

Review of “Ice-nucleating efficiency of aerosol particles and possible sources at three coastal marine sites”

Si et al. (2018) investigate sources of ice nucleating particles (INPs) from three coastal sites with a combined measurement-modeling approach. Measurements were taken with a suite of well-established instrumentation, that allowed quantification of INPs by an active site density function n_s . The results were compared with the output of a global INP model, and it was found that the two INP model of K-feldspar and marine organics missed a high temperature INP source. Speculation as to what this source is was carried out reasonably. The paper is well written and the figures are clear. I think this study merits publication in ACP after some minor concerns are addressed.

The first pertains to the global INP model. I would appreciate the inclusion of a more critical account of the limitations of the model when being compared to ground based measurements. There is a big jump in the conclusion that there is a missing source of INP. For example, how can the authors be sure the measurements aren't artificially inflating the INP activity at higher temperatures by sampling from the ground? The global INP model is supposed to shed light on what, statistically and on long/large enough scales, INPs matter. The measurements on the other hand are happening locally from boundary layer air. Please investigate this point further.

Other comments are specific to the text and are outlined below.

P2 L1-2: The studies cited do not conclude that INPs “significantly impact the frequencies, lifetime, and optical properties of ice and mixed-phase clouds”. Consider changing to something less assertive like “may impact”.

P9 L5: n_s as a function of size is a useful approach here. However, there are issues with surface area corrections that make n_s not without shortcomings. Studies by Beydoun et al. (2016), Emersic et al. (2015), and Hiranuma et al. (2015) discuss these shortcomings and should be included in an additional discussion on what kind of limitations the authors expect when analyzing n_s against surface area.

P11 L5: The authors can do a better job here of synthesizing their results and suggesting a way forward. For example, on the measurement side, samples can be investigated with a chemical composition analysis. On the modelling side, large eddy simulations can discern whether boundary layer INP are different than free atmospheric INPs simulated by the global model. So I think there's a bit more room here for discussing future efforts.

Technical correction:

n_s is a surface area density, not an efficiency. It has units of m^{-2} and does not range from 0 to 1 (like an efficiency would). You may also want to consider changing that in the title as well. Please refer to Vali et al. (2014) to ensure INP specific terminology is consistent.

References

- Beydoun, H., Polen, M., & Sullivan, R. C. (2016). Effect of particle surface area on ice active site densities retrieved from droplet freezing spectra. *Atmospheric Chemistry and Physics*, *16*(20), 13359–13378. <https://doi.org/10.5194/acp-16-13359-2016>
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- Hiranuma, N., Augustin-Bauditz, S., Bingemer, H., Budke, C., Curtius, J., Danielczok, A., et al. (2015). A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques. *Atmospheric Chemistry and Physics*, *15*(5), 2489–2518. <https://doi.org/10.5194/acp-15-2489-2015>
- Vali, G., DeMott, P., Möhler, O., & Whale, T. F. (2014). Ice nucleation terminology. *Atmospheric Chemistry and Physics Discussions*, *14*, 22155–22162. <https://doi.org/10.5194/acpd-14-22155-2014>