Summary

This manuscript ("Diverse mixing state and ice nucleation properties of aerosol particles over the Western Pacific and the Southern Ocean" by Xue et al.) investigates the identities of ice nucleating particles in marine environments from the Western Pacific to the Southern Ocean. They used CCSEM/EDX to characterize ambient particle composition and population mixing state. They used an ice nucleation cell to identify ice nucleating particles (INPs) at cirrus conditions, which were subsequently characterized using SEM/EDX. They also used the CCSEM/EDX results to characterize particle mixing state at the individual and bulk levels. They observed that sea salt particles were the most prevalent particle type, except for samples directly influenced by biomass burning and dust plumes. They also observed that fresh sea salt with organic coatings were the most efficient ice nucleators, while biomass burning particles were the least efficient. Mixing state depended strongly on the amount of sea salt measured.

Overall, I think this study is interesting and would eventually recommend publication in *ACP*. Detailed and direct chemical characterization of INPs is informative, especially in a relatively understudied environment like the marine boundary layer (and over such a varied set of sampling environments within the MBL). I thought they did a nice job of source apportionment with their measurements. I also appreciated their quantification of the particle mixing state. However, I believe that there are a few key issues with the manuscripts that need to be resolved before this manuscript is published.

First of all, the writing needs to be tightened considerably. There are many instances of poor grammar or confusingly worded sentences. I have noted some of these below, but I don't think that this list is comprehensive.

Second, I thought that the exploration of INPs and particle mixing state were both interesting, but I found that there was little link between these two aspects of the study. I presume that the authors were interested in quantifying particle population mixing state to derive some insights about the identities of INPs and/or the effect of mixing state had on INP propensity, but that was not clear throughout the manuscript. I would recommend that they try to strengthen the connections between these two aspects where possible.

Third, I am curious as to whether the authors compared between the intra-class particle composition for INPs and ambient particles. They have a relatively high number of INPs in their dataset, and it would be useful to determine whether INPs have particular physicochemical properties that distinguish them from non-active particles.

Fourth, I am not convinced by the kinetic analysis presented here. For example, the n_s shown in Fig. 12 increases for a given experimental temperature, as that temperature is dropped. However, the ns for each experimental temperature resets to roughly 10^2 cm^{-2}. This would suggest that what they are measuring is an experimental property and not reflective of the underlying particle properties.

Specific comments

Line 31: Meaning of sentence is unclear and should be re-written.

Line 33: Should I really be surprised tha the identified INPs are all major particle classes? Needs to be clarified, and either re-written or omitted.

Line 73: This sentence is confusing and needs to be re-written.

Line 81: Do not need this sentence.

Line 85: This sentence is duplicative of the one at line 73. Either remove, condense, or rearrange.

Line 90: This paragraph is confusing and should probably re-written. For instance, the authors discuss the importance of quantifying chemical mixing state, and then introduce it later. I think the logical flow would be to introduce it first and then discuss why it is important.

Line 117: I would note that this is only for deposition nucleation, and not immersion freezing where there is a roughly 100-1000x difference (McCluskey et al. 2018, DeMott et al. 2016).

Line 158: Similar phenomena was observed in Cornwell et al. (2019).

Line 170: I think it would be useful to have a brief description of this method.

Line 252 and 261: μ_i^a is defined by multiplying the molar fraction of *a* by μ_i, while μ_i is defined by summing the individual μ_i^a. This seems circular to me. From the text at line 243 I thought the authors were defining μ_i by the particle density and volume, but these equations (1) and (2) say differently. Please clarify.

Line 279: This sentence is confusing and needs to be re-written.

Lines 284 and 288: Write out the equation for D_i D_α, and D_γ explicitly.

Line 311: I thought that the mean wind was prescribed from the external domain for all simulations. Why does is the dispersion for the no-fire case seemingly isotropic?

Figure 4: What is the difference between Fig. 4 and Fig. 5a? It seems to me that they are providing the same information. If it is about the data presentation of Fig. 4, then I would recommend moving to the supplemental and providing a note somewhere in the text about it.

Line 395: From what to what are SS/Sulf and CNOS particles increasing? Over S8 to S10, or compared to the WP-1 region?

Line 404: This sentence is confusing and should be re-written.

Line 432: This sentence is mostly duplicative of the one prior to it. I would recommend removing or consolidating.

Line 449: This sentence is confusing and should be re-written.

Line 491: This sentence is confusing and should be re-written.

Line 516: I find much of this analysis to be self-evident and the end-goal of the analysis unclear. As one particle type becomes dominant, then by definition the χ will approach 100%. Similarly, the χ will decrease when there are more particle types in the population. I think this paragraph should be streamlined and it made more apparent to the reader why they should care about these results.

Line 539: The logic in this sentence seems circular to me. The contribution of these aged particle types can be treated as an indicator of aging. Re-write to clarify meaning. Also, I would say that aging would do more than potentially affect mixing state.

Line 583: How did the authors distinguish between immersion and homogeneous freezing? Whether it was below the homogeneous freezing limits?

Line 623: Would it be possible to quantitate the difference between the organic coating thickness between INPs and ambient BBA?

Line 625: It is confusing what the comparison is between. SS INPs from S11 to S4 and S12?

Line 643: This sentence is confusing and needs to be re-written.

Line 660: This sentence is duplicative to the one after it and should be removed.

Line 725, caption for Fig 11: Authors included sample numbers for each sample but didn't in the caption for Fig. 7. Would recommend changing captions to be consistent.

Line 756: Section title is generic and not very descriptive. Would recommend changing to something more descriptive, such as "Ice nucleation kinetics".

Fig 12: Are the samples the same ones presented in Figs. 7 and 11, or are these the particle classes presented in Fig. S10?

Line 812: The units for J_{het} are given as $cm^{-2}\ s^{-1}$, but listed in this sentence as cm^{-2}.

Line 903: This sentence is confusing and should be re-written.

Lines 923-927: This example does not clarify the authors first stipulation.

Line 928: Where was this shown? Some other paper?

Line 926: This sentence is confusing and needs to be re-written.

Lines 958-974: This paragraph needs to be more quantitative and specific with results.

Fig. S4 has (G) panel twice.

Fig. S10: The color border is difficult to easily distinguish given the close match between some of the particle types. I would recommend labeling each image with the particle type instead.

Technical comments

Line 21: as a question of journal style, is µm supposed to be italicized?

Line 29: "Dominate" should be "dominant".

Line 41: "Ice nucleating particles" should be "ice nucleating particles".

Line 52: Should "intergovernmental panel on climate change" be capitalized?

Line 71: "and have minimum **influence from** anthropogenic activities".

Line 117: "**Wagner** et al."

Line 122: "identified" and "found" are describing the same thing twice, recommend simplifying.

Line 137: "Residuals" is the preferred term.

Line 143: "Microscopic based" should either be "microscope-based" or "microscopic".

Line 161 and other places in the manuscript: "Rose sea" should be "Ross sea" (presumably).

Line 175 and many places throughout the manuscript: What are the journal conventions for italicizing variables with sub/superscript? I thought that the sub/superscript was supposed to be upright. In other words, "A_p" should actually be written as "A_p".

Line 176: I would recommend removing "Along with" from this sentence to improve readability.

Line 179: The "A" in "Ap" should be italicized.

Line 200: "for each sample depending **on** the particle loading".

Line 228: "The classification scheme ~~was~~ for INPs and non-INPs on the silicon wafer chips".

Line 289: "bulk population elemental diversity (D_γ) **are** calculated by taking".

Line 304: "the particle temperature **(T_p)** and relative".

Line 307: “and an optical **microscope**”.

Line 328: “conservatively” should be “conservative”.

Line 402: “The **average** BC concentration”, and “likely **originated** from combustion emissions transported from land”.

Line 411: “These two samples were collected **on** November 13th and 14th”.

Line 426: “the contributions of AgedSS,SS/Sulf, and CNOS type particles ~~to the total~~ from the middle of **the Ross** Sea”.

Line 454: “on pre-exi**s**ting particles”.

Line 465: “elemental composition and referred to **as** chemical mixing state”.

Line 540: “which potentially **a**ffects the mixing”.

Line 821: “Stati**sti**cal uncertainty”.

Line 907: “will reduce the mixing state index **as** the population becomes more **externally mixed**.”

References

DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., … Franc, G. D. (2016). Sea spray aerosol as a unique source of ice nucleating particles. *Proceedings of the National Academy of Sciences*, *113*(21), 5797–5803. https://doi.org/10.1073/pnas.1514034112

McCluskey, C. S., Ovadnevaite, J., Rinaldi, M., Atkinson, J., Belosi, F., Ceburnis, D., Marullo, S., Hill, T. C. J., Lohmann, U., Kanji, Z. A., O’Dowd, C., Kreidenweis, S. M., & DeMott, P. J. (2018). Marine and Terrestrial Organic Ice-Nucleating Particles in Pristine Marine to Continentally Influenced Northeast Atlantic Air Masses. *Journal of Geophysical Research: Atmospheres*, *123*(11), 6196–6212. https://doi.org/10.1029/2017JD028033