Answer to "Anonymous Referee #2"

1) **Applicable range of parameterization**

   The manuscript states that the parameterization of ns is valid between 226 and 250 K. In Figure 6, the experimental data starting at 223 K was excluded. Does that mean we need another parameterization for temperature lower than 226 K? Why the parameterization did not include the data at lower temperature? One of the purposes of parameterization is to use it in modeling studies for wider ranges of conditions, including temperature. As for the current parameterization (eqn. 16), the observed ns are within the 2 orders of magnitude. If include the data, it may go up to 3 or 4 orders of magnitude. If consider this variation/uncertainty, how would this affect on the results of the box model simulation? In other words, any sensitivity test in box model simulation to include uncertainty of the parameterization (two constants in eqn. 16)?

   The experimental data starting at 223 K was excluded because we observed that the humidity at which the ice nucleation onset is observed does not change much between 223 and 226 K. Thus, since the temperature dependence becomes less important we assume that ice nucleation should ideally be described by a different function for x_{therm}. Nevertheless, Fig.6 also demonstrates that ice formation can largely be described by our proposed ns function even below 226 K. Also, we would like to highlight that our study is mainly intended to provide a case study for investigating deposition nucleation properties of aerosol particles. The sensitivity of the two parameters in the ns equation can be roughly estimated from the experimental data. The individual experimental ns trajectories (see Fig.6) show that the two coefficients in the ns equation can be maximally varied such that the ns value itself varies by roughly one order of magnitude. For the first coefficient (a1,b1) this variation directly would be caused by a change within one order of magnitude, whereas for the second coefficient (a2, b2) this variation translates into a change within +/-15%.

   We would expect similar results from box model calculations investigating parameterizations taking this sensitivity into account, i.e. only variations within approximately one order of magnitude.

2) Estimation of the time dependence from classical nucleation theory

To investigate the time dependence, this study proposed another two equations (eqn. 19 and 20) for parameterization. These three (including previous one, eqn. 16) equations and three corresponding parameterizations were obtained for the same data set. When do the simulation for a reasonable range of pre-settings (temperature/RH/time), isn't it expected similar simulation results? To confirm the time dependence conclusions derived from box model simulation using ns parameterizations, one possible way is to do the same simulation using parameterizations based on classical nucleation theory (CNT).

The simulations that were performed for the three different parameterizations were intended to demonstrate how the ice crystal concentrations would develop if conditions were applied which differ from those which were investigated experimentally. Therefore, we chose starting conditions with very low aerosol concentrations and a wide variation of cooling rates. And even though the time dependence and thus the difference between the parameterizations was relatively small, we still wanted to estimate the variation of the predicted ice crystal concentrations in different scenarios.

We agree that extracting the time dependence by using classical nucleation theory might appear to be more promising in order to estimate the time dependence. However, there are three reasons why there are difficulties with this approach. First, the most important reason is that classical nucleation theory assumes that the temperature dependence is much weaker than we actually observed (compare to calculations presented in Fig. A1 in the review by Hoose and Möhler (2012)). Secondly, in the experiment the nucleation phase is associated with larger measurement uncertainties than later phases of the experiment. Thirdly, at different temperatures a variation in the contact angle is expected which adds another degree of complexity (Welti et al., 2012). These measurement uncertainties are caused by fluctuations in the observed ice crystal concentration at the beginning of ice formation. These two factors contribute significantly to the observed variation in contact angle distribution parameters which are estimated from the experimental data. Looking forward, using contact angle distribution parameters with large uncertainties will translate enormous deviations of the predicted ice crystal concentrations. Also, classical nucleation theory is apparently not able to describe deposition nucleation by ATD particles.

Minor comments

1) *p.18500, Eqn. 2,although used as a dimensionless parameter, is there any physical reason to directly add T and* S_{ice} *as a “thermodynamic” variable? The term of* x_{therm} *shouldn’t be identified as thermodynamic variable.*
   We will rename x_{therm} from “thermodynamic variable” into “temperature and saturation dependent function”.

2) p. 18500, define Sice.
   We will add on p.18500 l.20: “…with the saturation ratio with respect to ice $S_{ice}>1$…”

3) *p.18503-18504 and section 3.2.3, about the CNT and data analysis, what are the J values or how the J was used to derived theta?*
   Which value of surface tension at the ice/vapor interface was used? Please provide the values of the parameters used in CNT analysis.
   The Delta *gd in Chen et al. (2008) (Fig. 2 of the paper) was about 2E-20 J for temperature lower than 223 K. Why the Delta gd of 4E-20 J was used here?*
   - Regarding the heterogeneous nucleation rates, we would like to point out that we did not explicitly calculated heterogeneous nucleation rates. However, we implicitly used the heterogeneous nucleation rates J_{het} to calculate the ice fractions (eq. 11 from Wheeler et al. (2012)):

     $$\frac{N_f}{N_0} = 1 - \int_0^{\infty} \int_0^{\pi} \exp\left[-\pi D^2 J_{\mathrm{het}}(\alpha, T, S_{\mathrm{ice}})\, t\right] f_{\alpha}(\alpha)\, f_{\mathrm{num}}(D)\,\mathrm{d}\alpha\,\mathrm{d}D,$$
   - The surface tension was described as a temperature dependent function according to Pruppacher and Klett 1997 (see eqs. (5–46), (5–47a), and (5–12)).
     We will add on p. 18503 l. 18: “The surface tension was described as a temperature dependent function according to Pruppacher and Klett 1997.”
   - It is true that the Δg_d as directly inferred from the experimental data for ATD was at roughly $2.0\cdot10^{-20}$J (Chen et al., 2008). However, in the literature data cited in the study by Chen et al. (2008) for mineral dusts generally higher values were found, e.g. $8.7\cdot10^{-20}$J for ATD (Gustafsson et al., 2005). Therefore, we slightly adjusted the value of Δg_d used in our calculations. Also, Δg_d has only a limited influence on the predicted ice crystal concentrations compared to contact angle distribution parameters (Chen et al., 2008).

4) p. 18505, l.26, define AIDA when it was used for the first time.
   We will add: “…(Aerosol Interaction and Dynamics in the Atmosphere)…” on p.18505 l.23.

5) *p. 18508, l.11-14, does this mean that if there is sufficient water vapor and at high RH, particle with soluble materials will become aqueous droplets and could also be detected, especially at higher temperature, e.g. 250 K?*
   Yes, the WELAS instruments can detect droplets as well. For immersion freezing experiments, typically the formation of droplets is observed before these droplets are then converted into ice crystals at lower temperatures.

6) *p.18508, l.14-17, this manuscript focused on deposition ice nucleation, can you rule out the possibility of immersion freezing at high temperature, i.e., 250 K? If there is very small amount of soluble materials, once it takes up water, it will form aqueous particles then ice will nucleate through immersion freezing. It is not about the subsaturated conditions, it is about what are the soluble components and when it takes up water or deliquesces.*
We agree that immersion freezing cannot be ruled out completely but ATD particles contain only very small amounts of soluble components and thus droplet formation is very limited below RH_{wat}=90% (Vlasenko et al., 2005). Thus we assume that deposition nucleation is the dominant ice nucleation mechanism even at 250 K.

7) *p.18508, l.18-19, what is "a suitable size threshold"?*
With "suitable" threshold we wanted to express that the size thresholds that are used for the analysis of WELAS data are adjusted individually, depending on the aerosol size distribution and the ice nucleation mode. For immersion freezing, larger size thresholds may be used than for deposition nucleation experiments with the same aerosol types because for immersion freezing particles are activated to droplets which are larger than the aerosol particles.

8) *p.18508, define SIMONE when it was first used in the text.*
We will add: "…SIMONE (Scattering Intensity Measurements for the Optical Detection of Ice)" on p.18508, l.7.

9) *p.18511, l.22, how is the RHice uncertainty calculated? What are the uncertainties of gas and wall temperatures?*
RH_{ice} is derived from the absolute water vapor concentration as measured with the TDL (tunable diode laser) absorption spectroscopy and the water vapor saturation pressures with respect to ice at a certain temperature (Murphy and Koop, 2005). The deviation from the calculated saturation vapor pressures (Murphy and Koop, 2005) was less than 3% during the Aquavit campaign (Fahey et al., 2013). The measurement uncertainty regarding the well mixed cloud chamber is approximately ΔT=0.3 K which translates into an overall uncertainty of ΔRH=5%.

10) *p.18516, l.3-11 and Fig. 7, at 233 K, the RHice onsets are more than 10% lower than Kohler et al. (2010) and Welti et al. (2009), does that mean only the large particles nucleated ice in this study (polydisperse particles, see surface distribution in Fig.2)? How do these RHice onsets compare to the ice nucleation data by Knopf and Koop (2006).*
As assumed correctly by the referee, larger particles will initiate ice nucleation first. Thus, particle size distributions including larger particles (d<1µm) will indeed show ice nucleation onsets which are shifted towards lower relative humidities. This trend is also visible for our set of ice nucleation thresholds (see Fig. 4).
A comparison with the study by Knopf and Koop might not be appropriate because of differences in the experimental methods. Knopf and Koop (2006) used ATD solutions whereas in our experiments the dust was dry dispersed. Also, the range of humidities at which ice nucleation was observed by Knopf and Koop (2006) was very large. At 250 K, for example, ice nucleation occurred between RH_{ice}=105% and RH_{ice}=115%. So, even

though we observed ice nucleation at similar conditions, it would be difficult to draw solid conclusions from this comparison.

11) *p.18516, l.15-16, "devations" should be "deviations"? This statement didn't explain the deviation. If the ATD used in these studies are from the same source, the ice nucleation efficiency (RHice thresholds) by nature should be very similar and so the INAS at the same temperature. Does the statement in l.15-16 imply that the INAS parameterization provided here is only valid or limited to AIDA experiments? Then, how this parameterization can be applied for atmospheric application? Is there any other possible explanation for these deviations, what is the difference in surface area compared to the cited studies?*
We do not think that the INAS density approach is only applicable to AIDA results. We would like to point out that even though the particle diameters were given for the studies used for comparison, the aerosol surface area was not explicitly measured. Thus, some difference might as well come from differences between real and estimated aerosol surface areas. The differences between the results observed for different experimental setups highlight the necessity to achieve very small measurement uncertainties regarding temperature, relative humidity and aerosol surface area.
"Devations" will be changed into "deviations."

12) *p.18518,l.18-19, the manuscript didn't provide sufficient proof to support this statement.*
We would like to point out that there is a strong temperature dependence for deposition nucleation occurring between 235 and 250 K, because the ice nucleation onsets for similar experimental conditions vary strongly (see Fig.4). This strong temperature dependence is to our knowledge not reflected in current classical nucleation theory formulations.

13) *Table 1: It would be nice see the RHice threshold for each experiments.*
We can add the valued displayed in Fig.4. Thresholds in Fig. 4 are defined as humidity values at which ice crystal concentrations exceeding aerosol background concentrations were observed.

14) *Figure 5. please add description for the error bars showing in the figure.*
We will add the following sentence to the caption: "The error bars represent the measurement uncertainties in n_s with $\Delta n_s / n_s \approx 35\%$ and x_{therm} with $\Delta x_{therm} / x_{therm} \approx 5$."

15) *Since the parameterization is only valid for temperature above 226 K, it is misleading showing the blue solid line for 220 K. Where is the grey dashed line in the figure?*
The experimental results for 220 K and the two dashed lines (above and below the actual parameterization) are shown to illustrate that even though the x_{therm} formulation fits best between 226 and 250 K, deviations between experimental results and parameterization are within one order of magnitude even below 226 K.

16) *Any simulation at 250 K? Do they show similar results?*
We conducted selected simulations at 250 K. These simulations yielded conceptually similar results. However, we are only presenting results for 235 K, because for

atmospheric observations at 250 K, generally, there is a higher probability to find contributions by several ice nucleation modes. At 235 K, in contrast, deposition nucleation should be the dominant ice nucleation path for heterogeneous ice nucleation.

References

Gustafsson, R. J., Orlov, A., Badger, C. L., Griffiths, P. T., Cox, R. A., and Lambert, R. M.: *A comprehensive evaluation of water uptake on atmospherically relevant mineral surfaces: DRIFT spectroscopy, thermogravimetric analysis and aerosol growth measurements*, Atmos. Chem. Phys., 5, 3415–3421, doi:10.5194/acp-5-3415-2005, 2005

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