Response to Reviewer #2

General Comments:

The manuscript discusses two interesting dust-influenced cirrus events over the Pacific Ocean in the outflow regime of Eastern Asia. The remote-sending-based study makes use of spaceborne lidar and radar observations. The work is original and worthwhile to be published in ACP. The main effort is related to closure studies, i.e., deals with the question to what extent the ice-nucleation particle concentration (INPC, estimated from lidar observations) is in the same range of estimated ice crystal number concentration (ICNC, estimated from combined lidar-radar observations). Based on these ICNC-vs-INPC closure studies the authors also discuss to what extent homogeneous ice nucleation was involved in the (in situ) cirrus evolution.

Many aspects are unclear, a lot of speculative argumentation is given in the main result sections 3 and 4. A more careful discussion is requested. The uncertainty in all the retrieval products needs to be better considered. Therefore, major revisions are required.

One of the key points for me is: All the retrieval products (INPC, ICNC) can only be obtained within a large uncertainty range of an order of magnitude. ICNC cannot be obtained with an uncertainty of 25% as mentioned in the present study! Impossible! Even if you have well calibrated lidar and well calibrated cloud radar observations, and, in addition, Doppler information about fall speed of ice crystals (and thus shape and size information) the uncertainty can never be lower than expressed by a factor of 3-5 around the retrieval products. This is, e.g., shown in the reference, you provide (Ansmann et al., ACP, 2019). This is also discussed in detail by Buehl et al., AMT, 2019. Now, in the case of the CloudSAT radar we do not have Doppler information. So, the ICNC uncertainty is even higher. All in all, the uncertainty margins are roughly an order of magnitude for INPC and ICNC. That needs to be considered in the closure studies presented here and in all the conclusions drawn from the observations. To my opinion, this will make the full discussion easier and more straight forward.

Response: We appreciate the reviewer's thoughtful review and constructive comments. We have carefully checked the uncertainties in ICNC and changed the statement of '25% uncertainty in ICNC'. Accordingly, many revisions have been made in sections 3 and 4. All the comments have been addressed in the revised manuscript, and the responses to each comment are given below. We consider that this revised manuscript has been largely improved. 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.

Detailed Comments:

Title: This study is not a measurement report. This is an in-depth research approach (cirrus closure study) based on the analysis of spaceborne observations. The term 'Measurement report' suggests that just robust observations (measured data) are presented. But the study is widely based on estimated products and interpretation of the findings.

Response: 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.'

Abstract: The abstract needs to be rewritten after all necessary changes.

Response: We have modified the abstract according to the revisions in the text.

Introduction:

I would keep the introduction as short as possible. There are so many cirrus papers and all present lengthy paragraphs on the radiative impact. I would suggest that the importance of cirrus in the atmospheric system is just briefly described, followed by gaps in our knowledge regarding cirrus evolution, and then what you are going to present in this study.

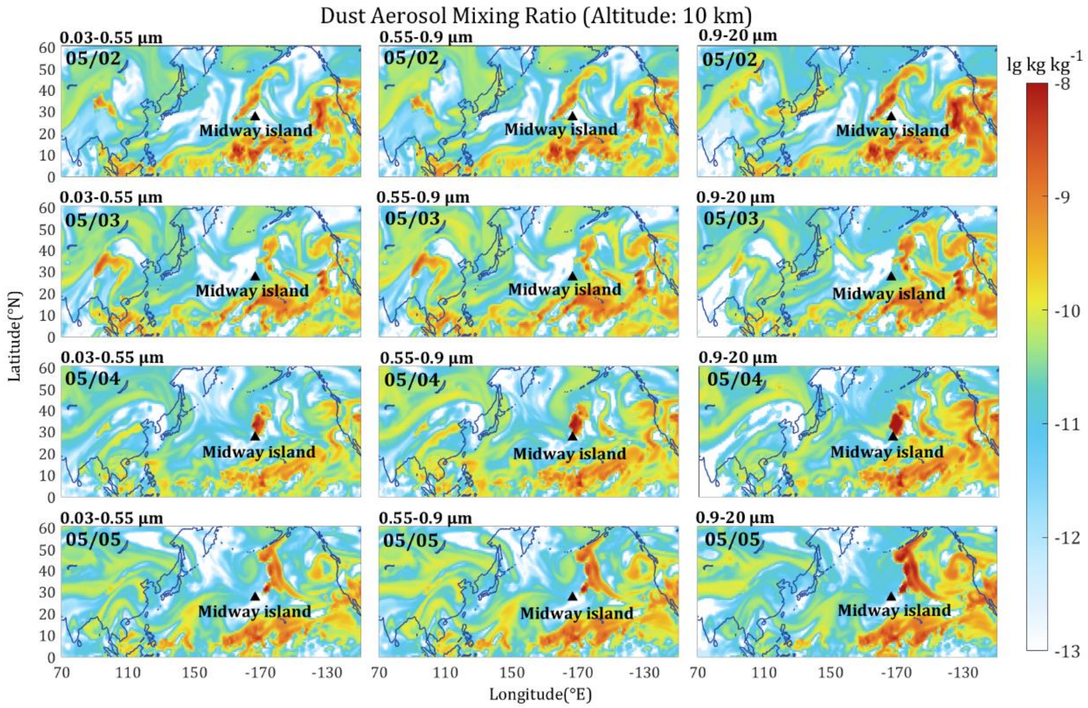
Response: Considering the reviewer's suggestion, we have shortened the start part of the introduction so that we can quickly enter into the research gap in the field and our research topic. The detailed contents regarding the radiative impact have been removed. In the revised manuscript, the first and second (just the beginning part) paragraphs have been rewritten. (please see lines 35-52)

Lines 41-42: Note that homogeneous freezing also needs aerosol particles, however pure liquid ones, such as sulfate aerosol (without any insoluble part). In the case of heterogeneous ice nucleation, one needs particles with an insoluble fraction (sites for ice nucleation).

Response: We have replaced '(spontaneous)' with '(e.g., sulfate aerosol solution)'.

Figure1: Does MERRA also deliver dust profiles? or only column mass values? Would be nice to have model dust profiles up to cirrus level.

Response: Thank you for the suggestion. The MERRA-2 data only provide column dust mass density. For dust situation at cirrus levels, here we utilize CAMS global reanalysis (EAC4) data to plot the daily-mean size-resolved dust aerosol mixing ratios on 2-5 May 2010 (as shown in the figure below). However, it does not trace the transport pathway of the dust plumes. Therefore, if possible, we would like to retain the use of MERRA-2 column dust properties to indicate that Asian dust may indeed be transported to the central of (or even across) the Pacific. In the case studies later, we will show the height distributions of the dust layers with CALIOP observations.



Method section:

Line 130: 25% uncertainty is unrealistic as discussed above. To use uncertainty margins of an order of magnitude (a factor of 3 around the retrieved value) makes sense. That is more realistic!

Response: Thank you very much for pointing out this issue. We have modified the uncertainty in ICNC to be a factor of 3 (Sourdeval et al., 2018) in section 2.2 as well as the rest part of the manuscript. (Please see lines 125-137)

Lines 140-145: I would remove all immersion freezing parameterizations. The two case studies deal with cirrus from 9 to 11.2 km height, and temperatures from -40 to -54°C. Furthermore, your Table 2 considers deposition ice nucleation, only (no immersion freezing INP values are given). Even in virga zones with higher temperatures, there is ice saturation or even ice sub saturation (and especially sub saturation with respect to water). Immersion freezing is impossible in the presence of in situ cirrus with well-developed virga structures.

Response: We have removed the immersion freezing from the text as well as from figures 6 and 10. Now the statements here are taken as '**Considering the temperatures at cirrus levels, we only considered deposition freezing in cirrus clouds (Kanji et al., 2017; Marcolli, 2014) by applying the parameterization scheme U17-D (Ullrich et al., 2017) for dust-related INPC computation. Here we disregarded contact freezing as well as immersion freezing, which needs an INP to collide with/immerse in a supercooled droplet.**' (please see lines 148-151)

Table 1: Are all the equations needed? A list of parameters and retrieval uncertainties makes sense. Again, please skip all immersion freezing parameterizations.

Response: We have removed the immersion freezing parameterizations in Table 1. As for the rest equations listed, if possible, we would like to retain them so that the readers without the background of the POLIPHON method can easily follow the work.

Result section:

The cirrus shown in Figure 3 is a classical in-situ cirrus, with a well-defined cirrus top region, obviously above 10 km height, and a virga zone from 10 km down to 6 km height. The cirrus is clearly dust influenced.

Let me explain how such cirrus layers develop: Ice nucleation starts at cloud top, at the coldest point of the cloud, here the ice nucleation probability is highest. And if dust particles are present, they will trigger ice

nucleation. Then diffusional growth of the crystals takes place, collision and aggregation. Sedimentation of ice crystals begins and the growth of the crystals leads to an immediate reduction in relative humidity (throughout the cirrus layer from 6 to 10.5 km height). There is almost no room for homogeneous ice nucleation (in the case of a well-developed cirrus system) because there is practically no potential to create a scenario with sufficiently high relative humidity over ice with values exceeding 150%, required for homogeneous ice nucleation. Homogeneous freezing is only possible at cloud top in the case of rather strong updrafts with large updraft speed so that super saturations levels develop faster than INPs can be activated (to reduce super saturation). All this is described in Kaercher et al., JGR, 2022. However, such a scenario with rather strong updrafts is not visible in Figure 3. A very harmonic cirrus development exclusively controlled by the activation of dust INPs seems to be realistic. Furthermore, radiative cooling at cloud top of a well-developed cirrus system will cause some amount of downward motion and may contribute to a suppression of such rather strong updrafts. If we keep an uncertainty of about an order of magnitude into account for ICNC and INPC, all observations support that heterogeneous ice nucleation on dust particles dominated. Because of the large uncertainty, I would not further analyze the defined observational periods (part A and part B), i.e., explain the differences in the ICNC values in terms of heterogeneous vs homogeneous ice nucleation. The differences in the results for part A and B just show, to my opinion, the uncertainty in the products.
Response: We are grateful for the reviewer's valuable comments and agree with the opinion that the possibility for homogeneous nucleation in this cirrus is rather small. Thus, we have revised the analysis area of the cirrus cloud by combining Part A and Part B in each case and explain the high ICNC values by the updated uncertainty in ICNC (a factor of 3 as also mentioned above). The subsequent calculations and analysis are all based on this update.

Back to the study:
Figure 3: The lidar is able to see the small ice crystals after nucleation with sizes of 1-5 micrometers at cloud top, above 10 km. The cloud radar seems to be not able to detect the ice crystals above 10 km height (Figure 5). The radar detects the ice crystals after diffusional growth and collision and aggregation processes, and after the start of sedimentation processes, i.e., several 100 m below cloud top, in the virga zone, as can be seen in Figure 5a and 5c. So, lidar-radar retrievals will not be able to exactly see the ice crystal number concentration of freshly nucleated ice crystals at cirrus top. And when radar comes into plays, aggregation took already place, and the number of crystals already decreased, may be already by a factor of 2, or even a factor of 3-5. Another point: Can we assume a 'classical' size distribution (as typically measured with aircraft instrumentation) in the case of freshly formed ice crystals? The size distribution is input in the DARDAR approach, and may be very narrow for the freshly nucleated crystals? All these unknown aspects cause the large uncertainty in the ICNC products (of at least one order of magnitude).
Response: Thank you for your valuable comments. Indeed, lidar is more sensitive to the small particles near the cloud top and radar is likely to be sensitive to the large ice particles that have already fallen for a while with the probable experience of collision and aggregation processes. For the DARDAR Nice data product, Sourdeval et al. (2018) mentioned that it is difficult to quantify the overall uncertainties in ICNC caused by instrumental sensitivity and physical assumptions (e.g., parameterization scheme for the scattering properties of ice crystals, particle size distribution of ice crystals, and lidar ratio for ice crystal). They instead evaluated the quality of ICNC by comparing them with in situ measurement and concluded that the PDS assumption contributes to the dominant error in ICNC (as seen in section 4.2 therein). It should be mentioned that for in situ measurements of ICNC, the two-dimension stereo (2D-S) probe for particle sizes of 5-1280 μm can be associated with large uncertainties in the first two bins (5-25 μm) as suffering from uncertainties due to instrumental response time and depth of field (Gurganus and Lawson, 2018) as well as the shattering effect (Korolev et al., 2015). Overall, in Sourdeval et al. (2018) about a factor of 2 overestimation was stated as the uncertainty in $n_{ice,5um}$ and $n_{ice,25um}$ due to a misrepresentation of the PSD shape by Delanoë et al. (2005) at warm temperatures. In this study, we conservatively consider the uncertainties in $n_{ice,5um}$ and $n_{ice,25um}$ to be a factor of 3-5 for the selected cases with both lidar and radar available. As for $n_{ice,100um}$, assumed PSD shape by

Delanoë et al. (2005), in principle, performs best at reproducing the concentration in large size, and in situ measurements also show better accuracy at this size range. We have added the related sentences in the second paragraph of section 2.2. (Please see lines 125-137)

References:

Delanoë, J., Protat, A., Testud, J., Bouniol, D., Heymsfield, A. J., Bansemer, A., Brown, P. R. A., and Forbes, R. M.: Statistical properties of the normalized ice particle size distribution, J. Geophys. Res.-Atmos., 110, D10201, https://doi.org/10.1029/2004JD005405, 2005.

Gurganus, C. and Lawson, P.: Laboratory and Flight Tests of 2D Imaging Probes: Toward a Better Understanding of Instrument Performance and the Impact on Archived Data, J. Atmos. Ocean. Tech., 35, 1533–1553, https://doi.org/10.1175/JTECHD-17-0202.1, 2018.

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.

Korolev, A. and Field, P. R.: Assessment of the performance of the inter-arrival time algorithm to identify ice shattering artifacts in cloud particle probe measurements, Atmos. Meas. Tech., 8, 761–777, https://doi.org/10.5194/amt-8-761-2015, 2015.

The ICNC values in Figure 5d vary strongly and indicate the uncertainty in all the retrieval products. Therefore, I would not introduce part A and part B and 'believe' that the rather different findings are caused by different ice nucleation processes. One may formulate hypotheses…, but one needs to consider the large uncertainty in the discussion. Solid conclusions are difficult to draw. And as I mentioned, I am skeptical that homogeneous ice nucleation has a chance to occur in the presence of a well-developed cirrus. The CALIOP lidar indicates dust around the cirrus and no indication that the dust INP reservoir was depleted. To my opinion, a depletion of the INP reservoir is unlikely during part A and B, and therefore homogeneous ice nucleation is unlikely.

Response: Considering the reviewer's suggestion, we have combined Part A and Part B and analyzed them as a whole. We agree with the reviewer that it is probably a heterogeneous nucleation case since it is a well-developed cirrus and the INP supply seems rather sufficient. Therefore, we have added some related sentences in the second paragraph of section 3.1 accordingly. (please see lines 215-217)

If we keep the uncertainty of one order of magnitude in mind, the ICNC values for part A and B shown in Figure 6e nicely indicate the ICNC uncertainty range. To repeat, to my opinion, only heterogeneous ice nucleation makes sense. In the presence of so many rather favorite dust INP particles (as shown in Fig. 6c, 4000 large dust particles with a diameter > 500 nm per liter were present, and the corresponding dust surface area concentration was high with values up to 40 μm^2 cm^{-3}) homogeneous ice nucleation is rather unlikely.

Response: Considering the reviewer's suggestion, we have combined Part A and Part B and analyzed them as a whole. The related statements have been modified now. (Please see lines 228-233, 252-262)

Figure 6 indicates that INPC values for S-ice =1.15-1.25 (Ullrich parameterization, deposition ice nucleation) seem to be very likely (or reasonable) and match roughly the ICNC values (n-ice for crystals with sizes > 5 µm, for the periods of part A and B), when keeping in mind that ICNC is probably underestimated close to the cirrus top (because DARDAR values are not very trustworthy here because of too weak or even missing radar reflectivity values). The best DARDAR values are shown after significant growth of crystals and after aggregation processes (from 9-9.6 km height), but then the ICNC values are already reduced compared to the number of freshly nucleated crystals at cirrus top, probably reduced by a factor of 2-5.

Response: We are grateful for the comments that are rather helpful for explaining the data and analyzing the physical process. We have added some sentences to discuss this issue as below '**… It should be also mentioned that ICNC values near the cloud top may be more uncertain because of the weak or missing radar reflectivity, which is not sensitive to the small ice crystals here…. and then undergo aggregation process with a reduction of ICNC by a factor of 3-10 (Field and Heymsfield, 2003)….**' (please see lines

249-252)

Line 242: As mentioned above, homogeneous freezing in an environment with already existing ice crystals (and thus super saturation values S-ice around 1.0) is very unlikely. When keeping the high dust load and the large uncertainty margins into account, the closure is fine, ICNC and INPC match reasonably well. This is ok! This is a good result.

Response: We have rewritten the related sentences in this paragraph. (Please see lines 252-262)

In summary here, please, do not compare part A and part B, just take the average of both periods and use the averaged values for comparison with the Ullrich results for INPC. Ice super saturation values of 1.15 are close to the values in the paper of Ansmann et al. (2019) for pure (unpolluted) dust scenarios. In the case of aged dust (or polluted dust, case study 2 in this study here) super saturations of 1.35 make sense. Such values are assumed to be realistic for aged, coated dust particles (see Kaercher et al, JGR, 2022). Homogeneous ice nucleation events do not make sense to me at all. However, I leave it open to you to find a proper and careful argumentation for the potential contribution of homogenous ice nucleation.

Response: We have combined Part A and Part B and analyzed them as a whole. Here we find good agreement between U17-D-derived INPCs with Si of 1.25 and $n_{\mathrm{ice,5\mu m}}$. We have removed the discussions about the possibility of homogeneous nucleation here.

In the discussions (sections 3 and 4) there are many speculative aspects. Speculations are not justified. A more careful interpretation of the results is needed. And if you have a hypothesis, start with: We hypothesize…. and then the hypothesis must be based on convincing argumentation. And please keep the large uncertainties in mind.

Response: We have rephrased many sentences and made significant modifications to these two sections to avoid the hypothesis of homogeneous nucleation, especially considering that the INP supply here is rather sufficient.

Case study 2:

Another nice case with an impact of dust. In this case, CALIOP aerosol typing seem to indicated polluted or coated dust. The ice nucleation efficiency of aged and coated dust particles may be reduced by a factor of 5-10 compared to the ice activity for pure dust. This may or could be considered when computing INPC values with the Ullrich parameterization by multiplying the Ullrich INP values by 0.1-0.2.

Response: We have added a sentence to mention that the dust particles included in the dust layer are mainly aged/polluted. Thanks to the reviewer's comments, we have also updated the analysis/discussion part of this case by multiplying the U17-derived INPCs by a factor of 0.1. (Please see lines 275-276, 312-322)

Again, I would not compare the results for part A and part B because of the large uncertainties. The results in Figure 10e support my comment here. The results as a whole (part A and B) are in good agreement with the Ullrich INP values for S-ice of 1.35 when considering a factor of 0.1-0.2 less INPC (in the case of polluted or coated, less ice nucleation efficient dust). Kaercher et al., JGR, 2022, used S-ice values around 1.35 for the activation threshold for polluted dust. In case 2, the ICNC values (from the DARDAR approach) increase up to cloud top. Obviously, the radar reflectivity values were strong enough to obtain reasonable ICNC values even close to the nucleation range at cloud top.

Response: We have combined Part A and Part B and analyzed them as a whole. Considering the 'modified' U17-D-derived INPC values by multiplying a factor of 0.1, good agreement between INPC and $n_{\mathrm{ice,25\mu m}}$, (or $n_{\mathrm{ice,100\mu m}}$) can be seen. The related statements have been added. (Please see lines 319-323)

To my opinion, there is again no room for homogeneous ice nucleation. There is dust 'before' and 'after' the cloud region, so a depletion of the dust INP reservoir is not visible. And at these conditions, homogeneous ice nucleation is unlikely.

Response: In the last paragraph of section 3.2, we have stated that this case is probably dominated by heterogeneous nucleation. (Please see lines 323-324)

Please avoid speculations on cirrus type, etc…. in sections 3 and 4. Just mention, what is really available from the observations.

Response: We have removed the speculations on the cirrus type.

Discussion section:

The first paragraph is not needed to my opinion.

Response: We have removed the first paragraph of the discussion section.

Line 346: Can we have longitude-latitude information (not only latitude). How long (in km) was the cirrus layer? The same for case study 1.

Response: The longitude ranges have been added to the text. We have also added the horizontal extent of the cirrus clouds in these two cases in section 3.3. (Please see lines 339 and 345)

Be careful with 'dominating homogeneous freezing' in environments with so much dust. It is simply difficult to produce high ice super saturation in the presence of favorable INPs.

Response: We have removed the statements regarding the possibility of homogeneous nucleation.

If you follow my suggestions, you can significantly 'improve' Table 2 by reducing the information content. I think ICNC for n-ice (> 5 μm) has to be compared to INPC, however information on n-25, n-100 is useful as well, especially to get a better feeling for the large uncertainties in all ICNC products.

Response: We have combined the Part A and Part B and updated the Table 2. And $n_{ice,25\mu m}$ and $n_{ice,100\mu m}$ are retained in the table as suggested by the reviewer.

In the case of the Ullrich INPC values, input is dust surface area concentration, S-ice as well as temperature! Temperature needs to be mentioned in Table 2 (Ullrich INP values).

Response: Thanks for your reminder. Temperatures (mean value as well as the maximum and minimum) for each case have been added in Table 2.