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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A study of uncertainties in the sulfate distribution and its radiative forcing associated with sulfur chemistry in a global aerosol model" *by* D. Goto et al.

Anonymous Referee #1

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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. 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. 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).

Specific comments: 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.

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.

Chapter 4.5: To evaluate the dry deposition of SO2, not only sulfate but additionally SO2 concentrations should be compared to observations.

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.

Table 5: According to the results of the COSAM comparison, wet deposition of SO2 is of relatively little importance in the regional budget of SO2, whereas in the current study (OS model version) 22% of the SO2 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 SO2 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.

Page 12290 ln 7-8: "larger sulfate distributions“ shouldn’t it be "larger sulfate concentrations“?

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

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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?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 12269, 2011.