Response to Reviewer #3

<u>Referee report for the manuscript entitled:</u>

"Measurement report: Influence of long-range transported dust on cirrus cloud formation over remote ocean: Case studies near Midway Island, Pacific"

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In this manuscript, remote sensing data has been used to determine whether long-range transported dust from Asia is active as INP in the formation of cirrus clouds. Data are collated from two space-borne instruments to calculate INP concentrations (INPC) and ice crystal number concentrations (ICNC). These values are compared at specific locations within an observed cirrus cloud that relate to where the dust would most likely be entrained. Where the values of INPC and ICNC are comparable, the manuscript concludes that this region is dominated by heterogeneous freezing due to the dust. Where the ICNC is much higher than the INPC, the manuscripts concludes that homogeneous freezing also takes place. Previous studies have shown that long-range transported dust acts as INP (Saharan dust in North America) and Asian dust has been shown to act as INP in China. This manuscript shows that Asian long-range transported dust also acts as INP. I am not an expert of INP, however having experience in adjacent aerosol fields, I am aware that there is still much to understand about INP sources and this manuscript confirms another significant source of dust and its far-reaching impacts.

When considering this manuscript for publication, there are several factors to take into account:

This manuscript is presented as a measurement report, however the data are retrieved from space-borne instruments rather than field or lab work. Additionally, there is analysis and interpretation of the results which potentially goes beyond the requirements of a measurement report.
This manuscript does not present novel ideas or methods, however it does establish evidence of long-range transported dust acting as INP in a new area.
For me, there are some major questions about the analysis of the individual boxes within the cloud. It appears that one of the boxes closest to the dust plume is described as lacking in INP and, as such, homogeneous freezing is dominant. This description requires some justification.

Response: We are grateful for the reviewer's constructive comments on the manuscript. All the comments have been addressed in the revised manuscript, and the responses to each comment are given below. Therefore, the specific discussions have been largely updated now. The main revisions are listed below:

(1) As strongly suggested by reviewer #2, here we decide to combine Part A and Part B and conduct the analysis by considering them as a whole, due to the uncertainty of DARDAR retrievals. The specific discussions have been largely updated now.

(2) As at least three reviewers suggest shifting this manuscript from 'Measurement Report' to 'Research Article', we would like to do so but also need to involve Handling Editor Prof. Krämer in the decision. In addition, considering the results have been largely rewritten by adding in-depth discussions, we also think it would be better to change it to 'Research Article'.

(3) As the cirrus clouds in the two cases have already formed for at least half an hour (as deduced from the vertical extent of the ice virga by assuming a falling velocity of 1 m/s), nucleated ice crystals may have undergone significant growth. Therefore, in the comparison between ICNC and INPC, we decide to mainly use the values of $n\_{ice,25um}$ and $n\_{ice,100um}$. The specific reason has been discussed in the text of the revised manuscript.

(4) As there is no evident indication of the depletion of dust INP, the possibility for occurring homogeneous nucleation is low. Therefore, we have removed all the discussions about the involvement of homogeneous nucleation in the cirrus clouds.

The methods would be enhanced by some more detailed description.

Response: Combining the comments from the four reviewers, we have realized that the uncertainties in DARDAR ICNCs are the most important concern that should be emphasized. Therefore, we have added a description of this issue in the second paragraph of section 2.2. (please see lines 125-137)

Restructuring of some paragraphs and tightening up the language in a few key sentences would greatly aid the reading and understanding of the manuscript.
Based on the above summary and the comments below, I recommend this manuscript is reconsidered after major revisions. I also recommend that this should be submitted as a research article if the following suggestions make the manuscript a more substantial contribution. Note that I do not have expertise in satellite products so am unable to comment on their descriptions here and the suitability of their use.
Response: Thank you for the valuable comments. We have largely revised the manuscript by considering the comments from you and the other three reviewers. Taking the reviewer's suggestion, we have removed 'Measurement report' in the title. Here we would also like to involve our Handling Editor Prof. Krämer to judge if it is justified to shift this manuscript from 'Measurement Report' to 'Research Article.'

Major points:

On the description of the method, the key aspects from He et al. (2022b) are mentioned throughout the manuscript, but it is quite scattered. I would suggest pulling this all together within the methods section so that the method is very clear for a reader who is unfamiliar with it. It is clear that if the ICNC is much higher than INPC then homogeneous freezing must be present, but it is not so clear how one knows that it is only heterogeneous if the ICNC is comparable with INPC. Why could it not still be both? I am not an expert in INP, but I think the key bit of information here is that the ice saturation will not reach the required 140 or 150% if heterogeneous freezing has already begun.
Response: Thank you for the reviewer's constructive comments. The details of the methodology were given in He et al. (2022). For this study, the significance is to reveal the potential influence of the long-range transoceanic Asian dust on the ice formation of cirrus clouds over the central Pacific, where is usually clean. Therefore, the methodology was not thoroughly described to avoid redundancy. It should be pointed out that, in either He et al. (2022b) or this study, the proposed method is only applied to case studies. One should note that the current space-borne remote sensing approach can help to find out only the dominant ice nucleation regime or the other due to the limited spatiotemporal resolution, insufficient sensitivity of radar signal at cloud top, and the relatively large retrieval uncertainty. Therefore, it may still count on in-situ measurements (Krämer et al., 2016, 2020; Sourdeval et al., 2018). The uncertainty in ICNC from the DARDAR product is estimated to be a factor of 3 due to instrumental sensitivity and physical assumptions; the uncertainty in INPC can be a factor of 0.5-5 due to the retrieval of dust-related INP-relevant parameters and limited accuracy of INP parameterizations. Besides, we also have to adjust the values of ice saturation and modified factor (polluted/aged dust will have weaker ice-nucleating efficiency) in INP calculations in different cases. As a result, we consider an ICNC-to-INPC ratio within one order of magnitude to be a good agreement.

For the second issue, if the ICNC is comparable with INPC, the most possible situation is that heterogeneous nucleation suppresses homogeneous nucleation by consuming the water vapor (i.e., reducing the ice saturation). This process can be continuous until the depletion of INP. In addition, there is another possibility that a very strong updraft is present so that homogeneous nucleation has a chance to immediately take place before the involvement of heterogeneous nucleation; in this situation, heterogeneous nucleation will finally occur later considering the sufficient INP supply. All in all, the actual physical process is rather complicated, the different types of ice nucleation mechanisms can take place one after another or simultaneously for a short while (i.e., the transition phase between the two mechanisms).

References:

Krämer, M., Rolf, C., Luebke, A., Afchine, A., Spelten, N., Costa, A., Meyer, J., Zöger, M., Smith, J., Herman, R. L., Buchholz, B., Ebert, V., Baumgardner, D., Borrmann, S., Klingebiel, M., and Avallone, L.: A microphysics guide to cirrus clouds Part 1: Cirrus types, Atmos. Chem. Phys., 16, 3463–3483, https://doi.org/10.5194/acp-16-3463-2016, 2016.

Krämer, M., Rolf, C., Spelten, N., Afchine, A., Fahey, D., Jensen, E., Khaykin, S., Kuhn, T., Lawson, P., Lykov, A., Pan, L. L., Riese, M., Rollins, A., Stroh, F., Thornberry, T., Wolf, V., Woods, S., Spichtinger, P., Quaas, J., and Sourdeval, O.: A microphysics guide to cirrus – Part 2: Climatologies of clouds and humidity from observations, Atmos. Chem. Phys., 20, 12569–12608, https://doi.org/10.5194/acp-20-12569-2020, 2020.

Sourdeval, O., Gryspeerdt, E., Krämer, M., Goren, T., Delanoë, J., Afchine, A., Hemmer, F., and Quaas, J.: Ice crystal number concentration estimates from lidar–radar satellite remote sensing – Part 1: Method and evaluation, Atmos. Chem. Phys., 18, 14327–14350, https://doi.org/10.5194/acp-18-14327-2018, 2018.

The methods could also contain a description of the two case studies before we dive into the results. In the discussion there is a nice point about the dust being from intense events where the dust is elevated by Mongolian anticyclones. Information like this would be better used in the beginning of the manuscript to set the scene. Is there other relevant meteorological information about the cases? Was there a particular reason for picking these two cases? What times and altitudes were used? Are the cirrus events very rare? A description of the two case studies would follow on nicely from the HYSPLIT model description in Section 2.

Response: There is some background information about the initiation of the Asian dust event (Sun et al., 2001). Asian dust plumes from the Gobi are always associated with the Mongolian anticyclone cases in which strong surface wind can blow up the dust particles and lift them to the troposphere, which then may be advectively transported. For the Asian dust plumes from the Taklimakan Desert, dust particles are elevated via the upward winds caused by the convergence of warm and cold air systems but most lifted dust below 5 km cannot be involved in the long-range transport due to the terrain confine from the mountains surrounding (i.e., West, North, and South sides). However, here we more focus on the dust-cirrus interactions rather than the initial mechanism of the Asian dust events.

It should be mentioned that it is not easy to find such clear-cut cases in which dust particles can be seen in the vicinity of (to connect with) the cirrus clouds. The cases presented in this manuscript can lead to a deeper analysis. And the dataset of collocated measurements from CALIPSO and CloudSat is limited. Hence, we do not conduct a statistical study. The launch of EarthCARE (with a 94 GHz Doppler cloud radar and a high spectral resolution lidar) in 2024 may enrich the measurements and provide many more cases for such a statistical study (Wehr et al., 2023).

References:

Wehr, T., Kubota, T., Tzeremes, G., Wallace, K., Nakatsuka, H., Ohno, Y., Koopman, R., Rusli, S., Kikuchi, M., Eisinger, M., Tanaka, T., Taga, M., Deghaye, P., Tomita, E., and Bernaerts, D.: The EarthCARE mission – science and system overview, Atmos. Meas. Tech., 16, 3581–3608, https://doi.org/10.5194/amt-16-3581-2023, 2023.

Sun, J., Zhang, M., and Liu, T.: Spatial and temporal characteristics of dust storms in China and its surrounding regions, 1960–1999: Relations to source area and climate, J. Geophys. Res., 106, D10, 10325–10333, https://doi.org/10.1029/2000JD900665, 2001.

The results could be restructured to make the storyline clearer. For each case, I would recommend starting with establishing the case using the HYSPLIT trajectory and the dust mass column density plots. Then the description of the cloud and the dust plume using the CALIPSO cross-sections would follow on nicely. Next, part A and part B need a clear introduction using the DARDAR product. Why were these regions in particular picked? The difference in the ICNC between the two boxes could be described and then finally use the profiles to compare the ICNC values with the INPC. Here, I would suggest stating the ICNC versus the INPC and the ratio for part A and B. Do these values alone indicate homogeneous or heterogeneous freezing? The main method used in the manuscript is this quantification so I would pull it out of the data and emphasise it. Then add the additional, contextual information from other papers about typical values for each mechanism.

Response: We are grateful for the suggestion. Just let us explain why we organize each case study as it is shown now. In this study, the most important thing is that CALIOP offers real observation to see that cirrus and dust are both there and have connected. Only with this fact, one can then use the MERRA-2 analysis data and HYSPLIT model as auxiliaries to trace/confirm the possible source and transport pathway of the air mass (i.e., dust plume). As MERRA-2 only shows the column information about the dust, we cannot use it to conjecture whether there are dust plumes at cirrus altitudes; also, no information on the presence of cirrus clouds can be provided with both MERRA-2 and HYSPLIT model. Therefore, it would be better to retain the structure as is and we hope the understanding from the reviewer.

As for the discussions of Part A and Part B, as suggested by reviewer #2, we have to combine them as a whole for further analysis. Thus, the result part has been largely modified. The reviewer may have to reevaluate it.

Table 2 shows the summary of ICNC and INPC for both cases split into the A and B boxes. Considering that the concentrations are defined as being “comparable” as within an order of magnitude, there does not seem to

be a large distinction between the A and B boxes. This is particularly true for the first case where the ICNC values are quite similar for both boxes. For the second case, there does indeed seem to be a distinction between the boxes, however the INPC values still seem much higher than both boxes. These values would benefit from some clarification, and perhaps this could come in the results section if it were restructured as recommended above.
Response: Thank you for your valuable comments. According to the reviewer's suggestion as well as the comments from reviewer #2, we have made significant modifications including merging Part A and Part B in each case, considering that the two parts merely reflected differences in retrieval uncertainty without substantial distinctions. The revised ICNC-INPC comparative results and analysis will be presented based on these modifications.

Minor points:

The title could state the outcome of the study since an effect has been found by this study. For example, "Long-range transported Asian dust plumes influence cirrus formation over remote ocean."
Response: We have changed the title a bit to **'Long-range transported Asian dust plumes influence cirrus cloud formation over remote ocean: Case studies near Midway Island, Pacific'.**

The abstract has a nice structure to it, but the sentence beginning "However, it is still not well…" (line 14) is confusing. This is a key sentence and rephrasing it would make the motivation of the research clear in the abstract. The results could also be stated more clearly, for example including the ICNC - INPC ratio.
Response: We have rephrased the entire abstract in the revised manuscript.

The comment on line 47 about geoengineering comes across as quite random and a bit of an afterthought. Perhaps it could be rephrased to tie in with the previous sentence about the uncertainty and how understanding of these clouds needs to be improved in order for cloud seeding to be done appropriately.
Response: Thank you for pointing out this. We have been requested to shorten the introduction by another reviewer so as to quickly enter into the main topics of this study. After consideration, we decide to remove the statements related to geoengineering and cloud seeding.

The introduction has good content to give context to and justify the study. It does very well to lay out why we care about cirrus and this long-range dust transport. However, a couple of paragraphs here could be restructured to improve the logic:
Response: We have made some revisions according to the reviewer's suggestions.

The paragraph beginning at line 50 could be split into first discussing the pristine environment, low AOD, and the related lack of observations of cirrus. It is also a bit unclear whether there are not many cirrus or there are many but it is unexpected because of the pristine environment. Then the question of whether this means cirrus are purely formed by homogeneous could be framed.
Response: We have revised the manuscript to more clearly state that there are many (occurrence of 0.3-0.4) but it is unexpected because of the pristine environment. (please see lines 58-60)

The ice saturation conditions for both mechanisms are a key part of the background science and this could be written in a separate paragraph. This could then link dust as an INP and give a description of why homogeneous freezing is much less likely to occur if dust is present (high ice saturation is prevented).
Response: We are grateful for the reviewer's suggestions about the organization of this paragraph. We have separated the different requirements of ice saturation conditions for heterogenous and homogeneous ice nucleation as a single paragraph (now the second paragraph of section 1), and have also made some adjustments. (please see lines 45-48)

As for the second issue, we consider, if possible, detailed physics such as '(high ice saturation is prevented in the presence of dust' would be better to provide in the results and discussions.

The paragraph beginning at line 67 could also be restructured. From the studies cited here, I have understanded that we know Saharan dust travels long distance, we know that Asian dust affects cirrus in China and also travels long distance to North America. So it seems to me that the question is not really whether the dust affects cirrus a long distance away, but rather is this long-distance transport creating a significant effect in an otherwise pristine environment? This paragraph could lead the reader more to this question about what is happening over the ocean.

Response: Thank you for the suggestion of the organization of this paragraph. Please also see the second sentence of this paragraph which is read as '*Hence, the possible influence of transoceanic dust particles on cirrus cloud formation should be considered.*' As requested by the reviewer, this sentence has raised a question about connecting the long-range transport dust to cirrus clouds. Afterward, we begin to introduce several very relevant studies regarding the influence of transoceanic dust on cirrus formation. Then, the topic of dust-cirrus interaction at the halfway of transpacific transport is raised at the end of this paragraph. Thus, we consider it would have already followed the reviewer's logic to structure the paragraph.

The paragraph beginning at line 83 could start with stating that He et al (2022b) determined the freezing mechanism by comparing INPC and ICNC before explaining why this works. Some clarification explaining how one knows that a mechanism is "dominant", rather than there just being a mixture of both, would help the reader. There is good placement of the descriptions of the products here and nice summary with referencing.

Response: For this method, we can only consider the possibility of '(1) sole heterogeneous nucleation' or '(2) homogeneous nucleation also involves (i.e., it can be considered as the mixture situation as the reviewer mentioned)'. This means one cannot judge whether there is a possibility for sole homogeneous nucleation. Because the actual cloud physical process is much more complicated. For (1), it is easy to determine. However, for (2), it may be more complicated. If dust INPs are provided, heterogenous nucleation can take place first, and then, along with the depletion of INP homogeneous nucleation may begin to be involved. Or, if a very strong updraft is present, homogeneous nucleation can rapidly take place regardless of a sufficient supply of dust INP, because there is not enough time for triggering heterogeneous nucleation. Considering the complexity, we would like to avoid discussing all of these details in the introduction and hope that the reviewer can understand.

The point about seeder feeder in stratocumulus clouds (line 80) could be made earlier because it is addressing the big question of why do we care about these cirrus? Perhaps it could be stated near the geoengineering comment as wider motivation for understanding cirrus.

Response: As requested by another reviewer, we have to shorten the introduction part. The statements regarding the geoengineering and seeder-feeder process have been removed now.

On line 82, there is the phrase "it is indispensable" but it is unclear what it is. I am not sure if this line adds anything.

Response: We have removed this sentence.

The overview of the paper (line 103) could be clearer and include "section 3".

Response: We have revised.

The manuscript tends to state what the figure is in the text, e.g. "Figure 5 shows the ice cloud properties, including cloud extinction coefficient, cloud particle effective radius, ice water content, 200 and ice crystal (with size <5 μm)....". They could consider removing this from the main text since it is already described in the figure caption. This may make the story flow better as the main text can then go straight into what is observed in the figure, e.g. "Figure 5b shows the region of dust-related cirrus... ".

Response: We have made the necessary adjustments in the manuscript to avoid this issue.

The introduction to the results sectioned could be cut as there is some repetition that one INP is generating one ice crystal and that secondary ice productions is not being considered to take place in these conditions. But the statement of what is required for "good" agreement and the definition of dust-cirrus interaction event are well placed here.

Response: We have made corresponding revisions for clarity. (Please see lines 180-182)

The authors might consider adding an explanation for why the description of the dust in the main text based on figure 3a and b (line 184) does not fit with where the dust is identified in figure 3c and e.

Response: In figures 3a, b, and d, we mark the same region to write the words beside 'Dust-related cirrus'. For figure 3c, since it is the vertical feature mask that can identify aerosol and cloud, we use a different rectangle to include the whole region of conterminous cirrus and dust and write the words beside 'dust-cirrus interaction'. For figure 3e, only aerosol subtypes are shown; thus, we just mark the region of dust aerosols and write the words beside 'Dust INP'. All in all, these different rectangles just aim to roughly indicate the regions where dust-cirrus interaction takes place rather than to show the specific regions employed for further quantitative study/analysis. The specific regions of cirrus and dust for analysis are given later in figure 6 as well as in Table 2. We have added a sentence to remind this in the first paragraph of section 3.1. (Please see lines 193-195)

In the main text about figure 5, the in-cloud averages are stated but there is no interpretation of these. In the introduction it is stated that homogeneous freezing produces more, smaller ice crystals. Do the in-box averages support the allocation of homogeneous/heterogeneous freezing based on the radius and ice water content?
Response: Indeed, extinction coefficient, effective radius, and ice water content can be considered the input parameters for calculating the ICNC in the DARDAR Nice product. However, it is difficult to just use these parameters to discuss which type of ice nucleation mechanism should take responsibility for the ice formation. Therefore, we only intend to provide all the available information in this case study. Detailed discussion regarding the ice nucleation mechanism will be left to the ICNC-INPC comparison part.

Some clarification on line 236 about "pristine ice crystals" in abundant INP near the top of the cloud could help the reader. Are these pristine ice crystals because they are from homogeneous freezing or because they are formed in pristine environments, where the INP are more numerous at cloud top? Perhaps changing the description of the INP from abundant to more numerous would help.
Response: The 'pristine' just means 'initial-formed' ice crystals formed near the cloud top regardless of the type of ice nucleation (usually considered within the range of 350 m below the cloud top, Bühl et al., 2016) and without further physical process, e.g., significant growth, aggregation, collision and so on. We have made extra explanations in the text. (please see lines 248-249)
Reference:
Bühl, J., Seifert, P., Myagkov, A., and Ansmann, A.: Measuring ice- and liquid-water properties in mixed-phase cloud layers at the Leipzig Cloudnet station, Atmos. Chem. Phys., 16, 10609–10620, https://doi.org/10.5194/acp-16-10609-2016, 2016.

In the discussions of figure 6e (and for 10e), it might not be clear to the reader how the calculations of INPC at different ice saturation ratios relate to the ICNC values. Does each saturation ratio relate to a different radius? A statement of which INPC and ICNC values are actually being compared for both part A and part B would make it clear to readers like me that are unfamiliar with the POLIPHON method.
Response: The saturation ratios (S_i) are endowed with 4 different values to simulate various degrees of heterogeneous nucleation, which allows us to infer the corresponding possible INPC values using parameterization U17-D. Note that part A and part B have been combined as a whole. A detailed explanation of computation and comparison has been added to the updated manuscript. Also, as suggested by reviewer #2, the Si used for pure dust (first case) and polluted/aged dust (second case) should be different, and the INPC for the polluted dust case is multiplied by a factor of 0.1 due to the weaker ice-nucleating efficiency of dust particles that have undergone the coating/aging process. Thus, we would like the reviewer to reevaluate the updated results and the related discussions. (please see lines 252-262, 309-324)

For part B starting at line 244, the message would be clearer if the comparison with the INPC came before putting it in the context of Ansmann et al (2019a), Cziczo et al (2013) and the interpretation of the dust acting as INP in the moist region. Additionally, if this is the moist region shown by the RH in figure 6d, this could be linked with a "as shown in figure 6d".
Response: We have made the necessary adjustments in the main text.

In the paragraph beginning on line 346 with "The overview of …. ", the authors might consider moving the sentences about the dust being uneven and the lack of INP resulting in homogeneous freezing near the start of the paragraph. They could then summarise the findings from part A and B for each case.
Response: As suggested by reviewer #2, homogeneous nucleation is rather unlikely as the presence of so many rather favorite dust INP particles as shown in figures 6 and 10, and the evidence for the depletion of INP is weak. Agreeing with this perspective, we have removed all the related argumentations regarding

homogeneous nucleation.

Related to the above point, a comment on why part B is shown to have lacking INP but is closest to the region of dust is perhaps warranted.

Response: As mentioned above, we have made major revisions to both cases by combining part A and part B as a whole as strongly suggested by reviewer #2. Now the results and discussions in section 3 have been significantly modified.

Line 375 states that cirrus would not form without these natural INP, but this conflicts with the establishment that homogeneous freezing does take appear to take place. Perhaps change to something similar to “would rarely form” or “there would be a much lower frequency of clouds”.

Response: Thank you for pointing out this. Also, according to the comments from reviewer #3, we have modified the related expression. (please see lines 323-324)

The authors have done well to suggest plenty of future work based on this study.

Response: Thank you for your encouraging comments.

Technical points

There is some inconsistency between using the terms secondary ice nucleation and secondary ice production in lines 84 and 174.

Response: We have adopted "secondary ice nucleation" consistently throughout the revised manuscript.

Blue and purple colours in the profile plots (figures 6e and 10e) are quite hard to distinguish. These could be more contrasting colours.

Response: Considering that Part A and Part B are combined in each case as mentioned in previous responses, Figures 6e and 10e have been updated in the revised manuscript.

In figure captions, the authors could consider separating out figure titles from the a), b) descriptions. For example, the figure 2 caption could be “Dust-related conversion…” and then a) and b) to describe each plot. Also in figure 6 and 10.

Response: The related modifications have been done for the captions of figures 2, 6, and 10.

On line 164, what does a “complete system” mean here?

Response: The phrase “complete system” refers to the comprehensive functions of the HYSPLIT method, including analysis for air parcel trajectories as well as simulations involving transport, dispersion, chemical transformation, and deposition. A more detailed description of the HYSPLIT model has been added to the main text. (please see lines 171-173)

Some typos:

The abstract contains some use of the word “dominated” rather than dominant. This might be present in other places too.

Paragraph starting at line 164 contains several cases of HYSPIT instead of HYSPLIT.

Line 63, what is the occurrence rate of cirrus measured in?

Line 73: “Saseen et al 2003” should be Sassen

Line 75: should “Sassen et al 2001” be 2002?

Line 119: “MEERA” should be MERRA

Line 169: “stimulated” → simulated

Response: Thank you for pointing out the issue with the typos. For line 63, the occurrence rate of cirrus is measured in units of 0.3-0.4, i.e., 30%-40%. In response to all the mentioned issues, the necessary corrections have been made in the updated manuscript.