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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Is aerosol formation in cirrus clouds possible?" *by* J. Kazil et al.

J. Kazil et al.

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We would like to thank the referees for their thoughtful review of our manuscript.

Reply to Referee 1

Referee: *Section 2, 1st paragraph.: This paragraph deals with the modification of the model by [Lovejoy et al. (2004)] by inclusion of new thermodynamic data for the neutral nucleation. It is stated that the model predicts more stable neutral clusters when the new thermodynamic data is used. It would be interesting to elucidate this in a little more detail: by how much are the results of [Lovejoy et al. (2004)] changed? In how far/where is ion induced nucleation still a relevant process in the atmosphere. But this could of course also be the subject of a separate paper.*

Authors: This question is addressed by [Hanson and Lovejoy (2006)]: For typical

atmospheric H_2SO_4 concentrations and at 50 % RH over water, ion-induced nucleation dominates over neutral nucleation for temperatures above $\sim$ 220 K, while below $\sim$ 210 K, neutral nucleation prevails. Although the relative strengths of neutral and ion-induced nucleation depend on relative humidity and on the ionization rate, this result constitutes a useful guideline. Hence ion-induced nucleation of water and sulfuric acid is a more efficient process in the lower and middle troposphere, compared to neutral nucleation of the same compounds, which will dominate at higher altitudes.

Referee: *p. 12187, l. 22: Here it would be interesting to present the average surface area of the cirrus ice as well, to clearly demonstrate the difference between condensational sink and surface area and to make it comparable to surface area and condensational sink of the preexisting aerosol.*

Authors: Implemented in the revised manuscript.

Referee: *Because the calculation of the condensational sink is quite important for this paper a reference to the concept should be given. A short description on how it was determined could be helpful as well.*

Authors: Implemented in the revised manuscript.

Referee: *For most of the figures, but especially for the cyan lines in Fig 5, increased line thickness and/or different coloring would be helpful.*

Authors: Implemented in the revised manuscript.

Reply to Referee 2

Referee: *The concept of condensation sink is used to explain the diffusion limited rate of condensation onto large ice crystals and for the importance of preexisting aerosol*

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surface area. It would be useful to define condensation sink.

Authors: Implemented in the revised manuscript.

Referee: *Is it possible to comment on whether cirrus clouds enhance or reduce nucleation?*

Authors: This is a very interesting question. At the very low temperatures of the upper troposphere, the aerosol nucleation rate is most sensitive to sulfuric acid concentration, while e.g. relative humidity plays a less important role. Therefore, one would assume that since cirrus ice crystals remove sulfuric acid and freshly nucleated particles from the gas phase, aerosol nucleation would be reduced in cirrus clouds compared to cloud free air, all other parameters being identical.

However, measurements show very high water vapor supersaturations (over ice) in cirrus clouds, although the mechanisms maintaining these supersaturations are not well understood [Peter et al. (2006)]. The high water vapor content might lead to a high production of the OH radical from ozone photolysis, and thereby to a strong oxidation of SO_2, resulting in a strong production of H_2SO_4. The result might be a more frequent occurrence and strength of aerosol nucleation within cirrus clouds compared to cloud free air with a lower water vapor content.

On the other hand, even cloud-free air can exhibit significant water vapor supersaturations (over ice) [Peter et al. (2006)]. Also, ozone photolysis is not the only source of OH in the upper troposphere: Acetone photolysis may contribute as well. The photolysis of acetone as a source of OH weakens the response of OH production to variations in water vapor. These two phenomena work against an enhanced H_2SO_4 production and thus aerosol formation in cirrus clouds compared to cloud-free air.

Hence answering the question whether cirrus clouds enhance or reduce aerosol formation requires a dependable representation of OH gas phase chemistry that captures the response of OH production to water vapor. In the present work, OH concentrations observed in the upper troposphere (including within cirrus clouds) have

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been used [Jaeglé et al. (2000)].

For these reasons we have excluded the question whether cirrus clouds enhance or reduce aerosol formation from the present work, and plan to address it in detail in a separate investigation.

Referee: *A useful addition to the paper might be a comment on whether it requires anthropogenic SO_2 for nucleation to occur in cirrus.*

Given some typical preexisting aerosol surface areas and cirrus ice crystal sizes what is the threshold SO_2 concentration at which cirrus nucleation is possible. This would help indicate how widespread globally cirrus nucleation is given predictions of upper tropospheric SO_2 from global sulfur models.

Authors: These are science questions of considerable relevance. We have not addressed them in the present work as we think that they call for an investigation that would best fit in a separate manuscript. We will try to give a rationale for this:

Our aerosol/cirrus model results show that SO_2 levels need to be elevated above the background median value so that aerosol formation proceeds in the considered cirrus clouds at an appreciable rate. The SO_2 enhancement required in the slow updraft cirrus is comparably modest and could be caused by variability in the background values or by SO_2 originating from volcanic or anthropogenic plumes. The SO_2 enhancement required in the fast updraft cirrus is much larger and is less likely to be caused by background variability. Hence in the case of fast updraft cirrus it is reasonable to assume that volcanic or anthropogenic emissions are required to initiate aerosol formation, while this is not necessarily the case for the slow updraft cirrus. However, it would seem premature to make a general statement based on the few cases considered in the present work, which, e.g., do not account for cirrus clouds forming at different temperatures.

Similarly, in order to assess the prevalence of aerosol formation in cirrus clouds on global scales, parameters in addition to aerosol and cirrus properties need to be

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accounted for, as a small change in one parameter in favor of aerosol formation, e.g. in temperature, can offset a change in an other parameter against aerosol formation, e.g. in aerosol surface area.

Referee: *Figure 1. Define RHi and RHw either in the figure caption or text.*

Authors: Implemented in the revised manuscript.

Referee: *Figure 2(a) appears to be Figure 1(b) repeated.*

Authors: Corrected.

Referee: *p. 12182, l. 1 and in Equations (1) and (2). Replace RH with RHw to be consistent with figure 1.a*

Authors: Implemented in the revised manuscript.

References

[Hanson and Lovejoy (2006)] D. R. Hanson and E. R. Lovejoy. Measurement of the thermodynamics of the hydrated dimer and trimer of sulfuric acid. *J. Phys. Chem. A*, 110(31):9525–9528, 2006.

[Jaeglé et al. (2000)] L. Jaeglé, D. J. Jacob, W. H. Brune, I. Faloona, D. Tan, B. G. Heikes, Y. Kondo, G. W. Sachse, B. Anderson, G. L. Gregory, H. B. Singh, R. Pueschel, G. Ferry, D. R. Blake, and R. E. Shetter. Photochemistry of HO_x in the upper troposphere at northern midlatitudes. *J. Geophys. Res.*, 105:3877–3892, 2000.

[Lovejoy et al. (2004)] E. R. Lovejoy, J. Curtius, and K. D. Froyd. Atmospheric ion-induced nucleation of sulfuric acid and water. *J. Geophys. Res.*, 109, 2004.

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Interactive Comment

[Peter et al. (2006)] T. Peter, C. Marcolli, P. Spichtinger, T. Corti, M. B. Baker, and T. Koop. When dry air is too humid. *Science*, 314(5804):1399 – 1402, 2006.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 12179, 2006.