

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

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Reply to referee #1

We thank the Referee for the review of our manuscript. We think that we can improve the quality of the manuscript according to the comments.

General comments

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.

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- The intercomparison was done running the aerosol microphysical models in 0-dimensional (box model) framework. We will include a description of this in the revised manuscript.
- *The neglect of "non-microphysical" sink processes such as gravitational settling is inappropriate for coarse particles ($> 1\mu\text{m}$) and integration time of 10 days. Uncertainties introduced by this neglect need to be estimated or the experimental design adjusted.*
 - We agree that neglecting gravitational settling should be accounted to describe more accurately the development of the aerosol size distribution. On the other hand, in the atmosphere, the sedimented particles will to some extent be replaced by coarse particles from the upper levels. More importantly, as can be seen from the manuscript, this approach shows how the modules perform in “extreme conditions” and reveals weaknesses in methods used in these modules.
- *The study focuses solely on volcanic SO_2 emissions. Is volcanic dust also relevant?*
 - Volcanic dust plays a role only in the first weeks after an eruption (e.g. Niemeier et al. (2009)). However, fine ash particles are large and sediment out very quickly so they are not relevant for the long term climate effect. In addition, our paper focuses on the ability of global aerosol modules to simulate the climate effect of an enhanced stratospheric sulphate load, either due to volcanic eruptions or due to geoengineering efforts. We therefore neglect volcanic dust in our paper.
- *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 findings to the assumptions made (e.g. size distribution of stratospheric background aerosol)?*

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- Under high concentrations of SO₂, the microphysical processes modify the size distribution to a bimodal shape with two very narrow modes, one in nucleation sizes and one in coarse sizes and since most of the particle mass comes from the gas-to-particle transformation, effective radii are very similar regardless of the initial background aerosol size distribution.

The aerosol effective radius is the mean radius of the aerosol size distribution weighted by the aerosol cross sectional area and a measure of which part of the wavelength spectrum is mostly affected. Particles with a larger effective radius ($R_{\text{eff}} > 0.7\mu\text{m}$) absorb more in the near infrared and infrared part of the spectrum than particles with a smaller effective radius eg $0.17\mu\text{m}$ (background) which leads to an increase in the aerosol induced radiative heating. Lacis et al. (1992) demonstrated that the climate forcing of stratospheric aerosol can be characterized with the aerosol effective radius. If the effective radius is equal or greater $2\mu\text{m}$ the global average greenhouse effect of the aerosol exceeds its albedo effect leading to net surface heating.

- *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.*

- We will investigate the total surface area given by different modules and will add a Figure showing the surface area in the revised manuscript if it gives more insight on the performance of the different modules. Nevertheless, we will cover the differences in the surface area calculated by the different modules in the revised manuscript.

Specific comments

We will include more specific values and specific phrases in the revised manuscript according to many of the comments by the Referee. Here we reply to the Referee comments that need more explanation.

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p.214, l.25-26, "...the model resolves the concentrations of aerosol particles containing up to 21 H₂SO₄ molecules individually." - What do you mean by this? Please clarify and consider rephrasing.

- We state more precisely in the revised manuscript:
 "In the kinetic part, the model solves the differential equations for the concentration of each aerosol particle containing up to 21 H₂SO₄ molecules. For particles with a larger H₂SO₄ content, the model uses geometric size sections: The aerosol size distribution is divided into size ranges; particles in neighbouring size ranges differ by a constant factor in their H₂SO₄ content. The model solves the differential equations for the concentration of aerosol particles within each size range."

p.215, l.9, "...uptake and loss by large aerosol particles derive from the liquid drop model and H₂SO₄ and H₂O vapor pressures over bulk solutions..." - I don't understand what you are trying to say.

- In aerosol models, the uptake and loss of gas phase H₂SO₄ and H₂O molecules by aerosol particles is described by the rate coefficients for these processes. These rate coefficients take into account the kinetics of the processes (details of the collisions between the gas phase molecules and the aerosol particles), such as treated by Fuchs (1964) in his widely used formulation of rate coefficients for Brownian coagulation, and the thermochemical data (enthalpy and entropy change, or change in Gibbs free energy) associated with the transfer of the molecules from the gas phase to the liquid solution of the aerosol. For aerosol particles, the thermochemical data for this transfer differ from those for a bulk solution, because adding a gas phase molecule to an aerosol particle increases its surface area, which, in the case of a non-negligible surface tension, requires energy. Hence if we want to calculate the rate coefficients for the uptake/loss of

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gas phase H_2SO_4 and H_2O by the aerosol particles we need a method to calculate the thermochemical data for these processes that accounts for the change in surface energy. This can be achieved as follows:

First we calculate the Gibbs free energy $dG_{\text{H}_2\text{SO}_4}$ for the uptake of H_2SO_4 by a $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ solution of given composition with a flat surface, over which the equilibrium vapor pressure of H_2SO_4 is H_2SO_{4p} :

$$dG_{\text{H}_2\text{SO}_4} = R_{gas} t \log(\text{H}_2\text{SO}_{4p}/(101325.0\text{Pa})) \quad (1)$$

H_2SO_{4p} is calculated by a computer code provided by Simon Clegg (U. of East Anglia).

Then we calculate the Gibbs free energy $dG'_{\text{H}_2\text{SO}_4}$ for the uptake of H_2SO_4 by an aerosol particle with a given radius and that same given $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ composition as

$$dG'_{\text{H}_2\text{SO}_4} = dG_{\text{H}_2\text{SO}_4} + 4\pi(R2^2 - R1^2)\sigma, \quad (2)$$

where $R1$ and $R2$ are the radii of the aerosol before and after the uptake, respectively, and σ its surface tension. The last equation is the classical liquid drop model; it is this last equation which we refer to when writing

"The thermochemical data for $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ 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, calculated with a computer code ..."

$dG'_{\text{H}_2\text{SO}_4}$ then goes into the calculation of the rate coefficients for uptake/loss of H_2SO_4 with the Fuchs formula for Brownian coagulation (Fuchs64). The uptake/loss of H_2O is treated analogously.

This liquid drop based method works only for particles down to a certain size, at which the concept of a surface tension breaks down because the composition of the particle and its internal structure are inconsistent with the composition and structure of a bulk liquid: Very small particles are molecular clusters, not

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spherical liquid droplets. This is the reason why for very small particles, the thermochemical data (enthalpy and entropy change, or change in Gibbs free energy) from the laboratory, measured using actual molecular clusters must be used. In an intermediate size range, where the particles cannot be identified as "clearly molecular cluster" or "clearly spherical bulk liquid with", we use an additional term in the calculation Gibbs free energy $dG'_{\text{H}_2\text{SO}_4}$ to smoothen the transition in this quantity from the very small to large aerosol particles.

We understand that the short overview of MAIA given in the present manuscript can leave many open questions, but we think that the focus of the manuscript does not require a detailed description of the model such as the one given here. The interested reader is asked to consult the papers that are given as references to MAIA.

p.215, l.10-11, "...that uses data from..." - What data? Please be more specific.

- Here we refer the data presented in the papers by Giauque et al. (1960); Clegg et al. (1994), which are used in the computer code mentioned above to calculate the vapor pressures of H_2SO_4 and H_2O over $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ solutions of given composition. These data include the relative molal free energies and entropies. Again, a detailed description of the procedure seems not required given the focus of the present manuscript, and we therefore replace

"...that uses data from..."

with

"...that is based on ..."

p.215, l.12, "The thermodynamic data..." - Again, data for what? Please be more specific.

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- Here we erroneously used "The thermodynamic data..." instead of "The thermochemical data...". In order to make clear what we mean we write in the revised manuscript:

"The thermochemical data for uptake/loss of gas phase H₂SO₄ and H₂O by intermediate sized particles are a smooth interpolation of the data for the small and large particles (see Lovejoy et al. (2004); Kazil et al. (2007))"

In addition, to be more clear, we have also replaced

"MAIA describes nucleation and growth of small neutral and charged molecular clusters based on laboratory thermochemical data"

with

"MAIA describes nucleation of sulfate aerosol and growth of the freshly nucleated particles using laboratory thermochemical data for the uptake/loss of gas phase H₂SO₄ and H₂O by small neutral and charged molecular clusters ..."

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.

- In order to test the validity of the equilibrium treatment of H₂O uptake/loss in MAIA for the stratospheric conditions used, we have run MAIA for 10 days at an RH of 1 % and an SO₂ mass mixing ratio of 3.9E-4 kg(S)/kg(air). This is the highest SO₂ mass mixing ratio used in our simulations. The other ambient conditions were taken from the manuscript. The gas phase concentration of H₂SO₄ peaks at ≈ 2E9/cm³ in this simulation, with a H₂O vapor concentration of 8E12/cm³. Hence in these conditions, [H₂SO₄] <= [H₂O]/4000, meaning that a collision with a water molecule is about 4000 times more likely than with a H₂SO₄ molecule. The assumption in MAIA that an aerosol particle will assume equilibrium with

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respect to H₂O uptake/loss before colliding with an H₂SO₄ molecule is therefore very plausible for these conditions.

We have changed the text to reflect the finding that that the equilibrium assumption for water uptake/loss in MAIA is valid even in the considered stratospheric conditions.

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 H₂SO₄ 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.

- Principally it might bias the results but since particle composition measurements were not carried out in the cloud, at least in the very first weeks after the stratospheric injection, one have to assume representative conditions. We choose not to deal with mixed or insoluble particles also to build a least common platform for our model intercomparison. We will address this issue in the revised manuscript

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.

- The size distribution in SALSA for particle sizes 50 nm – 700 nm is defined as four different size classes with two parallel size sections, soluble and insoluble. We will include a more detailed description in the revised manuscript.

Section 3 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

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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).

- In section 3 we show that the new time integration scheme performs at least as good or better than the one previously used, or than the Euler backward scheme: From the derivation of the new time integration scheme one can see that when the nucleation rate goes to 0, the solution of the differential equation for H_2SO_4 processes calculated with our new scheme converges towards the exact, analytical solution of that equation, as mentioned in the manuscript. The old time integrations scheme and the Euler backward scheme do not have this property, hence our new scheme performs better. This justifies the new scheme.

The three cases we present in the evaluation (Section 3.1) are meant to illustrate the improved performance of the new time integration scheme visually, without any focus on particular geophysical conditions or any claim of generality: There are three ways how H_2SO_4 can behave over a time step in a model. It can increase, remain about the same, or decrease. The three cases have been constructed by choosing the ambient conditions and the H_2SO_4 production/loss rate accordingly, but not with any particular geophysical conditions in mind: In the first case, production outweighs loss (condensation/nucleation), in the second production and loss are about the same, and in the third case loss dominates.

We agree that the pressure of 1013.25 hPa is inconsistent with the temperatures used in these cases from a geophysical point of view. However, the pressure (as well as the temperature, RH, and the ionization rate) enters the differential equation for H_2SO_4 only via the nucleation rate, as we set the production and condensation rates of H_2SO_4 to fixed values (which is commonly the case in global

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models over one time step). The nucleation rate is very insensitive to pressure, because the nucleating particles ($\approx 1-2$ nm) are much smaller than the mean free path of gas phase molecules (typically >100 nm in atmospheric conditions), when their H_2SO_4 uptake and loss as well as their coagulation take place in the free molecular regime. The results would be affected in an insignificant way by considering pressures other than 1013.25 hPa. But the calculation of the nucleation rates pressures other than 1013.25 hPa would require that we change the code that calculates the aerosol nucleation rate, and recalculate lookup tables for the rate coefficients of H_2SO_4 and H_2O uptake and loss by small aerosol particles for these pressures, which are currently calculated at 1013.25 hPa. We do not consider this additional effort justified by the anticipated result. However, we have changed the text to point out that the three cases are not meant to address any particular geophysical conditions, but to illustrate the behavior of the time integration schemes in different H_2SO_4 production/loss regimes.

If the reviewer is not satisfied with this point of view, we offer to remove the discussion of the three cases (Section 3.1) and Figure 1 from the manuscript. The new time integration scheme is sufficiently justified and shown to perform better than the old one and the Euler backward scheme by its convergence properties alone. A detailed evaluation of the new time integration scheme would then be published in a stand-alone manuscript.

p.221, l.6, "... ion pair production rate..." - What do you mean by ion production rate?

- The radioactive decay of Radon and galactic cosmic rays ionize the atmosphere, resulting in the formation of gas phase ions. The corresponding formation rate of ions is referred to as "ion pair production rate". The ions can initiate aerosol nucleation, see, e.g. Kazil et al. (2008) and references therein.

Section 3.1 - I suggest to show also relative errors for typical time steps of GCMs

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as those are more relevant than absolute errors to evaluate the performance of the different time integration schemes.

- The three cases in Section 3.1 and Figure 1 are meant to illustrate the improved performance of the new time integration scheme visually, without claiming to be a detailed analysis of the schemes performance. The relative errors for these three cases may not be representative for the overall performance of the individual schemes. The overall performance of the individual schemes, with corresponding relative errors would require a study covering a large number of different atmospheric conditions (temperatures, RHs, H₂SO₄ production/condensation rates etc.). Such a study, as pointed out before, could be published as a stand-alone manuscript.

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.

- The model setup was a box model using the initial parameters as given in the beginning of section 4. We will make a more precise description of this in the revised manuscript.

The diurnal cycle for OH was a square wave signal as described by the Referee. We will investigate the effects of using a sinusoidal functions when making changes in the manuscript.

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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.

- As stated in the manuscript that the value was taken from a MT. Pinatubo simulation. We rephrase the sentence to make it even clearer.
"The extreme case mixing ratio was derived from a MAECHAM5 simulation of the June 1991 Mt. Pinatubo eruption (Niemeier et al., paper in preparation, 2009), In this 3D simulation 17 Mt SO₂ has been initialized according to satellite observations after the Pinatubo eruption (Read et al., 1993)."

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.

- The size distribution is more heavily modified under high concentrations of gas phase sulphuric acid and increasing the time step length decreases the accuracy of the models to describe the change in the size distribution. We will rephrase this.

p.225, l.16, "...a detached bimodal..." - What do you mean by detached?

- Here we mean to say that the size distribution is distinctly bimodal.

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.

- We will change the figures to those calculated using a 15 minute time step, which is the default time step length of ECHAM5 in resolution T42.

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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"?

- A distinct bimodal size distribution with coarse mode is already visible one day into the simulation. We will explain this more specifically in the revised manuscript.

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 – 1µm only.

- When one likes to investigate why modules of different setups/definitions behave differently one has to look into the whole size range covered by the module. This is the main aim of our study. The size range of 0.05 – 1µm is relevant when comparing model results with satellite measurements, but if all the aerosol effects are concerned, we find it necessary to study the whole size range. Also, it has to be remembered that the calculated aerosol effects are derived using the whole size range when these aerosol modules are used in GCMs.

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.

- We will rephrase this and include a discussion on the surface area as mentioned above

p.229, l.16, "...SAM2 tends to bridge..." - What do you mean by "bridge"?

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- SAM2 generates a size distribution which is not separated into two distinct modes.

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.

- We find that showing the diurnal cycle gives more information on how well the processing of the aerosol size distribution is calculated by the different modules. Showing the diurnal cycle includes has basically the same information as daylight averages.

p.229, l.28, "This characteristic is pronounced..." - I do not understand. Which characteristic of what?

- The difference between the effective radii calculated using the two different integration ranges. We will rephrase this.

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 SO₂ concentrations. Please explain.

- What one can see from Fig 4 upper row compared to the rows below: The results differ strongly when the results are filtered/adapted to optical instruments detection range. Thus under weak "signal-to-noise ratios" the results differ more than under strong "signal-to-noise ratios".

C64

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.

- We explain the behavior later in section 5.3.4. We will rephrase this to make it more clear for the reader.

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.

- In the paper mentioned, we validate the SAM2 module coupled to a middle atmosphere GCM against aerosol size parameters (surface area, volume density, effective radius) derived from SAGE II extinction measurements and in situ measured particle number concentrations in the mid latitudes of the northern hemisphere. We will clarify this

p.231, l.2, "...shape of the evolution of the effective radius..." - What do you mean by "shape of the evolution"?

- The shape of the curve for the effective radius as a function of time.

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?

C65

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?

- Regarding the referee comments above: we will simplify and clarify the explanation of the differences in the diurnal cycle, which results from solving the microphysical processes by operator splitting.

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"?

- We will calculate the relative difference for the revised manuscript.

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?

- The concentration for SO₂ in the extreme case was estimated to be of the same order as during the Mt. Pinatubo eruption

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.

- To reduce the amount of tracers, for the three largest size sections in SALSA only the number concentration and the fixed mean radius of the size section are

C66

stored as tracers, so condensation and coagulation can not increase the size of these particles. Only the growth of particles from smaller subregions affects the number concentration.

p. 233, l.17, "Therefore, in the volcanic case, condensational growth is strongly underestimated in SAM2, ..." - This should only limit the distribution of sulfuric acid vapor among size bins but not the total uptake.

- Unfortunately it limits the total uptake, which is the reason why SAM2 fails in the volcanic case. Not all that could condense condenses, only the amount which is used by the advection scheme is taken from the gas phase reservoir. The CFL criterion is the limiter which adopts the growth rate in respect to the time increment and the width of the aerosol size sections.

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.

- We will rephrase this sentence, since our purpose was to say that while the aerosol particles in the "tail" are still in the stratosphere, the radiative response can be overestimated.

p.246, fig.4 - 1st row, right picture: What happens in SALSA at $t = 150$ h? This seems wrong. Also, the effective radius for the " $r > 0.05\mu\text{m}$ " case should always be larger than for integrating over all particle sizes. 3rd row, right picture: How do you explain the double peak feature in the diurnal cycle from SAM2?

- The coarse sectioning in SALSA delays the "drop" in effective radius when only particles larger than 50 nm are calculated in effective radius, but as can be seen

C67

on the left subplot, integrating R_{eff} over the whole size distribution reveals that it is an artifact caused by the coarse size sections. This also shows why it is important that the whole size range must be used in the integration when the actual climate effects are estimated

Unfortunately, the y-scales in the figure are different on the first row subplots and this is why it looks that effective radii are smaller in $r > 50\text{nm}$ although this is not the case. This will be fixed.

In the volcanic case in SAM2 the condensational mass flux onto the particles is limited through a scheme's definition which is stated in section 5.3.4 on p 233. This flux limitation accounts for a pronounced diurnal cycle in R_{eff} as well as for the formation of lower order oscillations preceding the increase in R_{eff} during night.

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