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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A novel model to predict the physical state of atmospheric $H_2SO_4/NH_3/H_2O$ aerosol particles" *by* C. A. Colberg et al.

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We thank Scot T. Martin for his generally positive comments on our study and appreciate his recommendations. We will take most of them into account when we revise the paper.

S. Martin's comments fall into two categories: (a) there are questions concerning our method of computing the deliquescence/efflorescence hysteresis as a process which we will try to fully answer below; (b) there is some criticism on the data base used in our specific treatment, and we agree with him in many of these aspects.

In our manuscript we state that we take "full account of the deliquescence/efflorescence hysteresis." S. Martin perceives this as a somewhat provocative statement and suggests a moderation, e.g. to "offer an initial treatment of...". What we mean to say is that our treatment is indeed the first to take the deliquescence/efflorescence hysteresis

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as a process fully into account. Of course the data base used for this process treatment is not perfect - and never will be. We would therefore prefer to keep our original formulation, but will add a statement of clarification to avoid misunderstandings.

In the following we respond to S. Martin's questions and suggestions in his order of mention:

Example #1: *Time resolution.* The ammonia-to-sulfate ratio, ASR(*x,y,z,t*), is indeed a fixed climatology for July and January (i.e. the variable t specifies only the month), while relative humidity, RH(*x,y,z,t*), is given on a 6-hourly basis. Reason for the coarser ASR resolution is data availability (Adams et al., 1999) at this stage of our study. Variability in ASR on shorter timescales will influence the physical state of the aerosol, which would be interesting to investigate in future calculations. On the other hand it is clear that the variability in RH is much larger than that of ASR, as precipitation affects RH, but only to a lesser degree ASR. Thus, variability in ASR may be expected to have a smaller influence on the physical state, but it will be interesting to see calculations improved in this respect.

Example #2: *Subgrid variability.* The grid size for the RH and T along ECMWF is $1^\circ \times 1^\circ$, which is state-of-the-art for global 3-D calculations. This has not been stated in the manuscript and will be implemented in the revised version. A subgrid analysis beyond $1^\circ \times 1^\circ$ is matter of regional modelling and out of the scope of this work.

Example #3: *Importance of particle phase for climate forcing.* We agree that compared to other uncertainties particle phase is of second order importance in accuracy of the globally and seasonally averaged radiative forcing. However, regionally for certain seasons large differences in radiative forcing are obtained depending on phase, e.g. of $1\ W \cdot m^{-2}$ (or 26 % of the total sulfate aerosol forcing) over Europe in July (Boucher and Anderson, 1995). In the revised manuscript we will stress the importance on the regional scale.

Example #4: All altitude bins have been analyzed equivalently (Figure 9 and Tables

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2-4). However, the lowest level (800-1000 hPa) is not shown as the assumption of a pure $H_2SO_4/NH_3/H_2O$ aerosol system is hard to justify. We will state this reason in the revised manuscript.

Specific points:

1. The $5^\circ \times 5^\circ$ refers just to the starting points of the trajectories and to the grid used to store the output data. Once released, the trajectories move freely and without reference to any grid. Every six hours the trajectory information is then interpolated to the $5^\circ \times 5^\circ$ grid for output. But this does not influence the motion of the trajectories, and therefore no reconciliation with the $4^\circ \times 5^\circ$ grid of the ASR climatology is necessary.

2. Thanks for the remark; we will change that in the finalized version.

3. Thank you, we will change this.

4. Thanks for the remark. "Temporal development of ASR" is misleading, we will rephrase this sentence.

5. With fraction we mean "fraction by number" as is stated on page 2459, lines 3-8. In the APSM on each of the 10080 trajectories the physical state of the $H_2SO_4/NH_3/H_2O$ aerosol particles is determined every 6 hours, taking full account of the hysteresis. This information is stored within the $5^\circ \times 5^\circ$ grid box, in which the trajectory happens to be. By the end of the month we obtain the information how many trajectories it contained and if the aerosol particles are totally liquid or contain solids. Hence the number fraction of particles containing solid ammonium sulfate [$(NH_4)_2SO_4$], letovicite [$H_2SO_4/NH_3/H_2O$], or ammonium bisulfate [NH_4HSO_4] relative to the total number of all $H_2SO_4/NH_3/H_2O$ aerosol particles can be determined for each grid box. We will check the possibility to replace "fraction" by "number fraction" throughout the whole manuscript and to supplement section 2.2.

6. We do not claim that soot particles definitely act as heterogeneous nuclei. We only discuss the possibility of heterogeneous nucleation and that implementing this in the

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model would require global concentration fields of mineral dust and soot.

7. It is the number fraction averaged over a whole month (January and July respectively). See issue 5 above.

8. Our lab experiments do not give evidence for heterogeneous nucleation on external condensation nuclei (e.g. dust particles). However, we cannot exclude the possibility of "internal" heterogeneous nucleation as we never changed the ASR during an experiment (e.g. letovicite might act as a condensation nuclei for ammonium sulfate). Therefore, we addressed these processes as a sensitivity study in section 3.5.1..

9. The curvatures and the discontinuities in the black dashed and solid lines (i) and (ii) in Figure 11 illustrate potential, however totally hypothetical pathways, which atmospheric aerosol particles might traverse during either a drying or a humidifying process. This is done in order to discuss the possibility of "internal" heterogeneous nucleation of a second solid once another ammoniated sulfate has formed at first. Such a process only can take place if the ASR is changing (i.e. NH_3 or SO_3 uptake) during an investigation period.

10. We rephrase the mentioned key sentence "To obtain a lower bound for the presence of solid-containing particles in the calculations described above we put RH to the value of the corresponding homogeneous ice nucleation point in the moment a trajectory intersects the ice saturation curve." to "Therefore we estimate a lower limit of solid containing particles in calculations analogous to the ones described above. For this purpose we set RH at the moment a trajectory intersects the ice saturation curve to the RH-value of the corresponding homogeneous ice nucleation RH." and hope that this clarifies the section.

11. Specific explanations of sections 2.1.3. and 2.2 are already made in the answers of examples #1 & #2 and the specific points 1, 4, 5 & 7.

11a. Although ASR is treated as a climatology, a (RH, ASR)-pair could be assigned to

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each grid box. This means that for each 6-hr time step of each trajectory RH and ASR values are spatially interpolated from ECMWF and GCM data, respectively.

11b. This sounds like a misunderstanding. We have 10080 trajectories and 12960 model grid boxes. (Trajectories: Three-dimensional, month-long trajectories are started on four levels on a $5^\circ \times 5^\circ$ grid. This results in $4 \times 72 \times 35 = 10080$ trajectories for each time period. Grid Boxes: Along with the 5 altitude bins the $5^\circ \times 5^\circ$ intervals yield a total of $5 \times 72 \times 35 = 12960$ model grid boxes.)

11c. Explained in issue 1.

11d. The functionality of the model is described in issue 5. Trajectories might intersect with each other and do this all the time. It is not necessary to average or interpolate RH between intersecting trajectories since for each 6-hr time step of each trajectory RH is known directly from the ECMWF field.

11e. Compare issue 5. Indeed the model offers an internally mixed aerosol.

11f. We will check the manuscript for consistency of agent and object. For the general understanding we try to explain here once again: Aerosol particles are tracked along trajectories from 6-hourly ECMWF fields. The RH and T are interpolated along these trajectories. However DRH and ERH values depend as well on ASR, which is interpolated from a monthly mean. This means that for each 6-hr time step of each trajectory RH and ASR, including the history of the air parcel, on the specific trajectory are known. The corresponding grid box position is memorized. For each grid box we obtain information how often a specific grid box is crossed by a trajectory and if the aerosol particles are totally liquid or contain solids inside this particular grid box. Hence the fraction of particles containing solid ammoniated sulfate particles can be determined in each grid box.

12. We will give some additional information in the manuscript which corresponds to the Eulerian, grid-based approach: (i) The RH which is used in Figure 7 results from

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a monthly averaged climatology of ECMWF RH-data. (ii) As DRH we denominate the RH at which aerosol particles are totally deliquesced. For purely solid particles this is identical to the starting point of the deliquescence process. However, since for mixed phase particles it is difficult to define the starting point of a deliquescence process, we decided to take the RH where the last solid constituent is dissolved completely. With this Eulerian, grid-based approach we can decide if a particle is liquid or solid (respectively contains solids), but we can neither specify a mass nor a number fraction. This exactly is the benefit of a trajectory study.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 2449, 2002.