

Response to Reviewer #1

General Comments:

This is an interesting study that relates mineral dust plumes from Asian deserts to the number concentration of ice particles in cirrus clouds over Midway Island in the Central Pacific. It does this strictly through satellite remote sensing, using retrieval methods for mineral dust concentration (i.e., ice nucleating particle concentration or INPC) and ice particle number concentration (ICNC). It is well written and organized, but some of the arguments do not appear to be supported by the data, and the results may be overinterpreted. Specifics are given below. I recommend major revisions in rewriting the article, but this may not require a great deal of additional work.

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 $n_{ice,25\mu m}$ and $n_{ice,100\mu m}$. 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.

Specific Comments:

Comments 1: Figure 4: The high column dust density measurements shown here in the back-trajectory do not ensure that air overlying these Asian deserts at ~ 10 km is having a relatively high concentration of mineral dust, since the column dust magnitude could result almost entirely from dust much below 10 km. This needs to be stated. On the other hand, it is commendable that the authors did this analysis since, although incomplete, it may be using all the available data and does provide important information.

Response 1: According to the suggestion, we have added the statements to mention that the MERRA-2 column dust density may be mainly contributed by the relatively high concentration of mineral dust at relatively lower levels. (Please see lines 197-198) Nevertheless, we just would like to employ the MERRA-2 column dust density to examine the rough transport pathway and possible original region of the dust plumes from a large-scale view.

Comments 2: Figure 5: There appears to be a problem with color legend. The plotting background is violet, which corresponds to an ice water content of 10-20 mg/m³. Not possible. The same problem occurs with Fig. 9c.

Response 2: Thank you for pointing out this. We have updated the color bars for Figures 5c and 9c.

Comments 3: Lines 234-235: For what purpose are ICNCs shown for $n_{ice,25\mu m}$ and $n_{ice,100\mu m}$? They provide

no closure information with respect to dust INP concentration since most of the ICNC can be associated with $D < 25 \mu\text{m}$ (Kramer et al., 2009, ACP). Only $n_{\text{ice},5\mu\text{m}}$ is relevant to the closure being sought with INPC.

Response 3: Thanks for the reviewer's comment. As suggested by reviewer #2, it is better to retain all of the information on $n_{\text{ice},5\mu\text{m}}$, $n_{\text{ice},25\mu\text{m}}$, and $n_{\text{ice},100\mu\text{m}}$, not only for the closure with respect to dust INPC but also for 'getting a better feeling for the large uncertainties in all ICNC products (as commented by reviewer #2)'.

Comments 4: Lines 236-237: Suggest citing Diao et al. (2015, JGR) to back up this statement.

Response 4: We have added it to the revised manuscript.

Comments 5: Lines 252-253: Figure 6e shows agreement between ICNC and INPC only near cloud top for $S_i = 1.15$, where ICNC is for $n_{\text{ice},5\mu\text{m}}$. From the previous page, it states that the layer average INPC is 7 L^{-1} and 96 L^{-1} for S_i of 1.15 and 1.25, respectively. Since the layer average ICNC is 111 L^{-1} for Part B, optimal agreement is for $S_i = 1.25$, with ICNC-to-INPC ratio of 1.16. Therefore, S_i here should be 1.25, not 1.15.

Response 5: We have modified the way we delineate our study regions by combining both Part A and Part B as a whole. As a result, the ICNC-to-INPC ratios have been updated. Now the results part has been largely modified.

Comments 6: Line 253: It is not clear how the ICNC-to-INPC ratio can be 0.9 for $S_i = 1.15$, based on the previous text. As noted above, this ratio appears to be 1.16 for $S_i = 1.25$, for which closure is optimal.

Response 6: As explained in Response 5, we have made the necessary modifications after combining Part A and Part B as a whole in the revised manuscript. Thus, we would like the reviewer can reevaluate the analysis.

Comments 7: Lines 317-320: Since a large portion of the ICNC resides at $D < 25 \mu\text{m}$, $n_{\text{ice},5\mu\text{m}}$ should be used rather than $n_{\text{ice},25\mu\text{m}}$ (as assumed in this study). This practice (of using $n_{\text{ice},5\mu\text{m}}$) was followed for the other May 5th case study. In Fig. 10e, $S_i = 1.25$ agrees best with $n_{\text{ice},5\mu\text{m}}$, consistent with the previous case study based on Comment 5 above.

Response 7: As previously mentioned in Response 5, we have revised the specified study areas. Also in both cases, the ice virgae exceed 2 km, which indicates that they have to be formed at least half an hour before (reasonably assuming a falling velocity of 1 m/s). In this situation, the pristine ice crystals nucleated at the cloud top should have already undergone significant growth. Therefore, to discuss these cases with an open mind, we would like to mainly use $n_{\text{ice},25\mu\text{m}}$. We have carefully discussed all these possibilities in the revised manuscript.

Comments 8: Lines 308-312: If $S_i = 1.25$ in the 2nd case study as indicated above, then the cirrus cloud of Part A could be produced only by heterogeneous ice nucleation since $n_{\text{ice},5\mu\text{m}} = 635 \text{ L}^{-1}$ and the mean dust-related INPC (U17-D) is 417 L^{-1} . This is contrary to what the article states here; that Part A is dominated by homogeneous ice nucleation.

Response 8: 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. Therefore, the specific discussions have been largely updated and we hope that the reviewer can reevaluate our analysis.

Comments 9: Lines 350-352: Alternatively, could this also be explained by variability in cloud updraft velocities?

Response 9: We appreciate your emphasis on this valuable perspective. Variability in cloud updraft velocities has the potential to influence the generation and distribution of ice crystals within clouds. In further detail, small-scale vertical velocity perturbations can impact both the generation of ice crystals and the levels of ice supersaturation (Diao et al., 2015). Moreover, as demonstrated by Spichtinger and Gierens (2009), variations in updraft velocities within clouds can influence the competition between homogeneous and heterogeneous

nucleation. More enhanced updraft velocities can intensify homogeneous nucleation, leading to a greater number of small ice crystals; while ice crystals produced by heterogeneous nucleation take a less important portion in these regions. In the first case, the uneven distribution of $n_{ice,5\mu m}$ should be attributed to the uncertainty in ICNC as suggested by reviewer #2. We have incorporated these insights by making revisions in the updated manuscript.

References:

Diao, M., Jensen, J. B., Pan, L. L., Homeyer, C. R., Homomichl, S., Bresch, J. F., and Bansemer, A.: Distributions of ice supersaturation and ice crystals from airborne observations in relation to upper tropospheric dynamical boundaries, *J. Geophys. Res.-Atmos.*, 120, 5101–5121, <https://doi.org/10.1002/2015JD023139>, 2015.

Spichtinger, P., and Gierens, K. M.: Modelling of cirrus clouds–Part 2: Competition of different nucleation mechanisms, *Atmos. Chem. Phys.*, 9, 2319–2334, <https://doi.org/10.5194/acp-9-2319-2009>, 2009.

Comments 10: Table 2: As mentioned earlier, for what purpose are ICNCs shown for $n_{ice,25\mu m}$ and $n_{ice,100\mu m}$? They provide no closure information regarding dust INPC.

Response 10: As previously mentioned in Response 5, we have decided to keep $n_{ice,25\mu m}$ and $n_{ice,100\mu m}$ based on our updated stratification for Case 1 and Case 2. This choice enables us to make more dependable comparisons between ICNC and INPC, especially when faced with considerable uncertainties in ICNC.

Comments 11: Lines 375–377: RH_i (relative humidity with respect to ice) rarely reaches 140–150% in cirrus clouds since het (heterogeneous ice nucleation) always occurs before hom (homogeneous freezing nucleation), and INP and/or pre-existing ice tend to prevent the RH_i from reaching the RH_i threshold for hom. But if INP concentrations were low enough, the RH_i threshold for hom would occur much more often to produce hom cirrus clouds, and their coverage could even exceed the coverage of het cirrus due to the smaller ice crystal sizes having lower fall speeds, as demonstrated in Mitchell et al. (2008, GRL). That is, lower ice sedimentation rates lead to longer cirrus lifetimes and greater cloud coverage. Moreover, the citation of Dekoutsidis et al. is misguided since that paper was showing hom is common in cirrus clouds and occurs mostly near cloud top where RH_i is greatest, consistent with the modeling study by Spichtinger and Geirens (2009, ACP). The reference by Cziczo et al. does not support the author’s claim either; rather it argues that most cirrus are het cirrus.

Response 11: Thanks for the constructive note suggested by the reviewer. As sufficient INPs are provided in this case, the situation in Mitchell et al. (2008) may not take place. According to the reviewer’s suggestion, we have also removed the citations of Dekoutsidis et al. (2023) and Cziczo et al. (2013). Thus, we have fully rewritten this sentence as ‘...**Apart from homogeneous ice nucleation, the supply of efficient INPs contributes to an additional opportunity for the formation of cirrus clouds over open ocean regions; ...**’ (Please see lines 369–371)

Comments 12: Lines 377–380: While changes in UT INPC may alter the microphysical properties of cirrus clouds, this may not result in an increase in cloud cover and associated albedo for the reasons stated above. Moreover, the net radiative effect of cirrus clouds considers the absorption/emission of LW radiation in addition to SW radiation, and whether a net cooling or warming effect occurs may depend primarily on cloud optical thickness, the season, and the latitude.

Response 12: According to the reviewer’s comments, we have rephrased the sentence as below ‘**the microphysical properties of cirrus clouds are modulated by differentiating ice-nucleating regimes, which may contribute different net radiation to the global climate.**’ (Please see lines 371–372)