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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Depositional ice nucleation on solid ammonium sulfate and glutaric acid particles" *by* K. J. Baustian et al.

Anonymous Referee #2

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This study presents heterogeneous ice nucleation on micrometer-sized solid ammonium sulfate and glutaric acid particles for temperatures between 214 and 235 K and ice saturation ratios between 1 and 1.7. Optical microscopy is employed to detect ice formation and Raman microscopy is used to verify the composition of the activated ice nuclei. This experimental approach allows direct comparison of the ice nucleation efficiency for both particle types. This study shows that solid ammonium sulfate particles are more efficient ice nuclei than glutaric acid particles indicating that solid ammonium sulfate particles can act, as previously suggested, as efficient ice nuclei for ice cloud formation.

The topic fits nicely within the scope of Atmospheric Chemistry and Physics. The manuscript is well written and the results are clearly presented. However, there might

be a fundamental problem with the experimental approach as outlined below which can affect the results of this study and thus should be addressed by the authors. The described issue is the main reason for the rating of the manuscript and if clarified I suggest the manuscript for publication.

This study employs a commercially available Linkam cryo-stage which is purged by a humidified flow of N_2(g) at 1 atm. Inside the sample compartment of this cryo-stage supply tubes for N_2(l) are located to cool the silver block (sample area) to low temperatures. If water vapor is introduced to this compartment which contains the ammonium sulfate and glutaric acid particles water vapor will always condense at the coldest point in that system (i. e. supply tubes at liquid N_2 temperature) and, thus, leading to a water vapor gradient within that compartment (e. g. similar to the Bergeron-Findeisen effect). This can significantly affect the desired relative humidity (RH) above the investigated particles within the compartment. High flows of water vapor into the cell may help but this would also increase the ice buildup continuously affecting the dew point of the humidified flow. Besides of this crucial issue, during the ice nucleation experiments not the entire particle sample is visible. If ice forms on particles outside the visible area, RH above the observed particles is most likely not the one adjusted for similar reasons given above. This may be indirectly corroborated by the ice nucleation experiments conducted on blank substrates. For temperatures between 214 and 235 K it is stated that ice nucleation occurred for saturation ratios between 2.33 and 1.6 which are significantly above water saturation at these temperatures. From this it can be concluded that water vapor was depleted by another sink within the Linkam cell and artificially high supersaturations were needed to induce ice nucleation. To circumvent these issues usually such "ice nucleation flow systems" (particles on cold surface and humidified flow above) possess very small sample areas to ensure complete visibility of the experiment and to ensure uniform RH fields (see e. g. Eastwood et al., JGR, 2008). Here, the deliquescence experiments of NaCl are successful since for temperatures above -30°C saturation with respect to ice is not achieved and thus water vapor condenses as liquid onto the hygroscopic particles. For these reason the presented

deliquescence experiments cannot ascertain the correct RH fields within the Linkam cell at lower temperatures. Having said this, can the authors prove experimentally and/or theoretically the correct RH above the particles. E. g. by repeating these experiments with highly efficient ice nuclei such as mineral dust particles (e. g. kaolinite etc.) or by other means?

Specific Comments:

Page 20953, line 18: Within the manuscript it is not mentioned when lasers with wavelength of 532 nm or 780 nm have been employed. Please indicate the excitation wavelength used.

Page 20955, line 19, 20: Hung et al. used FTIR for detection of the ferroelectric phase transition of ammonium sulfate. Knopf and Koop used a Raman microscope to detect the ferroelectric phase transition in ammonium sulfate particles applying it as a calibration point in their heterogeneous ice nucleation study similar to this study.

Page 20957, line 2-4: Later in the manuscript it is mentioned that there are mixed particles, i. e. ammonium sulfate containing glutaric acid. 1. Please explain how those mixed particles formed? 2. Can you distinguish between particles which may lie on top of each other, e. g. a 300 nm ammonium sulfate particle on top of a 10 micrometer glutaric acid particle or vice versa?

Page 20957, line 13: The dew point is changed by up to 2°C min^{-1}. How does this compare to atmospheric conditions?

Page 20957, last line: a reference for pre-activation is missing (e. g. Knopf and Koop, Wallace and Hobbs, or older studies)

Page 20963, line 1: The ice nucleation experiments in which Abbatt and coworkers were involved applied an IR flow tube to study ice nucleation on ammonium sulfate. Only few experiments referred to in Abbatt et al. 2006 were conducted in a cloud chamber.

Page 20964, line 13: 1. What is the atmospheric significance of perchlorate? 2. How do you know that its deliquescence point will be reached before ice nucleation occurs at such low temperatures? Please give a reference for this case.

Figure caption of fig. 5: Does the illumination of the particles with laser light during the ice nucleation studies affect the particle temperature due to absorption of these rather dark particles?

Technical Corrections:

Page 20950, line 111: Change “that” to “than”

Page 20963: line 28: missing comma after “glutaric acid”

Figure 3: Please indicate the vibration bands of the Raman spectrum. Also, which vibration bands indicate the para- to ferroelectric phase transition? Please make a mark and give corresponding wave number.

Figure 5: Please add Raman vibration bands for both ammonium sulfate, glutaric acid, and ice. 218.08 K can be rounded to first digit after comma.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20949, 2009.