calculations also document the altitude of trajectories (not shown) and we calculated the time that air masses reaching Dome C were traveling below 600 m elevation over oceans, distinguishing between sea-ice and open ocean areas. The elevation of 600 m asl corresponds to the middle of the boundary layer whose the thickness over oceanic regions is typically 1000-1500 m. Selecting the impactor runs corresponding to sampling time periods over which the 10-day backward trajectories indicate more than one day of travel over the ocean, we find an overall decreasing trend of R (i.e. a stronger sulfate depletion relative to sodium) when air masses arriving at Dome C (0 m agl) have a longer contact with sea-ice than with open-ocean (R^2 = 0.4). For example, from 9 to 23 September 2006, air masses reaching Concordia had travelled more than two days over oceanic sectors, with 40% and 60% of travel time below 600 m elevation over sea-ice and open-ocean boundary, respectively. The corresponding R value is low (R = 0.1, Fig. 11a), consistently suggesting a very significant contribution of sea-ice-related processes to the sodium level. At the opposite, from 28 August to 3 September 2008 (Fig. 11d), air masses arriving at Dome C have only travelled 5% of time below 600 m elevation over sea-ice (95% over open-ocean). Intermediate situations, from 26 July to 9 August 2007 (Fig. 11b) or 28

August to 11 September 2009 (Fig. 11c) are characterized by R values close to 0.15 and 20-25 % of travel time below 600 m elevation over sea-ice (75-80% over open-ocean)."

Line 30 on page 10 is confusing "A slight increase of the sulfate depletion in particles smaller than higher than one micron. . .."

We agree: this sentence is confusing and add nothing with respect to the preceding sentence. So we removed it.