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

Anonymous Referee #2

[C1] This paper describes the development of an improved representation of sulphur chemistry within a global model, and sensitivity tests to establish the relative importance of different simplifications for sulphur surface concentration, sulphate column burden and aerosol direct radiative forcing. The studies are conducted in a systematic way with good justification, and the conclusions reached are sound and quantitative. I therefore recommend it is published subject to some minor revisions. In particular I would recommend the authors consider carefully figure 1 and whether the information there requires a figure.

[A1] Thank you very much for reviewing our manuscript and giving us useful comments for improving the manuscript. For Figure 1, we have answered it in A5 below.

Specific comments:

[C2] Page 12270, line 15 change “led” to “brought”

[A2] Thank you, we have modified it as you suggest.

[C3] Page 12270, line 16 – please quote the uncertainty in these radiative forcing values in order to allow the reader to judge whether these values are really significantly different.

[A3] One of the large uncertainties in the radiative forincg is the emission inventory. As we mentioned in the text (L21-24, P12293), we also estimated it using different emission inventory. So we have modified the abstract as follows:

“The global annual mean radiative forcings due to the direct effect of anthropogenic sulfate aerosol was thus estimated to be -0.26 W m^{-2} (-0.30 W m^{-2} with a different SO_2

inventory), whereas the original SPRINTARS model showed −0.18 W m^{-2} (-0.21 W m^{-2} with a different SO_2 inventory)."

[C4] Page 12271, line 16: This sentence is not quite clear, do you mean that the sulphate forcing is larger magnitude than that due to BC or OC, or that the range of forcing from sulphates is larger than that due to BC or OC.

[A4] We wanted to say that the range of forcing due to sulfate is larger than that due to BC and OC aerosols. So we have modified the statement as follows:

"Also they showed that the radiative forcing due to anthropogenic sulfate aerosol is estimated to be from −0.16 W m^{-2} to −0.58 W m^{-2}, whose range is larger than those due to black carbon (BC) and organic carbon (OC) aerosols."

[C5] Page 12271, line 20-25. Please consider this figure. Surely it is obvious that the aerosol direct radiative forcing should be dependent on sulphate burden given the background in the first paragraph of your introductions. I don't believe this needs to be on Figure 1. Secondly, figure 1 does not convince me that the sulphate column burden increases with fraction above 5km – this relationship appears to be weak at best. I am not disputing that burden and distribution aren't important for producing model diversity but I don't think Figure 1 shows this. Please consider removing this and leaving only reference to the AEROCOM studies, or devising a figure that makes the point better. If you do include a similar figure with the new model data on it, please ensure consistency with OS and NS on this figure so as not to confuse the reader.

[A5] Before our explanation, we have to apologize for our mistake of Fig. 1 (including a point of 'this work' with the value of -0.58 W m^{-2}). We have modified Fig. 1 using two panels to clarify the relationship between the sulfate burden, fraction and radiative forcing.

In the manuscript, we mentioned about Figure 1 in terms of two points. Firstly, we stated that "the figure shows a tendency that the aerosol direct radiative forcing due to sulfate increase as the sulfate column burden increases". This statement is very trivial to be found in the figure as you suggested. Secondly, we stated that "the sulfate column burden increases as the sulfate fraction above 5 km increases". As you suggested, the correlation appears to be week. Strictly speaking, this statement is not correct for all the models but suitable for several models including SPRINTARS (OS and NS). Furthermore, we like to add the one more important point to the revised manuscript. That is, so far there is no figures to show how divergent are the model results of sulfate burden, fraction above 5 km, and the direct radiative forcing. In this regard, Fig. 1 of our manuscript is important to show differences in these values between models. Although we generally know the strong relationship between the burden and the aerosol radiative forcing, the figure clearly shows that the burden is not only key quantity, but we need to investigate the effect of the simulated vertical stratification of the radiative forcing. Several models including SPRINTARS seem to have a problem in suitable simulation of vertical transportation of gaseous and particulate materials to the upper atmosphere. So, we want to keep Fig. 1 to show in section 1. We modified the part as follows:

"Figure 1 shows scatter plots to show relations among global annual mean values of sulfate column burden, sulfate fraction above 5 km to its column burden, and aerosol direct radiative forcing due to anthropogenic sulfate aerosols using the data from Textor et al. (2006), Schulz et al. (2006), and the present study. The figure can help us to understand how relation among these key quantities is scattered showing that models still have problems in realistic simulation of the radiative forcing due to problems in modeling of both the aerosol burden and stratification. Firstly, the figure shows an obvious tendency that the aerosol direct radiative forcing increases as the sulfate column burden increases, though increasing rate is largely different among models. Secondly, even though not true for all models, the sulfate column burden tends to increase as the sulfate fraction above 5 km increases. This phenomenon is understood as that with increasing aerosol heights, the magnitude of the direct radiative forcing is getting large because the sulfate aerosol is more exposed to the incoming solar radiation above the Rayleigh and cloud scattering layers to generate the negative forcing. The figure also shows that such relation is also largely scattered among models. This point is important to be recognized, because there is another proposal that a value of the direct aerosol radiative forcing decreases with increasing heights of absorbing aerosols because they tend to lay over bright cloud layers to generate a positive value of the forcing through reducing the planetary albedo (e.g., Haywood and Ramaswamy, 1998). This figure shows the uncertainties of the negative direct radiative forcing due to problems in the model treatment of vertical stratification of sulfate is equally important to be improved as compared to the positive forcing caused by absorbing aerosols."

Furthermore, we would like to evaluate our model results (NS) with other modeling studies as mentioned in section 6 in terms of sulfate column burden and fraction above 5 km. Figure 1 shows that results in NS are much closer to those by other results compared to those in OS. This is also important point in this study to show.

Haywood, J. M., and Ramaswamy, V.: Global sensitivity studies of the direct radiative forcing due to anthropogenic sulfate and black carbon aerosols, J. Geophys. Res., 103, 6043-6058, 1998.

[C6] Page 12272 lines 20-25: This paragraph needs rewording. It would be sufficient to say that the GISS and SPRINTARS models show substantially different ratios to the other models.

[A6] We think it is important to point out that the two models (GISS and SPRINTARS) have not only low values of the ratio but also low sulfate burden suggested in Schulz et al. (2006). These two facts are linked with each other. As a result, we have modified this part as follows:

"In Fig. 2, the GISS and SPRINTARS models, which also have lower sulfate column burden as shown in Schulz et al. (2006), show substantially low values of the ratio to the other models. …"

[C7] Page 12273, lines 1-5. I think you could make your point more clearly here. "It is important to quantify the impact of this simplification by comparing against models with more physical, and/or complex, representation of the sulfur cycle".

[A7] Thank you for showing the clearer expression. We have modified them as you suggest.

[C8] Page 12273, line 7-8, “A discussion of the impact on aerosol direct radiative forcings is given in section 7”.

[A8] Thank you. We have modified them as you suggest.

[C9] Page 12274, line 6-10. It’s not very clear here which methods are more simplified and which are more physically based. For your experiments, tables 3 and 4 make it much clearer, is it possible to make this clearer in this paragraph when considering models in general?

[A9] Yes, we have added the following comments to section 2 in the revised manuscript:

“For timestep, physically based methods (e.g., Feichter et al., 1996; Boucher et al., 2002) set it in the aqueous-phase chemistry to be shorter than that in the transport model as physically based method, while simplified methods (e.g., Chin et al., 2000; Takemura et al., 2002) set the same time resolution in both the aqueous-phase chemistry and the transport model. For oxidants, i.e., O_3, H_2O_2, and OH radical, physically based methods (e.g., Easter et al., 2004; Tie et al., 2005) calculate them with online-coupling to chemistry, while simplified methods (e.g., Barth et al., 2000; Koch et al., 2006) use their offline distributions. For dry deposition, physically based methods (e.g., Liu and Penner 2002; Gong et al., 2003) treat all components of the resistance using Zhang et al. (2001), while simplified methods (e.g., Rasch et al., 2000; Pitari et al., 1993; 2002) assume the constant rate of the dry deposition.”

References

Gong, S. L., Barrie, L. A., Blanchet, J. –P., Salzen, K. V., Lohmann, U., Lesins, G., Spacek, L., Zhang, L. M., Girard, E., Lin, H., Leaitch, R., Leighton, H., Chylek, P., and Huang, P.: Canadian Aerosol Module: A size-segregated simulation of atmospheric aerosol processes for climate and air quality models 1. Module development, J. Geophy. Res., 108(D1), 4007, doi:10.1029/2001JD002002, 2003.

Pitari, G., Mancini, E., Rizi, V., and Shindell, D. T.: Impact of future climate and emissions changes on stratosphere aerosols and ozone, J. Atmos. Sci., 59, 414-440, 2002.

Pitari, G., Rizi, V., Ricciardulli, L., and Visconti, G.: High-speed civil transport impact: Role of sulfate, nitric acid trihydrate, and ice aerosol studies with a two-dimensional model including aerosol physics, J. Geophys. Res., 98, 23141-23164, 1993.

[C10] Page 12281, line 6: The use of a box model is mentioned here. Why is this necessary and what is it's formulation?

[A10] We used the box model to just calculate sulfate concentration formed from the aqueous-phase reaction. The formulation is described in section 2.1, so that we don't think we need to add more descriptions and formulations here. The box model was used to show the difference in the sulfate formation between the quasi first-order reaction and the second-order reaction in Figure 4. Furthermore, we used the box model to estimate the difference in the sulfate formation among different timestep as shown in section 4.2. In the manuscript (L11, P12283), we just mentioned 'we conduct sensitivity experiments using a box model'. So we have modified it as follows: 'we conduct sensitivity experiments using a box model to calculate aqueous-phase sulfur chemistry'.

[C11] Page 12287, line 17 and 18, I think NS and OS are used the wrong way around in this sentence!

[A11] Thank you very much. We had a mistake here. We have corrected it.

[C12] Page 12287, line 24: You state that the observations using in Fig 5c do not include China. This makes it hard to compare observations with simulations. It would perhaps be better to include only the simulation regions that compare with observations that are available?

[A12] Figure 5c includes available observation sites over East Asia. But unfortunately, EANET observation we used in this study is not available in Chinese's stations. Therefore, we compared simulations with observations only in available sites. In addition, we compared them over East Asia including China with ensemble results obtained by other models under MICS-Asia (Holloway et al., 2008), showing the results of sulfate surface concentrations in Figure S2 in the supplement, which has been added in the revised manuscript. So we have modified one point as follows:

"… EANET observation network here are not available in China where …"

[C13] Page 12288, line 1; Link back to your previous sensitivity tests here explicitly to help justify the claim that it is the suppression in sulphate production that improves the surface concentrations.

[A13] Yes, the sensitivity test is supporting this conclusion here. So we have modified it as follows: "The improvement of the surface sulfate concentration in NS is attributed to the suppression in the sulfate production rate under higher SO_2 concentrations, which is supported by the previous sensitivity tests."

[C14] Section 6: Much of the discussion in section 6 is designed to show how much improved the new version of the model is compared to the old version, and how it agrees better with other models. However, in many quantities, the simple range of other models is so wide that either OS or NS would appear to perform equally well. Could you consider whether there are better measures to agree with from other models than the range? Perhaps you were aiming to get your model closer to the centre of that range from other models for example?

[A14] The results shown in section 6 are basically difficult to validate, because the quantified values here cannot be observed and the estimation by other models cannot exclude uncertainties. In this case, however, we should assume that the results of ensemble models are the best even though the range of other models is very wide. So, as you suggested, our strategy is to get our results be within the range of ensemble results by other models. Although most results in both OS and NS are within the range of other models in terms of sulfur budgets shown in section 6, Figures 1, 2, and 9 clearly show that the results in OS are out of the range of other modeling studies, so that our first goal is to get our results in terms of sulfur components (sulfate concentration, vertical distribution, the ratios of the each process for sulfur species, and global budget) closer to those obtained by others. Therefore, we like to conclude that although simple comparisons of our model results with other model results could often show little differences, comparisons of the correlation between two parameters could show big differences as shown in Figures 1, 2, and 9.

[C15] Figure 12. The huge change in radiative forcing over SE Asia deserves more discussion in the text please.

[A15] The huge change in the radiative forcing is found over East Asia in Figure 12. The difference over East Asia is caused by differences in AOT shown in Figure 10. The change in AOT is caused by the change in the column burden of sulfate. Table 4 directly indicates that the difference in the column sulfate burden in China between the standard experiment and Q1ST (aqueous-phase reaction with quasi 1^{st}-order expression) is very large with the value of +37.4%. Therefore, the big change in the radiative forcing over East Asia is mainly caused by changing the solution in the sulfur aqueous-phase reaction. Therefore, we have added the following comments to the revised manuscript as follows:

"… North America and Southeast Asia with ranges of 0.5-1 W m^{-2} and usually over land with ranges of 0.2-0.5 W m^{-2}, respectively. <u>The big change in the radiative forcing over East Asia is mainly caused by the difference in the solution in the sulfur aqueous-phase reaction as shown in Table 4.</u> Over oceans, …"