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Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Depositional ice nucleation onto hydrated NaCl particles: a new mechanism for ice formation in the troposphere" *by* M. E. Wise et al.

Anonymous Referee #1

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This experimental study uses optical microscopy coupled with Raman spectroscopy to detect the formation of NaCl hydrates and ice nucleation. It is found that below 236 K all aqueous NaCl droplets effloresce to form a NaCl hydrate whereas between 236–252 K the aqueous droplets effloresce into hydrated and anhydrous NaCl. Above 252 K only anhydrous NaCl effloresced. In addition, this study evaluates the ability of the anhydrous and hydrated NaCl particles to nucleate ice via the deposition mode indicating that the hydrated NaCl particles are very efficient ice nuclei forming ice at about ice saturation which is about 0.1 lower than for anhydrous NaCl particles. These novel findings are applied to estimate the fraction of hydrated NaCl particles present in the atmosphere and corresponding radiative forcing.

The focus of this study falls well within the scope of ACP. The manuscript is well written and the experiments carefully conducted. However, I have a few comments which should be addressed before publication of this manuscript can be recommended.

General comments:

In the introduction, the first and second paragraph should include more recent literature on e.g. sea salt emission estimates and corresponding radiative forcing. See e.g. IPCC reports, etc. Also, the findings by the O'Dowd group that sea salt can consist to a major fraction of organic material should be mentioned.

In the experimental section it is mentioned that 50 randomly chosen particles were investigated. How many independently generated samples were used to account for biases in particle generation? Are the results of this study statistically significant? The study nicely shows the formation of a hydrated NaCl phase at low temperatures and its potential to act as deposition ice nuclei, however, is the data sufficient to extrapolate to the atmosphere? The linear relationship shown in Fig. 6 is based on how many experiments and samples? Related to this, representative uncertainties should be given in figures 5, 6, 8, 9, and 10.

No discussion of the purity of NaCl and water is given. Can this impact the nucleation/efflorescence behavior and corresponding phase? E.g. a study by Lee et al., currently in ACPD, shows that $NaNO_3$ powder with 99.999% purity most likely contains impurities which are to blame for the different efflorescence results in the previous literature. These issues should be addressed, so when applying these laboratory data to the atmosphere, these constraints are known.

The size effect on the phase transition is not discussed in this manuscript. The applied particles are about a factor of 10 larger in diameter than atmospherically relevant particles. For deliquescence this is not such a significant factor but for efflorescence. Does the fraction of effloresced hydrated to anhydrous NaCl particles correlate with particle sizes? The same for deposition ice nucleation. In most cases it is assumed that the larger the surface area the larger the ice nucleation ability. Are the onset ice nucleation conditions for hydrated and anhydrous NaCl particles conducted with particles of the same size? At which supersaturation will hydrated and anhydrous NaCl particles nucleate ice when 10 times smaller?

The atmospheric implications section refers to previous literature to understand the results. It would be easier for the reader to elaborate a bit more. E.g. if stated that below 236 K only hydrated NaCl forms, why does Fig. 10 indicate that there is only a fraction of those present at temperatures well below 236 K? Information on RH is missing to understand these results. Also with regard to the radiative forcing: there exists many different definitions. What is shown here? A ratio of radiative forcing compared to clear sky radiative forcing? Why is a positive value indicative of cooling? Figure 11 shows the radiative effect of hydrated versus anhydrous NaCl particles. However the text discusses growth factors derived from deliquesced droplets and respective hydrated or solid anhydrous NaCl particles.

Specific comments:

Page 23145, line 25: Please give reference for DRH at 244 K.

Page 23146, line 24-26: Does a size effect play a role in this range of efflorescence values?

Page 23148, line 9, 2nd paragraph: Further above in the text, a DRH value of 75.4% is reported for 244 K. It is compared to experimental data but here with theoretical results (which theoretical results, reference?). The hydrated NaCl particles deliquesce at 90% RH. How reliable is this number having potentially mass transfer of water vapor to the already deliquesced anhydrous particles?

Page 23148, line 27 and 28 and figure caption 5: I do not know what “accepted” DRH and ERH means and no references are given? Why not stating experimentally determined ERH and DRH values?

Page 23151, 2nd and 3rd paragraph: As mentioned above, what is the actual uncertainty in the average onset values due to uncertainties in RH? Also figure 8 could contain the freezing data of ammonium sulfate of Baustian et al., 2010. If figure 8 is plotted again with uncertainties included, how significant is the difference between ice nucleation onsets of hydrated and anhydrous NaCl particles (maybe using student’s t-test etc.)? Figure 8 shows that anhydrous particles also nucleate ice at $S \approx 1$ and that much less experiments were conducted using anhydrous NaCl.

Page 23152, line 4: What is meant by the data are consistent with one another? The deliquescence curve consists of two regimes with opposite slopes, neither of those are similar to the curve describing deposition ice nucleation.

Figure 1: Why is the Raman laser not focused on the particle but in some instances, e.g. panel c, is positioned quite far away considering the laser beam is about 500 nm in diameter? Information is missing to explain the data collection.

Figure 3: I suggest showing an enlarged view of the spectrum where the peaks are located and skipping wavelengths not important for interpretation.

Technical corrections:

Page 23140, line 27: Missing “1” in “1×10^{14} kg”.

Page 23142, line 3: Citation erroneous.

Page 23143, line 12: Is it meant to be: “is given here in detail when…”.

Page 23148, line 20: “The waters of hydration”… sounds awkward.

Page 23149, line 11: Maybe change “It was previously shown…” to “It was shown above…”.

Figure 5: Please use lighter shading. Crossed green circles could be a bit larger.

Figure 9: Crossed green circles could be a bit larger.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 23139, 2011.