We thank reviewer #3 for taking the time evaluating our manuscript. We provided a point-by-point response to the reviewer's comments given in bold faced letters below.

The topic of this manuscript is of interest to experimentalists and modelers working in the area of ice nucleation and cloud microphysics and, hence, fits well into the scope of ACP. Both the experiments as well as the data analysis appear to be carefully performed and executed. The manuscript is well written and the length of the text, the figures, and the supplement are all appropriate. In conclusion, I consider the paper being publishable in ACP, after the following minor comments have been taken into account.

Thank you for this evaluation.

Minor comments:
1. P. 4921, L.9-11: I think it would be appropriate to reference the paper by Karcher and Lohmann, JGR 108(D14), 4402 (2003) in this context.
Thank you for pointing out this omission. We include the study by Kärcher and Lohmann who first attempted to include a water activity based parameterisation of immersion freezing in a cloud resolving model. We add on line 11:
"Kärcher and Lohmann parameterised immersion freezing by scaling the a_w based homogeneous ice nucleation theory (Kärcher and Lohmann, 2003)."

2. P. 4922, L.13-15, 23-26, and Table 1: It was not clear to me whether the concentration of LEO and PP given in Table 1 is that after filtration or before (it is mentioned in the text that ~75% of the PP and LEO is lost during filtration).
The concentrations given in Table 1 represent the final LEO and PP concentrations accounting for the loss on the filter. To make this clearer we change the sentence starting on p. 4922, l. 26:
"Lastly, a known quantity of $(NH_4)_2SO_4$ was added to the suspension resulting in an aqueous solutions with known PP or LEO and $(NH_4)_2SO_4$ content."
To
"Lastly, a known quantity of $(NH_4)_2SO_4$ was added to the suspension resulting in an aqueous solution with known PP or LEO and $(NH_4)_2SO_4$ content as given in Table 1."

3. P. 4924, L.10-13: I do not understand why a partial solubility of PP may explain the difference in surface area measurements using SEM and BET.
Please see also response to referee #1 and #2. In brief, for BET derived surface areas, the PP and LEO remain anhydrous and thus their macromolecular size and orientation remains mostly unchanged. For SEM derived surface areas, the particles are placed in water, and as a result parts of these macromolecules possibly dissolve or rearrange thereby altering the size and orientation with subsequent effects on the visible condensed phase surface area upon water evaporation due to the high vacuum environment of the SEM.
We change the sentence on p. 4924, l. 11:"The reason for this difference may be due to the partial solubility of PP." to
"A possible reason for this difference is that PP and LEO remain anhydrous when examined by BET and thus their macromolecular size and orientation remains mostly unchanged. However, for SEM derived surface areas, these particles are placed in water and parts of these macromolecules may dissolve or rearrange thereby altering their size, orientation, or lead to breakup of particulate aggregates with subsequent effects on the visible condensed phase surface area."

4. P. 4925, L.16-19: I assume that the agreement of the measured ice melting points to the ice melting curve (from AIM?) implies a negligible solubility of LEO and PP. Is this correct? This would imply that the dissolved fraction of LEO and PP does not change aw significantly.

These conclusions are correct. The ice melting curve as a function of water activity is an updated parameterisation by Koop and Zobrist (2009). This will be included in the sentence. As shown in the supplementary file, the melting points as a function of weight percent are in agreement with AIM (Clegg et al., 1998). We will include this citation in the original sentence. We furthermore add one sentence to indicate that these results imply that water activity is not significantly affected by the presence of LEO or PP.

We change the original sentences on p. 4925, l. 16:

"Within the experimental uncertainty, mean melting temperatures of water and aqueous $(NH_4)_2SO_4$ droplets containing LEO and PP are in good agreement with the ice melting curve."

to

"Within the experimental uncertainty, mean melting temperatures of water and aqueous $(NH_4)_2SO_4$ droplets containing LEO and PP are in good agreement with the ice melting curve (Koop and Zobrist, 2009; Clegg et al., 1998)."

We add the following sentence:

"Agreement of measured ice melting temperatures with expected melting points (Koop and Zobrist, 2009; Clegg et al., 1998) indicates negligible effects on droplet a_w and composition due to the presence of LEO or PP."

5. P. 4926, L.1-2: I think it would be appropriate to reference the papers by Young and Leboef ES&T (2000) and by Koop et al. PCCP (2011) in this context.

Agreed. We include both references in the sentence. It now reads:

"At determined freezing temperatures and $a_w < 1$, these HULIS surrogates are most likely in a solid (glassy) state as inferred by a previous study employing fulvic acid particles as IN (Young and Leboef, 2000; Koop et al., 2011; Wang et al., 2012a)."

6. P. 4926, L.3-7: I think it would be appropriate to reference the paper by Karcher and Lohmann, JGR 108(D14), 4402 (2003) in this context, see above.

We respectfully disagree with the referee in this point. The text describes how the ice melting curve is shifted by Δa_w to obtain the immersion freezing curve, independent of homogeneous ice nucleation. However, Kärcher and Lohmann as shown in their Fig. 2, *shift the homogeneous ice nucleation rate coefficient*, J_{hom}, by Δa_w to derive the heterogeneous ice nucleation rate coefficient, J_{het}, under the assumption that the nucleation rate is 1 s^{-1} at a supersaturation of 1.2. As a result J_{hom} and J_{het} possess exactly the same temperature dependence. This is clearly a different approach compared to our description in this section of this paper. For this reason, the study by Kärcher and Lohmann (2003) is not mentioned when describing the water activity shift of the ice melting curve.

7. P. 4928, L.17: In Figs.2a and 3a, isn't Jhet(exp) increasing stronger than exponentially with decreasing T? Is it possible that the very steep increase of the last points at low temperature is already affected by homogeneous ice nucleation? May this contribution result in a potential bias in terms of the derived Jhet(exp)? Please proof that you can exclude such contributions from homogeneous ice nucleation.

We do not expect that homogeneous ice nucleation has a significant impact on derived J_{het} for the following reasons. Figure 1 gives the expected homogeneous ice nucleation temperature and corresponding J_{hom}=2E6 cm^{-3} s^{-1}. If we calculate J_{hom} for just 1.5 K

warmer conditions we obtain J_{hom}=3780 cm^{-3} s^{-1} (Koop et al., 2000). Assuming an average droplet diameter of 50 µm, this results in a nucleation rate of J_{hom}*vol=0.00025 s^{-1}. Thus, to observe 1 homogeneous ice nucleation event, the droplet has to stay at given temperature for about 4000 s. However, in our experiments the droplets experience a specific temperature only for a few seconds. For this reason we believe that homogeneous ice nucleation is unlikely to impact the freezing at the lower temperatures.

To make this point clearer we add the following information on p. 4928, l. 18:
"It should be noted that homogeneous ice nucleation is unlikely to have affected derived J_{het} since homogeneous ice nucleation rates 1-2 K above the expected homogeneous freezing curve shown in Fig. 1 are too slow to induce freezing under applied experimental time scales (Koop et al., 2000)."

8. P. 4930, L.25-26: Regarding the increase in alpha with decreasing temperature seen here (and also in previous studies): Can you speculate about any rational explanation/reason for this observation which implies that the compatibility between the IN and ice in the immersion mode becomes worse at lower temperature?
See also response to reviewer #2. The contact angle is defined according to Young's equation as the combination of three surface tensions relating to the three interfaces between the IN, ice, and liquid (Pruppacher and Klett, 1997):
$\gamma_{SI} + \gamma_{IL}\cos\alpha = \gamma_{SL,}$
where S: solid (IN), I: ice, L: liquid or aqueous phase. These surface tensions depend on temperature and composition of the aqueous phase. Thus it should come as no surprise that the contact angle defined in that manner may vary with temperature and water activity as previously discussed (Alpert et al., 2011, Zobrist et al. 2007). A recent study by Welti et al. (2012) found a similar relationship that the contact angle increases as temperature decreases. Furthermore, these authors, using physical parameterizations of the interfacial energies, taken from Helmy et al. (2004) and Pruppacher and Klett (1997), obtain a reasonable temperature dependent interfacial energy between the IN surface and the ice embryo corroborating the observed temperature dependency of the contact angle. Zobrist et al. (2007) also found that keeping the contact angle fixed with temperature and varying the pre-exponential factor in the equation of J_{het} did not result in an improved representation of the experimentally derived J_{het} compared to a temperature dependent contact angle. As pointed out in response to reviewer #2, this may be due to the unphysical temperature dependence of the interfacial energy when keeping the contact angle constant as derived by our sensitivity study.

We will add a very brief discussion on p. 4931, l. 9:
"The contact angle is defined by Young's equation which relates α to the three interfacial surface energies between the ice nucleus and ice, ice nucleus and aqueous solution (water), and ice and aqueous solution (Pruppacher and Klett, 1997; Alpert et al., 2011). These interfacial surface energies are temperature dependent and will be affected by the presence of solutes. A recent study by Welti et al. (2012) also found that the contact angle increases as temperature decreases. Using physical parameterizations of the interfacial energies taken from Helmy et al. (2004) and Pruppacher and Klett (1997), these authors derive a reasonable temperature dependent interfacial energy between the IN surface and the ice embryo and thus corroborate the observed temperature dependence of the contact angle being a physical phenomenon. Zobrist et al. (2007) have shown, that keeping the contact angle constant for all temperatures and varying the pre-exponential factor in Eq. 5, does not improve the representation of J_{het} with temperature compared to

the implementation of a contact angle that varies with temperature. Here, we have determined that when keeping the contact angle constant, σ_{sl} increases with decreasing T in contrast to the expected behavior (Pruppacher and Klett, 1997; Zobrist et al., 2007; Alpert et al., 2011b, Welti et al., 2012) further supporting a temperature dependent contact angle where decreasing temperatures yield increasing contact angles."

9. Fig. 4. and 5: The Jhet(exp) model seems to be significantly off the data in the panels for the lowest aw. Can you provide an explanation?

We believe that the referee refers to the α(T) model instead of the J_{het}(exp) model. The J_{het}(exp) model generates data points which lie almost perfectly on top of the measured frozen fraction points. The deviations in f between experimentally derived f and modeled f are due to the use of the fit to α(T). This fit yields a continuous function of J_{het} which lies within the experimentally derived J_{het}. For the lowest a_w, at higher freezing temperatures for Leonardite, the continuous J_{het} calculated from fitting α(T) deviates from the experimental values by about half an order of magnitude. When calculating the cumulative f, this leads to this apparent discrepancy between experimentally derived f and calculated f. In other words, the scatter in experimental data compared to the continuous model can lead to this unfortunate deviation. It is important to consider that this deviation first affects the continuous f curve derived from α(T) at the few highest freezing temperatures, but then propagates through the whole cumulative frozen fraction. Again, this is not a problem of the model, since J_{het}(exp) agrees perfectly, but just a matter of how α(T) is fitted.

To explain this issue we add on p. 4933, l. 12:

"f derived by the continuous function $J_{het}^{\alpha}(T)$ deviates in two cases at low a_w significantly from experimentally derived f. This is due to the application of a fitted α(T) curve that results in a continuous $J_{het}^{\alpha}(T)$. However, $J_{het}^{\alpha}(T)$ can deviate by half an order of magnitude from experimentally derived J_{het}^{exp}, depending on the scatter of the data. When calculating and integrating the cumulative f, this deviation in J_{het} at the beginning (high freezing temperatures) propagates through the whole dataset which leads to this apparently lesser fit quality. Again, this is not a problem of the model itself, since f derived from J_{het}^{exp} agrees almost perfectly with experimentally derived f, but is a matter of how α(T) is fitted."

10. P. 4938, L.10-11: I am puzzled by the statement that 'the alpha(T)-model yields alpha and Jhet values directly from the experimental data, thus, no fitting is involved.' Isn't alpha fitted such that Jhet matches the observed Jhet values?

We did not mean to cause confusion here. The α(T)-model is applied to derive α from experimentally obtained J_{het} in a discrete manner, i.e. data point by data point. In this case, no fitting is involved. These α in turn represent the experimentally derived J_{het} and/or frozen fractions – still no fitting involved. To obtain a continuous function $J_{het}(T)$, applicable e.g. in cloud resolving modeling studies, α can be expressed by a fit. The individually determined J_{het} values or the continuous $J_{het}(T)$ from fitted α(T) can then be applied to also determine frozen fractions. The α(T) model is entirely based on CNT and thus we do not fit/generate e.g. a α-data set that reproduces measured J_{het} or frozen fractions. In fact this is one of our main points in this paper, that the physical model underlying CNT is able to explain all data inclusive frozen fractions and thus allows application of the experimental findings to conditions outside of the experimental boundaries in contrast to fit-based analysis of the experimental data.

To make this point clearer, we change the original sentence

“It should be emphasized that only the α(*T*)-model yields α and *J*het values directly from the experimental data, thus, no fitting is involved.”
To
“It should be emphasized that only the α(*T*)-model yields α and *J*het values directly from the experimental data, thus, no fitting is involved except if a continuous function of J_{het}(T) and f(T) is desired.”

Technical comments:
11. P. 4927, L.20: Typo: ‘Jhet *is* calculated for all: : :’
Corrected as suggested.

12. P. 4928, L.17: I assume ‘a temperature decrease by *about* 10 K: : :’ is meant?
Yes, corrected as suggested.

13. P. 4931, L.12-14: I assume with ‘that can *lie* outside of the : : : probed in the laboratory’ you rather mean to say ‘that can *be predicted also* outside of the : : : probed in the laboratory’?
We agree that this is an awkward sentence. We meant to say that laboratory derived J_{het} can be applied to predict ice nucleation rates or frozen fractions for IN surface areas and nucleation times that were not probed in the laboratory.
We re-phrase the original sentence:
“J^{α}_{het}(*T*) can then be applied to given IN surface areas and nucleation (cloud activation) times that can lie outside of the IN surface areas and nucleation time range probed in the laboratory.”
To
“J^{α}_{het}(*T*) can then be applied to predict ice nucleation rates or frozen fractions for IN surface areas and nucleation (cloud activation) times that were not probed in the laboratory.”

14. Fig. 4. and 5: The frozen fraction f is without unit, hence I suggest to remove ‘a.u.’

Corrected as suggested.