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Geoscientific Model Development Discussions

Interactive comment on "The Sectional Stratospheric Sulfate Aerosol module S3A-v1 within the LMDZ general circulation model: Description and evaluation against stratospheric aerosol observations" *by* Christoph Kleinschmitt et al.

Anonymous Referee #1

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This paper describes a new microphysical aerosol module "S3A-v1" within the LMDZ GCM for stratospheric applications, and evaluates it against ambient and volcanic observations. This is valuable research, as stratospheric aerosols are important for climate and chemistry, and can have small (ambient) impacts or large perturbations such as volcanic eruptions or hypothetical geoengineering schemes. Additionally, as the authors note, due to the long lifetime, the growth and microphysical processes are complex and important, and most modeling studies lack at least one important process.

The model disclosed in this paper should be useful to advance our understanding of stratospheric aerosols.

This is a well-written paper with clear structure, meaningful utility, promising results, and good grammar. I have just a couple general suggestions and numerous minor specific suggestions to be considered before publication.

General Minor/Moderate suggestions:

1) Additional comparisons to observations can help strengthen the paper and understand how robust your model is. A few suggestions: a) Compare vertical profiles of sulfate aerosol mixing ratio taken by aircraft (Borrmann et al 2010; also applied in English et al. 2011 figure 10). b) Comparison to observations of sulfuric acid concentrations. (e.g. balloon data applied in English et al 2011 Figure 5c). c) Is there UTLS and stratospheric temperature data after Mt Pinatubo eruption (to test your aerosol radiative heating code)?

2) I suggest adding van der Waals forces to your coagulation scheme to improve simulations of ambient aerosol. In your Figure 5, small particles are overestimated and large particles are underestimated by about an order of magnitude in the middle/upper troposphere. Inclusion of van der Waals forces would significantly reduce this bias. This was investigated in the WACCM/CARMA model (English et al. 2011). WACCM/CARMA with van der Waals forces included in the coagulation scheme had a much better match to aerosol size distribution than your model (Fig 9 in English et al. 2011). This was concluded to be due to van der Waals forces; the experiment without van der Waals forces overestimated particle number (Fig 10 in English et al 2011), similar to your model. (Neither model includes meteorites, suggesting that not including meteorites is not the problem). This should improve your comparisons in your Figure 5 and 11, and would be a nice improvement to your model. Also, you could compare vertical profiles of mixing ratio (Borrmann et al 2010). Also, you could compare Pinatubo simulations to other studies that looked at van der Waals forces (English et al. 2013, Sekiya et al. 2016), al-

though as you mention, the observed variability is too large to conclude with confidence whether including van der Waals forces improves volcanic simulations. Most likely, van der Waals forces is important to match ambient observations of number concentration though.

3. Using prescribed chemical lifetimes is probably fine for ambient aerosol, but could be problematic with large volcanic eruptions or geoengineering. I suggest you clarify these caveats and either do some evaluations that conclude that prescribing the oxidants doesn't cause too many errors in your results, or state that this current model setup is not recommended for very large volcanic eruptions or geoengineering. Also, cite Bekki 1995 for evidence of limited oxidants. (It's reassuring that your results are reasonable for Pinatubo, but unclear whether your model is safe to use for eruptions larger than Pinatubo or for geoengineering)

Specific suggestions:

Abstract: 1) Add that stratospheric aerosols impact chemistry as well as radiative forcing 2) Describe your model in more detail (it computes aerosol radiative heating and QBO but has semi-prescribed chemistry). 3) Remove the word "all" from relevant microphysical processes, as certainly *some* are missing, such as van der Waals forces. 4) Add a few more details of your results, e.g. number of ambient large particles are underestimated/small particles overestimated (which may be due to not including van der Waals forces)

Introduction Line 21: Remove the word "quite"

2.1.1 Line 20: Does Hourdin et al 2006 evaluate the model for stratospheric applications? If not, I suggest adding a subsection where you describe and evaluate some of the stratospheric processes in your model to ensure it is behaving reasonably, such as looking at strat age-of-air, trop-strat transport processes, seasonal/latitudinal temperatures. It is nice that you looked at QBO.

2.1.2 Line 8: Say why you only compute nucleation, condensation etc in the stratosphere (to save computational time? or is a different aerosol module used in the troposphere?)

2.1.2 Line 10: Do you compute the tropopause level at each timestep? Please clarify.

2.2.1 Line 19: I suggest put these paragraphs in a new section called "Chemistry".

2.2.1 Line 31: Please add some discussion regarding my general concern #3 here.

2.2.2 Line 5: The statement that coagulation and growth, not nucleation, determines particle size distribution is a main conclusion from English et al. 2011.

2.2.5 Line 13: Please add discussion of the possible importance of van der Waals forces to accurately model ambient aerosol. Results by English et al. 2011 (see Figure 9) suggest van der Waals forces is very important to get ambient aerosol correct (likely more important for ambient aerosol than for large perturbations such as volcanic eruptions).

2.3 Line 23: "widely tested by the authors". are there any peer-reviewed papers to cite?

3.1 Line 29+: (discuss the likely importance of van der Waals forces). Also, since English et al. 2011 also didn't have meteoritic dust in their model, but the size distribution looks good, the lack of meteoritic dust is not likely a source of error for particle size/mixing ratio (but probably is a source of error for burden/extinction).

3.2 Line 21: Cite Aquila et al 2012 who also investigated the relationship between injection height and Pinatubo accuracy when aerosol heating is involved (How do your injection height results compare)

3.2 Line 26: Is this data published anywhere yet?

Conclusions Line 30: Please add discussion about whether your model is safe to use for perturbations larger than Pinatubo and/or for hypothetical geoengineering schemes

given that oxidants are prescribed.

Figure 5 and Figure 11: It is difficult to distinguish the model and observation lines, especially the dark blue lines. Can you change the colors or markers to better distinguish?

Figure 7: Add to the caption the source of the CMIP6 aerosol data set. Please add a header to each row and column describing each panel.

References

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Bekki, S. (1995), Oxidation of volcanic SO2: A sink for stratospheric OH and H2O, Geophys. Res. Lett., 22(8), 913–916, doi:10.1029/95GL00534.

Borrmann, S., Kunkel, D., Weigel, R., Minikin, A., Deshler, T., Wilson, J. C., Curtius, J., Volk, C. M., Homan, C. D., Ulanovsky, A., Ravegnani, F., Viciani, S., Shur, G. N., Belyaev, G. V., Law, K. S., and Cairo, F.: Aerosols in the tropical and subtropical UT/LS: in-situ measurements of submicron particle abundance and volatility, Atmos. Chem. Phys., 10, 5573–5592, doi:10.5194/acp-10-5573-2010, 2010.

Sekiya, T., K. Sudo, and T. Nagai (2016), Evolution of stratospheric sulfate aerosol from the 1991 Pinatubo eruption: Roles of aerosol microphysical processes, J. Geophys. Res. Atmos., 121, 2911–2938, doi:10.1002/2015JD024313.

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