

We thank the referee for the comments and suggestions. The Point by Point Clarifications to referees comments and suggestions are as follows;

Anonymous Referee #1

[C1] Twenty years ago Langner&Rodhe published the first global sulfur cycle model study. Although great efforts have been made since then to develop schemes of various degrees of complexity, there are still aspects that are not well understood. This includes, for instance, too slow oxidation of DMS and of SO₂ over higher latitudes in winter (e.g. Koch et al., JGR, 1999). Thus, the study of Goto et al, which investigates the effect of sulfur chemistry and of removal processes on the aerosol radiative forcing, deserves certainly publication in ACP.

[A1] Thank you very much for reading our manuscript and giving us useful comments for improving the manuscript. We already referred these studies in the manuscript, but we have added 'Langner and Rodhe (1991)' to section 1 as the first study of global sulfur model in the revised manuscript as follows:

L14, P12272 (modified):

Since Langner and Rodhe (1991) first published the global sulfur cycle model, sulfur chemistry modeling studies indicate that ...

[C2] My only point of critics is that often just the differences between sensitivity simulations are noticed but not really explained. Based on the sensitivity simulations performed it should be possible to elaborate clearer why (if so) the new scheme behaves better than the old one.

[A2] Thank you for your comments. We have performed various sensitivity tests of sulfur schemes to understand how large is the impact of the difference schemes on the sulfate burden in section 4. According to the results of the sensitivity tests, we added more explanations about the difference in sulfate concentration at the surface and in the vertical distributions in sections 4 and 5 as follows:

Section 4.1:

L1, P12282 (modified): ... the assumption of unlimited supply of H₂O₂ ...

→ ... the assumption of the open system that allows excess supply of H_2O_2

L3, P12282 (added): In case of low SO_2 concentrations, i.e., above boundary layers and/or over remote areas, on the other hand, the calculated sulfate concentrations by the second-order reaction are higher than those by the quasi first-order reaction (Fig. 4). This is because consumed H_2O_2 concentrations by the SO_2 oxidation in the aqueous-phase are larger than supplied H_2O_2 concentrations through Henry's law equilibrium before the step of the SO_2 oxidation. Therefore, the sulfate concentration by the quasi first-order reaction is underestimated compared to that by the second-order reaction.

L15-18, P12282 (added): ...These values are much larger than those in the results of C2ND, because of the assumption of excess supply of H_2O_2 in the aqueous-phase as shown in Fig. 4. For the sulfate column burden, on the other hand, the annually averaged RB is estimated to be minus almost over the world within the global mean value of -26.3% in Q1ST and -11.9% in C2ND, respectively, because of underestimated sulfate concentrations at upper heights (above boundary layers) caused by a lack of H_2O_2 in the aqueous-phase in Q1ST as shown in Fig. 4.

Section 4.2 and 4.3: we adequately mentioned explanations of the differences in the manuscript.

Section 4.4:

L1, P12286 (added): ... as CTL are generally positive as shown in Table 4, because of excess H_2O_2 especially in winter.

Section 4.5: Please see our answer 'A6' in this document.

Section 5.1:

L13-L16, P12287 (modified):

As mentioned in the previous section, the improved method of solving ...

→ The improved method of solving ... compared to the simplified model in OS, mainly because of the difference in the solution of the aqueous-phase sulfur chemistry as mentioned in section 4.

L19, P12288 (added):

...sulfate production rate under lower SO₂ concentrations, which is supported by the previous sensitivity tests.

[C3] I miss also a discussion about the impact of transport on aerosol distribution. In particular, aerosol concentrations -discussed in this study- in Polar Regions and in high altitudes might be significantly influenced by large-scale and small-scale transport (e.g. Rasch et al., JGR, 2001).

[A3] Thank you for the comment. We think this part is related to the results shown in the first paragraph in section 6. So we have added some discussion for them to section 6. Please see our answer 'A10' in this document.

Specific comments:

[C4] Page 12273 ln 18-20: A SO₂ turn-over time of 10 days is well above most estimates which range between 1 – 3 days (e.g. COSAM model intercomparison, Barrie et al., TELLUS B, 2001).

[A4] Thank you for correcting our mistake. We have changed the 'ten days' to 'one to three days'.

[C5] Figure 3 displays the differences between the old (OS) and the new (NS) sulfur scheme. NS includes the release of SO₂ from the aqueous into the gaseous phase if cloud droplets evaporate and the removal of SO₂ by wet scavenging. Can you, please, quantify the importance of these two processes for the SO₂ and SO₄ budgets.

[A5] The important things in this figure are (1) sub-cycle calculation and (2) the order of the processes to calculate. Firstly, we assume that the 'chemical' processes of both chemical reaction and Henry's law equilibrium are more rapid than 'physical' processes

such as evaporation of cloud droplets or cloud-precipitation conversion rate, so that the ‘chemical’ processes are calculated by shorter timestep as we mentioned. Secondly, we assume the ‘physical’ processes are calculated after the chemical processes.

Basically, both methods (OS and NS) include the release of SO_2 from aqueous-phase into the gas-phase in the last part of the sulfur calculation, because we do not predict the sulfur concentrations in the aqueous phase, that means we assume that cloud droplets can evaporate within one timestep of the GCM. As a result, all sulfur components in the aqueous-phase are released into the gas-phase, after the calculation of sulfur species. Therefore, we cannot quantify the contribution of the evaporation for the sulfur species in this study, although this assumption is common for GCM study.

In addition, wet deposition represents the removal of SO_2 from aqueous-phase of cloud droplets into the aqueous phase of rain droplets, which falls on the ground at the timestep. The importance of the wet deposition was already shown in Table 5.

[C6] Chapter 4.5: To evaluate the dry deposition of SO_2 , not only sulfate but additionally SO_2 concentrations should be compared to observations.

[A6] Thank you for your comment. We have added comments for differences in SO_2 concentrations among different processes of the SO_2 dry deposition to the second paragraph in section 4.5 as follows:

“We study the impact of SO_2 dry deposition using the improved method (as CTL) and the original method of SPRINTARS on the sulfate and SO_2 simulation. Table 4 shows that annually globally averaged RB values of surface sulfate concentrations and sulfate column burden are calculated to be -12.0% and -11.9%, respectively. In other areas, their values are estimated to be at most -20%. For SO_2 , annually globally averaged RB values of surface concentrations and column burden are calculated as -28.3% and -24.4%, respectively. In other areas, their values are estimated to be ranging from -30% to -10%. For gases, R_c has a great impact on the dry deposition rate. Therefore, ignoring the term R_c for SO_2 mainly causes an overestimation of dry deposition rate for SO_2 , and thus we found decreases in the sulfate and SO_2 concentrations. In conclusion, we find that differences in the dry deposition modeling also have relatively large impacts on the sulfur budget compared to differences in other parts of the sulfur process.”

We do not validate SO₂ concentrations using observations especially because of two major reasons. Firstly, available observations of SO₂ are limited especially over North America. Secondly, previous studies suggest the difficulty in the validation of SO₂ especially over industrial regions (e.g., Lohmann et al., 1999; Koch et al., 1999; Barth et al., 2000; Chin et al., 2000; Boucher et al., 2002; Easter et al., 2004; Liu et al., 2005; A de Meij et al., 2006; Berglen et al., 2007). The reason is probably that there are unavoidable errors caused by the coarse grids in the model as Chin et al. (2000) suggested. Generally, the lifetime of SO₂ in the atmosphere is approximately within 1-3 days and shorter than that of sulfate aerosols, so that the SO₂ concentrations in a monitoring site can be strongly affected by the nearest SO₂ emission sources in the simulation and probably also in the real atmosphere.

[C7] Page 12287 ln 26: "In conclusion, the results in NS are much better than those in OS." This statement is not supported by the results displayed in Figure 5, at least not in Europe and North-America.

[A7] Over East Asia, the available sites to compare with the simulated sulfate concentrations are limited, compared to Europe and North America. Furthermore, the sites used in this study over East Asia do not include Chinese stations where sulfate concentrations are probably large. So we did use results obtained by ensemble regional models shown by Hollway et al. (2008) to validate our results, and thus we found the results in NS over East Asia especially China are closer to the results by Hollway et al. (2008) compared to those in OS. Although the results of Hollway et al. (2008) are not observational but just 'modelled', we assume that they are more reliable than any other studies under the current situation (limited observations). Therefore, we concluded that 'the results in NS are much better than those in OS even in East Asia'. Please see the newly additional supplement (Figure S2) including monthly-averaged simulated sulfate concentration in March, July, and December to compare with the results in Fig.3 by Hollway et al. (2008).

[C8] Table 5: According to the results of the COSAM comparison, wet deposition of SO_2 is of relatively little importance in the regional budget of SO_2 , whereas in the current study (OS model version) 22% of the SO_2 emissions are removed by wet deposition. Is this caused by a different treatment of the processes or by a different method to calculate the budgets? Moreover, according to Figure 3 wet deposition of SO_2 in the aqueous phase is taken into account in model version NS but not in the version OS. Surprisingly, wet deposition in OS is much higher than in NS.

[A8] Some of your comments are related to those in comment 5 ‘C5’ in this document. Both OS and NS consider the wet deposition of SO_2 , although the order of the calculation is different from each other as shown in Fig. 3. The wet deposition in OS is considered after the first separation of SO_2 into gas and aqueous phases, whereas in NS it is considered after the step of chemical reactions. The difference in the budget of SO_2 shown in Table 5 is mainly caused by the differences shown in Fig. 3 (the method to calculate the budget is same).

In OS, because the wet deposition of SO_2 is calculated before the step of the SO_2 oxidation, the amount of SO_2 removed by wet deposition become larger than that in NS. Therefore, the budget of SO_2 by wet deposition in OS is larger than those in this study (NS) and COSAM study.

[C9] Page 12290 ln 7-8: "larger sulfate distributions" shouldn't it be "larger sulfate concentrations"?

[A9] Yes, thank you for your correction. We have corrected it in the revised manuscript as you suggest.

[C10] Page 12290 ln 9-11: "We are speculating ... " Please, give some reasons for the speculation or remove this statement.

[A10] The sulfate distribution depends on sulfur chemistry, deposition (mainly wet deposition), and transport. In this study, we modified only the scheme of sulfur chemistry, and thus the sulfate distribution in NS is different from that in OS. As we mentioned in section 6, the transport of sulfate to lower latitudes is still smaller compared to other results. The reasons are mainly caused by the differences in the wet

deposition and the transport. The wet deposition rate depends not only on the radius and wet growth of the particles but also on precipitation flux, cloud fraction, and advection of aerosols. As we mentioned in section 1, the difference in the radius and wet growth of sulfate aerosols among sulfur chemistry models is not so large. In conclusion, the differences in the sulfate concentrations over low latitudes are caused by (1) cloud and precipitation and (2) transport. In the manuscript, we attributed the possibility of both (1) to 'cloud and precipitation scheme' and (2) to 'boundary layer scheme'. But we also have to mention that the possibility (2) is caused by not only boundary layer scheme but also advection scheme as suggested by Rasch et al. (2001). We have modified this part of the manuscript as follows:

“The sulfate distribution depends on sulfur chemistry, deposition (mainly wet deposition), and transport (e.g., Rasch et al., 2001). After the modification of the sulfur chemistry, differences in the sulfate distribution still remain, so that they are probably caused by differences in wet deposition and transport. The wet deposition depends not only on the radius and wet growth of the particles but also on precipitation flux, cloud fraction, and advection of aerosol. As we mentioned in section 1, the difference in the radius and wet growth of sulfate aerosols among sulfur chemistry models is not so large. Therefore, the differences in the sulfate concentrations over low latitudes are caused by (1) cloud and precipitation, which are determined mainly by schemes of the boundary layer and/or the cloud and precipitation, and (2) transport pattern, which is determined by schemes of the boundary layer and advection; but a detailed study is our future work.”

[C11] Page 12291 ln 21-23: "In OS, aqueous-phase reaction fluxes are generally so large..." wouldn't it be correct "In OS, the winter-time aqueous-phase reaction ..." ? Why is the aqueous oxidation rate of SO₂ higher in winter than in summer?

[A11] Yes, thank you for your correction. We have added the term 'winter-time' to this part as you suggest. The reason is probably caused by the difference in the treatment of H₂O₂ as discussed in section 4.4. The offline use of H₂O₂ often causes unrealistic variability in the wintertime H₂O₂ near urban areas due to abundant H₂O₂ (e.g., Koch et al., 1999) as we mentioned in line 20-22, P12285. As a result, the amount of the

winter-time aqueous oxidation in OS (using offline H_2O_2) increases. Therefore, the relative values in the aqueous-phase reaction between summer and winter become small as shown in Fig. 9. We have added the reason to the part in the revised manuscript as follows:

“This low value in OS is caused by the treatment of use for offline H_2O_2 distribution as mentioned in section 4.4.”