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**Geoscientific
Model Development
Discussions**

Interactive comment on "Aerosol microphysics modules in the framework of the ECHAM5 climate model – intercomparison under stratospheric conditions" *by* H. Kokkola et al.

Anonymous Referee #1

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Referee report on the manuscript

Aerosol microphysics modules in the framework of the ECHAM5 climate model - intercomparison under stratospheric conditions

by

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I suggest to publish the manuscript in Geoscientific Model Development after addressing the comments and suggestions given below.

General comments

The authors present a comparison of aerosol size-distribution and effective radius of sulfate particles predicted by four different aerosol modules. They consider three different scenarios for volcanic SO_2 under stratospheric conditions. The study is interesting and worth publishing but needs to be revised carefully. Many things remained unclear to me and the current manuscript difficult to read. Imprecise statements need to be rephrased and missing information should be added. The language needs to be checked very carefully. American English and British English spelling should not be mixed.

- Model setup: It is unclear to me with what kind of model the experiments were performed. Boxmodel? Column model? Etc.? This is important and needs to be clarified.
- The neglect of "non-microphysical" sink processes such as gravitational settling is inappropriate for coarse particles ($>$ 1 μm) and integration time of 10 days. Uncertainties introduced by this neglect need to be estimated or the experimental design adjusted.
- The study focuses solely on volcanic SO_2 emissions. Is volcanic dust also relevant?
- What do the findings imply for application of the aerosol modules in climate models? How important are the differences in effective radii for calculation of radiative fluxes? How sensitive are the finding to the assumptions made (e.g. size distribution of stratospheric background aerosol)?

- It would be interesting to see the differences in surface area between the individual aerosol modules as surface area is highly relevant for heterogeneous chemistry.

Specific comments

p.212, l.11-14, "...relatively low...", "...are characterized by mean radii at least a magnitude smaller than..."

→ Please be more precise, give numbers.

p.212, l.27, "... human-induced artificial sulphate aerosol..."

→ There is no artificial sulfate aerosol even though it is formed by anthropogenic emissions of precursor gases or possibility emitted directly as primary particles.

p.212, l.29, "...larger particles scatter less visible light than smaller particles..."

→ This is not necessarily true. In fact, scattering of visible light has a maximum in the size range of the accumulation mode. Light scattering is a function of particle surface area, i.e. smaller particles (Aitken mode, nucleation mode) scatter less visible light than larger particles (accumulation mode). Total surface area is usually dominated by the accumulation mode.

p.214, l.25-26, "...the model resolves the concentrations of aerosol particles containing up to 21 H_2SO_4 molecules individually."

→ What do you mean by this? Please clarify and consider rephrasing.

p.215, l.9, "...uptake and loss by large aerosol particles derive from the liquid drop model and H_2SO_4 and H_2O vapor pressures over bulk solutions..."

→ I don't understand what you are trying to say.

p.215, l.10-11, "...that uses data from..."

→ What data? Please be more specific.

p.215, l.12, "The thermodynamic data..."

→ Again, data for what? Please be more specific.

p.215, l.17-18, "This simplification holds well in the troposphere..."

→ Your study focuses on the stratosphere. Is this approach still valid in the stratosphere? What are possible implications? This is important as chose this model as reference model.

p.216, l.7-8, "...only sulphate is treated, the insoluble modes are not used in the simulations."

→ The insoluble modes might provide surface area for condensation of H_2SO_4 vapor. Is insoluble aerosol from volcanic eruptions relevant in the stratosphere? If so, the omission of these modes might bias the results and needs to be addressed then.

p.216, l.24-26, "...four externally mixed size sections - soluble and insoluble - per size section consisting of sulphate, organic carbon, ..."

→ I don't understand what you are saying. Please rephrase.

Section 3

→ Please give also the original M7 integration scheme.

p.221, l.5-6, "The conditions for the three cases are given in Table 2."

→ These parameters rather reflect tropospheric than stratospheric conditions. Since this study focuses on the stratosphere and conditions after a massive volcanic eruption, one or two more cases reflecting stratospheric background conditions and a high volcanic emission case would allow to estimate how relevant the new time integration scheme for M7 is for this study. It also seems to make limited sense to me to use p=1013 hPa for all cases even though it is clear that you typically won't find T=225 K near the surface. I suggest to investigate five cases: troposphere (lower, mid and upper), stratosphere (background and high volcanic emission scenario).

p.221, l.6, "... ion pair production rate..."

→ What do you mean by ion production rate?

Section 3.1

→ I suggest to show also relative errors for typical time steps of GCMs as those are more relevant than absolute errors to evaluate the performance of the different time integration schemes.

Section 4:

→ The exact model setup is unclear to me. What kind of model did you use (box model, column model, ECHAM5 (stratosphere), etc.)? In case of a box or column model, what boundary conditions did you use? Was the box initialized and then isolated from its environment?

→ Which abstracted diurnal cycle for OH did you specify? Constant concentrations during the day and zero during the night? Such a square wave signal might introduce numerical oscillations and unrealistic behavior at the transition between night and day. E.g. a sinusoidal diurnal cycle might be more appropriate.

p.222, l.25-27, "The extreme case mixing ratio..."

→ How does your extreme case scenario compare to past volcanic eruptions? Please try to put your scenario into context.

p.223, l.20-21, "...gas-to-particle conversion of sulphur."

→ You probably mean sulfuric acid vapor?

p.223, l.24

→ Insert "ratio" after "mixing".

p.225, l.13-14, "Because the evolution of the size-distribution become more rapid yielding to steeper gradients in the aerosol concentrations."

→ I do not understand what you are saying. Please rephrase.

p.225, l.16, "...a detached bimodal..."

→ What do you mean by detached?

p.225, l.19-20, "Notable effects are seen here for fine particles and, in particular for M7 setup 1, also for medium size particles."

→ Which effects? Please be more specific. What do you mean by small and medium size particles? Please give size-ranges.

p.225, l.22-23

→ You probably mean the size distribution calculated by MAIA shows two narrow and well separated modes?

p.226, l.1, "Here we look in detail to the results for the results given by M7..."

→ ?? Please rephrase.

p.226, l.4, "Simulations with M7 were done using time step of 60s."

→ You are comparing aerosol modules available for use with the GCM ECHAM5. Thus, a time step of 10 minutes would be more relevant.

p.226, l.7

→ Change "value" to "concentration".

p.226, l.27-28

→ A unimodal size distribution has been used for initialization. How does a coarse mode form "after relatively short time scales" and what do you mean by "short time scales"?

p.226, l.29

→ What do you mean by "geometric specifications"?

p.227, l.21

→ Change "raise" to "increase".

p.227, l.24, "...visible size range of such remote sensing instruments..."

→ What do you mean by "visible size range"? Size range of aerosol particles detectable by remote sensing instruments?

p.228, 8-12

→ It is not surprising that evolution of the effective radii given by the individual aerosol modules varies significantly as the modules cover different size ranges. I suggest to look at the most relevant size range, e.g. 0.05-1 μm only.

p.228, l.16

→ This is not necessarily true for case 3.

p.228, l.22-23, "The aerosol mass in the modules constantly increases since we neglect the non-microphysical sink terms in this experiment."

→ The neglect sink processes such as gravitational settling is inappropriate for coarse particles (> 1 μm) and the integration times shown here (10 days). A growing coarse mode acts as condensational sink for H_2SO_4 possibly altering the evolution of the aerosol size distribution in an unrealistic way. The uncertainties introduced by this neglect need to be estimated or the experimental design should be adjusted. This is also important for calculation of the effective radii since a large fraction of total aerosol mass can be present in coarse particles.

p.229, l.1, "...the rates of change evolving R_{eff} are smaller during night."

→ I do not understand what you are saying. Please rephrase.

p.229, l.2-3, "...depends on module specific definitions."

→ Again, this is not surprising as the modules cover different size-ranges. I suggest to look at the most relevant size ranges for radiation only.

p.229, l.11, "for the calculation of the flux only the median radii of the modes are of interest"

→ This is not necessarily true. The condensational flux depends on total surface area and thus on particle number and median radii.

p.229, l.12-13, "When during night, the nucleation mode particle concentration tends to zero, the available gas is transferred to higher modes only."

→ How can this be if no OH is available during night hours? Oxidation of SO_2 by OH is the only production of H_2SO_4 taken into account. Without new production,

H_2SO_4 in the gas-phase should quickly condensate onto all pre-existing particles as condensation is usually faster than removal of nucleation mode particles by coagulation. Please be more specific and distinguish between processes occurring after "sunset" and later during the night.

p.229, l.15, "...when SO_2 increases..."

→ This is misleading as SO_2 is initialized in the beginning and should be decreasing afterward. Did you mean "when initial SO_2 concentrations increase..."?

p.229, l.16, "...SAM2 tends to bridge..."

→ What do you mean by "bridge"?

p.229, l.18-22, "Thus the lowermost range of the predicted size distribution strongly varies depending on the availability of sunlight (...)."

→ Another reason to focus on the relevant size range when calculating effective radii. If the diurnal cycle is so important, I would make sense to calculate effective radii only for daylight conditions as no scattering or absorption of visible light by aerosol particles takes place during night. Daylight averages (6-18h) instead of 24h averages would be more relevant then.

p.229, l.28, "This characteristic is pronounced..."

→ I do not understand. Which characteristic of what?

p.229, l.29, "...since the signal-to-noise ratio is much weaker than under volcanic conditions."

→ I would expect this to be the other way round as less very small particles are formed by nucleation in case of low SO_2 concentrations. Please explain.

p.230, l.7-8, "...this does not apply to SALSA and SAM2 in the volcanic case"

→ This needs to be explained. Please give reasons.

p.230, l.9, "...miscellaneous integration ranges..."

→ Please be more specific and give numbers.

p.230, l.14-18

→ What do you mean by "facing derived size parameters"? Comparison of modeled and observed size parameters? Which parameters? Why is the effective radius so sensitive to particle growth? You just showed that it is sensitive to the lower cut-off size because of new particle formation by nucleation. Please give more details and explain.

p.230, l.21, "...slightly increased stratospheric SO_2..."

→ It is misleading to call an increase in SO_2 concentrations of a factor of 2000 compared to background levels "slightly increased". Please avoid such phrases and give numbers instead.

p.230, l.23-24, "...the calculated effective radii are almost overlapping in the mode setups 1 and 2."

→ I do not understand your statement. Please rephrase.

p.231, l.2, "...shape of the evolution of the effective radius..."

→ What do you mean by "shape of the evolution"?

p.231, l.10-11, "This is accompanied by moderate numerical diffusion, resulting in a smaller diurnal cycle of the effective radius."

→ What do you mean by "accompanied by moderate numerical diffusion? Why should this have an impact on the diurnal cycle of the effective radius? How does the diurnal cycle of the effective radius look like?

p.231, l.12, "...the effect is reduced"

→ Which effect is reduced? By what amount? I also don't understand the reasons for this reduction. Please clarify and rephrase.

p.231, l.13-15, "In the latter case the hybrid scheme does not switch to upwind, so the diurnal cycle is represented in SAM2."

→ Again, I don't understand what you are trying to say. Why does it matter how the "equation of state" is solved numerically? This shouldn't be the case. And why does this imply that the diurnal cycle is captured?

p.231, l.18-19, "...before the evolution of this parameter decreases to a value which is approximately twice as high as when the whole particle size range is considered."

→ I don't understand what are you saying. How can an evolution of a parameter decrease? Please reconsider this sentence.

p.232, l.5, "...grown to fairly large sizes much faster..."

→ Please avoid such imprecise statements.

p.232, l.10, "overall good results...with almost overlapping results for the filtered parameter."

→ Once again, what are you trying to say and how do you define "good results"?

p.232, l.17, "...large tropical volcanic eruption..."

→ How does your "large eruption" case compare to estimates for past eruptions such as Pinatubo or El Chichon?

p.232, l.23/24/25, "...in time..." / "...a magnitude larger..." / "Early..."

→ Please be more specific.
→ Change "a magnitude larger" to "an order of magnitude larger"?

p.233, l.1/4, "...significant..." / "...pretty well..."

→ Again, please be more specific.

p.233, l.11, "...fixed size sections act only as a sink and thus prevent further growth of the particles."

→ I do not understand what you are saying.

p.233, l.14, "CFL"

→ Has not been explained.

p. 233, l.17, "Therefore, in the volcanic case, condensational growth is strongly underestimated in SAM2, ..."

→ Insert "high" before "volcanic case"?
→ This should only limit the distribution of sulfuric acid vapor among size bins but not the total uptake.

p.234, l.1

→ Change "emphasize" to "are emphasized"?

p.234, l.19-20, "...which might lead to an overestimation of the radiative response of a large volcanic eruption."

→ How can less particles lead to an overestimation of the radiative response? This should be vice versa.

p.243, fig.1

→ I suggest to include a reference to the definition of the 3 cases (tab.2) to the caption.

p.246, fig.4

$\rightarrow$ 1st row, right picture: What happens in SALSA at t=150h? This seems wrong. Also, the effective radius for the "r $>$ 0.05 μm" case should always be larger than for integrating over all particle sizes. $\rightarrow$ 3rd row, right picture: How do you explain the double peak feature in the diurnal cycle from SAM2?