Reply to ACPD-9-C7418-2009 comments

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 found 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 responses and internal variability, we increased the length of model results that analyzed from 5 years 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 budges 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 (in Italic) are given below.

1. Page 4: use fixed SSTs greatly limits the climate response in the model, including in particular the hydrological cycle, and this should be noted in section2.1

Reply: Yes, use fixed SST will limit the climate responses. On the other hand, without the convoluted ocean feedbacks, the relationship between sulfate forcing and climate response can be discussed more clearly. We have noted this at the end of section 2.1.

2. Page 5. Table 1: the sulfur mechanism presented in Table 1 seems very old. Isn't DMSO now thought to be a major product of DMS oxidation?

Reply: The reviewer is right in pointing out that DMSO is an important intermediate in the atmospheric oxidation of DMS. The reaction of OH with DMSO may lead to the formation of methane sulfinic acid (MSIA), and MSIA may react with OH to form SO_2 (Kukui et al. 2003). Unfortunately we currently do not have a good idea about how to model exactly the sources and sinks of DMSO. But we have acknowledged in the revision that our model did not consider these reactions, and neglecting them would lead to an underestimation of the SO_2 production from DMS. Since the reaction rate constant for MSIA to SO_2 seems to be of the same order as that for DMS to SO_2 (e.g., Kukui et al. 2003) and the limiting agent is OH, our results should be order-of-magnitude correct. In addition, as we focus on anthropogenic sulfate, the errors associated with this simplification tends to cancel out when the difference between simulations are taken (e.g. A2-N2 or A2-A1). We have noted these in the revision.

(Kukui, A., Borissenko, D., Laverdet, G., and Le Bras, G., Gas-phase reactions of OH radicals with dimethyl sulfoxide and methane sulfinic acid using turbulent flow reactor and chemical ionization mass spectrometry. J. Phys. Chem., 107, 5732-5742, 2003.)

3. Table 1: aqueous-phase reactions should not be represented as equilibrium.

Reply: Thanks for the correction. Revised accordingly.

4. Page 7: I'm confused as to whether the N and A simulations differ solely in their sulfur emissions or in their emissions of NOx, CO and VOCs as well. At several *points, the text makes mention of increased ozone in the A simulations due to anthropogenic emissions. However, the negative radiative forcing for A0-N0 (page 14, Table 4) would then not make sense if it's driven by the ozone increase (page 14) since ozone should have a positive radiative forcing.*

Reply: The N and A simulations are different in sulfur emission only and the NOx, CO, VOCs etc. are the same. To make it more clear, we added this sentence in the first paragraph of Sect. 2.4: "The emissions of other chemical species are kept the same for all simulations following the 1985 scenario so that their photochemistry is essentially identical except when the sulfur species is involved. Note that in the following discussions the term "natural condition" refers to natural sulfate, not the whole chemistry". As was mentioned on p. 22377, line 10-13, the changes of radiative forcing in A0-N0 are not only driven by ozone change but also due to model internal variability. As we have pointed out in Section 4, the responses from A0-N0 should be mostly noises.

5. *Page 7: Doing a single GCM 13-year simulation for each case, with analysis only of the last five years, does not provide very good statistics for climate response. Standard practice is to conduct ensembles of simulations to reduce the effect of noise. This should be at least acknowledged.*

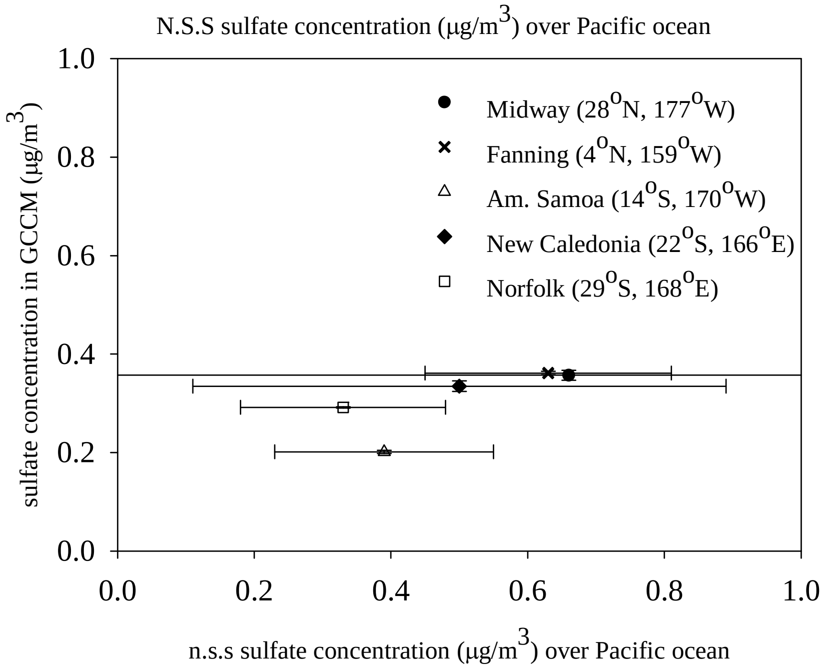
Reply: Yes, we will acknowledge that 5 years of simulation may be too short, and we did not perform ensemble runs. Note that the spin up time needed for GCCM in Wong (2004) is only 6 months. So we went back to re-check the results of the first 8 years and found that all variables remain quite steady from year to year. In order to increase the 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. But the 10-year results are still tentative, so we also commented that and results should be used with caution. Note that we have updated all the tables and figures as well as relevant discussions with the 10-year results.

6. *Page 8: section 3.1 on global distributions of sulfur species is very long and does not contain much that's new.*

Reply: For readers unfamiliar with the chemical component of the atmosphere, some of the information may be essential for their understanding of our later discussions on sulfur cycle and its response to climate feedbacks. In any case, we have tried to trim down the discussions. Note that we have divided Section 3.1 into two parts, with the first part describing the global distributions, and the second part focusing on the simulated sulfur cycle budges.

7. *Page 9: there are actually a number of long-term sulfate data sets in remote air that could be used for model evaluation and these are in fact routinely used by models for that purpose. For example the SEAREX Pacific data come to mind and would be well suited for a model using 1990 emissions.*

Reply: Thanks for pointing out such datasets. We have mentioned a comparison of SO_2 distribution with the GOME satellite data. The Pacific data is certainly a good complement. In the figure below we compared our results with the non-sea-salt sulfate data at the Pacific islands that listed in Saltzman et al. (1986). Our annual means are 12% to 48% lower than the observed values, which might be related to our omission of DMSO related reactions (among other factors). As this section is already quite long, we only included in Sect. 3.1 a short discussion on this and neglected the figure in our revision.



8. *Page 11: I was surprised by the dominant role of ozone as an aqueous-phase* SO_2 *oxidant. Models generally find* H_2O_2 *to be the dominant oxidant. At pH = 4.5 (Table 1), oxidation by ozone should be very slow. I am confused by the units given for kO3 (and the other aqueous-phase rate constants) in Table 1, as these units are for rates and not constants. The reaction between* SO_3^{2-} *and* O_3 *goes at a rate constant close to the aqueous-phase diffusion limit (_1.E9 M-1 s-1), but that's not apparent from Table 1.*

Reply: The reaction rate used in this study is based on Moller (1980) who compiled some published data for comparisons, including Penkett et al. (1974) and Penkett et al. (1979). The reaction rate is about 1.E9 M^{-1} s^{-1} in Penkett et al. (1974), but is 1.E5 to 1.E6 M^{-1} s^{-1} (depending on pH variation) in Penkett et al. (1979). Here we adopt the more recent value given in Penkett et al. (1979).

The units given for k_{O3} (and the other aqueous-phase rate constants) in Table 1 should be L/mol/s instead of mol/L/s. Thanks for pointing out the typo.

As for the production of sulfate by O_3 oxidation to be greater than that from

H_2O_2, we found that the original code that we adopted from Berglen et al. (2004) allows the aqueous reactions to occur also in cloud ice. This will result in more aqueous-phase oxidation by O_3, as H_2O_2 is concentrated in the warm-cloud levels, whereas O_3 concentration is higher at the upper troposphere where the ice cloud is located. We are grateful that the reviewer led us to find to this error. The simulations have been re-run with reactions in ice turned off. The new simulations give sulfate production of 10% by ozone, 52% by H_2O_2 and 24% by OH, which should be more reasonable. In the revised manuscript, all relevant values and discussions have been changed according to the new results, and the main results and conclusions on aerosol forcing and responses are not significantly affected.

9. Page 14, end of 1st paragraph: the argument about robustness seems circular.

Reply: Agree. The end of 1st paragraph on page 14 is rewritten as: Alternative estimation can be derived by (A2-A0)-(N2-N0) forcing for all-sky conditions, which gives a global indirect forcing of -1.87 W m^{-2}. This value is rather close to that from the A2-N2 results, indicating the robustness of its value.

10. Page 15: the authors find an anthropogenic sulfate forcing at the low end of the published range for the direct forcing, and at the high end for the indirect forcing. This seems to call for some explanation. The published AEROCOM intercomparisons provide some metrics for attributing model differences in forcings in terms of differences in model parameters.

Reply: The difference between GCCM and other models might be due to the diversity of models as discussed in AERO COM intercomparison (Textor et al, 2006). In particular, we think the discrepancy is mainly related to the differences in cloud fields, as well as the treatment of cloud effective radii in relation to sulfate loading (see

Section 4) among models. We have added such explanations in the same paragraph.

11. Page 18: a new aspect of this paper is the feedback of climate response on the sulfur cycle, although the use of fixed SSTs limits the response of the hydrological cycle. Page 18 reports very small effects on sulfate: are these effects actually significant (outside interannual variability)?

Reply: Yes, some of the responses (e.g. N2-N0, N2-N1, A2-N1, A2-A0) are significant according to the analysis given in Fig. 7 as well as the t-test results. In fact, these responses are actually quite strong regionally, but appeared to be small when taking global averages. Note that we recalculated these parameters by including simulations of the preceding 5 years (that is, as total of 10 years simulations), the results remain quite similar.

12. Page 24: climate responses given in the conclusions are greatly limited by the assumption of fixed SSTs and this should be stated.

Reply: Yes, we addressed this point again in the conclusion.