

## Anonymous Referee #2

GENERAL : The paper presents measurements, results and analyses of sulfur aerosols at the Concordia station. The work is carefully done, it is a valuable paper for the interpretation of Antarctic aerosols and ice cores. As a highlight I would mention the interesting result of the interpretation of the MSA/nssSO<sub>4</sub> and the photochemical destruction of MSA in summer. I can recommend publishing the paper in ACP, I only have minor revision suggestions.

*We first would like 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.*

### DETAILED COMMENTS

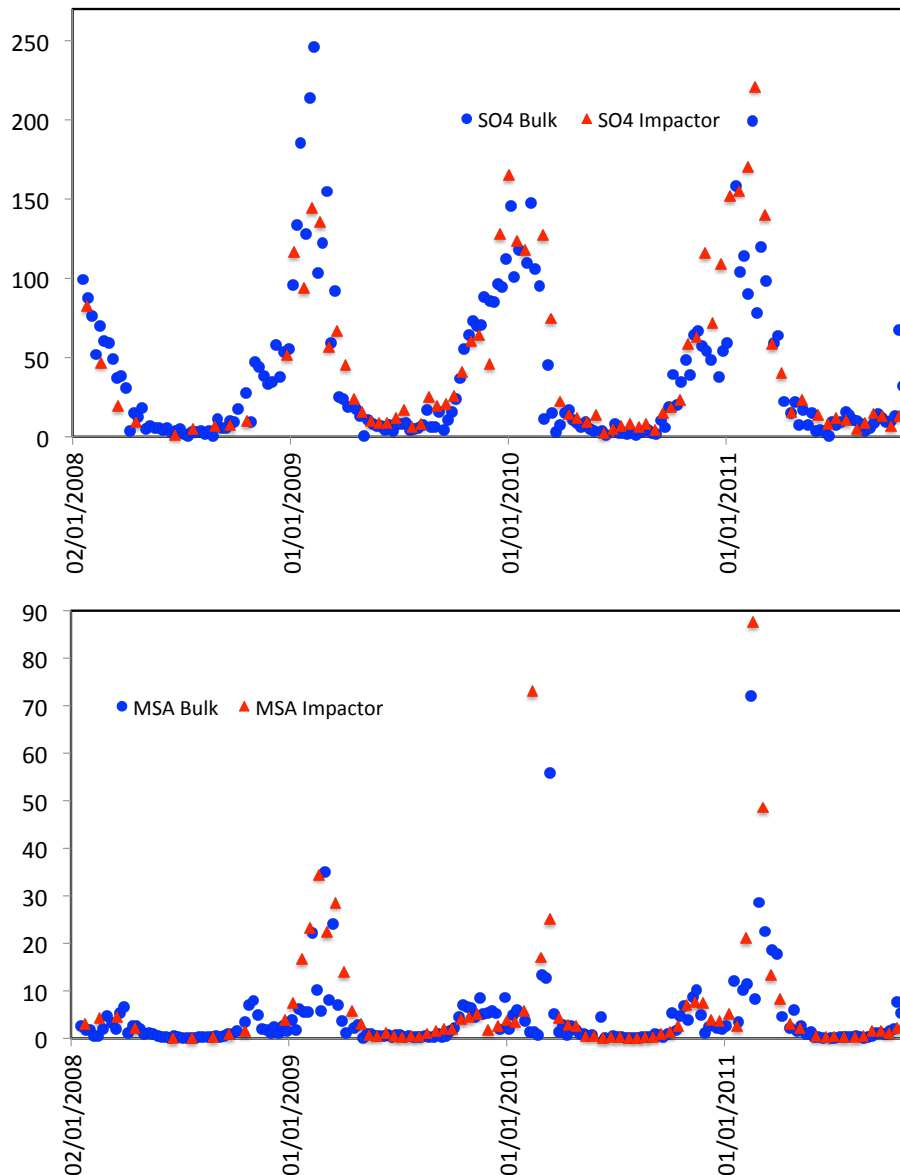
The time series is fairly long – are there any statistically significant trends? Yes or no, it would be potentially important. **Yes we now addressed this point as follows ” The aerosol record at Concordia now covers a decade. The long-term sulfate and MSA trends were examined by calculating the regression line slopes through annual and monthly mean values. No significant trend can be observed. For instance, a very weak annual increasing rate of  $1.9 \pm 5.6 \text{ ng m}^{-3} \text{ yr}^{-1}$  is calculated for sulfate in summer, however the regression line slope was found to be not statistically different from zero at the  $P > 95\%$  confidence level.”**

In the methods section: - sulfate might also come from the stations generator – could it? - was there any sector control? **Thank you for this comment and this point is now addressed in the text as follows: “The wind was occasionally blowing from the generator building of the Concordia station, disturbing measurements of atmospheric species like ozone (Legrand et al., 2016). Ozone measurements were also occasionally disturbed under very low wind speed conditions ( $< 2 \text{ m s}^{-1}$ ). The effect of such sporadic contamination of the station activities on the sulfate levels was here examined in the light of weekly denuder tubes sampling of acidic gases done at the site, as detailed by Legrand et al. (this issue) for HCl and HNO<sub>3</sub>. Indeed, the denuder tubes sampling of acidic gases conducted at Concordia also document SO<sub>2</sub> by measuring sulfate on the extracts. After subtraction of a mean blank of sulfate of  $1.5 \text{ ng m}^{-3}$  (i.e., 0.3 pptv of SO<sub>2</sub>), the average mixing of SO<sub>2</sub> collected from January 2013 to April 2016 (170 samples) is of  $0.7 \pm 0.6 \text{ pptv}$ . It is therefore unlikely that the station activities had emitted large enough amount of SO<sub>2</sub> to disturb the sulfate levels.”**

P4L8-11 "sulfate depletion relative to sodium with respect to the seawater composition .." there is the reference to the full paper but you could add a sentence or two as an explanation of the depletion here, too. **OK done as follows: “Examination of the size-segregated composition of aerosol present at Concordia indicates significant sulfate depletion relative to sodium with respect to the seawater composition from May to September (i.e. a  $k_{\text{SO}_4/\text{Na}}$  value of  $0.16 \pm 0.09$  instead of**

0.25 in seawater) (Legrand et al., this issue), resulting from the presence at the site of sea-salt aerosol emitted from both open ocean and sea-ice.”

Section 3.2.2 I am missing some comparison of HV and impactor data. I guess it has been done. A scatter plot with explanations would be nice. **Yes, we report the comparison in the figure below. We also add some sentences on the text: “Over the 2009 to 2011 years, aerosol was sampled on both bulk filter and impactor. A good agreement between the two data sets is found for sulfate as well as MSA (not shown). For sulfate, the relationship between the sum of concentrations observed on the impactor ( $[SO_4]_{\text{impactor}}$ ) and the concentration observed on the bulk filter ( $[SO_4]_{\text{bulk}}$ ) is  $[SO_4]_{\text{impactor}} = 0.91 (\pm 0.08) * [SO_4]_{\text{bulk}}$  with  $R^2 = 0.75$ . For MSA the relationship is  $[MSA]_{\text{impactor}} = 0.75 (\pm 0.06) * [MSA]_{\text{bulk}}$  with  $R^2 = 0.8$ . The slight difference between the two data sets is likely due to differences (up to a few days) in the sampling time intervals.»**



In Fig 4: there are the average size distributions of the respective seasons. How about showing there some range? **OK we change Figure 4 (a-d) showing the range for DC impactors.**

Also Becagli et al. (Atmos. Environ., 52, 98–108, 2012) showed size distributions measured at Dome C – make some comment on the main differences. **Yes but it is quite difficult to compare since Becagli and co (2012) used an impactor that samples submicron aerosol (above 0.4 micron) on two stages only (instead of 8 stages in our case above 0.028 micron) whereas most of sulfur biogenic mass is present between 0.1 and 1 micron. Nevertheless we have now referenced this study and results are reported in Table 2.**

P8,L6-8 "Impactor data corresponding to the March-November time period (Fig. 7) show that RMSA is very poorly related to the nssSO<sub>4</sub> content (R<sup>2</sup> of 0.01 and 0.06 for submicron and micron particles, respectively)". Fig 7 shows R vs MSA, not R vs nssSO<sub>4</sub>. I suggest adding subfigures where this is shown. **OK we agree and Figure 7 was modified accordingly.**

P10L9-10 "Assuming a sulfate concentration of 250 ng m<sup>-3</sup> for the continental free troposphere of the southern hemisphere, and applying a dilution factor of 18 based on <sup>210</sup>Pb data" Please explain how the dilution factor of 18 was obtained. Any uncertainty estimate for it? **OK we now specify in more details how these calculations were done: "Apart from marine biogenic emissions, sulfate present over Antarctica can also originate from southern hemisphere continents or the stratospheric reservoir. <sup>210</sup>Pb data permit to derive an estimate of the contribution of sulfate long-range transported from continents by comparing the <sup>210</sup>Pb concentrations at Concordia (27 μBq m<sup>-3</sup>) after having corrected them from marine <sup>222</sup>Rn exhalation (~ 15%, Weller et al. 2014) (i.e., 23 μBq m<sup>-3</sup>) with those observed at Chacaltaya (407 μBq m<sup>-3</sup>, Feely et al., 1988), a remote site located at 5220 m asl in Bolivia..... Assuming a sulfate concentration of 250 ng m<sup>-3</sup> for the continental free troposphere of the southern hemisphere, and applying a dilution factor of 18 based on <sup>210</sup>Pb data (407 μBq m<sup>-3</sup> at Chacaltaya compared to 23 μBq m<sup>-3</sup> at Concordia), we calculate a mean sulfate concentration of 14 ng m<sup>-3</sup>."**

P10L26-27 "Considering a mean sulfate mixing ratio of 0.3 ppb for the lower stratosphere, we estimate that stratospheric-tropospheric exchange may account for 0.4 ng m<sup>-3</sup> of sulfate" Is 0.3 ppb = 0.4 ng/m<sup>-3</sup>? **No, but may be the text was not clear enough that the value of 0.4 ng m<sup>-3</sup> was estimated by applying the dilution factor observed for <sup>10</sup>Be to the 0.3 ppb of sulphate present in the lower stratosphere. We have now reworded the sentence as follows: «Considering a mean sulfate mixing ratio of 0.3 ppb for the lower stratosphere, and the typical dilution factor observed for <sup>10</sup>Be between the lower stratosphere and the atmosphere at Concordia, we estimate that stratospheric-tropospheric exchange may account for 0.4 ng m<sup>-3</sup> of sulfate in winter at Concordia.**

Table 2 shows R in midsummer. March is not really midsummer any more. **Right and this was corrected in the Table caption as "in mid-summer and in February/March (if available).**

Figures with scatterplots: why don't you show the regressions there? **OK, we now report the regression lines in Figure 11 (corresponding to calculations reported in Table 4), as also required by the other reviewer. We don't do it for Figure 10 since that will overload the figure.**