

## ***Interactive comment on “Sulfur cycle and sulfate radiative forcing simulated from a coupled global climate-chemistry model” by I.-C. Tsai et al.***

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Received and published: 14 March 2010

We thank both reviewers for their thorough reviews and constructive comments, which have helped to improve the manuscript. The main changes to the manuscript include:

1. A question from one of the reviewers led us to find a potential error in the original model code which included cloud ice for calculating aqueous-phase chemistry. We have modified the code and re-run all simulations (which took us a bit longer to prepare for this revision). All values in the manuscript (including figures and tables) have been updated with the new results. Fortunately, our main conclusions on the radiative forcing and responses remain largely unaffected.
2. To increase the statistical significance of analysis on the relationship between re-

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sponses and internal variability, we increased the length of model results that being analyzed from 5 to 10 years. The conclusions remain the same.

3. The discussions in Section 3.1 have been trimmed down a little bit. In addition, the part on the budgets of sulfur cycle has been separated to form a new sub-section (i.e., Section 3.2: Sulfur cycle).

More detailed responses to the reviewers' specific comments are listed below.

1. p. 22373, l. 5: Could the relatively low sulfate loading in GCCM be responsible for the smaller direct effect versus many of the other models?

Reply: Yes, since the sulfate direct effect is related to the sulfate loading, the relatively smaller direct effect might be caused by the relatively low sulfate loading in GCCM. We add this statement in paragraph 2, Section 3.2, and re-emphasize it in the discussion section.

2. p. 22377, l. 9ff: This paragraph opens by stating that the direct effect can be estimated by comparing either simulations A1-N1 or A2-N2. However, nothing is mentioned after this about A2-N2 in this paragraph. Can the direct effect really be estimated from A2-N2? A2-N2 will include differences in meteorology due to the 1st indirect effect, which will feed back to the overall sulfate field. So, the difference in this case would include both the direct effect and some aspects of the first indirect effect. Since the comparison A2-N2 is not discussed in this paragraph, the reviewer suggests just removing the reference to A2-N2 in the opening sentence.

Reply: Thanks for pointing this out. We somehow deleted the relevant content and forgot to modify the opening sentence of this paragraph. The direct forcing from the A2-N2 results ( $-0.49 \text{ W m}^{-2}$ ) is a bit stronger than that from A1-N1 ( $-0.43 \text{ W m}^{-2}$ ). But, as the reviewer suggested, both the A1-N1 and A2-N2 results might contain some aspects of the first indirect effect. In addition, they also contain internal variabilities (which were not mentioned until Section 4). To avoid confusion, we agree with the

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reviewer to take out that sentence to simplify the discussion.

Also, while it is clear to most readers from the context, this paragraph would be a good place to mention that the “direct effect” referred to in this paper is the anthropogenic direct effect, i.e. it is the anthropogenic component of the total aerosol direct effect due to sulfate. This point should also be made here, or elsewhere as appropriated, for the indirect effect.

Reply: Agree. We have modified the sentence in line 15 to emphasize that it is the anthropogenic component that we were referring to.

3. p.22377, l. 24: The general statement is made that the indirect effect is substantially larger than the direct effect. This is based on the global average values for these effects. Are the authors sure that this is true regionally as well for all portions of the globe?

Reply: The indirect effect is obviously greater than the direct effect over the polluted areas. But because of dynamic feedbacks, the negative radiative forcing due to indirect effect may diminish or even turn positive at less polluted locations (see Fig. 6a and top of p. 22378). So we cannot say the indirect effect is stronger than the direct effect over all regions. A sentence is added here to make it clearer. We also emphasize in various places about the stronger regional values.

4. p. 22379, l.6: “apparently most of it is from the indirect effect”. . . Is this true? From Table 5, going from the differences for the 0 to 1 to 2 simulations,  $dT$  from the ozone effect is  $-0.04K$ ,  $dT$  from the (direct + ozone effect) is  $-0.02$ , and  $dT$  from (direct + indirect + ozone effects) is  $-0.09K$ . So, if we assume the effects interact linearly and back out the ozone effect and direct effect from the combined value, we get  $(-0.09)-(-0.04-0.02) = -0.03K$  for the indirect effect. This puts the value of for the indirect effect between that for the ozone and direct effects. Am I understanding this correctly?

Reply: The reviewer made a good point. This statement is not valid if these effects

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interact non-linearly. So we modified the sentence “and apparently most of it is from the indirect effect” into “But it is difficult to differentiate contribution of this temperature change from individual effects (e.g. ozone heating, aerosol direct and indirect effects) because of their nonlinear interactions. In addition, due to model internal variability which will be discussed later, the small forcing from ozone heating and aerosol direct effect might not be meaningful”. Note that in response to the other reviewer’s questions, we found that the original code that we used (from Berglen et al. 2004) allowed the aqueous reactions to occur also in cloud ice. This will result in more aqueous-phase oxidation by O<sub>3</sub>, as H<sub>2</sub>O<sub>2</sub> is concentrated in the warm-cloud levels, whereas O<sub>3</sub> concentration is higher at the upper troposphere where the ice cloud is located. So the simulations have been re-run with reactions in ice turned off. Relevant discussions have been modified accordingly. Interestingly, the interactions seem to be more linear in the new results.

5. p. 22380, l. 15: “A better comparison can be performed between either the A-series or N-series of simulations. . .” This statement is made in reference to understanding feedback onto the sulfur cycle from coupled chemistry. It is noted that the 0 and 1 scenarios are based on assuming a specified effective radius. It should be emphasized more in the text that the results of this section are going to be very dependent on the choice of effective radius and thus are just a representative example of the impact. If a different choice were made for the effective radius value, then the results could be different because the cloud fields would change. For example, the authors state rightly on p. 22380, l. 28 that the wet deposition has an impact. There will also be changes to local wind and temperature fields that will affect the sulfur as the clouds changes. An alternative would be to find a way to make the runs more comparable. This could be done by finding a way make the average effective radius the same between two simulations, which would then be used for comparison. For example, to decipher the indirect effect, Gustafson et al. (2007) used an initial simulation with the indirect effects turned on to get an average aerosol number and hygroscopicity that was then fed into a 2nd run for use in the comparison. This helps to minimize differences between cloud

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properties due to differences between cloud model assumptions.

Reply: It is interesting that the choice of effective radii does not significantly affect global cloud cover and precipitation, indicating that the model atmosphere tends to adjust itself in maintaining global stability of clouds and precipitation. This is probably why the variation in global (but not regional) sulfur loading is also small. In this regard, we have modified the sentence as: “Note that the results of N0, N1, A0, and A1 may dependent on the choice of effective radius. Yet, the global cloud cover and precipitation does not vary significantly among simulations, indicating that the dynamics of the atmosphere tends to adjust itself in maintaining global stability of clouds and precipitation (in the GCM). But regionally it is a different story”. We have thought about the idea of using average effective radius calculated from A2 for the simulation of A1 (or A0) in our original design. However, in doing so, we are in effect forcing the A1 (or A0) simulation to contain the indirect effect (as derived from A2). So all that left for the comparison is the direct effect, which has been demonstrated already by the A1-A0 results. The direct effect is relative easy to test by turning it on and off, but the indirect effect is so strong such that turning it off completely would cause a large shock in global radiation balance. Our approach is only a “somewhat better way”, and to this date we still cannot think of an ideal way to do such analysis.

6. p. 22381, l. 27ff: The use of Figure 7 in combination with the traditional statistics is a good way to present the internal variability issues. The visual comparison helps the reader comprehend the statistics. However, the authors should caution the reader that even though the statistics indicate some of the metrics reach the level of statistical significance; this is only based on a five year period. This is not long enough to encompass the full variability of the atmospheric system in terms of climate and year-to-year variability. Thus, the results should be understood as tentative and used with caution.

Reply: Originally, we did not use the results for the first 8 years to make sure the simulations reached their steady state. Note that the spin up time of GCM in Wong (2004) is only 6 months. We went back to re-check the results of the first 8 years

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and found that all variables remain quite steady year to year. In order to increase our confidence on the results, we re-analyzed the results from the fourth-year simulation for a total of 10 years. The results and conclusions are not significantly changed with the additional data. In any case, we mentioned in the revision that the 10-year results are still tentative and should be used with caution. Also, we have updated all the tables and figures as well as relevant discussions with the 10-year results.

7. p.22386, l. 16: “we demonstrated that climate signals from the direct forcing of sulfate are indistinguishable from the internal climate variability of the model...”. The authors should reword this to state that this is shown in the context of the variability of the 5-year simulation, not the full model variability.

Reply: Agree. Such statements have been added in line 18: Note that these results is shown in the context of the variability of the 10-year simulation, not the full model variability.

8. p.22386, l. 25ff: The reviewer does not understand the leap to needing the fully couple ocean-atmosphere model to understand the sulfur cycle on a regional scale. Is this because the internal variability of the model would change, possibly influencing the statistical significance of the signal? I do agree that changes to the sulfur cycle on a regional scale are going to be much more significant for some regions than for a global average. That point is worth making within this paragraph.

Reply: The ocean component may produce additional feedbacks (either positive or negative) which might be significant as the ocean occupies 70% of the global surface. This certainly adds to the uncertainty in model’s internal variability. We have added a sentence to make this a bit clearer. We also stress more on the higher regional responses of the sulfur cycle. Thanks for giving these nice suggestions.

Minor issues

9. In the introduction, a paragraph is devoted to discussing the emergence of on-

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line coupled aerosol-meteorology/climate models. As it stands, the discussion focuses solely on global models, which by necessity use a simpler sulfate cycle than is possible in regional online coupled aerosol-meteorology/climate models. Consider adding a sentence or two noting that regional models are also tending towards online coupling. In the final discussion at the end of the paper, this can then be tied into the need for improved aerosol-cloud interactions and also understanding the regional impacts. The regional models are able to use more complicated and physically based algorithms, and can serve as a way to develop more effective, simpler algorithms based on comparisons with the more complicated ones that are in the regional models. One example, though not the only one you could use, is WRF-Chem (Fast et al., 2006, Grell et al. 2005, Gustafson et al. 2007), which includes the direct and both indirect effects.

Reply: That is a good point. We have added a few sentences in both the introduction and conclusion to emphasize the regional model aspect.

10. p. 22370, Eqn.3: The c should be subscripted after the N.

Reply: Revised.

11. p. 22371, l. 10: “spin-off ” should be “ of spin-up”

Reply: Revised.

12. p22378, l. 5: “Similar to the approach applied earlier” is stated, but this reviewer does not see the earlier use of the alternative way to calculate the forcing using the difference of differences. Maybe I missed it?

Reply: Thanks for pointing that out. We forgot that the relevant earlier content has been removed. The statement is revised by deleting “Similar to the approach applied earlier.”

13. P 22378, l.17: The first sentence of section 3.3 is a little awkward

Reply: Thanks reviewer’s opinion. We have rewritten it as: “The dynamic response of

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the atmosphere to aerosol forcing will change the cloud fields which, in turn, feedbacks to the radiation fields. As the position of clouds is not directly controlled by aerosols, the indirect forcing may occur at locations different from those of the direct forcing.”

14. P22380, l. 1: the last sentence in section 3.3 could be stated a little better (the convoluted and tedious part). Would it be more accurate to state that the responses would be difficult to separate and cannot be deciphered from the given simulations alone?

Reply: Thanks for the suggestion. We have modified the sentence as: “The dynamic mechanisms of monsoons may differ significantly from region to region, and the responses would be difficult to separate thus cannot be deciphered from the given simulations alone. So we only take the East Asian region to exemplify the convoluted responses and feedbacks due to sulfate forcing.”

15. P23381, l. 5: refer to the “indirect effect” here as the “anthropogenic indirect effect” to give it context

Reply: Revised accordingly.

16. P22382, l. 16: “Note that the internal variability in shortwave radiation and surface temperature from simulations of the A-series are a bit smaller than from N-series, so the about discussion is based on a stricter standard.”... Consider stating this point up front, instead of stating “it will be discussed later” (p.22379, l. 13) earlier in the discussion. Having this knowledge before reading this section would help the reader to accept the statistical comparison more the first time they read it.

Reply: Revised accordingly.

17. P22391, l. 30: The reference for Jockel et al. is out of order.

Reply: Corrected.

18. Figure: the labels on the filled contour plots are very small. At least on the review

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manuscript they cannot be read. They should be larger.

Reply: Revised accordingly.

19. Figure 6: The caption refers to gray shading but the figure is in color. This should be corrected.

Reply: Corrected.

20. There are a significant number of minor grammatical mistakes in this paper. This reviewer suggests that the authors run the final manuscript past an editor for proper English grammar. Given the number of places that should be addressed, they will not be listed here.

Reply: Thanks for pointing that out. We have done our best to re-check and correct the grammatical errors with some help. Hope it is now more readable.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/9/C11505/2010/acpd-9-C11505-2010-supplement.pdf>

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 22365, 2009.

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