**Referee # 2**

We thank Referee #2 for the helpful comments. Please find our responses below.

One main point comes at the end of the paper. The main subject is the importance of halogen chemistry for the DMS –> sulphate oxidation pathway. New intermediates and reactions are included in a global chemistry-transport model. This gives better validations of intermediates, like MSA and MSA/nssSO4 ratios. Also, the main uncertainties in the reaction pathways are clarified. However, for the formation of sulphate aerosol the new pathways have limited impact, simply because this is the end-product. Only the speed of the DMS –> sulphate conversion may be affected, and maybe some intermediates have efficient dry- or wet deposition pathways. Thus the phrase "...with a simplified DMS oxidation scheme (gas-phase oxidation by OH and NO3 only) may overestimate sulfate abundances in the pre-industrial environment, potentially leading to underestimates in sulfur aerosol radiative forcing calculations in climate models." seems overstated. I expect no huge impact in sulphate forcing and some reflection on this is recommended. For the rest of my comments, see annotated manuscript. Please also note the supplement to this comment: https://www.atmos-chem-phys-discuss.net/acp-2018-410/acp-2018-410-RC2- supplement.pdf

**Response:** We agree that the new pathways added in model could have limited impact on the sulfate burden if the depositions of DMSO, MSIA and MSA are small so that DMS will be oxidized to form sulfate as an end-product. However, if a large amount of MSA produced from DMS oxidation is removed via deposition instead of being further oxidized to form sulfate, then it could potentially result in a non-negligible change in the sulfate burden. This is the case in this study when using a lower rate coefficient (updated) for the oxidation of MSA by $OH_{(aq)}$ in cloud droplets and aerosols. We think it is still necessary to mention that the updated sulfur chemistry scheme **may** be important for sulfur aerosol radiative forcing calculations in climate models.

**Changes to the manuscript:**

- At Page 14, Line 1: modified as "Compared to the standard GEOS-Chem model run, the updated sulfur scheme in this study decreases the conversion yield of DMS to $SO_2$ ($Y_{DMS \rightarrow SO2}$) from 91% to 75% and increases the conversion yield of DMS to MSA ($Y_{DMS \rightarrow MSA}$) from 9% to 15%. The remaining 10% of DMS is lost via wet and dry deposition of DMSO and MSIA. In order to gain insight into the impacts of our updated sulfur scheme on global $SO_2$, MSA and sulfate burden, we conducted two sensitivity studies by allowing DMS as the only sulfur source for both the standard model run $R_{std}$ ($R_{std\_onlyDMS}$) and full model run $R_{all}$ ($R_{all\_onlyDMS}$). Compared to $R_{std\_onlyDMS}$, the global DMS, $SO_2$, MSA and sulfate burden in $R_{all\_onlyDMS}$ decreases by 40%, 17%, 8% and 12%, respectively. The decrease in DMS is mainly due to DMS oxidation by BrO with the updated sulfur scheme. The decrease in $SO_2$ is due to a lower yield of $SO_2$ from DMS ($Y_{DMS \rightarrow SO2}$), but is partly compensated by the increase in the DMS oxidation rate. MSA decreases despite an increase in the yield of MSA from DMS ($Y_{DMS \rightarrow MSA}$) due to a shorter lifetime in $R_{all\_onlyDMS}$ (2.2 days in $R_{all\_onlyDMS}$ versus 4.1 days in $R_{std\_onlyDMS}$) that is caused by the aqueous-phase sink of MSA via MSA + $OH_{(aq)}$ and faster deposition of

MSA produced in the MBL. The decrease in sulfate is caused by the decrease in $SO_2$ but is partly compensated by the inclusion of MSA + $OH_{(aq)}$ as a sulfate source, which accounts for 4% of global sulfate production. The decrease in sulfate will be smaller if more MSA is oxidized into sulfate instead of being lost via deposition. In sum, climate models with a simplified DMS oxidation scheme (gas-phase oxidation by OH and $NO_3$ only) may overestimate $SO_2$, MSA and sulfate abundances in the pre-industrial environment, potentially leading to underestimates in sulfur aerosol radiative forcing calculations in climate models. Quantifying the impacts of our updated sulfur oxidation scheme on new particle formation is out of the scope of this study and should be addressed in the future."

Other comments from the second reviewer shown in the supplement have been accepted in the updated manuscript.

(1) **Comment:** Page 8, Line 7 "The difference between the results from Boucher et al. (2003) and this study is that Boucher et al. (2003) did not include DMS+BrO in their model simulation, 5 which could lead to an overestimate of the contribution of DMS+$O_{3(aq)}$ as both reactions are most important over the high-latitude oceans during winter.": since this channel only accounts for 12% of the DMS oxidation, I guess this argument is weak. I would expect that this "missing" oxidation would be divided equally over the remaining DMS oxidation channels.

**Response:** We agree with the reviewer that the smaller contribution of $O_{3(aq)}$ to DMS oxidation in our study (2%) compared to Boucher et al. (2003) (6%) is not solely caused by the inclusion of DMS+BrO in our study. The fraction of DMS oxidized by O3(aq) increases from 1.8% in $R_{\mathrm{all}}$ to only 2.4% in a sensitivity run without DMS+BrO reaction ($R_{\mathrm{noDMS+BrO}}$). We have updated the manuscript accordingly.

**Changes to the manuscript:**
- Page 8, Line 16, modified as "The difference between the results from Boucher et al. (2003) and this study could be due to the differences in oxidants abundances such as $O_3$, OH, BrO and Cl."

(2) **Comment:** Page 8, Line 27 "Multiphase oxidation is especially important over regions where clouds are frequent and OH concentrations are high, e.g. low- to mid-latitude oceans (Fig. 5).": somehow counterintuitive, since the gas-phase OH reaction pathway is more important than the multiphase oxidation.

**Response:** In our model, DMSO is removed from the atmosphere via deposition and oxidation by OH in both the gas phase and the aqueous phase. The aqueous-phase oxidation occurs faster than gas-phase oxidation when a lot of clouds are present (low- to mid-latitude oceans).

**Changes to the manuscript:**

- Page 9, Line 4: modified as "Multiphase oxidation mainly occurs over regions where clouds are frequent and OH concentrations are high, e.g. low- to mid-latitude oceans (Fig. 5)."
- Page 9, Line 17: modified as "Multiphase production of MSIA mainly occurs over low- to mid-latitude oceans where the OH abundance is high and clouds are frequent (Fig. 6)."

(3) **Comment:** Page 12, Line 16: Add the 23 stations description to a table.

**Response:** The manuscript is updated accordingly.

**Changes to the manuscript:**
- Add Table 5: The locations of the 23 stations that provide annual-mean MSA/nss$SO_4^{2-}$ observations.

(4) Other minor changes such adding "the" have been all accepted in the updated manuscript.