

***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 #3

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Review of Wise et al

Some unexpected and very interesting results are presented in this paper. Wise et al. use Raman microscopy to show that below about 250 K NaCl solution droplets effloresce to form hydrated crystals. On increasing RH these crystals only deliquesce at much higher RH than would be expected for anhydrous NaCl. Perhaps more exciting, is the result that the hydrated NaCl crystals nucleate ice at close to 100% RH_i – which as far as I am aware is unheard of. The only thing to nucleate ice at 100 % is ice itself! The paper is well within the remit of ACP as well as being well written and presented. I have a few comments and questions for the authors:

1) Title: 'hydrated NaCl' might imply liquid aqueous droplets. Possibly write: 'crystalline hydrated NaCl....'

2) How good a proxy for sea salt is NaCl? What justification is there? Could the other components influence the crystallisation kinetics? There is a Tang paper which may be of interest (Tang, I. N., Tridico, A. C., and Fung, K. H.: Thermodynamic and optical properties of sea salt aerosols, J. Geophys. Res., 102, 23269–23275, 1997).

3) What is the rate of change of RH in the experiments?

4) p23146, In6. The cubes do not look like poly-crystals, so Mikailov's capillary suggestion doesn't fit. Is there another explanation?

5) p23147, In 1-10. It's not clear why ice is being discussed as a possible phase which could be crystallising. At these low RH values, the solution is not supersaturated with respect to ice so it can't form. It's fine to mention that the spectra confirm this, but the discussion doesn't need to be as lengthy.

6) Are there hydrates of other materials which dehydrate on reducing RH? Is there precedent for this?

7) P 23150 and fig 5. It is claimed that the deliquescence data is inconsistent with the dihydrate. This is not clear to me. Extrapolating the dashed curve to lower T you'd expect the deliquescence to occur at ~85% at 240 K. By eye the experimental points are scattered around a value of 87 (+7)%. I don't think the authors should rule out the dihydrate on this basis; in fact, it seems to me that the data may be consistent with the dihydrate.

8) Fig 5: A key would be helpful.

9) P23151, In 8. But the ammonium sulphate data isn't compared in the plot. Was the ammonium sulphate data for the same size droplets?

10) For the ice nucleation work, is it possible to estimate ice active surface site den-

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sities? Quoting a threshold value for nucleation is semi-quantitative, but you can't compare this directly to other data. For example how does it compare to mineral dust experiments in the AIDA chamber? If possible it would be useful to normalise the result to surface area in some way.

11) I'd like to see some discussion of why the hydrate has a lower nucleation threshold. The hydrate particles appear to be rougher than the anhydrous particles. Could this account for the lower nucleation threshold rather than an inherent property of the hydrate? This idea is discussed by Zuberi et al. for ammonium sulphate in the immersion mode (GEOPHYSICAL RESEARCH LETTERS, VOL. 29, NO. 10, 1504, 10.1029/2001GL014289, 2002). Also, it would be useful to include a brief discussion about how well this material nucleates ice. Is there any other material that we know of that can nucleate ice at close to 100% other than ice?

12) P23154. In 14. The final comment on radiative forcing is too general. This uncertainty is for sea salt aerosol only. Also, how important is sea salt aerosol in the upper troposphere in terms of radiative transfer?

13) The modelling is for temperatures below 220 K, but the experiments were above this temperature. State the assumptions that were made in order to make this step.

14) Fig 1. Why are the crosses, which indicate the position the laser was focused, not on the particles?

15) Fig 2 c. The cross is missing in the picture.

16) Fig 5 caption. 'Wise et al. 2011'. Just state present study. I am also uncomfortable with the term 'accepted'. Just state where the parameterisation came from.

17) Fig 8. A key would be helpful

18) Figure 10 caption (and associated text). What is meant by fraction of hydrated particles? Is this the fraction of crystalline (or total) sea salt aerosol which are hydrated crystals?

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