Author responses regarding the second review from Reviewer 1

Format: The reviewers' comments are in black font while author responses are in red font. Text in red font italics indicates revised/added text in the revised manuscript.

We understand that reviewing this paper took a lot of time and effort, and we sincerely thank you for your comments that have improved this paper. Below are our responses to the general and specific comments:

Report by Referee #1:

While I find the paper to be improved, the authors have only marginally addressed most of my critical comments. However, since they did include a case study that is illustrative of the challenges they face with this algorithm, I recommend that the paper could be published with minor revisions if the editor deems appropriate. My fundamental criticism of the work can be summarized in my response to the authors noting how difficult it is to compare in situ data to a "layer" retrieval. I replied: That comparing "layer" retrievals with in situ data is difficult is exactly the point. If it is difficult to show validation, how can it be that the authors can develop a "layer" retrieval of Ni and IWC based on such in situ data? In the following are my responses to the authors' replies to my original comments:

Major Comment 1: My reply to theirs is as follows: In situ data collected over many campaigns show that middle latitude cirrus have a typical structure with high concentrations of small ice near the top and then depositional growth and aggregation towards the middle of the layer followed by a region of sublimation. Anvils have a slightly different vertical structure with size sorting and aggregation resulting in larger particles near the bases. Regardless, Ni and IWC vary strongly in the vertical implying that a "layer Ni" and "layer iwc" is not defined unless a region of the cloud where the observations are most heavily weighted is carefully defined.

After reading this second review from Reviewer 1, we now think that we have a deeper appreciation regarding this fundamental concern from this reviewer. This concern appears to pertain to the $X - \beta_{eff}$ relationships in Fig. 14, where X is N_i/A_{PSD} , N_i/IWC , or $1/Q_{abs}(12 \, \mu m)$. The reviewer correctly describes the typical vertical structure of Ni and IWC in cirrus clouds, especially geometrically thick cirrus. But there appears to be an implicit assumption that these $X - \beta_{eff}$ relationships depend on this vertical structure (i.e., the variation of X with height), but this is not the case. X, which is sampled from aircraft (i.e.,

calculated from the sampled PSD), can be sampled at any level in the cloud, and from this sampled PSD, β_{eff} is also calculated using the Modified Anomalous Diffraction Approximation (MADA) as described in Sect. 2.3 of M2018, and Eqns. (4) and (5) from M2018. When X = $1/Q_{abs}(12 \, \mu m)$, the same is true but now X is calculated more like β_{eff} is calculated. β_{eff} can be viewed as a radiative characterization or microphysical index of the PSD. Despite large environmental differences among samples, the X- β_{eff} relationships obtained are relatively tight (i.e., dispersion is not large). This enables them to be used whereby a given point on these X – β_{eff} relationships represents a cloud layer of arbitrary thickness where β_{eff} is related to the PSD. The retrieval then matches the β_{eff} from these in situ X – β_{eff} relationships with the IIR retrieved β_{eff} to obtain retrieved X. Since the IIR retrieved β_{eff} corresponds to the extinction-weighted PSD for the cloud layer, retrieved X corresponds to this extinction-weighted PSD.

A new second paragraph has been added to Sect. 3.2:

"Note that X, which is sampled from aircraft (i.e., calculated from the sampled PSD), can be sampled at any level in the cloud, and from this sampled PSD, $\beta_{\rm eff}$ is also calculated using the Modified Anomalous Diffraction Approximation (MADA) as described in Sect. 2.3 of M2018, and Eqns. (4) and (5) from M2018. When $X = 1/Q_{\rm abs}(12~\mu{\rm m})$, the same is true but now X is calculated more like $\beta_{\rm eff}$ is calculated. $\beta_{\rm eff}$ can be viewed as a radiative characterization or microphysical index of the PSD. Despite large environmental differences among samples, the X- $\beta_{\rm eff}$ relationships obtained are relatively tight (i.e., dispersion is not large). This enables them to be used whereby a given point on these X- $\beta_{\rm eff}$ relationships represents a cloud layer of arbitrary thickness where $\beta_{\rm eff}$ is related to the PSD. The retrieval then matches the $\beta_{\rm eff}$ from these in situ X- $\beta_{\rm eff}$ relationships with the IIR retrieved $\beta_{\rm eff}$ to obtain retrieved X. Since the IIR retrieved $\beta_{\rm eff}$ corresponds to the extinctionweighted PSD for the cloud layer, retrieved X corresponds to this extinction-weighted PSD."

Major Comment 2: My reply to theirs is a follows: I agree that the example at -80 in the tropical tropopause cirrus show no evidence of shattering because, I think, there are too few large particles to shatter. A single case study does not address my criticism however. What fraction of insitu data show an insignificant mode of likely shattered particles? For instance, I contend that even in fall streaks of mid-latitude cirrus where no small particles should be physically present still show a bimodal distribution with the small mode contributing significantly to Ni. The SPARTICUS 2DS data frequently show this artifact, albeit of much less amplitude than earlier campaigns, even though anti-shattering tips were used.

The first version of "Major Comment 2" asked "Can the authors explain the microphysical mechanism that would result in this (bimodal) behavior?" and "Can the authors point to in situ data that does not show this small mode?" We have responded to this question by providing an example PSD that does not show this small mode and that also shows strong evidence of homogeneous ice nucleation (henceforth hom), arguing that bimodal PSD may result when both heterogeneous ice nucleation (i.e., het) and hom are active. We cited Kärcher et al. (2025, npj Climate & Atmos. Sci., titled "Dissecting cirrus clouds: navigating effects of turbulence on homogeneous ice formation") for evidence that hom often occurs simultaneously with het, acting to broaden the PSD. Due to limitations in their modeling system, bimodal PSDs were not predicted in Kärcher et al., but the predicted PSD broadening by hom appears consistent with bimodal PSD behavior.

During ATTREX and POSIDON, both the 2DS and the FCDP were used, and they overlap between 5 and 45 microns. With the exception of the 1st size-bin (5 - 15 um) of the 2DS, agreement was generally very good in this overlap region (that should be affected by shattering if it was an issue). If random shattering was strongly biasing N(D) at these sizes, such agreement would be unlikely it seems. Note that the anomalous behavior of the 1st size bin may be unrelated to ice particle shattering based on conversations with Dr. Paul Lawson (who developed the 2DS probe along with others at SPEC, Inc.) and Gurganus and Lawson (2018, J. Atmos. & Oceanic Tech., titled "Laboratory and Flight Tests of 2D Imaging Probes: Toward a Better Understanding of Instrument Performance and the Impact on Archived Data").

Research on ice particle shattering might continue for years to come. However, it is worth pointing out that the ice PSD is a function of multiple physical processes including deposition growth, aggregation, sublimation, the Kelvin effect, and sedimentation. In Jensen et al. (2024, JGR, titled "The Impact of Gravity Waves on the Evolution of Tropical Anvil Cirrus Microphysical Properties"), it was found that "the combination of waves and the Kelvin effect drives growth of crystals with initial diameters of .3–10 µm to sizes of 20–30 µm." This may be one process contributing to bimodal PSDs.

The bimodality seen in fall streaks may be the result of two separate populations of particles produced through hom and het, respectively, falling at different mean velocities of course. The falling ice humidifies the adjacent atmosphere, allowing the smaller ice crystals to survive.

Major Comment 3: My reply to theirs is as follows: That comparing "layer" retrievals with in situ data is difficult is exactly the point. If it is difficult to show validation, how can it be that

the authors can develop a "layer" retrieval of Ni and IWC from such in situ data?

Also, there were several flight lines during SPARTICUS where the Lear Jet flew ramps profiling the cirrus layer. These ramps should reasonably allow the authors to quantify the vertical structure and compare "layer" quantities.

The rationale for developing a layer retrieval for Ni and IWC has been explained in our response to Major Comment 1 above.