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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.

C. A. Colberg et al.

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We thank referee 1 for his constructive comments.

Indeed, the emphasis in interpreting this study is placed towards free tropospheric conditions. 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 at that level. However, all altitude bins have been analyzed equivalently (Figure 9 and Tables 2-4). As already mentioned while answering Scot Martin's comment we will state the reason for not showing the 800-1000 hPa bin in the revised manuscript.

The referee is right with his comment concerning the use of the term "radiative forcing". It is a linguistic inaccuracy to use "radiative forcing" throughout the manuscript. Since it is not within the scope of our study to investigate whether the changes in aerosol radiative effect due to changing the state of the entire aerosol column is different since

pre-industrial conditions we should rather refer to the term "radiative effect".

We will add a more thorough description of available aerosol models to the manuscript. However, to our knowledge the complete efflorescence/deliquescence hysteresis has not been included in any of the existing global 3-D aerosol models (e.g. Adams et al., Jacobson, Metzger et al.) so far. Nevertheless we appreciate the excellent referee's comment that an Eulerian model that carries two tracers (one for solid and one for aqueous) in principle also can model hysteresis. For such a model the advection scheme would be of great importance (it should be avoided that e.g. the advection of solid particles "poisons" neighboring grid boxes). So a model simply has to track the fraction of particles that are in a given state and treat them separately within sufficiently small grid boxes.

In order to enhance the clarity of what happens when a crystal forms we will include a sentence in terms of what the referee suggested: "In these first simulations we assume only one solid phase to form (the one that first reaches its ERH). We then examine the effect of multiple solid phases".

We will incorporate a section that discusses what our study means to the real atmosphere, especially in respect to the global relevance of letovicite. We will also estimate what might happen under more complex conditions in the real atmosphere. As mentioned in the manuscript we are aware that our model just treats a simplified aerosol subclass, namely the $H_2SO_4/NH_3/H_2O$ system, and is therefore only a first step towards a more comprehensive modelling of the physical state of tropospheric aerosol particles.

In the following we want to respond to the referee's minor points in the order of mention:

1st sentence of abstract: We will check the use of "aerosol", "aerosols" and "aerosol particles" for consistency. With the particular use of one of these terms we do not only distinguish between singular and plural, but rather between specific properties. With aerosol particles individual aerosol particles which can perform phase transitions

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are meant. With aerosol an ensemble of aerosol particles (e.g. the $H_2SO_4/NH_3/H_2O$ aerosol is the ensemble of $H_2SO_4/NH_3/H_2O$ aerosol particles in a certain region) and with aerosols more than one ensemble (e.g. the $H_2SO_4/NH_3/H_2O$ aerosol and the $NaCl/H_2O$ aerosol) is meant.

2450.7. Thanks for the remark; we will change that in the finalized version.

2450.9. Thanks for the remark; we will change that in the finalized version.

2450.26. Besides this direct climatic effect, aerosol particles contribute also indirectly - as cloud precursors - to the terrestrial radiation budget.

2451.4. We will change this to: "The direct forcing by partially ammoniated aqueous sulfuric acid particles has been investigated in a number of studies ...."

2451.18. We will replace "amenable" by "susceptible".

2452.11. As mentioned above we are grateful for the comment and will change the manuscript in this sense.

2452.15. We will change this to: "Radiative transfer calculations at Julian ....."

2454.3. We will replace "that" by "by which".

2454.10. We will replace "In case" by "If the".

2455.5-15: We would like to leave the basic description of the used trap experiment in the paper since the reader gets at least a basic feeling about what we did. Further details could then be taken from the given references.

2456.15. Thanks for the remark; we will change that in the finalized version.

2456.20. We will replace "lack" by "absence".

2459.1 We will change this to: "....and ERH, which timely depend on ASR and T along the trajectories."

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2460.2. We will omit the "Clearly".

2460.10. As mentioned above we are grateful for the comment and will change the manuscript in this sense.

2460. To clarify this point we will add a paragraph at the end of section 3.1.: "The result that letovicite is the dominant phase could also be conceived by the phase diagrams displayed in Figure 2. In a drying process homogeneous crystallization does not occur before an investigated aerosol particle is sufficiently supersaturated (red ERH line in Figure 2). Whatever phase is sufficiently supersaturated first on an atmospheric pathway is being formed. This is letovicite for ASR$<$1.5 and ammonium sulfate for ASR$>$1.5. Since atmospheric ASR values are generally smaller than 1.5, letovicite is suggested to be the dominant phase."

2460.25. Within the limits of our study (H_2SO_4/NH_3/H_2O system, 200-800 mbar, two months July and January) this is a universally true statement. The exact values are given in Tables 2 and 3.

2462.4. It means a relative change in all of the input parameters. As mentioned above we will emphasize this section with further and more detailed sensitivity studies in the finalized version (compare our answers to the comments of the second referee).

2462.25. We will rephrase this sentence to: "....since according to our empirical extension the homogeneous ERH-value is lower than 0%"

2464.8. We think that massively is appropriate in this context and would prefer to keep it.

References:

Adams, P. J., Seinfeld, J. H., and Koch, D. M.: Global concentrations of tropospheric sulfate, nitrate, and ammonium aerosol simulated in a general circulation model, *Journal of Geophysical Research*, 104, 13 791-13 823, 1999.

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Interactive Comment

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