

## Summary

The authors generally did a good job of responding to my comments. However, I still see a few issues with the manuscripts that need to be resolved before this manuscript is published.

I still think the writing and logical flow of the introduction in particular needs to be improved. I have made suggestions where I could.

The authors have done a better job of linking IN activity and  $\chi$  (or rather not linking it, as it does not seem informative the way that it can be for CCN activity). However, I think that they could more explicitly state the reasons for the lack of connection. I think that it is probably due to the fact that IN activity is very sensitive to particle surface composition and morphology, and while  $\chi$  is influenced by surface composition, it is not wholly sensitive to it.

## Specific comments

Line 50: This sentence is duplicative of the one before, and should be removed.

Line 61: I think that the authors should note that they focus primarily on deposition, and to some degree immersion freezing in this study.

Line 85: This sentence overlaps closely with the one at 97, and they should probably be removed.

Line 89: This sentence overlaps with the one before it, and should probably be removed.

Line 93: I think that the authors need to highlight the necessity of using single particle techniques for quantifying mixing state index.

Line 97: I recommend moving this sentence before the previous one, to improve the logical flow.

Line 100: Doesn't this sentence directly contradict the one at Line 93?

Line 103: This paragraph on the identity of marine INPs is extremely disjointed. The authors list a number of different studies, but don't group them together in any way. For instance, a number of these studies looked at INP composition and determined that they were mostly derived from organics. Others looked at their IN activity relative to other particle types. Currently, this paragraph reads as a list of studies in no particular order, and I think the authors could improve this introductory material by grouping them in a coherent manner.

Line 129: This marks a transition in the overall topic, and I think should be split into its own paragraph.

Line 184: I just want to note that I was surprised by how short the sampling time for the first several samples was. Was there a reason for why these were so short?

Line 230: Fig. S2 is missing. This also means that the following supplemental figure references appear to be incorrect as well.

Line 381: Is the particle Dust or Dust-like? What is the distinction?

Line 419: Can you overlay the trajectories for S8 over Fig. S4C? It is difficult to assess the statement in this sentence with the current plot.

Line 450: This sentence and the next one can be combined.

Line 452: Is the study of Kunwar tied to a particular time of year?

Line 535: This is obviously true, and I am not sure it needs to be stated.

Line 550: I think that the section from this sentence to the end of the paragraph is quite obvious, and quite repetitive. I think the authors should shorten this sentence to improve readability.

Line 552: This sentence and the next one basically say the same thing. I would recommend to combine or remove one.

Line 591 and 593: The descriptions for *ii* and *iv* should both be condensed into one sentence.

Line 682: Fig. S15 shows the active site density for these samples, not the average elemental composition of INPs and non-INPs.

Line 924: I think the authors need to acknowledge that NaCl is not best proxy for sea spray due to the fact that it contains organics and may affect their interpretation of the results.

Line 936: Authors need to explicitly link the differences between the physicochemical properties that we know affect ice nucleation activity and what  $\chi$  actually measures.

Line 998:  $n_s$  has only been discussed in the supplemental, and thus I don't think it should be mentioned here in the conclusion.

Figure 10: The way this figure is visualized makes it difficult to tell how large the error bars are for the different particle types. Would it be possible to offset the markers and lines every so slightly, so that these lines can be observed?

Figure 14: Why do the authors include S1 and S10, and S14 into the analysis, which we know to not be sea spray influenced, when their reference particle is NaCl? Why not just limit the analysis to the SS samples?

Where is the reference for Fig. S14?

In the SI, the authors refer to the Ross Sea as the Rose Sea.

### **Technical comments**

Table S1: Should this go down at the bottom of the document? In other words, are supplemental tables supposed to be grouped together?

Line 49: "contribute" should be "contributing".

Line 82: "Due to the" should be "Due to their".