

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2022-61

Anonymous Referee #2

Referee comment on "Micro-spectroscopic and freezing characterization of ice-nucleating particles collected in the marine boundary layer in the eastern North Atlantic" by Daniel A. Knopf et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-61-RC2>, 2022

I support publication. The manuscript is well written and the data is well presented.

There is one point which I think should be addressed, but overall, the manuscript more than meets the standards for publication in ACP.

The surface area of the particles that are examined in this study is a critical part of the analysis, and there are some subtleties to this that need to be explained more carefully.

If I am reading the manuscript correctly, all of the particles are from stage 6 of a MOUDI, which has a cut size of 0.56 micrometers. I realize of course that the cut size isn't a step function. But that cut size and the size of the particles listed in Table 3 are not consistent. For example, the last particle listed (Night 2 #6) has an area equivalent diameter of 11.96 microns. The MOUDI's largest cut size is 10 microns, if I'm using the sizes for the right model here. I could certainly understand a particle that large being on the first stage. But the sixth? This is only the most egregious example. Most of the other particles have sizes larger than the cut size that's listed in the manuscript.

This discrepancy or uncertainty can also be seen in the figures, for example Fig. 3. The size distributions are shown as a function of area equivalent diameter. In that figure, the axes extend only to 2 microns or so, but the distributions are all showing particles larger than the MOUDI cut size. (See for example, Day 1, STXM mixing state... the mode of the distribution is at about 1 micron.) These discrepancies are not as large, but they are still puzzling.

I realize that the MOUDI cut size and the area equivalent diameter derived from SEM

measurements are not the same thing. But the authors do make an explicit point that these particles come from stage 6 of the MOUDI, and the cut size is specified. If you then go on to discuss particles much larger than the cut size, at least comment on it and perhaps provide some rationale for it.

The larger issue is the derivation of the surface area of the particles. (This issue is highlighted repeatedly in Knopf et al, 2020, which is cited in the manuscript.) Deriving the surface area of the particles from the SEM images (the area equivalent diameter) is problematic. Is the assumption that the particles are spherical, then using that to get a surface area once a diameter is derived? This is never stated.

And if that is, in fact, what is assumed, it almost certainly wrong. The SEM only "sees" the top of the particle. You can't access the third dimension. There's been work in recent years showing that dust, for example, is rarely spherical. See Huang et al, 2020 for example. I know that the INPs detailed here are not dust, but the point is still valid. None of the particles in panel A of Fig. 5, for example, look spherical to me.

I am not asking the authors to resolve these issues. I am asking that they more explicitly outline how the particle surface area is derived and discuss (at least) some of the uncertainties that may arise from that method. (I think quantifying the uncertainty may be beyond what's possible for this study.)

Minor points

Table 2, heading to column 6: "or" should "of"?

Figure 4: the blue open circle at ~ 231 K... The open symbol indicates that this is immersion freezing, but it is well below the water saturation line. Is this a particle that deliquesced, then froze from solution?

Reference:

Huang, Y., Kok, J.F., Kandler, K., Lindqvist, H., Nousiainen, T., Sakai, T., Adebisi, A. and Jokinen, O., 2020. Climate models and remote sensing retrievals neglect substantial desert dust asphericity. *Geophysical Research Letters*, 47(6), p.e2019GL086592.