Response to the Reviewers
Format: The reviewers' comments are quoted in italic
Section number in the response refers to the revised manuscript with tracked changes
Quotation in red stands for revised/added text in the revised manuscript

Overall comment:

We thank the reviewers for spending extra time providing more helpful comments. We provided two more time series in this response following the clarification of the reviewer and added one of them as the new supplemental Figure S8. In addition, we revised Figure 6 to include the entire X and Y axis range and temperature range as suggested by the reviewer. To be consistent with this update of Figure 6, we also updated Table S3, new supplemental Figures 9 – 12 that used linear regression to quantify aerosol influences. The nearly linear correlations between $d\log_{10}IWC$ and $d\log_{10}Na_{500}$ are consistently seen in the revised figures. The fact that correlations between $d\log_{10}IWC$ and $d\log_{10}Na_{500}$ are similar for various ranges of IWC as well as for in-cloud and clear-sky Na indicates minimal influences from ice shattering on the main conclusion.

Below are our individual responses to the reviewers' comments and the corresponding changes.

Response to Reviewer 2's comments:

Ref#2:

I have looked at the revision of Ngo et al. and my comments have mostly been addressed satisfactorily. However, the paper would be improved if these minor changes were made:

Lines 334-355: "with slight changes to Ni values" => with changes to Ni values

Revised.

Line 433: Mitchell and Garnier (2024) is cited but is not included under references. The reference is

Mitchell, D. L. and Garnier, A.: Advances in CALIPSO (IIR) cirrus cloud property retrievals. Part 2: Global estimates of the fraction of cirrus clouds affected by homogeneous ice nucleation, https://doi.org/10.5194/egusphere-2024-3814, 12 December 2024.

We thank the reviewer for these additional comments. We added the reference in the citation list mentioned above.

Ref#3:

Addressing Revisions

The authors have again provided much work attempting to validate their claim that large aerosol (D>500nm) measurements are not biased by small ice crystals, which are speculated to be intimately related to ice crystal shattering. This includes two additional time series, joint frequency occurrence plots and discussion of CDP particle concentrations with diameters less than 3 microns and sigma_Di (standard deviation of diameter). However, I remain unconvinced that large aerosol measurements are unbiased, or even minimally biased by the presence of ice crystals.

We thank the reviewer for spending extra time giving us additional feedback and helpful comments.

The authors revisit their original time series and claim "...we saw other spikes of Na500 at similar magnitude when IWC was much lower (by 1 order of magnitude) around UTC 25:38 or UTC 25:55 compared with the highest IWC at UTC 25:52." I don't see how this addresses the observed bias? I don't doubt that large aerosols will be occasionally observed at low IWC. We are speaking of a probabilistic bias rather than a deterministic one. Plus these alternative "spikes" are observably lower than the one at the highest IWC. The authors provided a new time series in response to my request: "...cases of level flight legs which have relatively similar aerosol concentrations within and right outside of the cloud – ideally where the cloud boundaries are observed at RHi>=~100." They provide the new time series and state "We chose another time series from NSF DC3 (Figure R2, below) to illustrate a relatively constant value of Na100 inside and outside of clouds as requested by the reviewer." This is not what I requested. The question is related to large aerosols, not those with diameters >100nm. Additionally, the large aerosol measurements for this and the following time series do not look correct. They are reporting quasi- steady values, whose minimal values appear to be a strong function of the level the aircraft is flying. Even assuming the data is correct, it is disappointing the authors have yet to provide at least one case where there are similar concentrations of large aerosols inside and outside of the cloud– ideally where the cloud boundaries are observed at RHi>=~100%.

Below we included two more time series from NSF DC3 to illustrate the type of time series that has relatively stable measurements of both Na_{100} and Na_{500}, RHi around ice saturation for in-cloud section, and relatively similar temperature range for the horizontal segment.

The first time series example is from NSF DC3 RF20 on June 21, 2012, added into supplemental as Figure S8. In this case, one can see much fewer Na_{500} samples with values > 0 compared with Na_{100} > 0 samples. The values of Na_{100} are higher inside the cirrus segment compared with the surrounding clear-sky samples, while the values of Na_{500} decrease at in-cloud condition between UTC 20:20 and 20:30. The decrease of Na_{100} values suggests that heterogeneous freezing may have activated some of the large aerosols as ice nucleating particles and formed ice crystals inside the cirrus segment.

Another time series example from NSF DC3 RF12 is shown in Figure R1. In this case, a similar decreasing trend of Na_{500} values for the in-cloud condition compared with the surrounding clear-sky condition is seen, while the Na_{100} values show increased values for the in-cloud conditions.

We would also like to mention the limitations of the field campaigns in this work to provide an ideal measurement of cirrus clouds that fits the description of the reviewer. Ideally, we would like to have cirrus clouds as the target for a field campaign, but most campaigns used in this study that had aerosol measurements were not targeting cirrus. The NASA ATTREX and POSIDON campaigns were cirrus-oriented but did not have aerosols measurements. In addition, the Na_{500} measurements have much fewer data points greater than 0 at the cirrus temperature range. Such imbalance of $Na_{500} > 0$ samples compared with other measurements also made it harder to find a perfect segment.

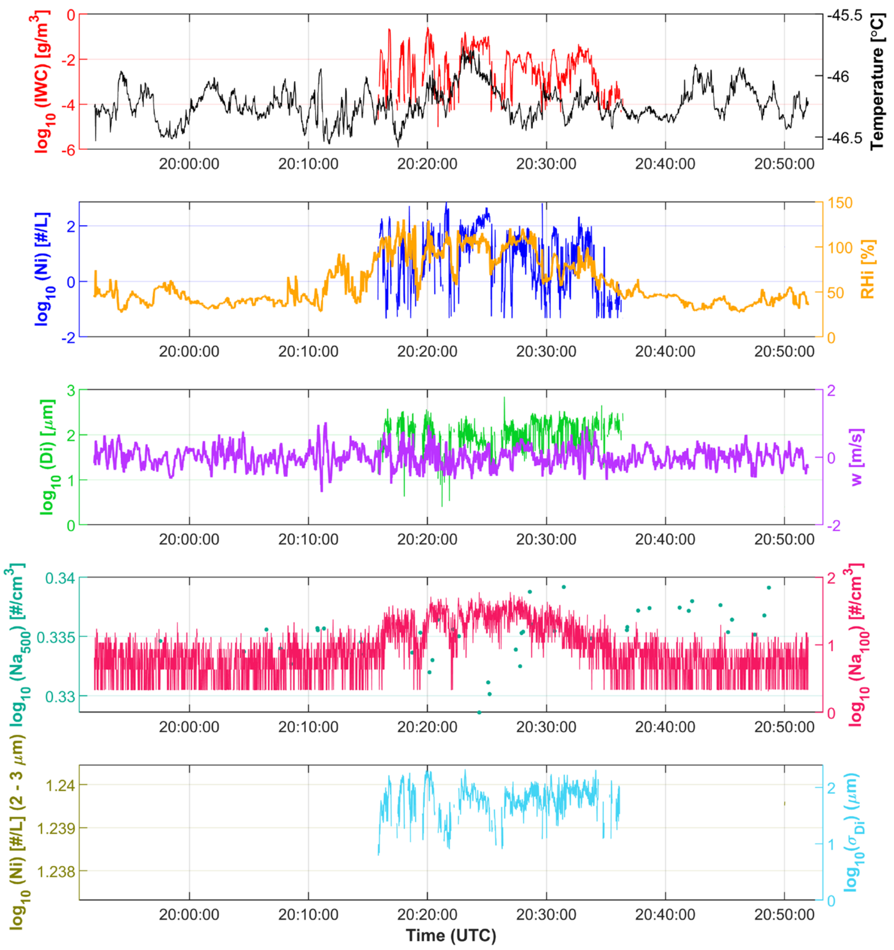

Figure S8. An example of time series from NSF DC3 RF20 on June 21, 2012. The Na_{500} values show a decreasing trend inside the cirrus segment at UTC 20:20 – 20:30 compared with the surrounding clear-sky conditions, while the Na_{100} values show an increasing trend for in-cloud segment. The Na_{500} samples also show much fewer data than the Na_{100} samples, indicating relatively fewer large aerosols at cirrus level.

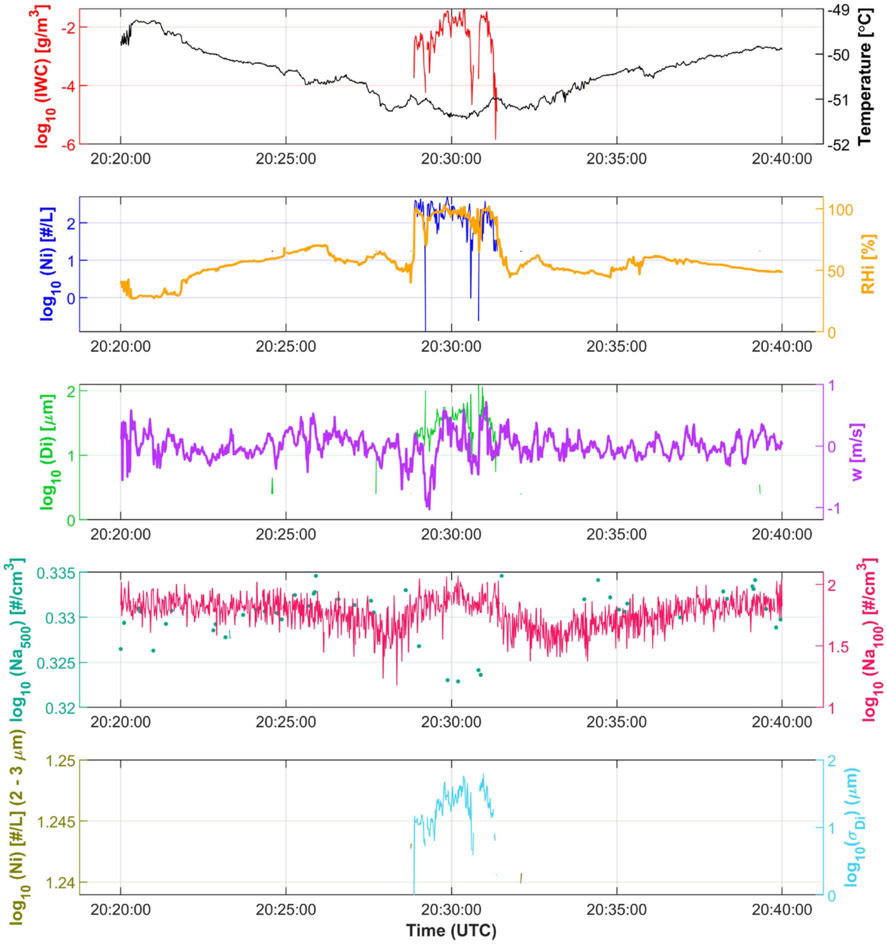

Figure R1. An example of time series from NSF DC3 RF12 on June 7, 2012. The Na_{500} values show a decreasing trend inside the cirrus segment at UTC 20:27 – 20:32 compared with the surrounding clear-sky conditions, while the Na_{100} values show an increasing trend for in-cloud segment. The Na_{500} samples also show much fewer data than the Na_{100} samples, indicating relatively fewer large aerosols at cirrus level.

Above and below, they argue using CDP concentrations of particles from ~1-3 microns should correlate with large aerosols as well, which is possible but I would not expect them to be 1:1 since they are different instruments and the bias is a probabilistic one. Additionally, the cloud droplet probe is also often fitted with anti-shattering tips similar to optical array probes, which would mitigate this bias assuming they are present in these field campaigns (the inlets are inherently different for both probes regardless). Also backtracking to Figure R3, note that these small particles are in fact restricted to the level leg within the ice cloud, and the approximate "spike" almost aligns with that of large aerosols (~25:30).

Figure S7 is similar to Figure R2 from their previous response, except now they adapted it to include occurrence frequencies. As hypothesized before, most of the samples where large aerosols are observed are at the relatively high Ni and Di, where IWC is speculated to be greatest (panel b). They now incorporate sigma_Di, arguing it should be larger when ice shattering is occurring. However, they show no evidence of the bimodal distributions they claim should exist, and they select an arbitrary threshold to use in panel d. Therefore I find this unconvincing evidence for an absent bias.

Responding to my concern that the large aerosol bias is likely only observed at large IWC, they show Figure 6 from the manuscript. They argue dIWC vs dlog10(large aerosols) trend lines fit similarly at positive and *negative delta values, meaning no bias is observed. First, it should be noted this isn't showing IWC and large aerosols, but their delta values. Maybe we can extrapolate smaller IWC from the lowest temperature range of -60 to -70C, but I now wonder if data is actually being cut off here. For a1, a5 and a9, the largest delta values clearly peak with "extra space" in the upper right portion of the panels. However, the lowest values sharply cut off at the bottom left of each panel. If I am interpreting these results correctly, this should be concerning since in Figure 5g dIWC values drop well below -2. Further, I wonder why data with temperatures lower than -70C is not included in a1, since there is a large amount of data here particularly having lower IWC (figure 4a).*

I don't wish to be stubborn or unmoved on this, but there has not been one time series presented meeting my request (I had hoped there would be multiple cases available), and other arguments have been made but unfortunately none convincingly. I wish to remind or inform the authors that this ice bias has been previously noted and is commonly accounted for, with examples specific to the UHSAS being Field et al. (2012), Froyd et al. (2022), Ladino et al. (2017) and Kupiszewski et al. (2016). I believe accounting for this bias in some manner will allow this study's findings to garner more attention from the research community.

Bibliography

Field, P. R., A. J. Heymsfield, B. J. Shipway, P. J. DeMott, K. A. Pratt, D. C. Rogers, J. Stith, and K. A. Prather, 2012: Ice in Clouds Experiment–Layer Clouds. Part II: Testing Characteristics of Heterogeneous Ice Formation in Lee Wave Clouds. https://doi.org/10.1175/JAS-D-11-026.1.

Froyd, K. D., and Coauthors, 2022: Dominant role of mineral dust in cirrus cloud formation revealed by global-scale measurements. Nature Geoscience, 15, 177–183, https://doi.org/10.1038/s41561- 022-00901-w.

Kupiszewski, P., and Coauthors, 2016: Ice residual properties in mixed-phase clouds at the high-alpine Jung fraujoch site. Journal of Geophysical Research: Atmospheres, 121, 12,343-12,362, https://doi.org/10.1002/2016JD024894.

Ladino, L. A., A. Korolev, I. Heckman, M. Wolde, A. M. Fridlind, and A. S. Ackerman, 2017: On the role of ice-nucleating aerosol in the formation of ice particles in tropical mesoscale convective systems. Geophysical Research Letters, 44, 1574–1582, https://doi.org/10.1002/2016GL072455.

We agree with the reviewer on the key statement that we are seeking a probabilistic rather than deterministic approach to quantify the potential influences of ice shattering on aerosol measurements. In our last revision, we realized that it is almost impossible to disentangle the causal relationship among aerosol-cloud-meteorology, especially from a time series perspective, because the positive correlation between aerosols and clouds could be potentially explained by many different reasons, such as a fraction of the large aerosols facilitated the formation of ice crystals, ice shattering leading to large aerosols, or meteorological or dynamical conditions that lead to higher concentrations of both, such as higher updraft that can lift up more large aerosols into upper troposphere and facilitate the formation of ice crystals. In other words, we cannot easily prove or disprove whenever there is a positive correlation between IWC and Na_{500}, whether it is caused by shattering or not. The aircraft measurements do not provide the history of air parcel development like a cloud model could do, which limits our interpretation of every positive correlation such as the part pointed out by the reviewer regarding the previous Figure R1 of NASA DC3 RF08.

Because we are seeking a probabilistic analysis, we are basically trying to quantify the impacts of possible ice shattering on the key findings. Thus, statistical analysis and linear regression in Figure 6 is indeed the most straightforward evidence to show that no matter IWC is lower or higher, no matter what RHi, w, or T ranges (except when sample is extremely low at -70 to -80C) are used, a nearly straight line is seen between

$d\log_{10}IWC$ and $d\log_{10}Na_{500}$. We also thank the reviewer for providing additional references on this topic, and we agree that ice shattering could happen, especially for the mixed-phase cloud temperature range reported previously with much higher IWC and Ni, such as the temperature range of several studies mentioned by the reviewer, including -16°C to -32°C in the study of Field et al. (2012), -8°C to -27°C in the study of Kupiszewski et al. (2016), about 0 to -15°C in the study of Ladino et al. (2017).

However, the main issue here is how likely it could lead to a biased analysis to the main conclusions of this study. Since Figure 6 and Figure S9 (100-s average clear-sky Na analysis) show no significant differences, the main conclusions regarding the correlations between $d\log_{10}Na_{500}$ (or $d\log_{10}Na_{100}$) and $d\log_{10}IWC$ remain the same. For example, the clear-sky Na_{500} at 100-s scale also shows relatively linear correlations with $d\log_{10}IWC$ in Figure S9. In addition, when restricting the IWC to $> 10^{-5}$, 10^{-4}, and 10^{-3} g m^{-3}, Figures S10 – S12 also show nearly linear correlations between delta values of IWC and Na_{500}. These results indicate that the influences of ice shattering are minimal on the key findings, because if that were the case, one would see visible differences in the correlations between lower and higher IWC, or between in-cloud and clear-sky Na.

We previously mentioned some drawbacks of using the clear-sky averaged Na, as it leads to a very sharp cutoff for the delta values of Na_{100} or Na_{500} seen in Figure S9, because the 1-Hz variability of Na is smoothed out on the 100-s scale. In addition, after thinking through the set-up of three ML tests (Test A, B, and C), we realized that if we switch all analysis to using 100-s average Na, the spatial scale sensitivity of 1-s, 50-s, 250-s, and 500-s scales shown by Test B in Table 3 cannot be examined anymore. It would remove the entire section about the influences of spatial scales of each predictor on the fluctuations of IWC.

We thank the reviewer for pointing out that some samples were cut off in the Figure 6 axis range. We agree that these points shouldn't be cut off. We also added a new temperature range of -80 to -70°C in the revised Figure 6 a1-a4 and b1-b4 to show the complete temperature range. We'd like to clarify that the reason there are a lot of low temperature samples in Figure 4 but not shown in Figure 6 is because NASA ATTREX and POSIDON provided most of the low-temperature cirrus samples in the tropics (as shown in Figure S1 panel f green and blue color markers). However, these two campaigns did not provide aerosol measurements and are excluded from Figure 6.

To be consistent with the updated axis range of Figure 6, we also updated Figure S9 – S12, as well as Table 3, which hosts the linear regression information for Figure 6. We copied the revised Figure 6 at the end of this document. These revised linear regression figures all show similar features compared with the last version, that is, nearly linear correlations are seen for delta values of IWC and Ni in relation to Na_{500}.

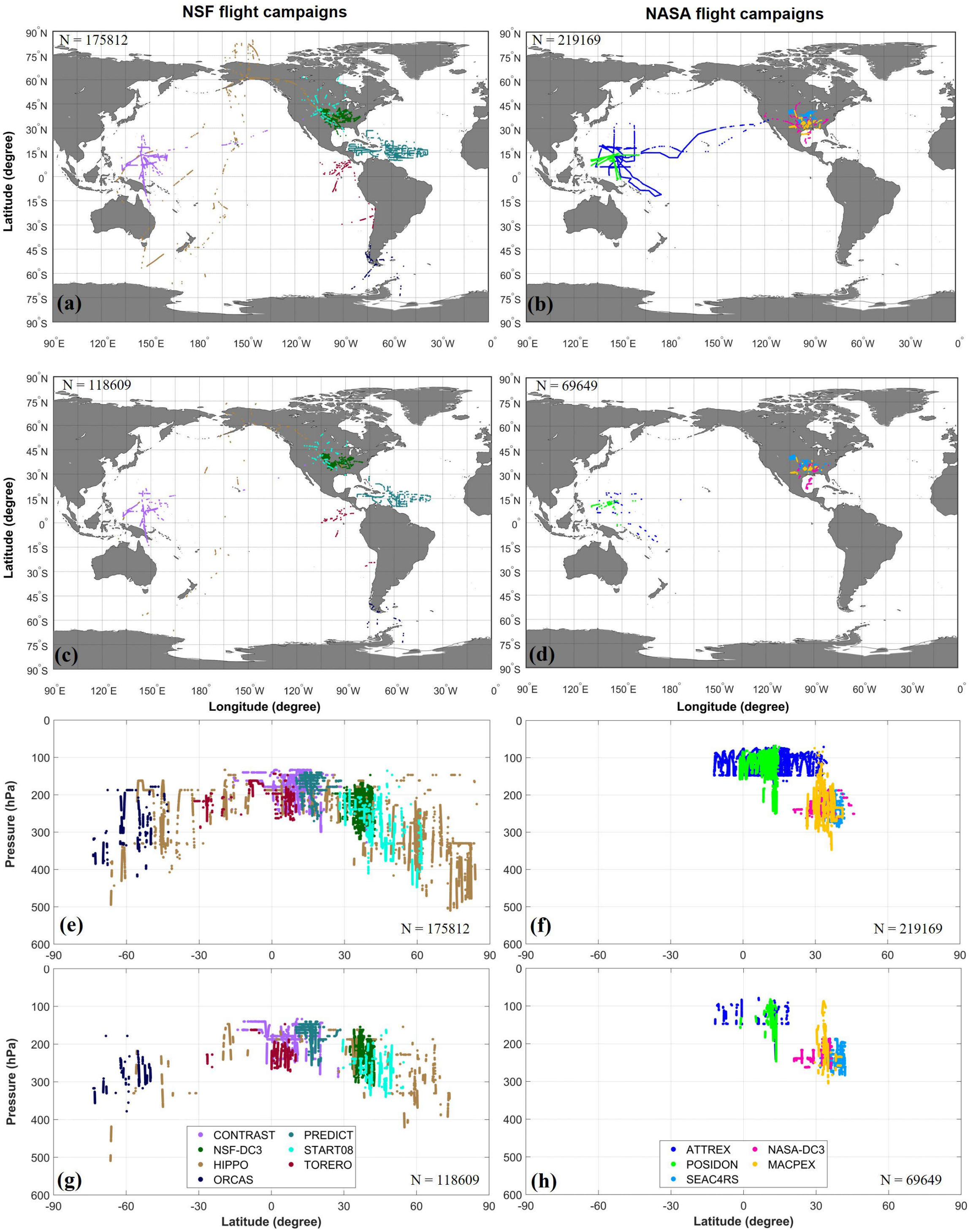

Figure S1. Global maps and vertical profiles of cirrus cloud measurements at temperatures ≤ -40 ºC based on seven NSF (left) and five NASA (right) flight campaigns. (a, b, e, f) Vertically quiescent cirrus clouds. (c, d, g, h) Non-quiescent cirrus clouds.

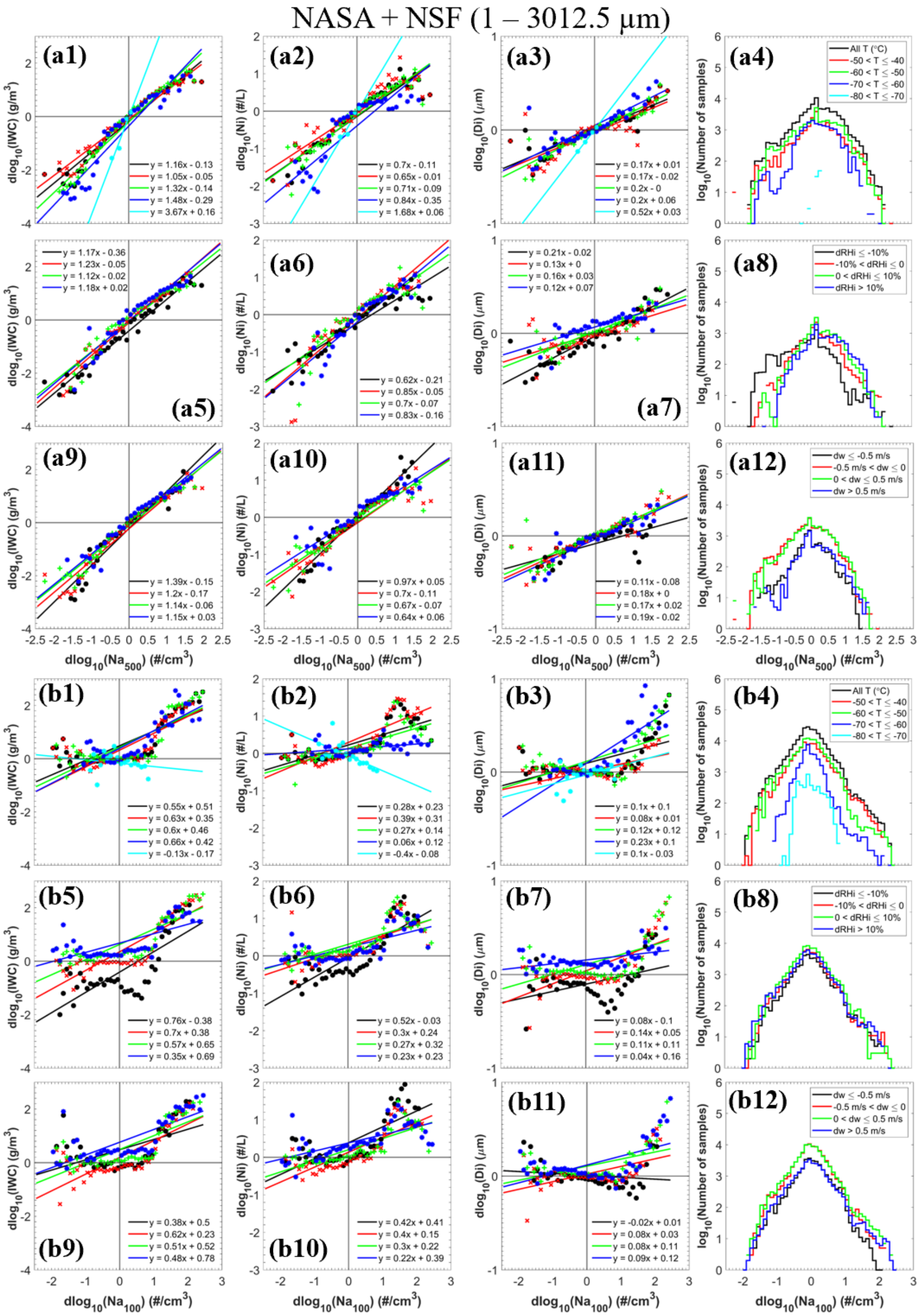

Figure 6. Linear regressions quantifying the correlations of $\mathrm{dlog_{10}IWC}$, $\mathrm{dlog_{10}Ni}$, and $\mathrm{dlog_{10}Di}$ with respect to $\mathrm{dlog_{10}(Na_{500})}$ in top 3 rows and $\mathrm{dlog_{10}(Na_{500})}$ in bottom 3 rows. The analyses in Figures 6 – 10 use the combined NASA+NSF datasets (1 – 3012.5 µm). ACI is examined for various ranges of temperature (in rows 1 and 4), dRHi (in rows 2 and 5), and d*w* (in rows 3 and 6). Colored dots represent geometric means of ice microphysical properties in each Na bin. Slope and intercept values are shown in the legend. The last column represents the distributions of the number of samples.