General comment:

In this paper the question is raised whether the number of droplets analyzed in experimental freezing studies is large enough to constrain uncertainties of experimental parameters sufficiently and how uncertainties in relative humidity, temperature, time, and surface area present in droplets affect interpretation of laboratory ice nucleation, corresponding ice nucleation parameterization and extrapolation to atmospherically relevant conditions. To do this, simulations of droplet freezing are carried out for recently published experimental freezing studies. The authors come to the conclusion that indeed the variation of heterogeneous surface present per sample often leads to strong uncertainties in J_{het} for the number of droplets investigated in experimental studies. However, in their analysis the authors explain all uncertainties in J_{het} by variations of ISA (ice nuclei surface area) per droplet and do not consider that a single J_{het} does not apply to a whole sample of INP when the sample composition is heterogeneous i.e. for multi component and inhomogeneous samples. In the introduction they state that J_{het} can be viewed as a material parameter, but did not specify that this is only the case for a homogeneous material or sample. In cases where their evaluation procedure derives a large value for the fit parameter σ_g, this condition is not fulfilled and their analysis leads to erroneous results when they apply a single J_{het} to the whole ISA present in a droplet. By using just one J_{het}, a non-linear slope $\ln J_{het}/T$ is ascribed to variations of surface area, while it is indeed caused by a variation of J_{het}. Therefore, they need to discuss for all studies whether it is justified to apply a single J_{het} and remove the ones for which this assumption is not fulfilled, which unfortunately will be the case for most datasets (the ones performed with ATD, K-feldspar, illite, and natural dusts). The assumption of a single J_{het} only seems to be valid for the kaolinite KGa-1b (see specific comments). Taking variations of J_{het} into account influences much of the conclusions drawn in this paper and make some even invalid. The implications of this study (Sections 3 – 5) need therefore to be reconsidered and rewritten. Such a revision is needed for publication in ACP.

Specific comments:

Page 13112, line 27: comparison with a second order rate constant is not very helpful and might be removed.

Page 13114, lines 3-4: The singular hypothesis can be easily combined with a freezing point depression by determining a Δa_w.

Page 13116, lines 9 – 11: Lüond et al. (2010) and Marcolli et al. (2007) do not assume that every droplet contains the same ISA. The citations have to be revised.

Page 13133, line 9: Such an increase due to surface roughness is not justified when one considers kaolinite particles with 300 nm diameters (e.g. Welti et al., 2009).

Experiments Iso1 – Iso4 shown in Figure 1a: This figure shows experiments from Herbert et al. (2014; Figs. 4b (KGa-1b, 16 droplets) and 7 (K-feldspar, 20 droplets). Herbert et al. state that K-feldspar is a multicomponent system and should therefore be represented by different J_{het}, not just one. They write: "For a uniform species the decay of liquid droplets over time will be exponential (as was the case for kaolinite KGa-1b in Fig. 4b), whereas a diverse species will result in a non-exponential decay. Inspection of the data in Fig. 7 shows that the decay of liquid droplets was not exponential, again consistent with a diverse population of INPs." In the present analysis, the parameter σ_g is used to account for droplet to droplet variability. This seems to work as fitting procedure but has no physical meaning. The authors should discuss this. I suggest that they remove these data from the paper.

Experiment IsoWR shown in Figure 1b: ATD is again a multicomponent system and should therefore be represented by different J_{het}, not just one. I suggest that this dataset is removed from the paper.

Experiments IsoBR and IsoHe2 shown in Figure 2: Broadley et al. (2012) use a multiple component stochastic model to describe their data (Murray et al., 2011). This model describes systems in which there is more than one nucleating species or type of nucleation site. Each nucleation site can be described by a single temperature dependent nucleation rate coefficient and the total absolute rate of freezing is a function of the distribution of nucleation sites. This seems to be the appropriate way to interpret the illite NX data. Assuming just one J_{het} does not seem to be justified. Moreover, Broadley et al. (2012) rule out different surface areas present in different droplets as a valid explanation for their experimental results: “One explanation is that different droplets may not have contained the same surface area, due to an inhomogeneous distribution of particles or particle sizes between droplets, which could have occurred during nebulisation. However, the surface area of NX illite in the droplets which nucleated in the first half of run 20 would have needed to be about seven times larger than the surface area in the droplets which nucleated in the second half if only one type of nucleation site was present, which seems unlikely. In addition, this did not appear to be the case when we applied the same experimental technique to ice nucleation by kaolinite (Murray et al., 2011b).” IsoHe2 was performed with K-feldspar which was considered by Herbert et al. (2014) as multicomponent sample, hence a single J_{het} is again not applicable. I suggest that these datasets are removed from the paper.

Experiments IsoDI1, IsoDI2, IsoDI3, Figure 3: These experiments were performed with illite NX, which is not a pure sample but contains only 60 – 69 % illite (Diehl et al., 2014 and references therein). Moreover, the large temperature range of freezing observed for illite NX suggests that a contact angle distribution has to be used to describe this sample as was done by Hiranuma et al. (2015) and a single J_{het} is not applicable. The authors should discuss how this affects the fitting parameters derived for illite. I suggest that these datasets are removed from the paper.

Spelling error:

Page 13134, line 8: „s“ has to be removed from “particles”.

Figure 3, Figure caption, second line: add “of” between “function” and “time”.