For clarity, below we list the referees comments in italics and our responses in normal font. Changes to the text are highlighted in blue. Page numbering refers to the ACPD manuscript.

General remarks:

We thank referee #2 for their comments and taking the time to review the manuscript. The major concerns listed by the referee are related to the transformations (converting active site mass to surface density) and scalings (scaling surface site density by Feldspar mass) not being discussed thoroughly enough (please see our reply to comment (10) below), as well as the "discussion" and "conclusions" sections being somewhat too speculative. We note that some similar issues were raised by referee #1. In our response to referee #1, we have detailed a number of changes to remove instances in the manuscript which were deemed too speculative by the referee (see replies 2, 8, 10, 11, 13, 14 to referee #1).

Specific comments:

(1) *Page 20277, line 7: The statement "These processes tend to shift the size distribution of hydrometeors in a cloud to larger sizes at lower concentrations." should either be explained in a little more detail or be removed.*

This has been removed.

(2) *Page 20278, line 2 ff: The statement ". . ., but it has been suggested that due to thermophoretic effects, contact nucleation is favourable only in water subsaturated regimes, where cloud droplets disappear rapidly prior to freezing (Philips et al., 2007)." should either be explained in a little more detail or be removed.*

Removed. Now reads "The relative importance of contact mode nucleation remains unclear (e.g. Hoose et al., 2010; Philips et al., 2007)."

(3) *Page 20278, line 6: The term "relative efficiency" needs to be defined.*

Added in brackets after relative efficiency "(as quantifiable, for example, by the determination of nucleation rates or ice active site densities)"

(4) *Page 20280, line 26: The consequences and possible artefacts due to the wet sieving process should be discussed in detail.*

This is related to the referee #1's major comment (5). In addition, we have added a discussion of the potential side effects of this treatment on the samples. At page 20281 line 5 we have inserted: "Possible consequences of the above treatments on the resulting particles sampled include a redistribution of dissolved organic carbon across these particles due to the wet sieving process, as previously highlighted by Conen et al. (2011). In addition, the act of sieving/filtration through a nylon net filter as a method of particle selection could result in changes to the distribution of particle shapes at a given size, as whether elongated particles make it through the square openings is dependent on particle orientation."

(5) *Page 20282, line 3ff: The uncertainties due to particle shape effects should be addressed and estimated.*

Issues due to particle shape effects, and how these can affect the intercomparison of results from different studies, have now been examined at page 20282 line 24:

"It should be noted that in expressing the particle size distribution in volume equivalent sphere diameters can result in uncertainties in that surface area if the particles deviate significantly from a sphere. Particles can be non-spherical, porous and have surface roughness. Typical values of particle sphericity (i.e. the ratio of the surface area of a sphere with the same volume as the given particle, to the actual surface area of the particle) can approach unity for rounded grains or cubic particles such as calcite, while highly irregular particles such as clays or mica can have sphericities as low as 0.2 (Yang, 2003; Holdich, 2002). As noted by Murray et al. (2012), for comparison of ice active site density data between studies the data treatment between the studies needs to be considered (Sect. 2.4)."

Further additional information on the model used in the analysis procedure has also been inserted in response to referee #1 minor comment (11).

(6) *Page 20283, line 19: Assuming that a variation in volume by six orders of magnitude corresponds to change in surface area by around six orders of magnitude is only correct if the change in volume is achieved by varying the number concentration, and assuming the shape of the size distribution to be the same. If I understood it correctly, this is not true for the picolitre droplet freezing experiments. This issue needs to be discussed in more detail and resulting uncertainties need to be addressed.*

Yes, the referee is correct. We have removed the sentence on p20283. We also appreciate that the reader may also benefit from having the issues surrounding the picolitre assay raised earlier, rather than later in the results section (as per comment (11) below). Accordingly, we have added at 20284, line 14:

"In contrast to the nano- and micro-DFEs, in the pico-DFE the size of the droplets examined in the experiment (total range = 10-22 µm) is so small that they do not each contain a representative distribution of dust particles. Accordingly, we do not attempt to quantify the nucleation efficiencies of soil particles normalised to surface area of nucleant per droplet in the case of the pico-DFEs. Instead, the data is presented using the cumulative nucleation spectrum in section 3.2 (nucleation events per unit volume of liquid, see section 2.4) which allows us to establish the influence of heat treatment on a sample's ice nucleating ability."

Then also in section 2.4 we reiterate the issues with the pico-litre experiments:

"As noted in section 2.3, the actual amount of nucleant per droplet in pico-litre experiments is expected to be a probabilistic function of the droplet volume. Accordingly, for the comparison of activities between the heat-treated and non-heat treated soil samples, exactly the same size bins were chosen to analyse both heated and non-heated samples"

(7) *Page 20286, line 18ff: The statement "... is expected to scale directly with surface area ..." is not necessarily correct for biological IN as outlined in Hartmann et al., 2013 and Augustin et al., 2012. They suggest that the number of ice nucleating entities per droplet is the controlling parameter which may not be related to particles surface area. I suggest to cite these publications and weaken the statement.*

Done. Rephrased to: "However, for cases where the surface area of the nucleating particles is readily definable, the number of active sites may be expected to scale directly with surface area (Niemand et al., 2012; Murray et al., 2011; Niedermeier et al., 2010; Augustin et al., 2012; Hartmann et al., 2013)."

(8) *.Page 20286, line 26ff: This description seems somewhat incomplete to me. Why are three bins used ? Why needs the bin centre to be estimated from the median of the droplet sizes enclosed within a bin ? Is the droplet population as polydisperse as suggested by the median been volumes? I think a little more explanation and discussion could be useful here.*

Changes to address these questions have been made below:

Why are three bins used ?, We have added "With three bins we were able maximise the number of droplets per bin (average per bin=17 in nanolitre experiments, 45 in picolitre experiments) while minimising the errors in droplet volume."

Why needs the bin centre to be estimated from the median of the droplet sizes enclosed within a bin ? The distribution is skewed to smaller sizes, so the median is more appropriate than the mean. This now reads: 'The droplet size distribution is typically skewed towards smaller sizes, hence we estimate the bin centre with a median volume rather than a mean'

Is the droplet population as polydisperse as suggested by the median been volumes? The median bin volumes were simply where the centre of the bins were located, and give no information on the widths. We have improved this section in response to referee #1 comment 6 and now explicitly state in the revised manuscript the maximum uncertainties in terms of volume owing to the size binning procedure.

(9) *Page 20287, line 15: Figure 4 could be larger.*

As per referee #1, major comment (15), this has been done.

(10) *Page 20287, line 23: Scaling ns by mass is an easy but maybe incorrect approach. To my understanding it at least implies that the particles are internally mixed and that the size distributions are similar. These assumptions should be discussed and proven if possible.*

The referee raises an important point regarding the assumption that the size distributions of feldspar is similar to that for the dusts which we have also discussed in response to referee #1. The pertinent section now reads:

'For comparison, we also show the parameterisation of n_s values for arid region mineral dusts presented by Niemand et al. (2012) as well as data for K-feldspar using the same experimental procedure as used for the soils. Recently, K-feldspar (microcline) has been shown to be a major ice-nucleating component in mineral dusts (Atkinson et al., 2013). The K-feldspar n_s data in Fig 4 have been scaled down to the feldspar content in each of the soils (≤11 %, see Table 1) for the purpose of comparison. In the absence of a mineral resolved size distribution for our soils we make the approximation that the K-feldspar mass fraction in the soils can be used to estimate the available surface area of this mineral able to nucleate (i.e. wt% K-feldspar = % of total SSA attributable to K-feldspar in the samples). It is important to note that, in this feldspar scaling approximation, it is assumed that the K-feldspar component of the soils has a similar size distribution as that measured for the bulk soils here and also that the feldspar in soils has not been chemically altered in a way which would change its ice nucleating ability. Inspection of Fig.4 shows that despite these assumptions the feldspar scaling approximation is a first order predictor of the ice nucleating ability of these soil samples in this temperature range, predicting n_s within an order of magnitude.'

(11) Page 20288, line 8: The authors state that "In this regime we estimate that most droplets do not contain particles with diameters above 0.4 µm despite these particles making up a significant part of the distribution. Hence, we were unable to determine nm or ns from these freezing data." This is correct, however in my opinion, the whole issue needs to be discussed earlier and in more detail (see above).

We have now discussed this much earlier in the manuscript – see our response to comment 6.

(12) Page 20289, line 14: At this point immediately the question arises, why the influences of the high temperature IN is not visible at lower temperatures, i.e., in the nano and picolitre experiments. A reference to this question being answered further down in the manuscript could be useful, here.

The possible answer to this question is addressed in the conclusions. Rather than reference the conclusions, we have explicitly given our suggested reason for this here "A potential reason for this contrasting behaviour may be the occurrence of rare, but highly active ice nuclei with abundances only detectable in microlitre drop freeze assays."

(13) Page 20291, line 18: Recent investigations by Hartmann et al. (2013) and Augustin et al. (2012) (please follow status of this paper) are newer publications which confirm these statements and were able to observe immersion freezing of droplets due to single macromolecules. It might be useful to cite these publications here as well.

Included these references here.

(14) Page 20294, line 19ff: I personally don't consider this paragraph important and suggest to remove it from the manuscript.

This has been removed, in line with response to referee 1, major comment (14).

References

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Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., and Skrotzki, J.: A particle-surface-area-based parameterization of immersion freezing on desert dust particles, J. Atmos. Sci., 69, 3077-3092, Doi 10.1175/Jas-D-11-0249.1, 2012.

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