

The authors thank each of the reviewers for their individual comments. We have made extensive changes to the manuscript according to the reviewers' suggestions. We feel that the final result is a significant improvement over the original version. Each of the authors agrees with the changes made to the original manuscript. Responses to individual comments are written below.

Reviewer 1 comment: 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.

Authors' response: The revised text now reads (page 2, line 45), "It is estimated that approximately 17 Tg of SSA enter the atmosphere per year (Textor et al., 2005). Of this amount 15 % is emitted as submicron size particles."

Furthermore, we have added (page 2, line 55), "O'Dowd et al. (2004) found that SSA can be enriched in organic material relative to bulk seawater. This enrichment increases with decreasing particle size."

Reviewer 1 comment: How many independently generated samples were used to account for biases in particle generation?

Authors' response: Each individual data point in the figures presented in the manuscript represents an independently generated sample. We generated approximately 150 different samples. The text now reads (page 5, line 122), "Each data point presented in this manuscript represents an independently generated sample, i.e. in total about 150 different samples were generated"

Reviewer 1 comment: Are the results of this study statistically significant?

Authors' response: We assume that the reviewer is asking if the results shown in Figure 6 are statistically significant. In Figure 6 we are not reporting an average or a standard deviation for each data point. Each data point represents the percentage of solid particles that were anhydrous NaCl_(s) at each temperature. Raman spectra of 50 different particles were collected randomly to determine this percentage for each point. Thus a total of 500 points were examined. We have now added an error analysis to this figure to account for random error. We used the expression for calculating standard error assuming random sampling ($\text{error} = \text{SQRT}(p(1-p)/n)$ where p is the percentage and n is the number of sample points). The text now reads (page 14, line 308), "Each data point utilizes 50 different particles and the error bars for each point represent the margin of error. "

Reviewer 1 comment: 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?

Authors' response: The RH and temperature conditions at which the experiments were performed are relevant to the troposphere. However, our lowest efflorescence temperature was 239 K and Figure 10 shows a scenario that uses temperatures from 180-220 K. Thus a significant temperature extrapolation was assumed. This could introduce error, especially at the lowest temperature. Between room temperature and 238K, we did not observe significant temperature dependence in the ERH, thus it is reasonable to assume a similar trend at lower temperatures. Similar behavior was observed in a previous study on ammoniated sulfate aerosols. Nonetheless, we have now added this caveat to our discussion of Figure 10 (page 19).

Our ice nucleation studies were performed at lower temperatures (down to 225 K). These are typical temperatures for cirrus cloud formation. We did not observe temperature dependence in Sice over the range studied. However, past work has shown that Sice values do increase at temperatures below 180 K, (Trainer et al., 2009), so an extrapolation would be needed for cirrus at the very lowest temperatures in the atmosphere. We have added a caveat to this effect in our discussion (page 19, line 426).

Reviewer 1 comment: The linear relationship shown in Fig. 6 is based on how many experiments and samples?

Authors' response: Figure 6 is based on 10 different experiments that utilized 50 different particles per point, for a total of 500 particles. Page 6, line 150 reads "After each particle effloresced following the second RH cycle, Raman spectra of 50 different particles were collected to determine the percentage of solid particles that were anhydrous NaCl_(s). In order to eliminate operator bias, 50 random particles were studied. Specifically, the substrate was moved in a straight line and each particle that was illuminated by the Raman laser was studied."

Reviewer 1 comment: Related to this, representative uncertainties should be given in figures 5, 6, 8, 9, and 10.

Authors' response:

Temperature error: The average error in the temperature calibration for all experiments is ± 0.2 K. Error bars for temperature cluttered Figures 5, 6, 8 and 9 so this value is included in words in the manuscript. The manuscript now reads (page 6, line 139), "The average error in the temperature calibration is ± 0.2 K for all experiments."

RH error: Additionally we calculated a range in RH error assuming that temperature was the only factor contributing to error. Over the temperature range studied it was found that the error in RH was always less than ± 2 %. Page 6, line 141 now reads, "The uncertainty in temperature corresponds to an error of less than ± 2 % RH over the

range of experimental conditions studied.”

S_{ice} error: Similar to error in RH, we calculated a range in S_{ice} error assuming that temperature was the only factor contributing to the error. Page 7, line 159 now reads, “The uncertainty in temperature corresponds to an error in S_{ice} of less than 0.025 over the range of experimental conditions studied.”

Figure 5: Data for the DRH, ERH and hydrate (green circles) have been put into bins that span 2 K and averaged. The plot now shows the average value for each temperature bin. The error range comes from the high and low value for each bin. Page 13, line 287 now reads, “All DRH and ERH data was put into bins that span two degrees Kelvin and averaged. Each data point represents the average value for each temperature bin. The error range is the high and low value for each bin.” Furthermore, page 14, line 317 now reads, “As before, the DRH data was put into bins that span two degrees Kelvin and averaged.”

Figure 6: We added uncertainties for the percentage of anhydrous particles to the figure. Page 14, line 308 now reads, “Each data point utilizes 50 different particles and the error bars for each point represent standard error assuming random sampling.”

Figure 8: The S_{ice} data shown in Figure 8 have also been put into bins that span 2 K and averaged. Each data point represents the average for each temperature bin and the error bars were created from the high and low value for each bin. Page 16, line 350 now reads, “All S_{ice} data was put into bins that span two degrees Kelvin and averaged. Each data point represents the average S_{ice} value for each temperature bin. The error range is the high and low value for each bin.”

Figure 9: The RH data in Figure 9 has been put into bins that span 2 K and averaged.

Reviewer 1 comment: 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.

Authors’ response: We agree with the reviewer that the solutions used in our experiments likely contain impurities. Not only can the impurities come from the solute but also from the atomization technique. Similarly, particles in the atmosphere contain “impurities”, many of which are uncharacterized. This fact makes the comparison of laboratory particles to “real” atmospheric particles extremely difficult. Every group that performs laboratory measurements deals with this difficulty. We now report the purity of our starting reagents (Page 5, line 118).

We also note that NaNO_3 is notorious for not crystallizing at the known ERH when

mixed with other species or even pure. There are reports of pumping NaNO₃ solutions to 10⁻⁷ Torr without causing crystallization. This is in contrast to the situation with NaCl. For NaCl there is general agreement between groups who determine the ERH. This suggests that NaCl is not nearly as sensitive to impurities as NaNO₃. More importantly, Tang et al. have studied both NaCl as well as artificial seasalt particles and observed very similar ERH and DRH values for the two systems.

Reviewer 1 comment: 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?

Authors' response: Past work has shown that the efflorescence of NaCl particles is not affected by size in the range 0.1 to 100 micrometers (see Phase transitions of single salt particles studied using a transmission electron microscope with an environmental cell, M.E. Wise, G. Biskos, S.T. Martin, L.M. Russell, and P.R. Buseck, *Aerosol Sci. & Tech.* 39, 849-856, 2005). There is a range of RH values at which the NaCl particles effloresce and that represents the stochastic nature of efflorescence, not size. Similarly, in the present study we do not see a correlation between particle size and the formation of hydrated/anhydrous particles. Interestingly, when 2 or more RH cycles are performed on the same sample, some particles that effloresce into the hydrate after the first cycle do not effloresce into the hydrate after the second cycle. We have now added this clarification to the text (Page 10, line 221).

Reviewer 1 comment: The same for deposition ice nucleation.

Authors' response: We do not see size dependence with depositional ice nucleation in our size range. Similarly, Baustian et al. (2010) did not see a size dependence for depositional ice nucleation on ammonium sulfate particles (Baustian et al., *Atmos. Chem. Phys.*, 10, 2307-2317, 2010). Furthermore, Froyd et al. (*Atmos. Chem. Phys.*, 10, 209–218, 2010) found that ice residuals were similar in size to unfrozen aerosol particles.

Page 16, line 354 now reads, “Additionally, there does not appear to be a temperature or size dependence on the ice nucleating ability of hydrated NaCl_(s) particles. This result is similar to Baustian et al. (2010).”

Reviewer 1 comment: 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?

Authors' response: The onset conditions for ice nucleation are conducted on particles in the same size range (~0.1-20 microns). However, on average, the hydrated

particles are larger than their non-hydrated counterparts. This is due to the fact that the hydrated particles effloresce into particles that have a greater size due to the waters of hydration. However, as stated above we do not see size dependence with particles in this size range. The size range includes both hydrated and non-hydrate particles.

Reviewer 1 comment: At which supersaturation will hydrated and anhydrous NaCl particles nucleate ice when 10 times smaller?

Authors' response: We do not have an answer to this question. However, we do know that small particles (less than one micron in size) sometimes nucleate ice first and sometimes bigger particles (5-6 microns in size) nucleate first. It appears that something other than particle size is controlling the nucleation in our size range. It has recently been suggested that perhaps surface defect sites are the controlling factor. We now discuss this in the text (page 17, line 379).

Reviewer 1 comment: 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.

Authors' response: We have added the following description to the manuscript to clarify Figure 10. Page 18, line 402 now reads, "The parcels were initially assumed to be aqueous NaCl with 100% relative humidity with respect to ice. The RH in each parcel was tracked on its path upward through the tropical upper troposphere. If the RH dropped below 35%, NaCl particles in the parcel were assumed to effloresce. They remained NaCl_(s) unless the RH increased above the deliquescence point (80%). By combining results from 648 trajectories throughout the tropics, statistics were generated of the time when NaCl particles were in aqueous or solid states."

Reviewer 1 comment: 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.

Authors' response: This section of the manuscript has been significantly reworked. We refer the Reviewer to this entire section for the response to this comment.

Reviewer 1 comment: Page 23145, line 25: Please give reference for DRH at 244 K.

Authors' response: The manuscript now reads (page 9, line 190), "The DRH of NaCl_(s) particles is extrapolated from higher temperature data (Tang and Munkelwitz, 1993 and Koop et al., 2000) and is ~ 76.7 % RH at 244 K. This value is higher than the RH

observed here for the onset of water uptake”

Reviewer 1 comment: Page 23146, line 24-26: Does a size effect play a role in this range of efflorescence values?

Authors’ response: This question was answered previously.

Reviewer 1 comment: 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?).

Authors’ response: As discussed above, the DRH is compared to other experimental results.

Reviewer 1 comment: 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?

Authors’ response: The reviewer brings up a good point. Mass transfer of water vapor during the deliquescence experiments was of concern. This is especially true with the hydrated particles that deliquesced at 90 % RH. However, if mass transfer were a problem, all deliquescence data for the hydrated particles would have been skewed towards high RH values. As shown in Figure 5, as temperature decreases to 235 K, the DRH reduces to approximately 80 %.

Reviewer 1 comment: 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?

Authors’ response: The “accepted” DRH and ERH come from Tang and Munkelwitz (1993) and Koop et al. (2000) which is now referenced.

Reviewer 1 comment: Page 23151, 2nd and 3rd paragraph: As mentioned above, what is the actual uncertainty in the average onset values due to uncertainties in RH?

Authors’ response: See previous discussion on uncertainties.

Reviewer 1 comment: 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.

Authors' response: In order to keep Figure 8 less cluttered, we did not include the ammonium sulfate data from Baustian et al. (2010). We performed student's t-test with the binned anhydrous NaCl and hydrated NaCl S_{ice} data. For a two-tailed T test, the P-value is 0.0012. Therefore, there is a statistical difference between the S_{ice} values for the anhydrous and hydrated NaCl particles. Page 16, line 363 now reads, "Student's t-test was performed with the binned anhydrous NaCl_(s) and hydrated NaCl_(s) S_{ice} data. For a two-tailed T test, the P-value is 0.0012. Therefore, there is a statistical difference between the S_{ice} values for the anhydrous and hydrated NaCl_(s) particles."

Reviewer 1 comment: 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.

Authors' response: By consistent, we meant that there is a smooth transition between ice nucleation at the lower temperatures and deliquescence at the higher temperatures.

Reviewer 1 comment: 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.

Authors' response: It should have been explained that during Raman collection, the cross hairs were positioned on the particle. During image collection, the cross hairs were moved from the particle so the particle could be seen well. The last line of the caption on Figure 1 is removed from the revised manuscript.

Reviewer 1 comment: Figure 3: I suggest showing an enlarged view of the spectrum where the peaks are located and skipping wavelengths not important for interpretation.

Authors' response: The middle portion of the spectrum (which has no peaks) has been removed from the figure in the revised manuscript.

Reviewer 1 comment: Page 23140, line 27: Missing "1" in " 1×10^{14} kg".

Authors' response: This line has been removed in the revised manuscript.

Reviewer 1 comment: Page 23142, line 3: Citation erroneous.

Authors' response: The reference is to a book which shows the NaCl phase diagram. We have

changed the reference in the revised manuscript.

Reviewer 1 comment: 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.

Authors’ response: The manuscript now reads (page 5, line 116), “Additional details are provided when the current experiment differs from that of Baustian et al. (2010) and Wise et al. (2010).” We have changed “waters of hydration” to simply say “water”

Reviewer 1 comment: Page 23149, line 11: Maybe change “It was previously shown. . .” to “It was shown above. . .”.

Authors’ response: The manuscript now reads (page 15, line 296), “It was shown above (Figure 1) that at 244 K a mixture of NaCl_(s) and hydrated NaCl_(s) particles form upon efflorescence of solution droplets.”

Reviewer 1 comment: Figure 5: Please use lighter shading. Crossed green circles could be a bit larger.

Authors’ response: While the PDF used to create Figure 5 had lighter shading, we agree that it looks too dark in the published version. We will lighten it further. We have made the green circles slightly larger to make them more visible.

Reviewer 1 comment: Figure 9: Crossed green circles could be a bit larger.

Authors’ response: This has been changed in the revised manuscript.

Reviewer 2 Comment: In the Abstract, I suggest adding the size of particles that are used and referring to how ice deposition onset is defined. In particular, there is no unique onset value for deposition ice nucleation, being dependent upon particle size, total surface area, numbers of particles that are being observed, observation time, etc. The onset RHs for deposited particle experiments such as that used here, with particles a few microns in size, tend to have substantially lower ice onset supersaturation ratios than do continuous flow diffusion chamber results that use smaller particles. This is important because modellers may be tempted to take the value quoted in the Abstract at face value, without noting the context within which the experiments were conducted.

Authors’ response: The manuscript now reads (page 1, line 24), “NaCl_(s) particles (~1 to 10 μm in diameter) deliquesced at 75.7±2.5 % RH which agrees well with values previously established in the literature.” Furthermore, the manuscript now reads (page 1, line 33), “Here, depositional ice nucleation is defined as the onset of ice nucleation and represents the conditions at which the first particle on the substrate nucleated ice. Thus the values reported here represent the lower limit of depositional ice nucleation.”

Reviewer 2 Comment: A few experiments details: How was the quartz substrate made hydrophobic? Does the Raman laser probe the full depth of the particle? How is the RH known? I realize some of these points may be in earlier publications but just a sentence on each would help the reader.

Authors' response: The reviewer is correct, the information about the preparation of the quartz substrate is detailed in the earlier publication referenced. However, the manuscript now reads (page 5, line 120), "The particles exiting the atomizer were then impacted onto a hydrophobic quartz disc (silanized with RainX prior to experimentation) for analysis."

We can obtain Raman spectra of the entire depth of the particle. It really depends on where the Raman laser is focused. The manufacturer of the Raman microscope claims a confocal resolution of 2 microns. Page 6, line 134 now reads, "The focal point of the Raman laser can be adjusted. Thus, spectra representative of the chemical constituents contained throughout the depth of the particle can be obtained."

In response to how the RH is known, the manuscript now reads (page 6, line 136), "Frost point measurements from the hygrometer allow determination of the water partial pressure within the cell. The sample temperature is measured using a platinum resistor sensor embedded in the sample block. Temperature calibrations are performed as described in Baustian et al. (2010). The average error in the temperature calibration is ± 0.2 K for all experiments. The water partial pressure and sample temperature are used together to determine the RH during experimentation."

Reviewer 2 Comment: P23146. Concerning water uptake below the DRH for NaCl. I seem to recall that George Ewing at Indiana was the first to observe this. If so, then his work might deserve a reference.

Authors' response: The manuscript now reads (page 9, line 201), "Furthermore, Ewing (2005) shows that at water vapor pressures of ~ 20 mbar at 24°C (67% RH), water adsorbs to the surfaces of NaCl crystallites with a surface coverage of ~ 4.5 monolayers (see Figure 9 of Ewing 2005)."

Reviewer 2 Comment: P23148. What is meant by the "cracking of a particle"?

Authors' response: "Cracking" can be seen in the last panel of Figure 4. The particle starts out round and then appears to break up into smaller pieces after efflorescence. In order to make this more clear to the reader we have added (page 13, line 275), "The cracking phenomenon, which is the physical separation of a particle into multiple pieces, is evident in the last panel of Figure 4."

Reviewer 2 Comment: Figure 8. This comment follows on from Point 1 above.

The definition of ice onset needs to be presented in the Figure caption and the associated text.

Authors' response: The caption for Figure 8 now reads, “**Figure. 8.** S_{ice} versus temperature for the onset of depositional ice nucleation on NaCl (solid squares) and hydrated NaCl particles (open circles). An S_{ice} of 1 is denoted with the dotted line. Here, depositional ice nucleation is defined as the onset of ice nucleation and represents the conditions at which the first particle on the substrate nucleated ice.”

Reviewer 2 Comment: In the Dubessy et al. 130K work that measured the spectrum of the dihydrate, how was it known that the material under study was the dihydrate? i.e. is it known that both your experiment and the Dubessy et al. work are indeed looking at the same substance?

Authors' response: It is not known that the material in the present study is the dihydrate. From the Raman spectrum, we do know that the particles contain water. However, the DRH of the hydrate in this study does not match up with the theoretical DRH for the dihydrate. Thus we are not sure which hydrate of NaCl is being formed.

Further, we do not know that our material is the same substance as in Dubessy et al. In Dubessy et al, 1982, there was not an independent method used to determine the composition of the hydrate. Rather, they state that “solutions were prepared with such concentrations that the studied hydrate was almost the only phase to crystallize on cooling” Since neither experiment was able to independently confirm the hydrate formed, we simply do not know if the studies were examining the same hydrate.

We have rephrased the sentence accordingly. Page 11, line 234 now reads, “Dubessy et al. (1982) used the Raman microprobe MOLE to collect the Raman spectrum of a hydrated crystalline form of NaCl, supposedly $\text{NaCl}\cdot 2\text{H}_2\text{O}_{(s)}$ at 103 K.”

Reviewer 2 Comment: P23153. Typo: Q_{exp} or ext ?

Authors' response: The reviewer is correct, this is a typo and it will be changed in the revised manuscript.

Reviewer 2 Comment: P23154. Why are the radiative forcing calculations done at 532nm? It would be a much more convincing calculation (and presumably not so hard to do), to simply do the calculation over all solar visible wavelengths, weighting the results by the solar flux at each wavelength at the top of the atmosphere. As presented, it is a bit unusual, showing the Mie resonances in

Figure 11 that arise because the calculation was done at only one wavelength and particle size. These resonances are not easily observable in the atmosphere. To have more atmospheric relevance, I strongly suggest that a more representative calculation be done.

Authors' response: This section of the manuscript has been significantly reworked. We refer the Reviewer to this entire section for the response to this comment.

Reviewer 2 Comment: Figure 10. On that note, providing a few more details about the trajectory analysis would also be useful. As presented in the paper, there is really not much detail given for an atmospheric dynamics novice to know how the calculation was done.

Authors' response: The manuscript now reads (page 18, line 400), “The result of the model is shown graphically in Figure 10. Specifically, the temperature, relative humidity, and NaCl phase was tracked along parcel trajectories after they were detrained from deep convection (at 100% RH). (See (Jensen et al., 2010) for details.) The particles were initially assumed to be aqueous NaCl with 100% relative humidity with respect to ice. The RH in each parcel was tracked on its path upward through the tropical upper troposphere. If the RH dropped below 35%, NaCl particles in the parcel were assumed to effloresce. They remained NaCl_(s) unless the RH increased above the deliquescence point (80%). By combining results from 648 trajectories throughout the tropics, statistics were generated of the time when NaCl particles were in aqueous or solid states. It was calculated, at temperatures below 220 K, that hydrated NaCl_(s) is present 40-80 % of the time in the troposphere.”

Reviewer 2 Comment: Just wondering ... was it ever observed that ice nucleation could induce conversion of an anhydrous particle over to a hydrate? i.e. Did the Raman spectrum ever change after a cycle of ice formation and evaporation?

Authors' response: Interesting question! In all the experiments we performed, we never observed the conversion of an anhydrous particle to a hydrate due to ice formation and subsequent evaporation. We only observed the hydrate forming from efflorescence of a liquid droplet at low temperature.

Reviewer 3 comment: Title: ‘hydrated NaCl’ might imply liquid aqueous droplets. Possibly write: ‘crystalline hydrated NaCl...’

Authors' response: This suggestion has been incorporated into the manuscript.

Reviewer 3 comment: 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).

Authors' response: The reviewer is correct; components other than NaCl could influence the crystallization kinetics/optical properties of sea salt. Thus the complex sea salt composition should be included in laboratory experiments and models similar to the one in the Tang et al. paper. This manuscript represents the first attempt to characterize the crystallization and ice nucleating ability of sea salt at low temperatures using NaCl as a proxy for sea salt. As a first attempt, we do not think using NaCl as a proxy for sea salt is unjustified because NaCl is the dominant ionic compound present. There is precedence for using NaCl as a proxy for sea salt. For example, Cziczo et al. (*J. Phys. Chem. A* 2000, *104*, 2038-2047) study particles composed of NaCl and MgCl₂ to better understand the behavior of particles of marine origin. With that being said, further experimentation on crystallization and ice nucleation needs to be carried out with mixtures that more closely approximate the actual composition of sea salt particles. We now state in the manuscript (page 2, line 61) that, "Although natural SSA are chemically complex (which can affect water uptake properties) NaCl has been widely used as a proxy for SSA. The use of NaCl as a proxy for SSA is appropriate because many of the physical properties of SSA are controlled by NaCl. For example, Tang et al. (1997) found that for a specific dry particle size distribution, NaCl aerosol scattered light as efficiently as freshly formed sea salt aerosol (per unit mass)."

Reviewer 3 comment: What is the rate of change of RH in the experiments?

Authors' response: The rate in change of RH is ~1-10%/minute. We change the rate more slowly near phase transitions and more rapidly at all other times. The manuscript now reads (page 6, line 142), "During experimentation, the rate of RH change ranged from 1-10 % RH per minute."

Reviewer 3 comment: p23146, ln6. The cubes do not look like polycrystals, so Mikailov's capillary suggestion doesn't fit. Is there another explanation?

Authors' response: We agree with the reviewer that the capillary suggestion presented in Mikailov's paper isn't necessarily at play in the present work. We now state (page 9, line 199), "While the NaCl particles don't appear polycrystalline, perhaps microscopic cracks or fissures could cause similar water uptake."

Reviewer 3 comment: p23147, ln 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.

Authors' response: The discussion of ice in this section has been removed from the revised manuscript.

Reviewer 3 comment: Are there hydrates of other materials which dehydrate on reducing RH? Is there precedent for this?

Authors' response: The Reviewer brings up an interesting question. Yes, indeed there are

studies in an electrodynamic balance that show the transition between dry and various crystalline hydrates upon drying and humidifying for LiI crystals. Page 13, line 276 of the revised manuscript now reads, “A similar behavior of a hydrated crystalline solid losing hydration water to form a less hydrated or dry crystalline form has also been observed in single aerosol particles consisting of LiI (Kurtz and Richardson, 1984).

Our lab has done similar experiments with perchlorate hydrates and we do not see the dehydration phenomenon. This may be due to slow dehydration kinetics that does not exist with NaCl hydrates. For example, Vaniman et al. (*Nature* 431, 663-665) “found that crystalline structure and H₂O content (of magnesium sulfate) are dependent on temperature–pressure history” and “that an amorphous hydrated phase with slow dehydration kinetics forms at <1% relative humidity.”

Reviewer 3 comment: 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.

Authors’ response: We agree with the reviewer that the experimental points are scattered around a value of 87% RH. This value is close to where the dihydrate would deliquesce at 240 K. However, the scatter in the data for the dihydrate DRH is large when compared to the scatter for dehydrated NaCl DRH. This gives us reason to pause when assigning the identity of the hydrated NaCl. We are not ruling out the dihydrate on this basis. Rather we are saying that the identity is not fully understood. On page 15, line 320 of the revised manuscript we state, “However, the experimental points are scattered around a value of 87% RH. This value is close to where NaCl·2H₂O_(s) would deliquesce at 240 K (~85 % RH; Koop et al., 2000). However, the scatter in the DRH data for NaCl·2H₂O_(s) is large when compared to the scatter for NaCl_(s) DRH. Therefore, we cannot definitively confirm the identity of the hydrate.”

Reviewer 3 comment: Fig 5: A key would be helpful.

Authors’ response: We have incorporated this suggestion into the revised manuscript.

Reviewer 3 comment: P23151, ln 8. But the ammonium sulphate data isn’t compared in the plot. Was the ammonium sulphate data for the same size droplets?

Authors’ response: This comparison is removed from the revised manuscript.

Reviewer 3 comment: For the ice nucleation work, is it possible to estimate ice active surface site densities? 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.

Authors' response: The Reviewer brings up a very good point concerning the estimation of ice surface active densities. Unfortunately, we have no way of estimating surface active densities given our experimental set-up.

Reviewer 3 comment: 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?

Authors' response: We agree with the reviewer that the hydrated particles appear to be rougher than the anhydrous particles. The article meant by the referee is not Zuberi et al. GRL 2002, but rather Zuberi et al, J. Phys. Chem. A 2001. We now discuss this in the text.

Page 17, line 379 now reads, "The hydrated $\text{NaCl}_{(s)}$ particles prepared in this study appear to have a rougher surface than that of the dehydrated $\text{NaCl}_{(s)}$ particles. This surface roughness might be one reason for the very low ice nucleation threshold for such particles. It has been shown previously that ammonium sulfate crystals can act as IN in an immersion ice nucleation process (Zuberi et al., 2001). In these experiments it was observed that crystals with a polycrystalline structure, i.e. a rougher surface, nucleated ice at very low supersaturation ($S_{\text{ice}} \sim 1.08\text{-}1.18$) while smooth single crystals required significant supersaturation ($S_{\text{ice}} \sim 1.64\text{-}1.67$).

It has been suggested that heterogeneous ice nucleation is initiated when an ice embryo forms at an ice-active surface site. Sullivan et al. (2010) tested this hypothesis by "processing" Arizona Test Dust with sulfuric acid. They found that the processed dust had a decreased ice nucleation efficiency compared to the unprocessed dust. This result was attributed to the acid digestion of ice surface sites. Although we do not have any direct evidence for hydrated $\text{NaCl}_{(s)}$ particles having more ice-active surface sites than dehydrated $\text{NaCl}_{(s)}$ particles, it is a plausible hypothesis given the morphology of the particles. Another plausible hypothesis is that the hydration waters may be good sites to adsorb further water and nucleate ice"

Reviewer 3 comment: Also, how important is sea salt aerosol in the upper troposphere in terms of radiative transfer?

Authors' response: It is known that sea salt can reach the upper parts of the troposphere and participate in cirrus cloud formation. For example, Cziczo et al. (2004) showed sea salt residuals in cirrus clouds over Florida. However, we are uncertain about what percentages of aerosol particles in the upper troposphere contain sea salt. Thus we are also uncertain about the importance of sea salt aerosol in terms

of radiative transfer.

Reviewer 3 comment: The modeling 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.

Authors' response: The manuscript now reads (page 19, line 411), “The lowest efflorescence temperature utilized in the laboratory studies presented in this manuscript was 239 K. Figure 10 shows a scenario that uses temperatures from 180-220 K. Thus a significant temperature extrapolation was assumed. This could introduce error in the model, especially at the lowest temperature. Between room temperature and 238K, we did not observe significant temperature dependence in the ERH. Thus it is reasonable to assume a similar trend at lower temperatures.

In a similar study to model the deliquescence and efflorescence behavior of ammoniated sulfate particles, Colberg et al. (2003) assumed a constant offset between the DRH and ERH independently of temperature based on available data for ammonium sulfate. We note that if the DRH of the hydrated form of NaCl increases slightly with decreasing temperature similarly to that suggested for NaCl·2H₂O_(s), one would expect an increasing ERH at the lowest temperatures. This would lead to a larger fraction of particles being in a hydrated crystalline form. Hence, our approach of a nearly constant ERH is a conservative estimate of the fraction of crystalline particles.

Further, we did not observe temperature dependence in S_{ice} over the experimental range down to 225 K. However, past work has shown that S_{ice} values do increase at temperatures below 180 K, (Trainer et al., 2009), so an extrapolation would be needed for cirrus at the very lowest temperatures in the atmosphere. “

Reviewer 3 comment: Fig 1. Why are the crosses, which indicate the position the laser was focused, not on the particles?

Authors' response: It should have been explained that during Raman collection, the cross hairs were positioned on the particle. During image collection, the cross hairs were moved from the particle so the particle could be seen well. The last line of the caption on Figure 1 is removed from the revised manuscript. The position of the crosshairs is now explained in the figure caption.

Reviewer 3 comment: Fig 2 c. The cross is missing in the picture.

Authors' response: As with Figure 1, the cross hairs were moved from the particle so that the particles could be seen well. It just so happens that in this image, the cross hairs were out of the field of view.

Reviewer 3 comment: 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.

Authors’ response: This suggestion has been implemented in the revised manuscript.

Reviewer comment: Fig 8. A key would be helpful

Authors’ response: This suggestion has been implemented in the revised manuscript.

Reviewer comment: 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?

Authors’ response: This has been clarified in the revised manuscript. Page 18, line 402 now says “The parcels were initially assumed to be aqueous NaCl with 100% relative humidity with respect to ice. The RH in each parcel was tracked on its path upward through the tropical upper troposphere. If the RH dropped below 35%, NaCl particles in the parcel were assumed to effloresce. They remained NaCl_(s) unless the RH increased above the deliquescence point (80%). By combining results from 648 trajectories throughout the tropics, statistics were generated of the time when NaCl particles were in aqueous or solid states. It was calculated, at temperatures below 220 K, that hydrated NaCl_(s) is present 40-80 % of the time in the troposphere.”