

Anonymous Referee #1

This manuscript discusses the sources and processes affecting sulfate and MSA concentrations, and the MSA to sulfate concentration ratio, in the continental Antarctica. This is a very important topic, since the MSA to sulfate ratio has been widely used in estimating biogenic vs anthropogenic sulfur source sources as well as in interpreting ice core data. The measurement data used in this paper appears to be of good quality and the analysis itself is, in most part, scientifically sound. I have a few minor issues to be considered before accepting this paper for publication.

We would like first to thank the reviewer for its helpful comments (see our detailed answers below). We identify 10 bulk sulfate and MSA erroneous values (in January/March 2009 and 2010). They were corrected and all figures and Tables accordingly. These erroneous values however did not change the overall results and conclusions.

Main comments:

End of section 3.2.2 and Figure 7: separating the data into those with sulfate concentration lower and larger than 100 ng/m³ seems very artificial. What was the bases for this specific border? Do the authors have any idea why the two subsets showed much higher correlations than all the data together?

We updated Figure 7 that now also shows the plots versus sulfate.

There is no strict border but as now explained in the text separating data with high and low sulphate permit to check the drop of R_{MSA} from November to December and from February/March to January. "Note that most of samples containing less than 100 ng m⁻³ of nssSO₄ correspond to the November-December period, whereas those containing more than 100 ng m⁻³ of nssSO₄ to the January-March period. In this way, the drop of R_{MSA} from November to December and from February/March to January is examined separately. In both cases, the R_{MSA} drop is mainly due to a decrease of MSA. »

The discussion in section 3.2.4 is highly speculative. Furthermore, the authors only mention the destruction of MSA in cloud water in this section, even though multi-phase reactions have also reported to be an important source of MSA as also mentioned in section 3.2.1. The subsection 3.2.4 should be partially rewritten.

Well, we stated at the end of Section 3.2.3 that "The fact that the drop of R_{MSA} values in mid-summer at Concordia is mainly due to the disappearance of MSA in the fine aerosol and not to an increase of sulfate, permits to reject the assumptions of (1) a selective deposition of MSA with respect to sulfate or (2) a preferential production of sulfate with respect to MSA during transport between the coast and the inland Antarctic plateau. As seen in Fig. 8, the only significant change that coincides fairly well the drop of R_{MSA} in mid-summer is the occurrence of the secondary maximum of ozone mixing ratio that is attributed to a local photochemical activity driven by NO_x emissions from the snowpack of the Antarctic plateau." It is therefore legitimate to question the occurrence of a chemical destruction of MSA as we did in this section 3.2.2.

But you are right we forget to mention that, in contrast to the fast heterogeneous production of MSA, under conditions encountered in the marine boundary layer its destruction is slow. But once formed, MSA can be subsequently destroyed over the Antarctic plateau given the high levels of oxidants there and the few days during which the air mass travels over this region. We updated the discussion here as follows: “The preceding observations of a drop of R_{MSA} driven by a decrease of MSA level in submicron particles around beginning of November and its recovery in February, simultaneous to the high photochemical activity at mid-summer at Concordia, suggests the occurrence of a (photo-)chemical destruction of MSA taking place in submicron particles at that time. Under conditions encountered in the marine atmosphere, in contrast to its fast heterogeneous production, a significant (but slow) in cloud destruction of MSA is suspected to take place (Von Glasow and Crutzen, 2004; Barnes et al., 2006; Hoffmann et al., 2016). After its production, MSA present in air masses travelling inland Antarctica will encounter more oxidative conditions, specially when air masses remained for a few days over the high plateau and is thus characterized by high ozone mixing ratio (see discussions in Legrand et al. (2016)).”

Page 10, lines 11-14: I have some difficulty in following the logic here. Please clarify, especially what you mean by overestimation here and what is the actual reason for it. **As we stated, it is due to possible impact of pollution from the city of La Paz.**

Page 12: lines 5-13: The linear fits given in Table 4 should also be presented in Figure 11. **OK, we now report the regression lines in Figure 11 (corresponding to calculations reported in Table 4), as also required by the other reviewer.**

Besides correlations, how significant were the derived relations? **The standard error of the estimates of the slope and the y-intercept are presented in Table 4.**

A statement such as "the linear relationship . . . even less linear. . ." is meaningless/incorrect in a statistical sense and should be modified. **We agree and the text was modified as follows: “In fall, the relationship between $nssSO_4$ and MSA becomes even less linear than in spring.....”**

Minor issues:

Page 3, line 12: I do not think that this is an acceptable way of citing other ACP papers (Legrand et al. this issue). **Yes, it is, since it is a companion paper.**

Page 5, line 15: weakness in abundance? Strange wording. **Ok we reworded this sentence as “We then focus discussions on the striking drop of the MSA to $NssSO_4$ ratio observed during mid-summer over inland Antarctica (Sect. 3.2).”**

section 3.2.2: should it be supermicron particles rather than micron particles? Page 8, line 8: . . .the higher. . .the higher. **OK Done.**

Please check out the grammar Page 9, line 4: des-appearance. Please check out the wording. **OK we corrected this.**