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.