Dear editor,

We thank editor and the referees for their further comments and suggestions to improve our manuscript. After carefully considering these comments, we provide point-by-point responses and made the corresponding changes in both the main text and supplemental file. As suggested by editor, the title is adjusted to "Diverse sources and aging change the mixing state and ice nucleation properties of aerosol particles over the Western Pacific and Southern Ocean". We hope that these responses and the revised manuscript are acceptable and meet the requirements for publication.

Thank you for your consideration.

On behalf of all co-authors,
Bingbing Wang

In the following point-by-point responses, line numbers in the responses (in light blue fonts) refer to the revised manuscript and those referring to the previous version of manuscript are specified. The line numbers in the referees' comments (in black fonts) refer to the previous version of manuscript.

Point-by-point responses:

I thank the authors for addressing most of the comments raised by the three reviewers; however, there are additional comments (listed below) that need to be properly addressed before the manuscript can be accepted for its publication.

Reviewer #1

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.
Thank you for the comments. We revise the manuscript as suggested.

The authors have done a better job of linking IN activity and χ (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 χ is influenced by surface composition, it is not wholly sensitive to it.

We fully agree with reviewer on the explanation for the lack of connection between ice nucleation activity for deposition ice nucleation and χ. We also have mentioned this in the main text in L955-966.

We revise the following statement to explicitly state these reasons in L961: "In addition, no direct connection between *Err* (i.e., θ) and χ for DIN is likely attributed to the fact that ice nucleation propensity for DIN depends on the particle surface composition and morphology on the individual particle basis, whereas χ characterizes the overall mixing of species (i.e., composition) within the particle population which may not sensitive to the particle surface characteristics."

Specific comments

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

Line 61: I think that the authors should note that they focus primarily on deposition, and to some degree immersion freezing in this study.
We make this clear by revising the statement in L158 as follows: "We measure the ice nucleation onset conditions and identify the nucleation pathways (DIN or IMF) for representative samples. In this study, we focus primarily on DIN and in some cases IMF below 240 K. Individual identified INPs are characterized and compared with the particle population to relate ice nucleation ability to the mixing state.".

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

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

Line 93: I think that the authors need to highlight the necessity of using single particle techniques for quantifying mixing state index.
We add the following sentence in L93: "Thus, chemical characteristics of individual particles should be achieved by single-particle techniques to quantify χ."

Line 97: I recommend moving this sentence before the previous one, to improve the logical flow.
We revise the text as suggested.

Line 100: Doesn't this sentence directly contradict the one at Line 93?
We revise the sentence in L98 as follows: "However, the impacts of particle sources and atmospheric aging on the chemical mixing sate of marine particles are not well understood.".

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.
We reorganize this paragraph and separate it into three short paragraphs in L102-149. These three paragraphs focus on the lab or mesocosm studies with DIN and IMF, field studies on INP concentrations and IMF, and field studies on DIN and INP identification, respectively.

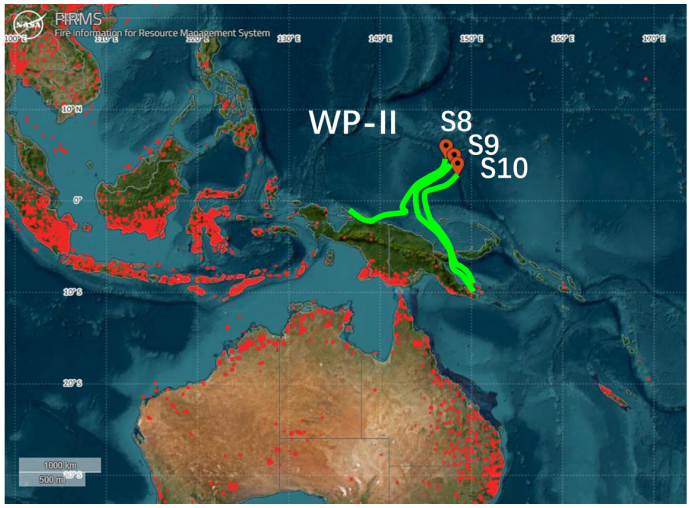
Line 129: This marks a transition in the overall topic, and I think should be split into its own paragraph.
We reorganize this paragraph, please see response above.

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?
To get suitable samples for both single particle analysis and ice nucleation experiments, we adjusted the sampling time according to the aerosol concentrations in different regions and by inspecting the particle loading on the substrates. This is described in the Text S1. The main reason is that, at the beginning of the cruise, the first several samples were influenced by a dust storm (S1 and S2) and likely influenced by the continent outflow of East Asia (S3). After that, the particle concentration decreased dramatically as shown by the BC concentration in Table S1. After S4, we increased the sampling time by about 20 times. In fact, after collection, the estimated particle surface area for S4 is a factor of 8 less than S1 and indeed about 13 to 26 times less than S10 to S14 as shown in Table 1.

Line 230: Fig. S2 is missing. This also means that the following supplemental figure references appear to be incorrect as well.
We check that the SI file now provides all the correct supplemental figures.

Line 381: Is the particle Dust or Dust-like? What is the distinction?
We correct this typo and refer only to “Dust” particle.

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.
We overlay the 120 h backward trajectories for S8, S9 and S10 as shown below. The backward trajectory of S8 originated from the areas near the fire spots of New Guinea. We revised the statement in L433 as follows: “As shown in Fig. S4C, this is further supported by the air mass for S8 originated around the region where fire spots were detected in NASA Fire Information for Resource Management System (FIRMS).”



Line 450: This sentence and the next one can be combined.
We combine these two sentences as follows in L463: “The formation of polynyas during the austral summer allowed phytoplankton to grow and produce DMS which can be transferred into the atmosphere and oxidized.”.

Line 452: Is the study of Kunwar tied to a particular time of year?
The particle samples for Kunwar’s study were taken from 22 November, 1994 to 11 February, 1995, which is close to the sampling period of this study. We add this information in L466: “Previous work found that the MSA concentration over the Southern Ocean from November 1994 to February 1995 was about two times higher than that of the Western Pacific.”

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

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.
We delete the sentence in L554 of the previous revision. We also shorten the sentences in L570-572 of the previous revision, now in L568 as follows: “These results verify the schematic descriptions on the evolution of the mixing state described by Riemer et al (2019) after adding new particles or particles of a dominant type.”.

Line 552: This sentence and the next one basically say the same thing. I would recommend to combine or remove one.
We delete this sentence in L554 of the previous revision.

Line 591 and 593: The descriptions for ii and iv should both be condensed into one sentence.
We revise the descriptions as follows: in L599, “ii) BBA influenced sample (S14) in which CNO and AgedSS particles contributed 43% and 51%, respectively.” And in L602, “iv) FreshSS and AgedSS dominated samples (S4, S11, and S12) with a total number percentage of these two particle types great than 70%.”.

Line 682: Fig. S15 shows the active site density for these samples, not the average elemental composition of INPs and non-INPs.
In the current version of SI file, Fig. S15 provides 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.
We agree with reviewer on this. Future investigations using better proxies are needed. We add the following statement in L953 to make this clear: "Instead of pure NaCl, better proxies for SSA should be used in future studies to obtain better estimations in *Err* and thus J_{het}."

Line 936: Authors need to explicitly link the differences between the physicochemical properties that we know affect ice nucleation activity and what χ actually measures.
To the best of our knowledge, there is no a single physicochemical variable that is used to describe the physicochemical properties on the individual particle basis which affect ice nucleation activity, such as cracks, pores, and surface composition. That is why we use θ to characterize the ice nucleation activity based on CNT. χ reflects the overall composition of particle population.
We revise the following statement in L961 to explicitly state these: "In addition, no direct connection between *Err* (i.e., θ) and χ for DIN is likely attributed to the fact that ice nucleation propensity for DIN depends on the particle surface composition and morphology on the individual particle basis, whereas χ characterizes the overall mixing of species (i.e., composition) within the particle population which may not sensitive to the particle surface characteristics. There is a large gap in the understanding of how physical and chemical mixing state affects ice nucleation potential of particle population."

Line 998: ns has only been discussed in the supplemental, and thus I don't think it should be mentioned here in the conclusion.
We remove or revise these statements related to n_s.

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?
Thank you for the suggestion. We revise Figure 10 as shown below:

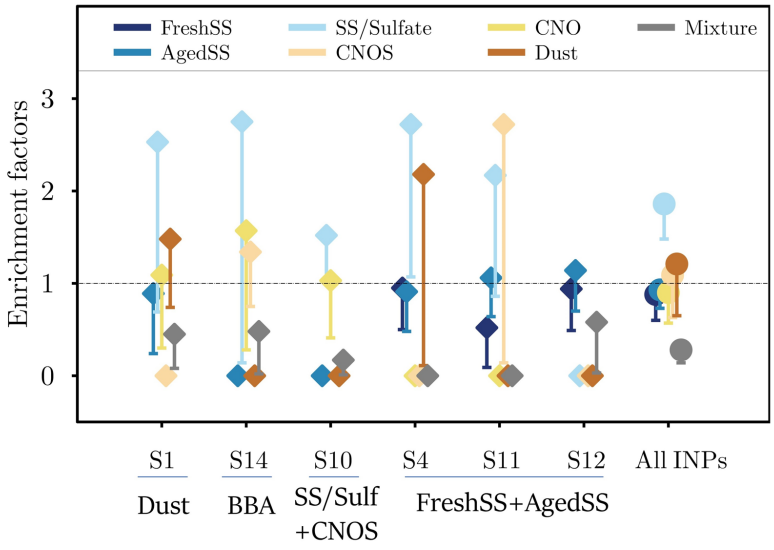


Figure 14: Why do the authors include S1 and S10, and S14 into the analysis, which we know to not be sea spray influenced, when their reference particle is NaCl? Why not just limit the analysis to the SS samples?

There are two purposes for this analysis in the section 3.7. The first one is to probe the potential connection of χ to the overall ice nucleation ability of individual particles (means of θ for 15-26 INPs) for each sample. The second one is to illustrate the potential differences in J_{het} or θ for oceans with complex particle sources as compared to open oceans dominated by SSA, especially for coastal areas. We revise the sentence to emphasize this in L950 as follows: "This suggests that the effects of aging processes and mixing state need to be considered in the ice nucleation modeling for different oceans with complex particle sources, such as coastal regions."

Where is the reference for Fig. S14?

The Fig. S14 were referred in L670.

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

Corrected.

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?

In the revised supplemental material, we grouped the supplemental tables and placed them right after the supplemental texts.

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

Corrected.

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

Corrected.

Reviewer #2

I appreciate the authors' efforts to respond to the reviewers' comment about connecting mixing state and INP propensity, but I don't think the new section 3.7 makes any sense.

There are two purposes for this analysis in the section 3.7. the first one is to probe the potential connection of χ to the overall ice nucleation ability of individual particles (means of θ for 15-26 INPs) for each sample. The second one is to illustrate the potential differences in J_{het} or θ for oceans with complex particle sources as compared to open oceans dominated by SSA, especially for coastal areas. We think that section 3.7 is necessary for this manuscript.

We revise the sentence to emphasize this in L950 as follows: "This suggests that the effects of aging processes and mixing state need to be considered in the ice nucleation modeling for different oceans with complex particle sources, such as coastal regions.".

The mixing state metric chi based on elemental composition has nothing to do with INP prospensity, and therefore it's not surprising that they don't find any relationship in Figure 14.

The mixing state metric needs to be chosen so that it connects to the underlying physical process. For example, in Ching et al. (2017), where the target quantity was CCN concentration, chi was defined based on hygroscopic and non-hygroscopic species. For the INP application, I would think one needs to look into how "good" INP species are mixed (or not mixed) with "bad" INP species. But there is an additional complication, namely with INP it really depends on what is on the surface of the particles. This morphological aspect is not captured with the chi metric.

Response to two comments above:

We agree with the reviewer that the χ based on the elemental composition which also does not capture the morphological characteristics is not the best mixing state metric for the current study. We also agree that mixing state metric reflecting the underlying ice nucleation process needs to be chosen. However, 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. We have mentioned this in L960-961. In future studies, one will need to come up with a mixing state metric that reflects the physicochemical properties controlling ice nucleation ability.

We revise the following statement in L961 to explicitly state these limitations: "In addition, no direct connection between Err (i.e., θ) and χ for DIN is likely attributed to the fact that ice nucleation propensity for DIN depends on the particle surface composition and morphology on the individual particle basis, whereas χ characterizes the overall mixing of species (i.e., composition) within the particle population which may not sensitive to the particle surface characteristics. There is a large gap in the understanding of how physical and chemical mixing state affects ice nucleation potential of particle population."

We add the following suggestion in L969 for future studies: "Mixing state metrics reflecting the underlying physical process and the particle physicochemical properties controlling ice nucleation ability are needed in future studies, for example, metrics measuring the morphological characteristics of particles.".

In addition, contact angle is not a good choice as error metric in this context. Instead, it should be something like INP concentration (i.e., answering the question how wrong are our INP

predictions if we assume that the population is internally mixed while in reality it is not internally mixed).

To the best of our knowledge, there is no single physicochemical variable that is used to describe the physicochemical properties on the individual particle basis which affect ice nucleation activity, such as cracks, pores, and/or surface composition. In our study, we focus on detecting and identifying the first ice nucleation events for deposition ice nucleation, not to fully capture the INP concentrations at different temperature and RH_{ice} conditions. That is why we use θ, not the INP concentration, to characterize the ice nucleation activity based on CNT. We agree with the reviewer that INP concentration or other parameters can be used and maybe a better proxy, for example, for immersion freezing.

We add the following sentences in L967 to include this suggestion: "Other ice nucleation variables, e.g., INP concentration, can be used to estimate the potential error in cloud modeling if internal mixing is assumed for the population. Mixing state metrics reflecting the underlying physical process and the particle physicochemical properties controlling ice nucleation ability are needed in future studies, for example, metrics measuring the morphological characteristics of particles.".

(Note that it is fine to report how different the contact angle is from pure NaCl, but the dependence on chi is confusing and potentially misleading).

One of the purposes of Section 3.7 is to probe the potential connection of χ to the overall ice nucleation ability of individual particles (means of θ for 15-26 INPs) for each sample, not the dependence. We go through the main text to avoid any statement saying the dependence of θ on χ.

Even though I had asked the authors to strengthen the connection between the two topics, I think it makes more sense to keep the two topics separate. It would have been nice to have this connection established, but as it stands there is more work to get there. The findings individually are still interesting and merit publication. It would be good to emphasize in the introduction that the point of the paper is not to establish the connection between the topics but to report on them individually.

We agree with the reviewer that there is more work to do before we fully understand what controls the ice nucleation ability of individual particles and how the physical and chemical mixing states affect the ice nucleation potential of particle population. We add the following sentence in the last paragraph of introduction (in L151) to state our main goal of this paper: "In this study, our main objectives are to quantify the mixing state and ice nucleation ability of marine particles over the Western Pacific and Southern Ocean."

Editor

Minor Comments:

I invite the authors to adjust their title to follow the ACP guidelines:

"Titles should be concise and consistent with the content and purpose of the article. For

research articles, ACP prefers titles that highlight the scientific results/findings or implications of the study. Examples of preferred result- and implication-based titles:
- Observed relationship between aerosol emissions and cloud albedo over the Atlantic (neutral)
- Increases in aerosol enhance cloud albedo over the Atlantic (definite)
- Increases in aerosol since 2010 enhanced global mean cloud albedo by 20% (quantitative)
- Aerosol-cloud brightening and the implications for climate sensitivity (neutral)
- Recent changes in aerosol-cloud brightening imply reduced climate sensitivity (definite)
Examples of less-preferred titles that highlight only the topic or method are given below. Authors may be asked to convert articles with a methodological title to a Technical Note:
- An exploration of the effect of aerosol on cloud properties
- Machine learning to understand aerosol effects on clouds
- Aerosol effects on clouds in the European climate model"

We now change the title to "Diverse sources and aging change the mixing state and ice nucleation properties of aerosol particles over the Western Pacific and Southern Ocean".

2. L140-L151: I think this should be written in the present tense.
We change to the present tense.

3. L169-170: "This study focuses on deposition ice nucleation at low temperatures and INP identification." The authors also reported information related to IMF. Given that deposition nucleation is the main topic of the present study, this should be stressed out in the introduction referring to relevant maritime studies on deposition ice nucleation
We revise the literature review on marine INPs and emphasize the deposition ice nucleation part in L134-149

Figures S17 and S18 are not called in the main text.
We now refer Figs. S17 and S18 in L808-810 in the main text as follows: "n_s based on the singular hypothesis (Vali, 1971; Connolly et al., 2009) and its parameterizations are also presented and discussed in Text S2, Fig. S17, and Fig. S18."

Technical comments:

L30: "Ice nucleation onset conditions". Please include the corresponding ice nucleation mode.
We include the nucleation mode in the sentence in L31: "Ice nucleation onset conditions primarily for the deposition mode were measured and the investigated particles showed diverse ice nucleation abilities."

L52: Define "INPs"
It is now defined.

L57: Replace "when droplets" with "when supercooled droplets"
L58-59: Replace "water vapor condenses on droplets or particles." with "water vapor condenses on droplets or particles at temperatures below 0C."

L78: Replace “changes in the physicochemical properties of particles” with “changes in their physicochemical properties”
L83: Replace “become mixtures” with “become a mixture”
We change the corresponding texts for the suggestions listed above.

L83: Add a reference after “morphologies”
We add the following references for this statement.

Riemer, N., Ault, A. P., West, M., Craig, R. L., and Curtis, J. H.: Aerosol mixing state: measurements, modeling, and impacts, Rev. Geophys., 57, 187–249, https://doi.org/10.1029/2018RG000615, 2019.
Li, W., Shao, L., Zhang, D., Ro, C.-U., Hu, M., Bi, X., Geng, H., Matsuki, A., Niu, H., and Chen, J.: A review of single aerosol particle studies in the atmosphere of East Asia: morphology, mixing state, source, and heterogeneous reactions, J. Clean. Prod., 112, 1330–1349, https://doi.org/10.1016/j.jclepro.2015.04.050, 2016.

L108-109: “are efficient INPs below 225 K”. Please indicate the ice nucleation mode
The ice nucleation mode is IMF. According to the comments of Reviewer #1 above, we adjusted this paragraph and revise this sentence in L110 as follows: “SSA can be efficient INPs via IMF before complete deliquescence (Schill and Tolbert, 2014)”.

L125-126: “observations. INP concentrations were lowest in the polar regions and highest in the temperate climate ocean”. Please indicate at what temperature range this is true
We add the information in L128-129 as follows: “INP concentrations were lowest in the polar regions and highest in the temperate climate ocean at about 258 K”.

L127: Should “oceans focus on” be “oceans have focused on”?
L134-135: Replace “depends on the details of composition” with “depends on the composition”
L198: Replace “microscopy” with “microscope”
L226-227: Replace “and the Cl/Na ratio is > 0.8” with “and with the Cl/Na ratio > 0.8”
L304: Replace “Mixing state index” with “The mixing state index”
L339: Replace “that immersion freezing” with “that IMF”
L373: Replace “its core surrounded” with “its core is surrounded”
L378: Replace “showed in” with “shown in”
L483: Replace “Mixing state described” with “The mixing state described”
L495: Replace “were” with “are”
We change the corresponding texts for the suggestions listed above.