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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hygroscopic behavior of NaCl–$MgCl_2$ mixture particles as nascent sea-spray aerosol surrogates and observation of efflorescence during humidifying process" *by* D. Gupta et al.

Anonymous Referee #2

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General comments

In Hygroscopic behavior of NaCl–MgCl2 mixture particles as nascent sea-spray aerosol surrogates and observation of efflorescence during humidifying process, the authors measure water uptake and phase behavior of aqueous droplets containing sodium and magnesium chloride salts. The work is motivated by the need to characterize the hydroscopic behavior of salt systems that represent sea spray aerosol particles better than pure NaCl aqueous systems alone. Measurements are performed

using optical microscopy, with supporting characterization with Raman microspectrometry and, for a limited number of cases, X-ray spectrometry.

The aqueous solutions studied include the two pure salt systems, as well as 12 distinct mixed chemical compositions of 0.01 to 0.9 solute mole fraction NaCl, with measurements performed over a 3% to 90% relative humidity range. The amount of experimental characterization is impressive. The only other similar NaCl-MgCl2-H2O laboratory single particle study (Chan et al. 2000) is limited to one molar ratio, with a more restricted relative humidity range. In addition to new experimental characterization and comprehensive phase diagrams, the authors also find scientifically interesting results, including a kinetically favored MgCl2.4H2O structure in pure MgCl2 aqueous systems instead of the thermodynamically predicted MgCl2.6H20.

Publication in ACP is recommended, after the authors address the comments below.

Specific comments

- More discussion is needed on if a (quasi-)equilibrium state is truly reached at each RH step change. For levitated single particle studies, an incremental change in the RH value is held until there is no change in mass at that value. Is a similar procedure used here for OM? Is the RH value held until this is no change in area? If not, kinetic artifacts may arise. For example, a rate dependence may explain the reported particle size dependence in during dehydration, with the ERH and MERH RH values lower with larger particles (lines 8-10, p17812).

- There is significant structural rearrangement of the particles represented in Figures 3 (0.9 solute mole fraction NaCl) and Figure 5 (0.2 solute mole fraction NaCl). Do these observations of rearrangement occur consistently at the same RH values for all particles of the same composition and type?

- Too much attention may have been given to the three particles types in Section 3.2.2, given the overlapping range in critical RH values in both hydration and dehydration

behavior for each of the three types (Figures 7 and 8). Some statistical analysis is needed to convince the reader that there truly are three distinct behaviors observed. If the differences in the three states are robust, more discussion is needed for why these three occur.

- The RH step change is 0.3% in RH for OM, but around 3% in the Raman characterization. Why is the procedure different? If OM is run at 3% step changes to match the procedure used to obtain the spectroscopy results, would the same efflorescence/ deliquescence curves result as with 0.3% changes?

- It would be beneficial if the author could connect the area ratio vs. RH results from OM (e.g. Figure 4a) to the more traditional mass fraction solute or mass ratios vs. RH results from EDB experiments (e.g. Fig 1 in Chan et al., 2000).

Technical corrections

- The readability of the manuscript could be improved significantly by fixing run-on paragraphs (e.g. line 5 p17807 to line 20 p17808) and compound sentences.

- (Line 2, p17830) a space is needed between salts and with

- (Line 16, p17802) Co-efficients should be Coefficients

- (entire manuscript) Use ‘hydration’ or ‘humidification’ instead of ‘humidifying’

- (introduction) Use the ion types, instead of electrolyte names, when discussing the sea water composition (note, it was done correctly in lines 16-18, p17822 of section 3.6).

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 17797, 2015.