List of relevant changes

In the revised manuscript, changes in the text (except typos and small changes like commas) are marked in red.

Relevant changes can be find in

- Title
- Section 1 Introduction
- Section 2.3 Particle Samples
- Section 3.1.3 Comparison to literature data
- Section 3.2 Application of heterogeneous freezing models
- Section 4 Summary and Conclusions

Detailed responses to the referees are listed in the following.

Responses to Zamin Kanji (Reviewer 1)

First of all, we would like to thank the reviewer for the useful elaborated comments and suggestions which help to improve the new version of the manuscript.

All comments have been treated carefully by the authors, rewordings and deletions as suggested by the reviewer have been conducted, mentioned references have been added, and critical formulations have been reformulated or removed.

Where it is required, replies to some points are listed here:

Introduction: *This maybe true, but if you are going to mention it, you must provide a reason as to why is it important to have the freezing process observed while the droplets are already at their terminal velocity.*
That is not true for Ladino et al 2011 - drops are at their terminal velocity.
These critical statements have been removed from the revised manuscript.

Section 2.1.1: *This is unclear what the authors are trying to say, elaborate more?*
The ceramic surface of the Pt100 sensor does not show the same emissivity as an ice surface, this part has been elaborated in the revised manuscript.

Section 2.1.1: *Comment on at the atmospheric relevance of this stationary drop?*
The fact that the drops are very stationary in the acoustic levitator was described as an advantage because this allows to directly measure the surface temperature of the freezing drops. This is not possible with drops floating in the wind tunnel because of their movements in the air stream. Of course drops in the atmosphere are not stationary but freezing as a hydrodynamical process should not be affected. A comment has been added to the paper.

Section 2.1.2: *you should report how long it took the pure water drops to freeze above -35°C*
We never observed pure drops freezing above -35°C for time periods as long as 60 min.

Section 2.2: *Why choose 4s, when from your figures 5, it is clear that the time is closer to 5 seconds rather than 4?*
The reviewer is right, this has been changed in the revised manuscript and corresponding Figures 6a and 6b have been modified accordingly.

Section 2.3: *Mont K10 from sigma-aldrich are known to be acid treated and have different properties from untreated montmorillonite. This sample is not representative of any atmospheric* *dust. Something to this effect is mentioned in Pinti et al (2012), why was this not taken into consideration?*

These experiments were performed first in the very beginning to use montmorillonite K10 as a test substance to prove the function of the levitator. At that time we were not aware that it is not representative of atmospheric dust.

Section 2.3: *I think we know that sometimes the manufacturer's quote inaccurate compositions for various reasons. I am certain upon talking to various people from the INUIT community that there is more than just a trace of Feldspar. I suspect there is more Feldspar in this sample and this should be confirmed with XRF or XRD, should be easy to get a sample analysed.*

Based on analyses from an INUIT group, the numbers have been changed in the revised paper and the references are corrected.

Section 2.3: *Shouldn't this be (Hiranuma et al., 2014, in Prep) I think his publication is almost ready to submit.*

In the mentioned paper, illite IMt1 is not included but only illite NX.

Section 3.1.3: *This is very vague - the lines intersect at one point, but do not overlap, the slope is completely different therefore the temperature T50 dependency on SA is different too. You have to comment on this and offer some explanations as to why this may be.*

According to another reviewer's suggestion, the comparison with the median freezing temperatures of Broadley et al. and previous Figure 12 have been omitted and replaced by comparisons with the results of Knopf and Alpert (2013).

Section 3.2.2: *Why third order regression curve? Why not use the exponential function for Ns(T) that previous studies have used before, like Niemand et al., or Tobo* et al (2014) ACP)

We had the impression that the data points are represented best by a third order regression curve and followed Broadley et al.

Section 3.2.3: *Shouldn't you discuss on the suitability of which model is better used to represent your results, the time dependent or independent model?*

Some remarks about this issue have been included in the revised paper, in Sections 3.2.1 and 3.2.2.

Responses to Reviewer 2

First of all, we would like to thank the reviewer for the useful elaborated comments and suggestions which help to improve the new version of the manuscript.

General remarks:

1. According to the reviewer's suggestion, the title of the paper has been changed as follows:

   Particle surface area dependence of mineral dust in the immersion freezing mode: Investigations with freely suspended drops in an acoustic levitator and a vertical wind tunnel.

2. Due to the reviewer's comments, remarks about effects of charge on freezing have been removed.

3. The authors fully agree with the reviewer that the calculation of the specific surface area includes some errors due to the BET method which was used for illite NX and IMt1. The surface density of active sites has been selected to compare the results obtained with the same particle sample but with different methods. In a paper which will be submitted soon immersion freezing of illite NX of several techniques from the INUIT research group and others will be described. It includes discussions about the weakness of BET surface area, therefore, this issue has not been included in the present manuscript. The suggestion to use n_s in cloud models has been removed. All about parameterizations based on illite NX data will be included in the forthcoming paper (Hiranuma et al., ACPD, will be submitted end of August 2014).

4. Regarding Figures 10 and 11 vs. Figures 14 and 15, the reviewer is right that there is actually provided twice the same, one f_{ice} itself, the other $\ln(1-f_{ice})$. Therefore, Figures 10 and 11 have been omitted in the revised manuscript to reduce the number of figures.

Specific comments:

Section 2.2, line 10: We have include references of previous measurements in the revised paper.

Section 3.1.3, Fig. 12: We agree with the reviewer's comment that Figure 12 should be omitted as it does not have any added value and have remove it from the revised manuscript.

Section 3.2.1, line 22: According to the reviewer's remark, the somewhat vague formulation of the sentence has been changed into "…that the nucleation probability is the same for all drops of a population."

Section 4, line 14: The reviewer is right, the mentioned sentence does not make sense and have been removed from the revised paper.

Section 4, line 25: The mentioned issue is discussed in the revised paper.

Responses to Reviewer 3

First of all, we would like to thank the reviewer for the useful elaborated comments and suggestions which help to improve the new version of the manuscript.

Major comments:

Abstract, l. 4-5: Due to the reviewer's comments, remarks about effects of charge on freezing have been removed.

Abstract, l. 14-16: Statements about the time dependence of freezing are added to the revised paper.

Introduction, p. 12889, l. 13: According to the reviewer's comment, any valuation of the different experimental techniques has been avoided in the revised manuscript. Some sentences are added about the interpretation of the results by two freezing models. These are described in more detail in Section 3.2. However, the authors decided not to include too many details about the two freezing models as the paper is already rather long and those details are available in a number of previous references which are mentioned in the paper.

Knopf and Alpert (2013): In the revised version, the present results are compared to these previous results for illite immersed in pure water drops (water activity = 1), see Section 3.1.3. As the present measurements were undertaken only with pure water drops and not with solution drops of different activities, to our opinion, it might not make sense to apply the water activity model of Knopf and Alpert (2013). Therefore, only the median freezing temperatures of pure water drops are compared.

Experiment, p. 12890, l. 14: Previous Figures 1 and 2 are put together in one figure. In general, as suggested by the reviewer, two or three figures have been combined so that the number of figures is reduced to 10 in the revised paper.

p. 12892: Homogenous freezing was already investigated with the acoustic levitator and published in Ettner et al., 2004, and Diehl et al., 2009, which is mentioned in section 2.1. In Diehl et al. (2009), homogeneous freezing of sulphuric and nitric acid solution drops was investigated while the acid concentrations were varied. The minimum parameters for drop volume and time were 0.1×10^{-3} cm^3 and 90 s, respectively, thus, resulting in a nucleation rate of 111 cm^{-3} s^{-1}. The measured freezing temperatures followed the trend of the homogeneous freezing curve of Koop et al. (2000) and matched a nucleation rate of 100 cm^{-3} s^{-1} indicating that the Koop formulation is valid as well for large drop sizes as used in the acoustic levitator (Diehl et al., 2009). The data were also compared to previous measurements of Chang et al. (1999) and Vortisch et al. (2000).

A sentence mentioning the validation of homogeneous freezing in the levitator has been added in Section 2.1. To include a complete new discussion of homogeneous freezing in the present paper, as suggested by the reviewer, is not possible because of the experimental techniques. With the present set-up of the levitator in the walk-in cold chamber experiments regarding homogeneous freezing cannot be achieved as the ambient temperatures are not low enough (> -35°C). To measure homogeneous freezing at the vertical wind tunnel is in general not feasible as well as the ambient air temperatures in the tunnel reach -30°C only. Thus, the present investigations are limited to heterogeneous freezing.

p. 12893, l. 21-22: Following the reviewer’s suggestion, statements that ventilation and heat transfer do not significantly affect ice nucleation have been added to the revised paper (sections 3.1.2. and 4).

Results, p. 12897, l. 25: In the revised version, the present results are compared to the previous results of Knopf and Alpert (2013) for illite immersed in pure water drops (water activity = 1), see Section 3.1.3.

p. 12898, l. 16-19: In section 3.2 more detailed descriptions of the freezing models have been included as suggested by the reviewer.

p. 12900, l. 10-12: The mentioned phrases have been re-formulated according to the reviewer’s suggestion.

p. 12902, l. 5-7: In Section 3.2.2, it has been mentioned that the singular model has been used to interpret the data, in particular for comparisons of the two methods, although it neglects time dependence.

Conclusions, p. 12904, l. 5-6: Due to the reviewer’s comments, any remarks about effects of charge on freezing have been removed.

p. 12904, l. 7: The statement mentioned by the reviewer is included in the paper.

p. 12905, l. 4-15: Some phrases about the experimental techniques have been included in the Introduction. Line 9: This just means that if someone would like to perform experiments with an acoustic levitator a complete walk-in cold chamber is not required.

Conclusion section: The section was re-written as suggested by the reviewer except point 1). Details about homogeneous nucleation measured with the acoustic levitator were published already in Diehl et al. (2009), and to measure homogenous nucleation at the wind tunnel is not possible as the required low temperatures are not reached there (see also the remark to p. 12892, above).

Minor comments:

All minor comments have been treated carefully by the authors and changes have been included in the revised paper. Where it is necessary, to some points replies are listed here:

p. 12891, l. 13: The levitator was not flushed with particle-free air, but purity checks before starting each set of heterogeneous freezing experiments ensured that no undesired particles were present, i.e. if required cleaning was repeated until pure water drops did not freeze.

p. 12891, l. 11-12: As mentioned in Section 2.1.1, drops as small as 100 µm could be floated in the acoustic levitator but the measurements of the drop surface temperatures require larger drop sizes.

p. 12891, l. 18-20: Some more technical details regarding the electrical power output of the HF generator and its frequency are included in the revised paper, but the exact power levels in the acoustic field are not given by the manufacturer.

p. 12892, l. 2-3: To avoid confusions, the concentrations of the bulk solutions were not listed in Table 1 but only the particle surface areas per drop. The bulk solutions were selected in the way that in spite of the different drop sizes in the levitator and the wind tunnel the particle masses and surface areas per drop were similar.

p. 12892, l. 6-10: The images of the video camera were used afterwards to determine the drop sizes before freezing.

p. 12892, l. 16: From the levitated drops during the experiments, residues remain in the levitator which might disturb following freezing experiments. Therefore, purity checks were made with pure water drops (see Section 2.1.2). Furthermore, drop residues might pollute the oscillator and the reflector so that the function of the levitator is disturbed.

p. 12893, l. 26-27: As mentioned above, homogeneous freezing cannot be measured at the wind tunnel because the temperatures are not low enough. Checks were made before each series of experiments to ensure that pure water drops did not freeze by any pollutions.

p. 12894, l. 16ff: In the wind tunnel, the drops do not float in a stationary fashion as in the levitator. Therefore, measurements with the infra-red thermometer are not possible and any photo or film shooting is very difficult. For that reason drop sizes and surface temperatures have to be determined from the terminal velocities and ambient temperatures, respectively, as described in Section 2.2. How fast the drops evaporate is dependent on the daily status of the wind tunnel air, i.e. temperature and dew point. In general, it takes around 3 minutes to evaporate a 350 µm radius drop to a 150 µm radius drop and within the 30 to 40 s observation time, the drops shrink within the range of the size error.

p. 12896, l. 5: The mentioned paragraph has been included in Section 2.3 as suggested by the reviewer.

p. 12897, l. 11: As mentioned in Section 2.2, in the wind tunnel 40 to 50 drops for each temperature and particle type were investigated. During the observation time, some drops froze and some did not. Therefore, the numbers of *observed* drops per one single data point was 50 but the number of *frozen* drops per one single data point can vary from 1 to 50. Errors were determined due to counting statistics.

p. 12901: The problem was to account for a non-linear cooling rate, during the experiments of Murray et al. (2011) and Zobrist et al. (2007) the cooling rate was constant.