

## ***Interactive comment on “Tropospheric aerosol microphysics simulation with assimilated meteorology: model description and intermodel comparison” by W. Trivitayanurak et al.***

**W. Trivitayanurak et al.**

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Thank you for your valuable comments. We would like to address the comments by first giving an overall response, which clarifies our intercomparison objectives, and then we respond to each comment.

There are mainly two styles of model intercomparison. In the first, all models are constrained in all inputs/processes except the one part being focused on. In the second, each model is taken "as is" and the differences represent range of uncertainty the modeling community are currently facing. Each has its own merits, although the first type is obviously more time consuming. It requires more simulations, more diagnostics, and more analysis to nail down all the differences. Ultimately, such an intercomparison

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would be useful but would be most beneficial when ~6 or more models participate.

The major goal of this paper is still to document the inclusion of TOMAS into GEOS-CHEM. Therefore, the limited model intercomparison we did was of the second type for logistical reasons: we want to use model results readily available from similar two-moment aerosol microphysics models. Another reason is we want to sample real models, each developed independently and producing plausible predictions, and analyze what are the differences and similarities of their predictions.

Another overall response is we appreciate the referee's attention on the lack of discussions about several differences among models which affect the intercomparison, for example, the different oxidant fields. Therefore we revise our manuscript by adding corresponding discussions in the model intercomparison section.

Now we will address each comment.

For comments by Referee 1:

Point 1. "...for the model intercomparison, similar oxidant fields for each model would suit better, since there are already a number of other differences in the models. If simulations are done with different fields, there could be a few words about possible differences caused by these fields."

The simulations were done with different oxidant fields, namely GLOMAP uses mean oxidant fields archived from TOMCAT tropospheric chemistry simulation, GISS GCM-II' uses tropospheric chemistry simulations (C. Spivakovsky pers. comm. referenced in Koch et al., 1999; Wang et al. 1998), and GEOS-CHEM simultaneously calculates oxidant concentrations together with production and loss of other species and represents realistic oxidant concentrations. We added the following discussion of these details in Section 2.1.7, which now reads:

... Additionally, the GISS GCM II' uses prognostic H<sub>2</sub>O<sub>2</sub> calculated from archived average photolysis rates and uses archived average oxidant fields for OH, HO<sub>2</sub>, and NO<sub>3</sub>

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(Koch et al., 1999 and references therein) while GEOS-CHEM, in this work, uses the option to calculate and update the oxidant fields simultaneously with photochemistry.

...

and

... In this version of GLOMAP, oxidant (OH, H<sub>2</sub>O<sub>2</sub>, HO<sub>2</sub>, and NO<sub>3</sub>) concentrations are specified using monthly mean fields archived from TOMCAT detailed tropospheric chemistry simulations. ...

On a short (~hourly) time scale, some differences will be caused by having realistic (interactive) oxidant availability versus mean oxidant values. Since our focus is not on such short time scale but for annual average, the different oxidant fields may result in overall faster oxidation in one model than others if there are generally higher concentrations of oxidants. This average time scale difference is summarized in the form of SO<sub>2</sub> lifetime with respect to oxidation and these values (shown below for GEOS-CHEM with TOMAS, GEOS-CHEM with bulk aerosol, GISS GCM-II', and GLOMAP, respectively) are added to the global annual sulfur budgets (Table 2) under SO<sub>2</sub> "Oxidation lifetime (days)".

We present these calculated lifetimes to show the abundance of oxidants where they participate in gas and aqueous phase reactions with SO<sub>2</sub>. Oxidation is fastest in GLOMAP implying that oxidant concentrations are highest in regions with abundant SO<sub>2</sub>.

We also added a discussion about different oxidant fields in the context of sulfur budgets in the second paragraph of Section 5.3, which now reads:

... The effect of different oxidant fields is demonstrated in the oxidation lifetime of SO<sub>2</sub> showing that GLOMAP has the most abundant oxidants in regions with high SO<sub>2</sub>, both OH and H<sub>2</sub>O<sub>2</sub>, for reacting with SO<sub>2</sub> while GISS GCM-II' has the slowest oxidation. The fact that GLOMAP is the only model in this intercomparison that uses prescribed

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average H<sub>2</sub>O<sub>2</sub> concentration and has the fastest oxidation agrees with the findings by Roelofs et al. (1998; 2001) that models applying full chemistry or prognostic approach tend to have lower sulfate oxidation by H<sub>2</sub>O<sub>2</sub> because they represent oxidant limited conditions more realistically.

Point 2. "In section 4 (and Fig. 5) comparison with observations shows a clear underprediction of CN at latitudes 15S-60S. It is pointed out that this could be due to sea-salt emission or the lack of carbonaceous aerosols, and that addition of carbonaceous would only have a minor effect. The sea-salt emissions of GEOS-CHEM are already higher than those in other two models (Table 2), still GEOS-CHEM has generally lowest CN in this comparison. Is the sea-salt emission parameterization the main error source, or is there another source of CN that is missing?"

After additional investigation, we can confirm that the different sea-salt emission rates in the different models is the main source of discrepancy. While GEOS-CHEM has the highest sea-salt emission globally, we found that GEOS-CHEM sea salt emission exceeds the other two models primarily in areas not included in these grid boxes selected for comparison with the Heintzenberg et al. (2000) dataset. Therefore, it is not expected that CN<sub>10</sub> concentrations in GEOS-CHEM will be greater than the other models. Additionally, deposition and vertical transport play roles. For example, vertical transport in GLOMAP seems to bring significant amount of CN<sub>10</sub> from the tropical FT down to the surface in the southern latitudes as presented in Figure 7c.

Point 3. "Why are the GEOS-CHEM results for year 2001 and GLOMAP results for year 2006? It is a good idea to include a GCM with its own meteorology in the comparison, but for CTM comparison the use of different meteorological fields takes away the benefit of using a CTM. In addition, the used meteorological fields should be from the year when most of the observational data was collected."

I think referee#1 meant GLOMAP results for year 1996. For an ideal controlled inter-comparison, we would want to compare with the same simulation year for GLOMAP

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and GEOS-CHEM, but we felt it is an acceptable compromise to use results of GLOMAP with sulfate-seasalt aerosols from 1996 which was already available and was published. GEOS-CHEM's choice of year 2001 is for a later comparison with ACE-Asia field campaign carried out in 2001. Being aware of the different years, we tried to focus on the annual average results but still aware of possible causes of differences, e.g. one being a wetter year as added in the discussion in paragraph 6 of Section 5.3 which now reads:

Again faster wet removal in GLOMAP suggests that year 1996 may be a wetter year than 2001 of GEOS-CHEM simulation. Low microphysical growth in GEOS-CHEM is ...

We also cite a finding from a new GLOMAP study that finds 7-14% fractional difference of CCN(0.2%), comparing different years, due to interannual variability in wind speed and thus sea salt. Now the end of the second paragraph of Section 5.1 reads:

... Interannual variability in wind speed can contribute around 7 - 14% difference in CCN(0.2%) over SH oceanic area, as found in a different study using GLOMAP (Korhonen et al., in press). Thus 30% discrepancy shown in marine ML CCN(0.2%) ratios of GLOMAP-to-GEOS-CHEM, despite comparing different years, suggests involvement of several factors, e.g. meteorology, emission parameterization, aerosol microphysics.

Point 4. "...it would be beneficial to include detailed nucleation scheme in a global model. The nucleation treatment presented in the paper is based on rather old nucleation rate calculations, and could easily be replaced with for example Vehkamäki et al. (JGR, 2002) binary sulphuric acid-water nucleation parameterization, that can be used for a wide range of atmospheric conditions..."

Implementing a more sophisticated nucleation scheme into TOMAS involves overcoming a significant computational challenge compared to the current scheme. Separate work in our research group has developed a novel approach to speeding up the nucleation-condensation calculation. This is being described in a separate paper.

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Point 5. "Figures 6 e-f present an interesting result: for the Antarctica region, GLOMAP seems to predict a magnitude more of CN than GEOS-CHEM, but a magnitude less of CCN(0.2%). It was not mentioned if GLOMAP CN in this figure includes the nucleation mode (1-10 nm) particles, which would partly explain the figure."

The comparison in Figure 6 is for the same size range for all models. To prevent confusion about the size range of CN, we defined CN10 as total number concentration of particle with  $D_p > 10$  nm and use CN10 throughout the document. "CN10" is defined in the beginning of Section 3.3. The inter-model differences in Antarctic CN and CCN is partly due to the vertical transport of Aitken mode particles from the tropical FT down to the Antarctic region, which is particularly strong in GLOMAP (Figure 7c and Figure 7g). Another reason is the aerosol microphysics of GLOMAP that calculates high nucleation rate over the Antarctic where there is high elevation and low temperatures. However, this nucleation over Antarctic surface does not happen in TOMAS microphysics in GISS GCM-II' and GEOS-CHEM. The particularly lower CCN(0.2%) over the Antarctic in GLOMAP then is a consequence of having high ultrafine particle concentrations but low growth rates to CCN sizes because of low SO<sub>2</sub> concentrations. The added discussion is in the first paragraph of Section 5.1 which now reads:

... GLOMAP exhibits notably higher CN10 and particularly lower CCN(0.2%) over Antarctica compared to GEOS-CHEM and GISS GCM-II'. These high CN10 concentrations are due to high elevations over Antarctica with corresponding cold temperatures resulting in high nucleation rates calculated by GLOMAP especially in the winter months as presented in Figure 11(a) of Spracklen et al. (2005a). The limited availability of condensable vapor means that these particles do not grow significantly to become CCN(0.2%). Elevated ultrafine concentrations over the Antarctic surface is a behavior that GLOMAP exhibits but not present in TOMAS as shown in Figure 7. ...

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For comments by Referee 3:

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"1. Gaining insights from model comparison is a challenge because models can have very different treatment of physics, chemistry, numerics, etc. This study makes the task even more difficult by using different emissions and different simulation years (in the case of GEOS-CHEM and GLOMAP). I recommend the authors redo the intermodel comparison using the same emissions (at least for non sea-salt) for all three models and simulation year (i.e. same meteorology) for GEOSCHEM and GLOMAP. In addition, the intercomparison would be more informative if the same oxidant fields are used for all three models."

Along the same line as the overall response, we preserve the intermodel differences for practical purposes as well as to investigate the real model differences that reflect the range of uncertainties of global aerosol models. The added discussion is in the sixth paragraph of Introduction, which now reads:

... An exhaustive model intercomparison activity would involve numerous models and tightly constrained scenarios designed to isolate the effects of different processes and inputs. A more limited model intercomparison compares models as they are and looks at the intermodel discrepancies to indicate the range of uncertainty currently facing the scientific community. This work presents this more limited style of intercomparison. ...

"2. A table listing the similarities and differences between GEOS-CHEM, GISS GCMII', and GLOMAP models would be helpful to the reader. Some differences are never mentioned in the text (though some can be inferred from Table 1, e.g. GLOMAP does not include MSA); the differences can be listed explicitly in the new table."

We have added a table listing the major differences and similarities of the three models in Table 1 and introduce Table 1 at the end of Section 2.1.7.

"3. The paper should address the differences between using monthly-averaged oxidant fields and using online photochemistry in GEOS-CHEM. Also, it is not clear if the monthly-averaged oxidant fields used for GISS GCM-II' and GLOMAP are averaged from the online results of the GEOS-CHEM simulation or from some other sources.

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If from other sources, how different are the oxidant fields (in particular OH and O<sub>3</sub>) among the three models?"

Please refer to the response to point 1 of Referee 1 above. Since the referee#3 mentioned O<sub>3</sub>, we should point out that each model applies different set of chemical reactions and GEOS-CHEM is the only model in this comparison that includes the oxidation by O<sub>3</sub>, which in fact is a minor pathway.

"4. The lifetime of DMS calculated by GISS GCM-II' is about 3 times greater than the results of GEOS-CHEM and GLOMAP. Is the difference due to different OH fields or something else?"

The difference is due mainly to the different OH and NO<sub>3</sub> fields and partly to the rate constant used. This discussion is added to the end of the second paragraph in Section 5.3 and now reads:

... Also, DMS oxidation in GISS GCM-II' is about three times slower compared to other models and the majority of the difference in lifetime is the different oxidants while the different rate constants over different temperatures contribute to about 10% discrepancy. Nevertheless DMS emissions between models are similar therefore the SO<sub>2</sub> production from DMS is also similar.

"5. A detailed discussion of the similarities and differences of the moving-center scheme used in GLOMAP and the TOMAS module is needed. What are the pros and cons of the two modules? How much of the differences in the results between GLOMAP and GISS GCM-II' are due the difference in the simulation of aerosol microphysics?"

The difference between TOMAS two-moment sectional scheme and the moving-center scheme is essentially how the aerosol size distribution is represented \*within\* each size section. We have added a short discussion regarding the different schemes. We feel it is out of our scope to discuss in detail the similarities and differences of the two mi-

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crophysics schemes. Both the TOMAS microphysics and the moving-center schemes have been evaluated in the past by comparing against analytical solutions and proven to provide excellent accuracy as well as conserve the two moments (Tzivion et al., 1987; Jacobson et al., 1995; Jacobson, 1997). Note that for TOMAS, the evaluation in Tzivion et al. (1987) was for the numerical solution to the stochastic collection equation developed for rain drop microphysics in that study but later applied to work for aerosol microphysics by Adams and Seinfeld, 2002. It is fair to say that the discrepancy caused by these different microphysics schemes is far smaller than discrepancies from other model elements, e.g. chemistry, precipitations. The added discussion is in the forth paragraph of Section 2.1.7 and now reads:

Although GLOMAP and GEOS-CHEM with the TOMAS microphysics are models developed independently, there are several similarities. Both use assimilated meteorology. While both TOMAS and the moving-center algorithms are two-moment sectional approaches, they differ in the closure assumptions used to solve the aerosol general dynamic equation. TOMAS treats the aerosol size distribution within a section with either a "top hat" or "linear" approximation (Stevens, 1996; Tzivion et al., 1987; Tzivion et al., 1989) but the moving-center approach treats the size distribution as monodisperse within the bin (Jacobson, 1997b; Jacobson et al., 1994). Both the two-moment sectional treatment of TOMAS and the moving center treatment in GLOMAP are high-resolution sectional treatments of the aerosol size distribution that have been evaluated against analytical solutions and found to be very accurate and guarantee that both number and mass balance equations are satisfied (Jacobson, 1997b; Jacobson et al., 1994; Tzivion et al., 1987). Any difference in aerosol prediction caused by these different microphysics schemes should be insignificant compared to the uncertainties of other model components, e.g. chemistry, deposition.

"6. The three models used in the study have very different vertical resolutions, which can lead to very different vertical transport of species from the boundary layer to the free troposphere. The paper needs to address how these differences impact the model

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intercomparison."

There are two issues involved here, vertical resolution and vertical transport scheme. Having higher vertical resolution may translate into the ability to present details and variability but more importantly vertical transport of species is transport scheme dependent. To address differences of the vertical transport schemes of each model, which encompass several processes, e.g. vertical advection, diffusion, dry and moist convection, and scavenging with convective updrafts and precipitations, we turn to published studies. Though several studies in the past evaluated vertical transport of each model using one or more or the combination of  $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$ , and  $^7\text{Be}$  as tracers (Liu et al., 2001; Koch et al., 1996; Rind and Lerner, 1996; Allen et al., 1996; Stockwell and Chipperfield, 1999; Lohmann et al., 2001; Jacob et al., 1997), it is still difficult to summarize the performance of each vertical transport scheme for several reasons including the uncertainties in the sources of these species, uncertainties in the measurement, the limited coverage of measurements at altitudes, and the imperfection in the meteorological inputs. Nevertheless, all models were able to reproduce measured concentrations to within a factor of 4. The added discussion regarding this issue is in the third paragraph of Section 5.2, which now reads:

... Different vertical transport schemes definitely play a part in the discrepancies. Several studies in the past evaluated vertical transport of each model using one or more or the combination of  $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$ , and  $^7\text{Be}$  as tracers (Allen et al., 1996; Jacob et al., 1997; Koch et al., 1996; Liu et al., 2001; Lohmann et al., 2001; Rind and Lerner, 1996; Stockwell and Chipperfield, 1999) and yet vertical transport remains an important uncertainty. Nevertheless, all models were able to reproduce measured concentrations to within a factor of four. Though we cannot postulate the exact causes of discrepancy here, several factors definitely contribute including microphysics schemes, precipitations, and meteorological inputs.

One of the main conclusions from the COSAM model intercomparison was that surface concentrations predicted by different models agree mostly within a factor of two but

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column burden and vertical profiles vary greatly among models (Lohmann et al., 2001), similar to our results. We added this discussion and a citation in the third paragraph of Section 5.2, which reads:

Model-to-model differences increase as one moves upward from the surface; a similar result was found in the COSAM intercomparison of sulfur models (Lohmann et al., 2001). ...

We also add a discussion regarding the different locations of in predicted peak CN10 in Figure 7 that associates with the different vertical transport of SO<sub>2</sub>. This discussion is added in the first paragraph of Section 5.2 and now reads:

...Also, the peak CN10 occurs at different altitudes for each model, i.e. 150 and 100 mbar for GEOS-CHEM and GISS GCM-II', respectively, and 100 and 300 mbar for GLOMAP; this could be because the different temperatures and relative humidity predicted in each model as well as the ability of different vertical transport schemes of each model to transport SO<sub>2</sub> up to the free troposphere.

As for the impact of different vertical resolutions, we observe the difference in SO<sub>2</sub> dry deposition that can be explained by the different vertical resolution and is consistent with the comment by Roelofs et al. (2001). This discussion is added in the third paragraph of Section 5.3, which now reads:

Dry deposition is the most important sink for SO<sub>2</sub> and is modeled using similar resistant-in-series approach in all models. Despite similar treatment of gas dry deposition, the GISS GCM-II' shows a higher dry deposition lifetime compared to GEOS-CHEM and GLOMAP. This difference can be attributed to the lower vertical resolution in GISS GCM-II' and is consistent with the findings in the COSAM intercomparison that models with higher vertical resolution apparently weaken mixing between the boundary layer and the free troposphere, thereby enhancing the impact of dry deposition of sulfur species (Roelofs et al., 2001).

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"7. Wet deposition is the dominant sink for sulfate; and wet deposition is strongly dependent on the modeled cloud and precipitations fields. A comparison of the modeled cloud and precipitation fields is needed."

The net effect of precipitation on our aerosol simulations is reflected in the wet deposition lifetime. The line shown below with values for GEOS-CHEM with TOMAS, GEOS-CHEM with bulk aerosol, GISS GCM-II', and GLOMAP, respectively, has been added to Table2 under SO42- "Wet deposition lifetime (days)".

The wet deposition lifetime of the GEOS-CHEM bulk aerosol model is shorter because bulk aerosol was subjected to more removal compared to the size-resolved aerosols; this discussion was already in Section 3.1. Additional discussion is added to paragraph 4 of Section 5.3 which reads:

... As for SO42- sinks, all models consistently indicate that wet deposition is dominant. The SO42- wet deposition lifetime, summarizing the effects of different cloud and precipitation fields in each model, is shown in Table 2. GLOMAP has the most active removal by precipitation and GISS GCM-II' has the slowest wet removal despite the largest removal rate.

"8. One of the stated motivation of the intercomparison is to suggest observations required to eliminate intermodel discrepancies, but the paper makes no recommendations regarding what data are needed to help evaluate and improve the models." As is generally true, models agree better near the surface because there are more surface observations to constrain models. Observations that can help constrain models and eliminate intermodel discrepancies are aircraft measurements of aerosol microphysics, including size distributions and compositions. There should be more semi-long term aircraft data sets with appropriate microphysical measurements (e.g. CN, size distributions). This discussion is added to the last paragraph of Section 6, which now reads:

These intermodel discrepancies at altitudes point out the need for more and longer term aircraft measurements of aerosol microphysical properties. Global annually aver-

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aged budgets show....

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