Dear editor and referees,

Please find below the responses and revision of our manuscript entitled “Diverse mixing state and ice nucleation properties of aerosol particles over the Western Pacific and the Southern Ocean” by Xue et al.

We thank the referees for their insightful comments and for providing suggestions to improve our manuscript. Considering the reviewers’ suggestions, two major changes have been made: (1) adding a discussion on the connection between mixing state and ice nucleation propensity; (2) simplifying the discussion on the ice nucleation kinetics. We provide point-by-point responses to the comments made by the referees. The corresponding changes are marked in the revised manuscript. We hope that these changes are satisfactory and that the revised manuscript is acceptable.

Thank you for your consideration.

On behalf of all co-authors,
Bingbing Wang

In the following descriptions of the major changes and the point-by-point responses, line numbers in the responses (in light blue fonts) refer to the revised manuscript and those referring to the original manuscript are specified. The line numbers in the referees’ comments (in black fonts) refer to the original manuscript.

Two major changes:
(1) We conducted additional experiments on NaCl particles and added a discussion section on the connection between mixing state and ice nucleation propensity. As suggested by the reviews, we made the following changes. We added a new Section 3.7, L911 and Figure 14 and Figure S20.
“**3.7 Ice nucleation propensity and mixing state of particle population**

Previous studies have used χ to estimate the error in predicting aerosol CCN activity and optical properties when assuming idealized mixing states (Ching et al., 2017; Yao et al., 2022). For example, Ching et al. (2017) quantified the error in predicting CCN concentration as a function of χ. When internal mixing was assumed, errors of up to 100% overpredicting CCN concentration were found for populations with more external mixing (χ < 60%). However, CCN concentrations were well predicted for populations with χ > 75%. Inspired by these studies, we used θ as a measure of particle ice nucleation propensity to discuss the potential link between the mixed state of population and particle ice nucleation propensity. We investigated the relative difference (error) in θ for the field samples compared to pure NaCl particles. Ice nucleation experiments were conducted on the laboratory-generated NaCl particles (Text S3) and θ was calculated for NaCl particles (see Section 3.6.2 for the calculation details). Pure NaCl particles were used as a surrogate of complete internally mixed marine aerosols (χ = 100%). We analyzed the relationship between the relative error in θ of INPs (*Err*) and the mixing state of the population in which the

INPs were identified. *Err* of each INP from a field sample with a given mixing state (χ) was defined as:

$$Err = \frac{\theta(\chi) - \theta(100\%)}{\theta(100\%)} \times 100 \quad (15)$$

where $\theta(100\%)$ is the average θ for the NaCl standard. As shown in Fig. 14, χ for the six investigated samples changed from 61% to 86%. Five samples (S1, S4, S10, S12, and S14) exhibited positive mean *Err* ranging from 2.3% ± 7.5% to 16.3% ± 6.6%, while S11 had a negative mean *Err* of -18.6% ± 6.2%. The FreshSS and AgedSS dominated samples, S4 and S12, have a mean *Err* of 6.9% ± 12.4% and 13.0% ± 9.2%, respectively.

The θ for particles in the marine environment is different from that for NaCl particles. This can lead to errors in J_{het} of several orders of magnitude if the model assumes an internally mixed particle population. According to CNT, we calculated DIN J_{het} for NaCl and particles with *Err* of 5% and 10% under different temperature and humidity conditions (Fig. S20). Between 135% and 180% RH_{ice}, *Err* of 5% and 10% will result in 1.5 – 4 and 2 – 10 orders of magnitude differences in J_{het}, respectively. Moreover, the lower the temperature and RH_{ice}, the larger the error in J_{het}. This suggests that the effects of aging processes and mixing state need to be considered in the ice nucleation modeling. No specific correlation or trend was found between χ and the *Err* from this data set with the limited number of samples. This may be expected since ice nucleation is controlled by various physiochemical properties of individual particles, particularly for DIN. For example, one crucial factor is the physical mixing state of particle, such as coating. Organic coatings play a significant role in altering the θ of sea salt particles, as exemplified by S11 compared to S4 and S12. In contrast to CCN, which in general can be described by the κ-Köhler theory, there are still no appropriate physical models to describe INPs. In addition, ice nucleation propensity for DIN is determined on the individual particle basis, whereas the chemical mixing state (i.e., χ here) characterizes the overall mixing of species within the particle population. There is a large gap in the understanding of the relationship between INP propensity and χ."

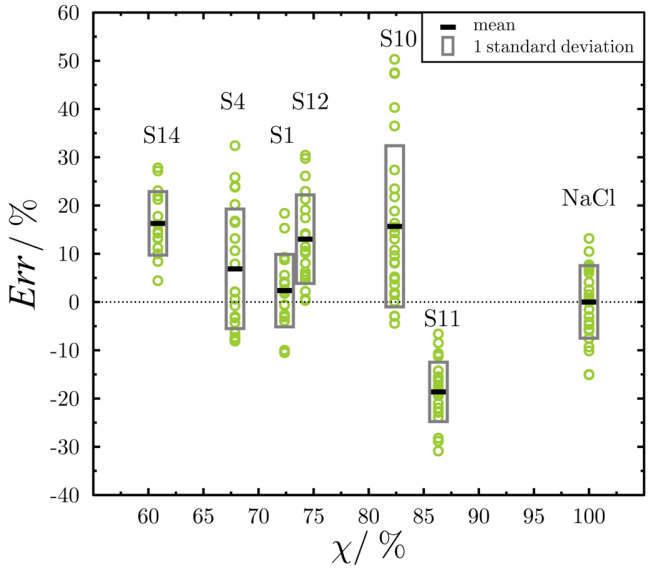

Figure 14. *Err* in θ for the investigated samples relative to the internally mixed particles (pure NaCl particles, χ = 100%). Black thick lines indicate the average θ and the gray boxes represent one standard deviation. Green circles represent θ for individual INPs.

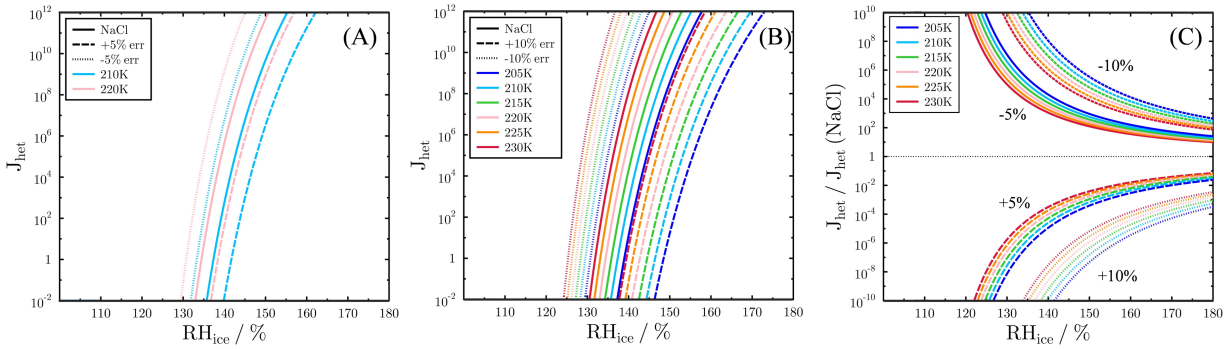

Figure S20. The DIN J_{het} calculated based on CNT for NaCl, *Err* of ±5% (A) and *Err* of ±10% (B), and their ratio to J_{het} of NaCl (C) at different temperature and humidity conditions.

(2) We shorten Section 3.6 on ice nucleation kinetics. we decided to focus on the discussions on J_{het}, contact angle, and the corresponding parameterizations in the main text. Some of the discussions, parameterizations, and figures were moved to the supplemental information. The changes are listed below:

- Section 3.6.1 "Ice nucleation activated fraction", Section 3.6.2 "Ice nucleation active site density", and Figure 12 in the original manuscript were moved to Text S2 and as Fig. S17.
- Section 3.6.3 is now Section 3.6.1, "Heterogeneous ice nucleation rate coefficient": L816-830 (uncertainty calculation for J_{het}), L883-891 and Figure 15 (parameterization of contact angle) in the original manuscript were moved to Text S2 and as Fig. S19.
- L796 is revised: "Classical nucleation theory (CNT) has been widely used in the cloud models (e.g., Pruppacher and Klett, 2010; Khvorostyanov and Curry, 2004; Liu and Penner, 2005). In this study, heterogeneous ice nucleation rate coefficient (J_{het}), contact angle (θ),

and their parameterizations are presented and discussed. Ice nucleation activated fraction of each sample was listed in Table 1 and discussed in Text S2. n_s based on the singular hypothesis (Vali, 1971; Connolly et al., 2009) and its parameterizations are also presented in Text S2."

Point-by-point responses:

Review #1

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.

Thank you, we checked the grammar and improved the writing throughout the text.

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.

Please see the major change #1 listed at the beginning of this document.

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.

We conducted two-sample t-test comparing the particle composition for INPs and non-INPs. There is no sufficient evidence showing that the INPs and non-INPs are different based on the elemental composition. We added the following text in Section 3.4, L682 and Fig. S15: "We calculated the average elemental composition of INPs and non-INPs for each particle type as shown in Fig. S15. A two-sample t-test was also performed on the elemental compositions of INPs and non-INPs for each particle type. At the significant level of 0.05, there is no sufficient evidence showing that the elemental compositions of INPs and non-INPs are different."

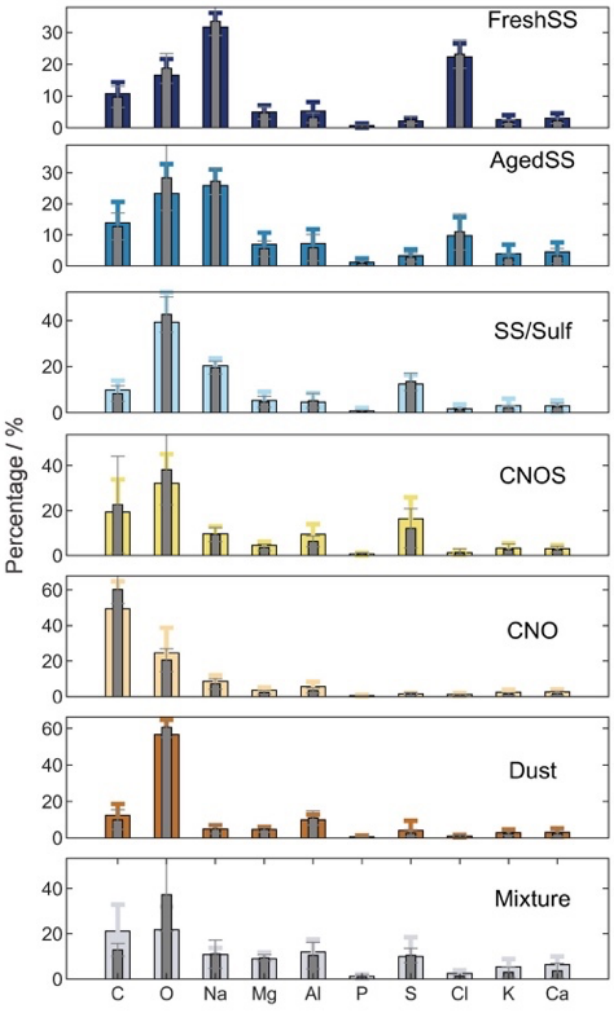

Figure S15. Elemental composition of INPs and non-INPs for each particle type (mean with one standard deviation). Narrow gray bars and wider color bars represent INPs and non-INPs, respectively.

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.

For the kinetic analysis presented here, we believe that n_s and J_{het} reflect both the experimental property and the underlying particle properties for the following reasons: (1) One of the important factors affecting the values of n_s and J_{het} is the particle surface area. As shown in Table 1, the particle surface areas for the investigated samples are similar (within a factor of 5) except for WP-I-S4. This is likely the main reason why the samples have similar n_s. (2) We used the same cooling rate for all ice nucleation experiments. The experimental cooling rate can also affect the nucleation kinetics. J_{het} reflects the nucleation time, but n_s does not. This also leads to similar n_s. (3) In this study, we focus mainly on the deposition ice nucleation and report only the first ice nucleation event(s) for each experiment. Ice nucleation activated fraction at the onset conditions likely is lower (less than 0.02%) compared to other ice nucleation measurement techniques that typically report temperature and RH conditions at higher activated fraction, e.g., 1%. The n_s reported here for deposition ice nucleation at the onset conditions does not extend over several orders of magnitude over a wide temperature range.

Following the suggestion of another reviewer and to keep the main text concise, we moved the discussion of n_s and Fig. 12 from the original manuscript to the SI. Please see the major change #2 listed at the beginning of this document.

Specific comments

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

We revised the sentence in L28 to emphasize new findings as follows: "Multiphase processes on sea salt particles resulted in chlorine deficiency. This selective aging process made the marine particle population more externally mixed."

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

We omitted this sentence.

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

We changed the sentence in L68 as follows: "Other sources may also contribute to marine aerosols, such as ship emission (Ault et al., 2009, 2010) and long-range transport of aerosols from industrial emission, biomass burning, dust storms, and fossil fuel combustion (Han et al., 2006; Fu et al., 2013; Geng et al., 2019)."

Line 81: Do not need this sentence.

We combined this sentence with the one at L77 of the original manuscript and revised in L71-L74 as follows: "In addition, atmospheric oxidation of volatile organic compounds from the ocean or anthropogenic activities results in gas-particle conversion, multiphase reactions, and the formation of secondary materials on particles (Cochran et al., 2017)."

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

We removed the last two sentences in L85 of the original manuscript.

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 wouldbe to introduce it first and then discuss why it is important.

We rearranged the sentences in this paragraph as reviewer suggested.

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).

Yes, we changed the sentence in L109-L113 to: "Wagner et al. (2018) showed that SSA and desert dust particles share the same order of magnitude of ice nucleation active surface site density (n_s) for deposition nucleation. Previous studies have also shown that n_s for SSA is 2-3 orders of magnitude lower than that for dust particles in immersion freezing (DeMott et al., 2016; McCluskey et al., 2018a; Cornwell et al., 2019).".

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

See the response to the previous comment above.

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

We added the following text in L174 to briefly describe the method: "When the relative wind direction against the ship heading is between 110◦ and 260◦ and the relative wind speed is below 2 m s^{-1}, samples may have been influenced by the ship exhaust (Park et al., 2020). Potentially contaminated samples were excluded from the analysis, resulting in a total of 29 selected samples."

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.

For clarity and to keep it concise, we deleted the original Eq.2 and move the definition of μ_i in L261 after Eq.1.

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

We revised the sentences in L288 as follows: "H_i and H_γ are used to describe the mass distribution of species (elements) with the i^{th} particle or particle population, respectively. A higher entropy indicates a more uniform distribution of elements in the individual particle or

particle population whereas a lower entropy towards a non-uniform mass distribution."

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

We added the following equation in L301-303:

$$D_i = e^{H_i} = \prod_{a=1}^{A} (p_i^a)^{-p_i^a} \quad (10)$$

$$D_\alpha = e^{H_\alpha} = \prod_{i=1}^{N} (D_i)^{p_i} \quad (11)$$

$$D_\gamma = e^{H_\gamma} = \prod_{a=1}^{A} (p^a)^{-p^a} \quad (12)$$

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?

We couldn't relate this comment to L311. We think the reviewer is likely referring to L411. The backward air trajectories were calculated using GDAS (1 degree, global) meteorological data. The backward trajectories for S13 to S16 arriving at 100m from Southwest look similar. However, the trajectories at a lower level (the sampling height of 13 above sea level) might be somehow different. In addition, the in-situ BC measurement showed that the non-fire cases had relative low BC concentrations of ~161.5 ng/m^3 for S13 and 31.1 ng/m^3 for S16 comparing to 674.7 ng/m^3 for S14 and 356.8 ng/m^3 for S15. Thus, we think S13 and S16 might have minor influences from the fire emission.

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.

Figure 4 and Fig. 5a show the same information. We removed the Fig. 5a and remove or revised the related text. We deleted L442-443 in the original manuscript.

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

We revised the sentence in L412-414 as follows: "S8 – S10 were dominated by AgedSS, SS/Sulf, and CNOS particles. The percentage of SS/Sulf and CNOS particles increased from 18% to 36% and from 17% to 39%, respectively, from S8 to S10."

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

We revised the sentence in L419-422 as follows: "As shown in Fig. S4C, this is further supported

by the air mass for S8 passed through the region where fire spots were detected in NASA Fire Information for Resource Management System (FIRMS)."

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

We deleted the sentence.

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

We revised the sentence in L466 as follows: "The sea salt containing particles including FreshSS, AgedSS, and SS/Sulf classes had higher percentages in the super-micron size range. The majority of CNOS particles were in the submicron size range."

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

We revised the sentence as follows in L507: "χ first decreased from 72% to 50% (S1 to S3) and then increased to 68% (S4) as the dominant particle type switched from Dust to sea salt containing particles."

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.

Yes, reviewer is right that as one particle type dominates, χ will reach 100%. Also, additional particle types make the population more externally mixed, thus decrease in χ. This is what we found and is conceptually consistent with the scheme proposed by Riemer et al.(2019a). In this study, we also found that as the fraction of the aged particles (AgedSS and SS/Sulf particles) increased, χ decreased. This is contrast to the conception that aging makes particle population towards internally mixed, i.e., higher χ (Riemer et al., 2019a). As suggested by other reviewers, we redo the data analysis and emphasize this new finding. Please also see the response to the reviewer #3 (Specific comments No.20) on Figure 6.

We revised the paragraph to make the purpose of this analysis clear. The paragraph in L535 now starts with "The changes in the particle composition may affect the mixing state of the particle population. To focus on the effect of particle aging on the mixing state of marine aerosols, we excluded four samples from two events from the analysis, i.e., the Dust-dominated samples from a dust storm (S1 and S2) and the BBA-dominated samples from wildfire emissions (S14 and S15). Figure 6 shows the relationship between χ and the percentages of FreshSS, AgedSS and SS/Sulf, and CNOS particles in different size ranges."

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 domore than potentially affect mixing state.

We revised the sentence in L569 as follows: "Atmospheric aging processes can change the composition and mixing state of aerosol population. In marine environments, the degree of aging on sea salt particles can be reflected by the contribution of AgedSS and SS/Sulf particles in the population."

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

In this study, we won't be able to visualize and distinguish the immersion and homogenous freezing for our samples with small particle/droplet size due to the limited spatial resolution and imaging speed of the optical microscopy. We assume immersion freezing occurred if RH_{ice} onset of freezing is lower than the homogeneous freezing limits.

We added this information in L337 as follows: "The freezing mechanism within the small droplets after the water uptake could not be visualized due to the limited spatial resolution and imaging speed of the OM used in this study. Thus, we assume that immersion freezing has occurred if the RH_{ice} onset of freezing is lower than the homogeneous freezing limits."

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

We calculated the coating thickness and did the t-test for the difference between INPs and non-INPs in BBA. We add the following text in L657: "The organic coating thickness of BBA INPs (1.07 ± 0.68 µm, n = 13) is significantly thicker than that of other BBA (0.53 ± 0.37 µm, n = 112) at the significant level of 0.001."

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

We revised this sentence in L659 as follows: "Different number fractions of INPs identified in the FreshSS and AgedSS dominated samples (S4, S11, and S12) have organic coatings. More than 80% of INPs in S11 were coated with organics compared to S4 (33%) and S12 (30%)."

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

We revised this sentence in L677 as follows: "Taking S1 as an example, six particle classes were identified in the particle population and the 19 identified INPs are from five particle classes except for the CNO class."

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

We deleted one of sentences which are duplicative.

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.

We made the changes.

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

Thank you for the suggestion. We changed it to "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?

Yes, the samples presented in Fig. 12 of original manuscript are the same ones presented in Fig. 7 and 11 of original manuscript; and the INP images and particle class information are presented in Fig. S10, S11, and S12 of original manuscript. As suggested by another reviewer, we shorten the Section 3.6 and moved the Fig.12 of original manuscript to the supplemental information (Fig.S17).

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

We made the corrections.

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

We deleted this sentence since it is duplicative to the one before it.

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.
To make the manuscript more focus and concise, for the three comments listed above, we decided to delete this discussion and the example from Lines 920-930 of the original manuscript.

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

We added or revised the following sentences:
in L1015, "Except for the samples impacted by dust storm, fresh sea salt, aged sea salt, and sea salt mixed with sulfate particles were the most prevalent particle types with the total number percentage ranging from 48% to 99%.";

in L1019: “Significant contributions of sulfur-containing particles up to 93% in number were found in the Ross Sea, which was affected by phytoplankton blooms in the austral summer.”;
in L1021: “The chemical mixing state index of particle population ranges from 50% to 95%.”.

Fig. S4 has (G) panel twice.

We corrected this typo.

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.

We changed the figure as suggested.

Technical comments

Line 21: as a question of journal style, is µm supposed to be italicized?
When it is used in a unit, it should be roman.

Line 29: “Dominate” should be “dominant”.
We moved this sentence to shorten the abstract and to emphasize new findings.

Line 41: “Ice nucleating particles” should be “ice nucleating particles”.
Corrected.

Line 52: Should “intergovernmental panel on climate change” be capitalized?
Corrected.

Line 71: “and have minimum **influence from** anthropogenic activities”.
Corrected.

Line 117: “**Wagner** et al.”
Corrected.

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

Line 137: “Residuals” is the preferred term.
Corrected.

Line 143: “Microscopic based” should either be “microscope-based” or “microscopic”.
We changed to “microscope-based”.

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

Corrected.

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, "Ap" should actually be written as "Ap".
Yes, the referee is right. The overall rule is that symbols representing physical quantities (or variables) are italic, but symbols representing units, or labels, are roman. When sub/superscript is a variable index, it should be italic. For example, the total mass of the i^{th} particle would be denoted μ_i, where the i is italic. We went through the manuscript and made corrections.

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

Line 179: The "A" in "Ap" should be italicized.
We didn't use "Ap" in Line 179. We went through the manuscript and made corrections regarding the correct format for the variables. Please see the response to the similar comment above.

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

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

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

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

Line 307: "and an optical **microscope**".
Corrected.

Line 328: "conservatively" should be "conservative".
Corrected.

Line 402: "The **average** BC concentration", and "likely **originated** from combustion emissions transportedfrom land".
Corrected.

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

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

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

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

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

The sentence has been rewritten in L569 as follows: “Atmospheric aging processes can change the composition and mixing state of aerosol population. In marine environments, the degree of aging on sea salt particles can be reflected by the contribution of AgedSS and SS/Sulf particles in the population.”

Line 821: “Stati**sti**cal uncertainty”.
Corrected. This sentence has been moved to Text S2.

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

Corrected.

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 ofice 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

Review #2

This paper presents an analysis of individual aerosol particles that were collected on a cruise that extended from South Korea to Antarctica, altogether 29 samples. The authors use micro-spectroscopic techniques to determine the particle composition and morphology. The analysis

has two themes, the first is to determine quantitatively the chemical mixing state using diversity metrics, and the second is to quantify the particles' ice nucleation properties at cirrus conditions.

This is a rich dataset from an area of the world where particle samples are scarce, and I want to commend the authors for the work they have done in their analysis. The paper fits within the scope of ACP, and merits publication. As it stands, the paper is quite long and reads more like a report of "what is out there" (with some additional pieces of information such as the ice nucleation rate coefficients/INAS densities). Given that we don't have much data of this detailed level from this region of the earth, this may be fine, however I think the impact of the paper as it's presented is limited and could be strengthened by emphasizing the new contributions.

We made following revisions to emphasize our new findings:
(1) in L26, "The mixing state index of the particle population ranged from 50% to 95% over the northern and southern hemispheres."
(2) in L28, "Multiphase processes on sea salt particles resulted in chlorine deficiency. This selective aging process made the marine particle population more externally mixed."
(3) in L30. "Ice nucleation onset conditions were measured and the investigated particles showed diverse ice nucleation abilities. The fresh sea salt particles with organic coatings exhibited the highest ice nucleation ability at a relative humidity with respect to ice as low as 121%. The sea salt mixed sulfate particle was enriched in INPs by a factor of 1.9."
(4) in L35, "Our analysis shows that assuming an internally mixed particle population in the marine atmosphere can lead to errors of several orders of magnitude in predicting ice nucleation rates."

Major comments:

- Currently the mixing state analysis and the ice nucleation analysis are disconnected. When I started reading the paper, I expected that the paper would elucidate the connection of the two, i.e., how does mixing state impact ice nucleation properties (which would be a very exciting topic). However, the connection is very weak, basically remaining at a level of speculation (line 753) "These variations are likely contributed to not only the complex compositions but also the physical and chemical mixing state of these particles." There are several studies out there that already show this. Is there any way to connect the two topics more closely? This may not be easily possible because I'm not sure if the chi metric is suitable to predict errors in ice nucleation abilities when assuming some idealized mixing state (this has been shown to be possible to some extent for CCN activity (Ching et al., 2017, 10.5194/acp-17-7445-2017.) and optical properties (Yao et al., 2022, 10.5194/acp-22-9265-2022.)). Considering this, I wonder if it would be better to separate the two topics into two separate (but shorter) papers.

  Thank you very much for the suggestions. We did analysis and tried to link the contact angle for INPs with mixing state index for particle population. Although we didn't find a strong correlation between these two variables, this is one step further to gain a better understanding in how mixing state impact ice nucleation. More studies are needed. Please see the details in the major change #1 listed at the beginning of this

document. We have decided to simply some of the discussion, especially on the ice nucleation kinetics (the major change #2 listed at the beginning of this document). Thus, we prefer to keep these in one manuscript.

- Even when considering the mixing state topic and the ice nucleation topic separately, I'm left with the question "so what"? For the mixing state topic, the authors could strengthen their discussion if they connected their observations to what models currently assume. Is this consistent or not with their findings? What could models learn from these observations? For example, it is interesting that the authors find partially externally mixed populations in these regions, and the fact that they identified aging mechanisms that lead to a more external mixture is also very interesting. For the ice nucleation topic, I'm not sure what to do with the analysis in section 3.6.2-3.6.4. These parameters could be useful for modelers, but other studies have been putting forward parameterizations like these for similar particle types – how are the results presented here similar or different from previous studies? Or, if this is the first time such parameterizations are derived for a given particle type, say so.

  On the mixing state topic, we made the following revision in Section 4 on the atmospheric implication. In L970, we revised the statement to "We showed that multiphase processes lead to the aging of fresh sea salt particles resulting in chlorine deficiency and thus changing the mixing state of the population (Fig. 5). This is a selective process that occurred on the sea salt particles and is different from the aging by condensation of secondary materials. As discussed in Section 3.2, these types of selective or inhomogeneous aging on particles move the population toward a more externally mixed state. To the best of our knowledge, this is not considered in the models predicting the mixing state of aerosols, even for the particle-resolved model. Our study suggests that, when discussing the influence of aging on the evolution of the mixing state, it is necessary to consider whether the aging processes occur uniformly on all particles."

  On the ice nucleation kinetic, we decided to focus on J_{het} and contact angle in the main text and kept the discussion on ice nucleation activated fraction and n_s in the supplemental information. Please see the major change #2 listed at the beginning of this document. We added a brief discussion on the comparison of J_{het} with other studies and revised the Fig. 13 of the original manuscript as follows:

  in L845, "In Fig. 12, we compared our J_{het} parameterizations with previous studies. DIN J_{het} of the Dust-dominated sample is similar to the particles collected during the Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE ENA) field campaign within the marine boundary layer (ACE ENA MBL) (Knopf et al., 2022, 2023). It is about 1-2 orders of magnitude lower than particles collected from the free troposphere (ACE ENA FT). DIN J_{het} of the other particle types investigated in this study are lower than particles collected from ACE ENA FT, ground site (ACE ENA GD), and ACE ENA MBL. This is consistent with the results showing that our particles nucleated ice at higher RH_{ice} than

particles collected during ACE ENA (Fig. 11). DIN J_{het} for the FreshSS and AgedSS samples is similar to the SSA investigated by Alpert et al. (2022) in the lower Δa_w range (less than 0.25). This is likely because S11 nucleated ice in this Δa_w range (i.e., RH_{ice}) is similar to the SSA INP investigated by Alpert et al. (2022) as they have similar morphology and composition. IMF J_{het} of the FreshSS and AgedSS samples is slightly higher than particles from ACE ENA GD and 1-2 orders of magnitude higher than the other field samples (Fig. 12C). The IMF J_{het} of the FreshSS and AgedSS samples displays a smaller slope with Δa_w compared to the laboratory-generated particles, such as illite (Knopf and Alpert, 2013), natural dust (Alpert and Knopf, 2016; Niemand et al., 2012), leonardite particles (Knopf and Alpert, 2013), and diatomaceous material (Knopf and Alpert, 2013; Knopf et al., 2011; Alpert et al., 2011a, b). This is consistent with the study by Knopf et al. (2022) which suggested that the diversity of ambient particles exhibit different ice nucleation efficiencies compared to single-component INP types studied in the laboratory at different temperature ranges. Our results emphasize the importance of studying the ice nucleation ability of particles and their parameterizations from different geological areas with different diversities in particle characteristics.".

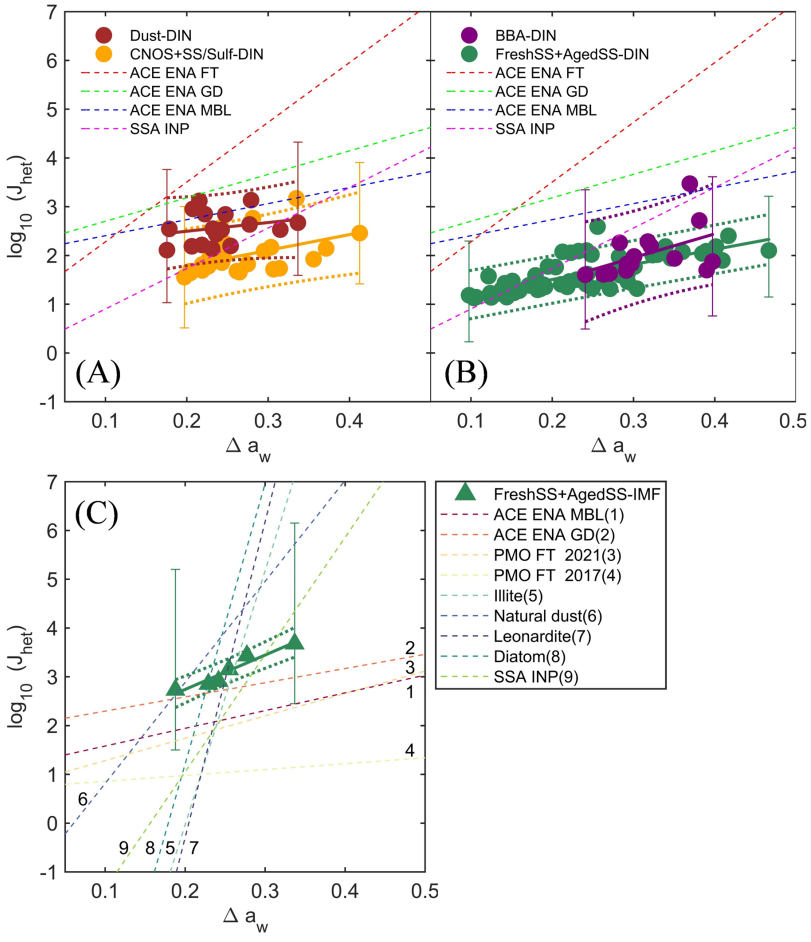

Figure 12. J_{het} as a function of Δa_w. Circles represent DIN J_{het} for (A) Dust, CNOS and SS/Sulf dominated samples, (B) BBA, FreshSS and AgedSS dominated samples. Triangles represent IMF J_{het} (C) for FreshSS and AgedSS

dominated samples. Solid lines indicate the fittings according to $\log(J_{\mathrm{het}}) = c \times \Delta a_{\mathrm{w}} + \mathrm{d}$ with associated dotted lines representing the 95% prediction intervals. Representative ΔJ_{het} (error bars) are plotted (Text S2). (A-B) Dashed red, green, and blue lines represent DIN J_{het} parameterizations of particles collected from ACE ENA FT (Knopf et al., 2023), ACE ENA GD (Knopf et al., 2022), and ACE ENA MBL (Knopf et al., 2023), respectively. Dashed magenta line indicates J_{het} for SSA INPs (Alpert et al., 2022). (C) Dashed lines with the number label of 1-4 represent IMF J_{het} for particles from ACE ENA MBL (Knopf et al., 2023), ACE-ENA GD (Knopf et al., 2022), and the Pico Mountain Observatory (PMO) under free-tropospheric (FT) conditions in the Azores (PMO FT 2017 and PMO FT 2021) (China et al., 2017; Lata et al., 2021), respectively. Dashed lines with the number label of 5-8 represent IMF J_{het} for illite particles (Knopf and Alpert, 2013), natural dust (Alpert and Knopf, 2016; Niemand et al., 2012), leonardite particles (Knopf and Alpert, 2013), diatomaceous material (Diatom) (Knopf and Alpert, 2013; Knopf et al., 2011; Alpert et al., 2011a, b), and SSA INP (Alpert et al., 2022), respectively.

Minor comments:

- The English language needs some polishing throughout the paper (typos and grammar).

  We checked the grammar and improved the writing throughout the text.

- Looks like Figure 4 and 5A display the same information? Omit Figure 4?

  Yes. We decided to remove the Fig. 5a.

- Line 512: the difference in mixing state of super-micron and submicron particles is interesting. When discussing this, it would be helpful to state what the species/elements are that are internally (or not internally) mixed. Information like this would be helpful in other places of the paper as well.

  We revised some of the statements to add this information, for examples:

  In L515, "The highest χ of about 95% for S16 and S17 indicated that these two samples dominated by Na and Cl were largely internally mixed."

  In L520, "This was reflected in χ of about 58% for S14 and S15. The main elements of Na and S in these two samples are more externally mixed."

  In L530, "This suggests that the main elements of Na, S, and Cl and the minor elements of Mg, K, Fe, Ca, and Si in super-micron particles tend to be more internally mixed than in submicron particles (Fig. S8B and Fig. S9B).

- What is the duration of each ice nucleation experiment?

  We stop the experiment after first ice nucleation occurred. Thus, depending on the onset of ice nucleation temperature, the duration of each experiment ranged from 20 min to 70 min at the cooling rate of 0.2 K/min.

Review #3

In the manuscript submitted by Xue et al., the authors conducted a comprehensive investigation into the mixing state and ice nucleation abilities of ambient particles collected over the Western Pacific and the Southern Ocean. Considering that the aerosol samples were collected on a broad spatial scale and there were limited studies on ice nucleating particles (INP) over the covered regions, I suggest that this paper fits well within the scope of ACP. Nevertheless, there is a need for overall improvement in the English language expression, and a major revision is recommended before publication.

Major comments:

1. The manuscript needs overall improvement in English language expression.

   We checked the grammar and improved the writing throughout the text.

2. In the introduction section, there is a lack of discussion on the state-of-the-art information regarding how the mixing state influences the ice nucleation of particles. The limited studies in the literature could serve as a significant motivation for the present study.

   We added the following text in L132 to strengthen the motivation of our study: "INPs and ice residuals are often mixtures of chemical species. Ice nucleation is inherently related to the mixing state of particles because it depends on the particle surface properties. Quantifying the ice nucleation rate also depends on the details of composition, surface area, and the mixing state of the particle population. The importance of mixing state for INPs is well recognized, but there are limited studies focusing on its quantification which is poorly understood (Kanji et al., 2017; Knopf et al., 2018; Riemer et al., 2019)."

3. The method section lacks details concerning the sampling time, cut-off size of particles used in different analyses, and operational procedures.

   During the field campaign, we collected particles on the third and fourth stages of the impactor with 50% collected efficiency at the aerodynamic size of 0.5 µm and 0.25 µm, respectively. In this manuscript, we only used the particle samples on the fourth stage with a smaller size range. We added the following text related to these experimental methods:
   In L157: "Particles were collected on the third and fourth stages of the impactor with 50% collection efficiency at aerodynamic sizes of 0.5 µm and 0.25 µm, respectively."
   In L166: "Particles were collected simultaneously on these two substrates which were placed side by side in the same impactor."
   In L171: "Therefore, we limited our analysis to the samples collected on the fourth stage."
   In L174: "When the relative wind direction against the ship heading is between 110° and 260° and the relative wind speed is below 2 m s^{-1}, samples may have been influenced by the ship exhaust (Park et al., 2020). Potentially contaminated samples were excluded from the analysis, resulting in a total of 29 selected samples."

In Text S1: “As shown in Table S1, close to the continent of East Asia, the sampling time was 10 min for S1 to S4. From S5 to S20, the sampling time was about 240 min. Over the SO region, the sampling time was about 360 min for S21 to S29.”. The detail sample information including exact sampling times are listed in Table S1.

4. One of the major conclusions on coating thickness or coating compositions may influence the ice nucleation efficiency of collected particles is not sufficiently supported by the present work. The discussion focuses on one sample (S14) with a low ice nucleation efficiency.

   In the manuscript, we agreed that the statement on coating compositions influence the ice nucleation efficiency of the collected particles in our study is not sufficiently supported since we didn’t know the coating composition. So, we revised the statement in L749-751 in the original manuscript, “The reason of this discrepancy at lower temperatures is not clear, but as previously mentioned, we speculate it has something to do with coating thicknesses or coating composition.” to L787, “The reason for this discrepancy at lower temperatures is not clear. Future work on the coating composition may provide more insight.”.

   However, we still think the coating have potential impacts on ice nucleation which are supported by our observations listed below.

   (1) We showed that the FreshSS and AgedSS dominated sample, S11, have clear difference in the number fractions of coated INPs, in L659 “Different number fractions of INPs identified in the FreshSS and AgedSS dominated samples (S4, S11, and S12) have organic coatings. More than 80% of INPs in S11 were coated with organics compared to S4 (33%) and S12 (30%).”

   (2) We found that the coating also impacted the ice nucleation of dust particles and showed similar effects, in L727, “The DIN onset conditions of S1 are similar to the M5/illite (10:1) mixtures (Fig. 11A). We also found that coated particles (Fig. S16) in S1 serving as INPs have a similar coating thickness (Fig. S11) as organic coatings on M5/illite greater than 1:1.” and also the related discussion in the same paragraph.

   (3) We calculated the coating thickness and did the t-test for the difference between INPs and non-INPs in BBA. Although the S14 has a low ice nucleation efficiency, as described in L657, we found that the organic coating thickness of BBA INPs (1.07 ± 0.68 µm, n=13) is significantly thicker than the other BBA (0.53 ± 0.37 µm, n=112) at significant level of 0.001.

5. The author has developed several parameterizations to predict the heterogeneous ice nucleation of marine aerosols. However, the motivation behind deriving these

parameterizations is not clearly presented. The derived parameterizations have different assumptions, and their forms vary among different particle classes. How to apply these parameterizations and to which aspect can they contribute to the prediction of particle ice nucleation are not clearly explained. I would suggest focusing on one or two parameterizations that are atmospheric-relevant and feasible.

We shorten Section 3.6 on ice nucleation kinetics. Some of the discussions, parameterizations, and figures were moved to the supplemental information. Please see the details in the major change #2 listed at the beginning of this document.

Specific comments:

1. Quantified results need to be included in the abstract. For example, the percentage of different aerosol sources that contribute to the collected particles (L27-L28).

   We added or revised the following sentences:
   in L24, "Most of the samples were dominated by fresh sea salt, aged sea salt, and sea salt mixed with sulfate particles, with total number percentages ranging from 48% to 99% over the Western Pacific and the Southern Ocean.".
   in L26: "The mixing state index of the particle population ranged from 50% to 95% over the northern and southern hemispheres."
   in L31: "The fresh sea salt particles with organic coatings exhibited the highest ice nucleation ability at a relative humidity with respect to ice as low as 121%."
   in L33: "The sea salt mixed sulfate particle was enriched in INPs by a factor of 1.9."

2. Some statements in the abstract are unclear. For example, the statement "We tested different ice nucleation parameterizations of marine atmospheric particles for their applicability. Finally, we discuss how the mixing state of particle populations impacts ice nucleation in the atmosphere" lack specific outcomes. Could you provide more details on the results of these tests and discussions?

   We deleted this statement in the abstract since we revised the discussion on the parameterization and deleted the example from Lines 920-930 of the original manuscript.

3. L85-L87 are repeating with L73-L75.

   We removed the sentence in L85-L87 of the original manuscript.

4. L96-L97: Consider deleting L96-L97, as the main focus in this paragraph is on chemical composition and mixing state.

   We deleted this sentence.

5. L138-L140 should be moved to the following paragraph.

   We removed this sentence and revised the following paragraph.

6. The collection time for each sample exhibits large variation (Table S1). Could the authors provide an explanation for this and include these details in the SI? This information will help indicate the atmospheric representativeness of the aerosol samples.

   For the single particle analysis and ice nucleation experiments, we need a suitable and sufficient particle on the substrates. Particle loading should be good enough to obtain statistically significant information by single particle analysis. Over-loaded sampling and particle aggregation should be avoided. Thus, during the field campaign, we adjusted the sampling time according to the particle concentrations in different regions.

   We add the information in the SI Text S1 as follows: "During the sample collection, we need a suitable particle loading on the substrates for the single particle analysis and ice nucleation experiments. A good particle loading with sufficient particles should be achieved for single particle analysis to obtain statistically significant information. Over-loaded sampling and particle aggregation should be avoided. Thus, we adjusted the sampling time according to the particle concentrations in different regions during the field campaign. As shown in Table S1, close to the continent of East Asia, the sampling time was 10 min for S1 to S4. From S5 to S20, the sampling time was about 240 min. Over the SO region, the sampling time was about 360 min for S21 to S29."

7. L161-L163: Were both the TEM grid and silicon wafer chips collected using the SKC sampler? It is not clear whether they were collected in parallel or not. This should be clarified for better understanding.

   Yes, the samples were collected in parallel on both substrates. We added a sentence in L166 to clarify this: "Particles were collected simultaneously on these two substrates which were placed side by side in the same impactor."

8. L167-L168: Are only samples in the fourth state used for further measurements? Could the authors explain the rationale behind applying such a small cut-off size?

   In this manuscript, we mainly focus on INP identification and deposition ice nucleation at low temperatures. Thus, we only used the particle samples on the fourth stage with a smaller size range that may have a longer lifetime in the atmosphere and can potentially transport to higher altitudes at lower temperatures. We are working on a separate manuscript focusing on the immersion freezing of particles collected on the third and fourth stages of the impactor from this field campaign.

   We added this information in L169: "This study focuses on deposition ice nucleation at low temperatures and INP identification. Particles with a smaller size range may have a longer lifetime and can potentially be transported to higher altitudes in the atmosphere. Therefore, we limited our analysis to the samples collected on the fourth stage."

9. L196: What kind of particle size do you mean here. Is it referring to the aerodynamic size (AD) determined by the SKC sampler, or is it the electrical mobility diameter (ECD)? This should be clarified in the main text.

   We revised the sentences in L201 and make it clear that it refers to size determined by SEM: "The particle size reported here is the equivalent circle diameter (ECD) based on the two-dimensional projected area of the particle as determined by CCSEM/EDX analysis. Particles with a size (ECD) between 0.2 µm and 3 µm were included for analysis in this study."

10. L222: Any citations for using 0.8 as a specific threshold value for fresh and aged SS?

    We conducted CCSEM/EDX analysis on fresh sea salt particles generated by nebulizing sea water. As shown in the figure below, the Cl/Na was slightly higher than 0.8 for particles at about 0.2 µm. This is partially due to electron beam damage on small particles. This is consistent with previous study using CCSEM/EDX analysis on NaCl particles (Laskin et al., 2012; Ghorai et al., 2014). Thus, in this study, we use Cl/Na of 0.8 as a threshold value to distinguish the fresh and aged sea salt particles since the samples we investigated have large number of small sea salt particles.

    We added the following sentences in L228: "CCSEM/EDX analysis of fresh sea salt particles generated by nebulizing sea water shows that the Cl/Na ratio is slightly higher than 0.8 for particles at about 0.2 µm (Fig. S2). The samples we investigated contain a large number of small sea salt particles. Thus, we use a Cl/Na ratio of 0.8 as the threshold value to distinguish between the fresh and aged sea salt particles."

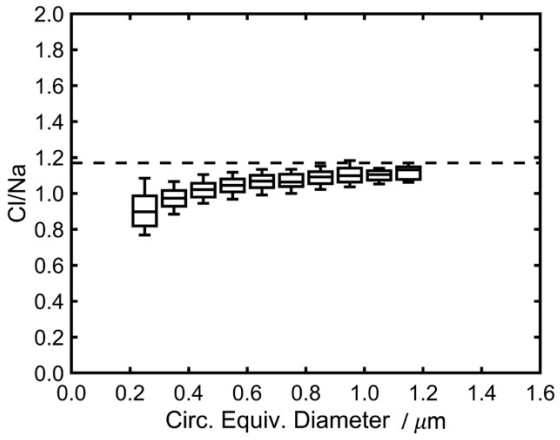

Figure S2. The Cl/Na of NaCl particles and fresh sea salt particles by nebulizing sea water based on CCSEM/EDX analysis. The Cl/Na was slightly higher than 0.8 for particles at about 0.3 µm. This is partially due to electron beam damage on small particles. The Cl/Na increases as particle size increases close to the Cl/Na of 1.17 in sea water. This is consistent with previous study using CCSEM/EDX analysis on NaCl particles (Laskin et al., 2012; Ghorai et al., 2014).

11. Line 229: There is a missing period in this sentence.

    Corrected.

12. L244: Why was a hemispherical shape of particles assumed rather than a sphere?

    Due to the impaction on the substrates during the sampling, particles may have deformed and no long exist as sphere on the substrates, for example, aged sea salt and sulfate particles. Thus, we assume the particles are in hemispherical shape on the substrates to have better estimation on the particle volume.

13. L308: Which particle sample, TEM or silicon wafer?

    We used the sample on silicon wafer for ice nucleation experiment. We added this information in L320 as following: "Prior to ice nucleation experiments, a particle sample collected on a silicon wafer was placed in the INC."

14. L316: Ice formation of one particle or all particles on the grid/chip? I assumed you have many particles in one sample?

    We only report the first ice nucleation event. Usually, at the ice nucleation onset, there is only one ice crystal formed on one particle. Sometimes, two ice crystals may form at the same time. If continue to cooling the sample, more particles may form ice crystals, but we don't report these because the water vapor will go to ice crystals and the RH field over the whole sample may not be uniform anymore. We have mentioned this in L340. We reported the particle number available during the experiments and activated fraction at the ice nucleation onset conditions in Table 1.

15. Figure 3: Is the scale unit on TEM images missing?

    The scale bar for all images is 1 µm. We added this information in the figure caption.

16. L431: Citations are needed here regarding particle formation from biogenic emissions in the Rose Sea.

    We added the references in L445-449: "The increased contribution of these three sulfur-containing particle classes may be related to the biogenic sulfur emission from polynyas (areas of open water surrounded by sea ice) in the central Ross Sea (Fig. S7) (Brean et al., 2021; Baccarini et al., 2021; McCoy et al., 2021; Jang et al., 2019; Zhang et al., 2015)".

17. L488-L489: Not sure if I followed this sentence.

    We deleted this sentence to keep the text concise and avoid more confusion.

18. Table 1: BBA exhibits relatively larger particle size compared to particles from other sources. Could the author provide an explanation for this?

    The particle size reported here is the circular equivalent diameter based on the 2-D projected area measured by SEM. We suspect that the organics in BBA may present in a liquid or semi-solid state and spread on the substrate due to the impaction during sampling. Thus, this increases BBA's circular equivalent diameter. We don't have further support on this speculation that we won't have additional discussion in the text on this topic.

19. L540-L541: The negative correlation between the number percentages of AgedSS and SS/Sulf particles and χ is not supported by the low R^2 (<0.11) in Figure 6B. Therefore, the statement on "The negative correlation between the number percentages of AgedSS and SS/Sulf particles and χ indicates that aging resulted in a more externally mixed particle population." needs justification.

20. Figure 6C: The negative correlation between the number percentages of CNOS and χ needs justification, as it is likely caused by a few extreme points. I would suggest repositioning these extreme points and conducting the fitting again.

    Response to comment #19 and #20: To focus on the impact of particle aging on mixing state, we excluded two events, i.e., the Dust-dominated samples from a dust storm (S1 and S2) and BBA-dominated samples from wildfire emission (S14 and S15), from the analysis (Figure 6). We use the data from 25 samples for the new Figure 6. To minimize the potential influence of extreme data points on the regression, we binned the data sets based on the number percentage of particle type. Linear regression is now based on the binned data. In the figure, p values are also provided. As shown in the revised Figure 6, the p values are all smaller than 0.05 and the R^2 for each regression is greatly improved.

    We revised the sentences in L535-550 (the L516-L521 in original manuscript) as follows:

    "The changes in the particle composition may affect the mixing state of the particle population. To focus on the effect of particle aging on the mixing state of marine aerosols, we excluded four samples from two events from the analysis, i.e., the Dust-dominated samples from a dust storm (S1 and S2) and the BBA-dominated samples from wildfire emissions (S14 and S15). Figure 6 shows the relationship between χ and the percentages of FreshSS, AgedSS and SS/Sulf, and CNOS particles in different size ranges. To minimize the potential influence of extreme data points, the linear regression was performed using the binned data based on the number percentage of particle type. We found a strong correlation between the percentages of FreshSS particles in the samples and their χ (Fig. 6A, $R^2 = 0.66$, $p = 0.008$), especially for submicron particles ($R^2 = 0.85$). χ increased as the contribution of FreshSS particles increased. There is a negative correlation between χ and the total percentage of AgedSS and SS/Sulf particles (Fig. 6B, $R^2 = 0.46$, $p < 0.05$). We also found a strong negative correlation between χ and the contribution of CNOS particles

(Fig.6C, R^2 = 0.86, p = 0.002). The results of the above-mentioned relationship also hold for both submicron and super-micron size range particles."

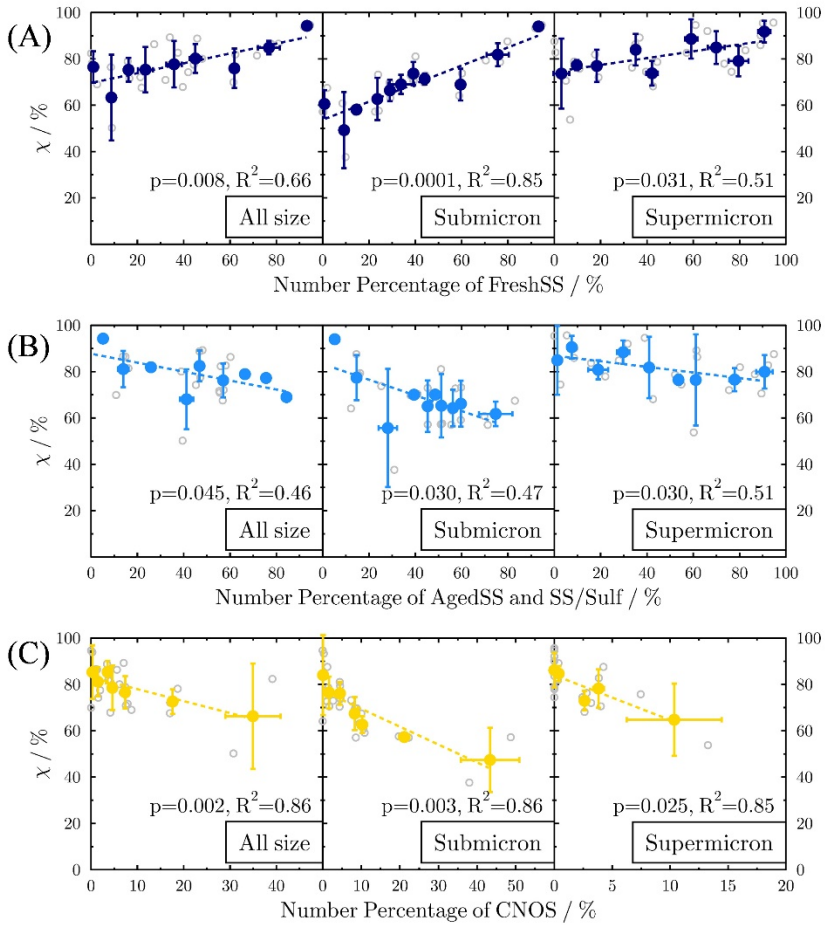

Figure 6. Variation of mixing state index (χ) with the number percentages of different particle classes: (A) FreshSS, (B) AgedSS and SS/Sulf, and (C) CNOS. Panels from left to right are for particles with different size ranges: all particle size (All size), 0.2 – 1 μm (Submicron), and larger than 1 μm in diameter (Super-micron). Original and binned data are shown in open circles and solid color circles, respectively. Linear regression (dash line) is based on the binned data.

21. Equation 12. The legend used here is confusing. If A represents a certain class of particles and N is the number of particles in that class, it may make more sense for N_A^{INP} to be represented as N_{INP}^A ?

    We corrected the equation, now Eq. 14, as reviewer suggested.

22. L723-L724. I would refrain from asserting that S14 is ice-active given the large uncertainties in its onset conditions (Figure 11). This is also contrary to the author's earlier statement (L578) that "The RH_{ice} onsets were only about 3% lower than the homogeneous

nucleation limits between 228 K to 220 K, and thus samples dominated by BBA may not have been efficient heterogeneous ice nuclei."

We deleted the sentence and only keep the comparison with SRFA in L758 as follows: "Suwannee River standard fulvic acid (SRFA) and leonardite were used as surrogates of atmospheric organics from biomass burning (Wang and Knopf, 2011). Ice nucleation onset conditions of S14 are very similar to those of SRFA and ozone aged leonardite particles above 220 K but higher at lower temperatures."

Reference：

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