Authors' replies to the reviews of the ACP manuscript acp-2020-1241

"The ATAL within the 2017 Asian Monsoon Anticyclone: Microphysical aerosol properties derived from aircraft-borne in situ measurements"

Please notice that the title was changed to: "The Asian tropopause aerosol layer within the 2017 monsoon anticyclone: Microphysical properties derived from aircraft borne in situ measurements"

In the name of all authors, we would like to thank the two reviewers for their very detailed, diligent reviews and for providing their constructive, helpful suggestions to our submitted paper. These comments significantly contributed to the improvement of the paper. We hope to have adequately addressed all comments and hereby submit a revised version for re-evaluation. Thereby, we also thank the reviewers in advance for their renewed efforts.

Anonymous Reviewer #1 (Rev1)
Review

The manuscript describes the characterization of aerosols within the Asian Tropopause Aerosol Layer measured with a combination of in situ instruments onboard the M55 Geophysica research aircraft during the StratoClim field experiment 2017. The in situ data is compared to data from two near-range remote sensing instruments as well as satellite-borne lidar observations. The data presented here represent a valuable contribution elucidating aspects of the nature of this phenomenon so far detected only by means of remote sensing methods and balloon experiments.

The manuscript is well written and concise, the research is sound and in line with the overall subject areas of ACP. I would recommend the manuscript to be published after some minor points have been addressed.

General points:

[Rev1]: It would be good to add a slightly more detailed characterization of the UHSAS-A measurement and the data analysis (potentially as a supplement to the paper) given that it represents the central measurement for this study. This instrument fairly complex and the results are sensitive to environmental influences such as low temperature and pressure as well as the assumptions on refractive index of the aerosol. The authors discuss tests of the stability of the sample flow in a low pressure chamber. Were the uncertainties in the flow during ascents and descents introduced by the layout of the flow system investigated as well (see Kupc et al 2018, doi:10.5194/amt-11-369-2018)? Have there been any experiments checking the counting efficiency in different size ranges? And which uncertainty is "estimated to be up to 25%" (l.124)

> [Authors]: We agree that this should be discussed in more detail and added an Appendix (Appendix A) to cover these topics.

[Rev1]: By default the instrument can measure sizes in up to 99 size bins, what were the considerations for the binning used to represent the size distributions?

> [Authors]: We decided to use this binning to find a compromise between the size resolution and a reasonable averaging time at low number concentrations, as well as the ability of the UHSAS-A to resolve the signal response (most relevant for particles with diameter > 600 nm). See also Appendix A2.

[Rev1]: Similarly, the size information from the NIXE-CAS instrument was not used fully but only as a single bin (Figure 6).

> [Authors]: We agree and added the overlapping NIXE-CAS bin for a better comparison with the UHSAS to Figure 4 and Figure 6.

[Rev1]: For the derivation of optical properties such as the backscatter ratio assumptions made for the shape of the input particle size distribution might be important. Therefore I would ask the authors to extend the description of those calculations and give an estimate for the uncertainties introduced by those assumptions.

> [Authors]: A sensitivity study to determine the influence of shifts in the bin-limits of the size distribution as well as the influence of different refractive indices on the backscatter calculations was added to the appendix.

[Rev1]: I was a little confused by the term "Scattering Ratio". To my knowledge the lidar community commonly uses the more explicit term "Backscattering Ratio" for this quantity. Although I see that the cited reference also uses this term I would suggest renaming this throughout the text for clarity.

> [Authors]: As noticed by the referee, we wanted to stay consistent with the cited literature. But we agree with the referee and changed to the term "backscatter ratio" throughout the manuscript.

Specific points:

Title:

[Rev1]: I would highly recommend writing out the acronym ATAL in the title so readers not directly familiar with the topic have a chance of understanding what this paper is about.

> [Authors]: The title was changed to "The Asian tropopause aerosol layer within the 2017 monsoon anticyclone: Microphysical aerosol properties derived from aircraft borne in situ measurements"

Abstract:

[Rev1]: The abstract is relatively long for a not very long paper. It might be good to shorten that a bit. The measurement values in the abstract are given with a precision that is not likely reasonable. Throughout the paper, the authors should carefully revise all numerical values for stating a reasonable number of significant digits.

> [Authors]: The abstract was shortened a bit, while trying not to lose information. The particle mixing ratios of atmospheric aerosol discussed in the literature are generally reported in mg^-1 (Wilson et al. (1992); Brock et al. (1995); Curtius et al. (2005);…). We decided that for the general reader and for direct comparability with the literature it is better to report the particle mixing ratios also in mg^-1 and not in µg^-1. The presented values are specified to the last significant digit depending on the measurement errors.

[Rev1]: p7, l192. Check the precision of numerical values of MR. See above.

> [Authors]: See previous comment

[Rev1]: p7-8,l 210ff: I am not convinced that the total number of 1Hz-data points makes the median more robust here: At a given theta the possible values of those data points are not continuous but limited to certain values of MR because of the integer nature of the underlying count values which follow a Poisson statistics. The median cannot take any other value than one of the "stripes", therefore the slope of the median MR with theta in this upper region above 440K is primarily determined by the pressure/temperature structure of the atmosphere and even below that, between 420 and 440K, it will already be affected by the insufficient counting statistics. For the comparison to other instruments later in the manuscript this caveat should be added.
Possibly, resampling the data to longer time intervals might help to improve that statistics, though that depends on the detailed flight conditions in how far that would be meaningful. Resampling requires that the atmospheric conditions are quasi-homogeneous over that longer sample interval.

> [Authors]: We agree and resampled the data to 0.1 Hz (see red dots in Fig. 3a). We also calculated the median profile above 420 K using the 0.1 Hz data. For this we have to assume that above 420 K the atmospheric conditions are quasi-homogeneous within a 10 second interval (about 1.7 km flight distance).

[Rev1]: Fig 3: I am not sure this figure needs two panels given that the UHSAS-A and COPAS 2017 line are identical in both plots anyway and the UHSAS data is a repetition of Fig 1. By enlarging the figure the comparison to the other experiments should be sufficiently visible in just one panel.

> [Authors]: We think that it makes sense to show the StratoClim related measurements separately, especially as we now also added the resampled 0.1 Hz data-points to Fig 3 (a).

[Rev1]: p8, l239: I think "noticeable" might be the wrong word here.

> [Authors]: This paragraph has been rephrased.

[Rev1]: Fig 4: Figure labels and the text in the legend are very small and hard to read. Please enlarge the labels and legend. Possibly the information inside the legend could be put elsewhere to reduce the size of the legend overall.

[Authors]: The figure was modified to improve readability.

[Rev1]: p9, l265ff: The comparison to data from other campaigns in this section is certainly interesting from a point of view of atmospheric physics but is not a strong argument to prove the performance of the modified UHSAS-A since those measurements were taken at different times and locations. I think the statements in this direction should be removed from this section, the findings regarding the agreement with previous measurements and the size distributions added by this measurement should be the main topic of this paragraph.

[Authors]: We edited this section, to better transport the message as suggested by the referee and added a quantitative comparison based on the aerosol surface area and volume concentration calculated from the size distributions. To compare the measurement performance of the UHSAS-A (for large particles) at this altitude, instead we added the overlapping size bins from the NIXE-CAS to Fig. 4, as suggested by the referee.

[Rev1]: p10, l302: Although the last sentence in this section might be true it seems out of place here.

[Authors]: We agree that the sentence seems out of place here. The sentence was removed, and parts of its message were included into a sentence earlier in this section.

[Rev1]: Sec 6.1: Also referring to the comment above about the choice of size bins for UHSAS and NIXE-CAS it would be good to see how well those instruments match in the overlap regions of both size ranges. In addition, as mentioned above, a discussion of the uncertainty in the backscatter ratio introduced by the assumption of those fairly large bins should be added here. The discussion in the paper by Cairo et al 2011 cited here refers to measurements of cirrus cloud particles which have different size ranges and optical properties and may not be directly transferable.
The refractive indices used to derive the size distributions assume purely scattering particles. Given the influence of convection on the ATAL discussed later the presence of absorbing material such as BC cannot be excluded. How would the uncertainty estimates on the derived quantities change if this cannot be ruled out?

[Authors]: We added the overlapping NIXE-CAS size bins to Fig04 and Fig06. Between 1 and 3 µm is only one NIXE-CAS size bin available.
As stated at one of the general comments, a sensitivity study for the backscatter calculations was added to the appendix (and referred to in the main manuscript) showing a variability that is in agreement with Cairo et al 2011.
Concerning the sizing of BC particles the following paragraph was added to the UHSAS-A characterization section in the appendix (Sec. A2):
“Based on limited laboratory studies, Kupc et al. (2018) reported that black carbon particles might incandesce and vaporize due to the particles’

absorption of energy of the UHSAS-A detector laser (optical cavity laser power: ∼ 1 kW cm −2 at 1054 nm). This effect and the complex refractive index of black carbon would alter the sizing of black carbon particles significantly. While particles might be undersized because of the complex refractive index, the incandescing of black carbon particles could potentially result in an oversizing or an undersizing of these particles."
Referring to that, in the method section for the backscatter calculation (Sec. 6.1) the following sentences were added: "Additionally, black carbon particles might alter the result of the backscatter calculations, due to their complex refractive index and the uncertainties for their size representation in the particle size distribution measured by the UHSAS-A (see Appendix A2). Even though the presence of black carbon particles in the ATAL altitudes is enhanced during the ASM season, it's contribution to the overall aerosol particle mass concentration (for particle diameters < 2.5 µm) is reported to be only about 1.3 % at the 100 hPa pressure level (Gu et al. (2016)).". Another aspect is that the concurrently conducted in-situ particle mass spectrometry measurements on the M55 Geophysica indicated a very low presence of BC particles, probably even below 1.3%. This is subject of current data analyses and a forthcoming manuscript on the chemical composition of the ATAL.

[Rev1]: p11, l319ff: Check precision of numerical values (see above).

[Authors]: Throughout the manuscript we now report diameters equal or lager 1000 nm in µm.

[Rev1]: Fig 6: Like for Fig 4, consider enlarging the axis labels.

[Authors]: The figure was modified accordingly.

[Rev1]: Sec 6.2, Fig 7: Are there any uncertainty estimates for the various lidar measurements that could be added in this plot? In the range above 19km the yellow line of the MAL measurements is obscured by the in-situ-derived data.

[Authors]: We think adding the additional bars to this figure would overload it in the main part of the manuscript. We added the detection limits of the MAS and MAL to the respective instrument descriptions. Additionally, we added a version of Fig. 7 including the variability as bars to the appendix.

[Rev1]: p13, l374: The in-situ-derived scattering data are potentially affected by the sampling issue mentioned above. Although the trend is likely robust the exact slope might not be. This should be stated as a caveat.

[Authors]: We assume this comment is referring to the comment on p7-8,l 210ff. For the calculation of the backscatter data the size distributions were averaged over 100 second intervals.

[Rev1]: Sec 6.3: The authors should state how many CALIOP profiles are available in this region for the given time period and show a measure of variability in addition to the mean for those as well. Can the CALIOP profiles be split in time corresponding time periods as well?

> [Authors]: The shown CALIOP profile is based on 311100 profiles (information also added to the manuscript). Because there is no CALIOP data available for the first campaign period, it is not possible to split the profile. Because we do not want to overload the figure in the main part of the manuscript, we added a figure including the variability bars of the CALIOP BR profile to the appendix.

[Rev1]: Fig 8: Make sure the labels are sufficiently large to be readable in the final production. If there is only a single CALIOP profile to show consider merging the panels into a single figure.

> [Authors]: We agree and merged the three panels into one figure.

[Rev1]: Sec 6.4: As mentioned above detection of CO makes the presence of BC in the aerosol layer conceivable. How would the size distributions change if the assumption of a purely scattering refractive index is relaxed and would that have an effect on the derived scattering properties?

> [Authors]: Please see authors response to previous comment [Rev1]: Sec 6.1.

[Rev1]: p15, l450: were in situ measured -> were measured in situ ...

> [Authors]: Changed as suggested.

[Rev1]: p15, l473. The statement of slow vertical ascent is conceivable but not shown by the data presented here. Therefore, a reference should be given to this statement.

> [Authors]: Corresponding references to Vogel et. al. 2019 and von Hobe et. al. 2021 were added.

[Rev1]: p16, l475: Is there an "and" missing before "removal"?

> [Authors]: Corrected.

[Rev1]: p16, l480: The mention of the box model come somewhat surprising here given that it has not been mentioned in the main part of the paper. Maybe rephrase the sentence to place the reference to Weigel et al 2020a more prominently.

> [Authors]: The sentence was rephrased to: “Weigel et al. (2021a) showed that the freshly nucleated aerosol particles (as observed from COPAS) coagulate onto the background aerosol (as observed by the UHSAS-A) within a few hours by using simple box model simulations (adopting the SOCOL (SOlar Climate Ozone Links (Stenke et al. (2013)) coagulation subroutines).”

Anonymous Reviewer #2 (Rev2)

Review of "The ATAL within the 2017 Asian Monsoon Anticyclone: Microphysical aerosol properties derived from aircraft-borne in situ measurements" by Mahnke et al.

This manuscript reports important, high-quality results from unique stratospheric flights into the Asian tropopause aerosol layer (ATAL). The data demonstrate that the ATAL is a modest increase in aerosol number concentration and scattering ratio beyond the background stratosphere. The manuscript is well laid out, and the data presentation is mostly clear. However, there are some portions of the manuscript that are not precise, and the analysis and discussion needs to place the results in the broader context of the influence of the ATAL on the stratosphere. The numbers are presented, but there is limited discussion about whether the documented enhancements in aerosol concentration are significant to stratospheric processes such as radiative transfer and chemistry. Further, the comparison with previous balloon-borne data needs to be improved. I recommend that the manuscript undergo a major revision to address these two primary issues. In addition, there are several spots in the manuscript that need improvement for clarity and precision. The measurements are great; the analysis and presentation just need some improvement.

Major comments:

[Rev2]: 1) Discuss the relevance of the findings. The manuscript presents some very interesting, well-made, and unique measurements made within the heart of the ATAL. It has been very difficult to get in situ airborne measurements over the Indian subcontinent, and the investigators are to be commended for their persistence in accessing the airspace to make these important observations. Because these measurements are so unique, they should be placed in the context of their relevance to stratospheric processes. Currently the manuscript compares the data with balloon-borne observations, earlier airborne measurements, and lidar observations. But there is no real scientific take-home message: does the ATAL matter much to stratospheric processes? The best way to test this question would be to use a global model, adjusting it to match the observed ATAL characteristics, and then examining impacts on stratospheric chemistry, circulation, and radiative transfer. This is clearly beyond the scope of this manuscript. However, it should be possible to detail the fractional increase in aerosol surface area and say something about its relevance to heterogenous chemistry, and to estimate the radiative effect induced by the particles. What fraction of the Junge layer does the ATAL represent, both locally and globally? Does the light scattering from the ATAL represent a significant perturbation to the stratospheric radiation budget? (Does a backscatter ratio of 1.08 (8% above molecular backscatter) matter much?) These are the questions that need to be discussed in the context of these new and exciting observations. I recommend that a discussion section be added to the manuscript to address these topics.

> [Authors]: We agree with the referee that future studies are needed, utilising our datasets in combination with detailed global modelling to answer the question about the local and global effect of the ATAL on aerosol chemistry and radiative forcing. We added discussion within the main parts of the manuscript (Sec. 5: ”…This shift of the distributions main mode to larger particles is even more prominent in the surface area (Fig. 5 (b)) and volume

size distribution (Fig. 5 (c)). This increase in aerosol surface area can have a local effect on heterogeneous chemical processes. Additionally, Yu et al. (2017) reports based on model simulations that these particles spread throughout the entire northern hemispheric lower stratosphere and contribute annually with about 15 % to the stratospheric column aerosol surface area in the northern hemisphere and set a lower limit of the ASM contribution to the global stratospheric aerosol surface area of about 7 %." and Sec. 6.2: "The regional total sky radiative forcing caused by the ATAL was reported by Vernier et al. (2015) to be around - 0.1 Wm^−2 since the late 1990s, which corresponds to one third of the reported total radiative forcing (0.3 Wm^−2) from the global carbon dioxide increase during the same time period (Vernier et al. (2018)).") and in the conclusion section by setting our observation in context to previous observations of the ATAL and modelling results from the literature.
There are several studies ongoing within the StratoClim modelling community where exactly this point of the reviewer is addressed. The authors behind will use our measurements and need a published reference for these. For this reason, we would like to stay for our manuscript with a now better placing of our measurements into the framework of the published literature.

[Rev2]: 2) Comparison with Wyoming balloon measurements. The comparison with the Wyoming balloon-borne size distributions is limited to visual examination of the size distributions, and then saying, "sufficient agreement of the measurement results can be seen". This is an extremely subjective and unsatisfying comparison. First, based on the launch site of Hyderabad, the measurements from the Wyoming sensor were in southern India, even though no latitude or longitude for the sampling location is given. It's not clear that the Wyoming measurements were within the ATAL, even if they were within the ASM period. Second, the size distributions displayed in Fig. 4., and the remainder of the analysis throughout the manuscript, does not make use of the 3 CPCs in the COPAS. Differencing the concentrations in the 3 channels should yield size bins from 6-10, 10-15, and 15-65 nm, which is useful information on the recency of NPF and growth to larger sizes. Third, the comparison does not included any quantitative evaluation. Are the integrated number (over the relevant size range for the balloon measurements), surface, and volume comparable? If not, why not? To me, the size distributions display obvious discrepancies on a log-log plot, which suggests they are not very close in these integrated parameters.

[Authors]: We widely agree with the referee. The discussion was rephrased and extended to include the suggested information, quantitative comparisons, and to redirect the message of this part of the section as suggested.

Here and elsewhere it is asked why the COPAS data shown here are only presented in a single size interval, although the COPAS measurement with three different cut diameters (Dp50) could provide a higher size resolution: The COPAS measurement of each individual channel always provides a total number concentration of particles in the submicrometre range. Finer resolution of the COPAS size range could only be achieved by subtracting these total number concentrations. This works very well whenever, for example, in new particle formation (NPF), the concentration of particles with

Dp>6nm is excessively much higher than the concentration of particles with Dp >10nm or with Dp >15nm. But even then, a conservative criterion is applied to interpret an NPF event as such (cf. Weigel et al. (2011); Weigel et al. (2021a)).
Outside NPF, the scatter of the individual detector signals (about +/- 20 %) leads to comparatively inaccurate results when subtracting the concentrations with different cut diameter, which would lead to a distorting contribution in the size distribution shown. Such an increased uncertainty would minimise the informative value of the size distribution. The influence of the scatter on the solidity of the resulting values becomes even more critical if the COPAS bin is divided not only into two, but into three sub-bins. The smaller the distance between the cut diameters, the more uncertain the resulting difference in concentrations. This is complicated by the fact that the cut diameters represent the particle diameter for which a 50% detection efficiency (η) was determined according to a sloping η-curve (cf. Weigel et al. (2009)). It must be taken into account that the sub-20nm particles (in contrast to the direct optical scattered light detection of the larger particles with Dp>65nm) must be artificially made to grow before their detection with COPAS. The cut diameters are therefore also subject to an uncertainty that makes a Dp_{50} too inaccurate to hold as a distinct bin boundary in a size distribution. To detect a recent NPF event, the investigation of the differential concentration is done by subtracting very large numbers from each other. The uncertainty of this subtraction outside of an NPF event does not appear sufficient for a meaningful representation of the difference data in a size distribution."

[Rev2]: 3) A new Section 7 is needed to discuss the results in the context of stratospheric processes. Does the ATAL matter, or is it just of peripheral interest? How large is the perturbation to the radiation budget of the stratosphere, compared with the Junge layer? What fraction of the total stratospheric columnar loading is present in the ATAL? What is the estimated (calculated) amount of scattering and absorption? What is the surface area, and how does it compare with the literature? Is it important for stratospheric chemistry? Some discussion and evaluation would help make this manuscript much more relevant to the general reader of ACP.

[Authors]: Please see response to major comment 1).

Minor and Technical comments:

There are a number of places in the manuscript where more precise use of language would add clarity and reduce confusion. In addition, there are some technical corrections that need to be made.

[Rev2]: a) Line 9: Please don't use "density" when you mean "concentration" here, and elsewhere in the manuscript.

[Authors]: We changed this throughout the manuscript.

[Rev2]: b) Line 10: What is "NIXE"?

[Authors]: The full name “New Ice eXpEriment” for NIXE was added.

[Rev2]: c) Line 41: Does deep convection really provide "efficient" transport of aerosol particles and precursors (this implies low losses)? Or do you mean "rapid"?

[Authors]: Instead of "efficient" the term "rapid" is now used in the manuscript.

[Rev2]: d) Line 51: Replace "production" with "emission".

[Authors]: Replaced as suggested.

[Rev2]: e) Line 55: Nitric acid is needed as well as ammonia.

[Authors]: Agreed and adapted in the manuscript.

[Rev2]: f) Line 57: Emissions of what? Are particles transported to the ATAL, or just gas-phase precursors? I can't answer this question after reading this section.

[Authors]: We specified this reference to Fairlie et al. (2020) to:”…emissions of particle precursors like sulfate, nitrate, and ammonia, but also aerosol particles (e.g. like primary organic aerosol)...”.

[Rev2]: g) Line 61: "In this paper we examine the vertical distribution. . . ."

[Authors]: Changed as suggested.

[Rev2]: h) Line 65: Change "calculated" to "calculate".

[Authors]: Changed as suggested.

[Rev2]: i) Line 70: The title of the field campaign should be capitalized, even if it doesn't match the acronym.

[Authors]: Here we decided to stick with the spelling of the title as it is in the title of the special issue and most if not all of the other literature in ACP/AMT referring to this campaign.

[Rev2]: j) Line 70: Throughout this paragraph, please use past tense verbs when discussing StratoClim.

[Authors]: Corrected as suggested.

[Rev2]: k) Line 78: Is there a reference for the Geophysica and the basic payload?

[Authors]: Currently, there is not yet a general reference for the Geophysica and the basic payload during StratoClim available. We added Borrmann et al. (1995) and Stefanutti et al. (1999) as references for the Geophysica.

Additionally, there is a reference for the UCSE (Unit for Connection with Scientific Equipment) for the avionic and meteorological parameters in Sec. 3.

[Rev2]: l) Line 80: Remove "flight paths see" when referring to Fig. 1.

[Authors]: Removed as suggested.

[Rev2]: m) Line 82: Change "were headed to India" to "were over northeastern India".

[Authors]: Changed as suggested.

[Rev2]: n) Line 100: Change "wing-sonde" to "underwing".

[Authors]: Changed as suggested.

[Rev2]: o) Line 105: Change to ". . . version of the UHSAS-A were necessary: integrating a. . ."

[Authors]: Changed as suggested.

[Rev2]: p) Line 106: Change to ". . . of the UHSAS-A and installing a new pump system. . . ."

[Authors]: Changed as suggested.

[Rev2]: q) Line 107: Were these instrument changes not made prior to the deployment in 2016 in Greece? If not, were the high-altitude data from Greece valid given the pumping problems?

[Authors]: The new pump system could only be integrated after the deployment in Greece. The data from Greece for pressure < ~ 120 hPa could not be validated, due to the intense drops in sample, sheath, and purge flow. Appendix A1 was added to the manuscript to show the difference of the UHSAS-A internal flows before and after the new pump system was integrated.

[Rev2]: r) Line 109: Remove "Also, ".

[Authors]: Removed as suggested.

[Rev2]: s) Line 112. Change to "characterized as a function of pressure."

[Authors]: Changed as suggested.

[Rev2]: t) Line 115: Change "has been" to "was" and Polystyrol Latex spheres" to "polystyrene latex spheres." (note use of lower case)

[Authors]: Changed as suggested.

[Rev2]: u) Line 117: Add ". . . to remove doublets and contamination particles." (Why else use a DMA?)

[Authors]: Added as suggested.

[Rev2]: v) Line 118: Add ". . . without the DMA". I assume these sizing checks were performed without the DMA.

[Authors]: This assumption is correct. Added as suggested.

[Rev2]: w) Line 119: What was the reference standard from which you determined 10% uncertainty in counting efficiency. A CPC?

[Authors]: Yes, we used a TSI 3025A CPC for these measurements. We added Appendix A3 for more details.

[Rev2]: x) Line 120: I'm confused by the uncertainties. It sounds like there is a base counting uncertainty of 10% (due to knowledge of flow rate?) and an additional statistical (Poisson) uncertainty that is the square root of the number of counts in a given sampling interval (1s). If this is correct, please explicitly state this and give a representative total uncertainty given the observed number of counts per second in the ATAL.

[Authors]: Concerning the 10 % counting uncertainty see previous comment. The paragraph was rephrased to make it clearer.

[Rev2]: y) Line 127: Please remove the entire sentence beginning "The upper limit of the particle diameter. . . ." and change the next sentence to "Particles with diameters <1 µm were aspirated. . . ."

[Authors]: Changed as suggested.

[Rev2]: z) Line 135: "NIXE" again.

[Authors]: The full name “New Ice eXpEriment” for NIXE was added.

[Rev2]: aa) Line 140: Change to "et al. (2017). More detailed descriptions. . . ."

[Authors]: Changed as suggested.

[Rev2]: bb) Line 141: Remove "have been used to"

[Authors]: Removed as suggested.

[Rev2]: cc) Line 147: Change "close to" to "from".

[Authors]: Changed as suggested.

[Rev2]: dd) Line 148: Change to "which translates into a horizontal resolution of 1-2 km at the M55. . . ."

[Authors]: Changed as suggested.

[Rev2]: ee) Line 150: Please provide the detection limit in m^-1 sr^-1, since you are examining a small signal.

[Authors]: We added the detection limit of the MAS for the aerosol backscatter coefficient of 5 * 10^-10 m^-1 sr^-1 for a single 10 seconds data point.

[Rev2]: ff) Line 158. Also provide the detection limit for the MAL.

[Authors]: We added the detection limit of the MAL for the aerosol backscatter coefficient. For a 900 second flight interval (at cruising speed of about 170 $m\ s^{-1}$) probing an atmospheric layer at 17 km altitude from a distance of $\sim$ 1500 m the detection limit of the aerosol backscatter coefficient is $5 \cdot 10^{-10}\ m^{-1}\ sr^{-1}$.

[Rev2]: gg) Line 160: "carbon monoxide" is not capitalized.

[Authors]: Agreed and changed.

[Rev2]: hh) Line 165: Change to "and updated electronics".

[Authors]: Changed as suggested.

[Rev2]: jj) Line 166: Do not capitalize "tunable diode laser spectroscopy". It's a method, not a product name.

[Authors]: Agreed and changed.

[Rev2]: kk) Line 171: Same for "new particle formation".

[Authors]: Agreed and changed.

[Rev2]: ll) Line 185: Change "altitude" to "theta" (the Greek character).

[Authors]: Changed as suggested.

[Rev2]: mm) Line 187: Change "inclines" to "increases". An "incline" is an upward slope from horizontal.

[Authors]: Changed as suggested.

[Rev2]: nn) Line 193: "NPF" is already defined.

[Authors]: Re-definition of NPF removed.

[Rev2]: oo) Line 193: I would imagine that convective outflow in laminae is also a major source of variability at this altitude.

[Authors]: We agree and changed the sentence accordingly.

[Rev2]: pp) Line 203: Change "begins to abate" to "decreases with increasing (theta symbol)."

[Authors]: Changed as suggested.

[Rev2]: qq) Line 210: Change "weak" to "poor".

[Authors]: Changed as suggested.

[Rev2]: rr) Line 211: Remove the unnecessary sentence, "However, due to the high number. . . ."

[Authors]: The sentence was rephrased, accounting for the resampling to 0.1 Hz of the UHSAS-A data for Theta levels < 420 K and the accordingly recalculated median profile. See also the authors reply to Rev1 comment p7-8,l 210ff.

[Rev2]: ss) Line 215: The flights from Greece may or may not have been in the "extratropics", depending on the meteorology and direction of flight. Please state the season and brief evidence (e.g., north of the subtropical jet) for this statement.

[Authors]: The dates and the information about the latitudinal and longitudinal extend, as well as the location of the flights from Greece relative to the strong subtropical potential vorticity gradient were added to the campaign description (Sec. 2).

[Rev2]: tt) Line 217: Change "read out of" to "digitized from".

[Authors]: Changed as suggested.

[Rev2]: uu) Line 226: Change "that was also" to "was".

[Authors]: Changed from "A maximum (…) that was also observed by Brock et al. (1995)…" to "A maximum (…) in this Θ range was also observed by Brock et al. (1995)…"

[Rev2]: vv) Line 230: Change to ". . . from about 10-1000 nm, and those of Brock et al. (1995) were from 8-3000 nm (or whatever), while the UHSAS-A . . . ."

[Authors]: Changed as suggested.

[Rev2]: ww) Line 233: Change "densities" to "concentrations".

[Authors]: Changed to particle mixing ratios.

[Rev2]: xx) Line 233: How does it follow that 10-65nm particle concentrations demonstrate "fresh nucleation"? If the particles were ~50 nm, this could be several days old given low coagulation rates. Why do you not report the concentration of the 6-10 COPA channel difference to provide evidence for recent NPF?

> [Authors]: We intentionally used the formulation "indicates…the influence of…". But we agree with the referee that it indicates the influence of NPF and removed the term "fresh".
> The identification of NPF during StratoClim, utilising the difference of multiple COPAS channels was already comprehensively discussed by Weigel et al. (2021a) and Weigel et al. (2021b). To avoid publishing repetitive results, we referenced to these publications.

[Rev2]: yy) Paragraph beginning line 235: I find this paragraph confusing. Are you saying that the UHSAS mixing ratio (>65 nm) over this theta range is greater than the canonical "background" values of reported by Brock (>8 nm)? If so, just say that.

> [Authors]: We rephrased the paragraph to make this clearer.

[Rev2]: zz) Line 253: I don't know that a change from 470 to 170 per mg is "subtle".

> [Authors]: We agree that "subtle" is not describing the change correctly and removed it.

[Rev2]: a1) Line 263: Evident from what technical parameters? I'm not sure what this means.

> [Authors]: We rephrased the sentence and referred to the new Appendix covering the characterization of the UHSAS-A.

[Rev2]: a2) Line 264: Replace "profoundness" with something else. Accuracy?

> [Authors]: The sentence was rephrased.

[Rev2]: a3) Line 268: Please note the latitude and longitude of Hyderabad and note if the measurements were made in the ATAL or not.

> [Authors]: The latitude and longitude of Hyderabad (17.47°N, 78.58°E) were added. Because we discuss here measurements at about 20 km altitude, none of these measurements we made within the ATAL. But Vernier et al. (2018) showed that the balloon-borne measurements from Hyderabad were made above the ATAL.

[Rev2]: a4) Line 274: Change "the data set" to "the balloon data set" to identify which measurement you're speaking about.

> [Authors]: Changed as suggested.

[Rev2]: a5) Line 287: Please specify quantitatively what "sufficient agreement" means. Are they within stated uncertainties in concentration and size? Comparing integrated number, surface, and volume is a good way to provide a quantitative evaluation, at least over the size range where the instruments overlap.

[Authors]: The discussion was widely rephrased and extended to include the suggested quantitative comparisons and to redirect the message of this part of the section as suggested in major comment (2).

[Rev2]: a6) Line 306: Change "could already confirm" to "confirmed".

[Authors]: Changed as suggested.

[Rev2]: a7) Line 307: Change "To go one step further" to "To compare with these observations".

[Authors]: Changed as suggested.

[Rev2]: a8) Line 307: Here and throughout the manuscript. I find it odd to call the ratio of total to molecular backscatter the "scattering ratio". This should be the "backscatter ratio". See, for example, https://doi.org/10.5194/amt-12-4065-2019.

[Authors]: Agreed and changed throughout the manuscript.

[Rev2]: a9) Line 316: Change "flight segment a UHSAS-A measured" to "100-s interval the UHSAS-A-measured".

[Authors]: Changed as suggested.

[Rev2]: a10) Line 333: I recommend calculating backscattering properties using the same refractive index as the calibrant (PSL spheres). The reason is that each bin of the UHSAS represents a certain amount of light scattering, in this case the amount of light scattered by PSL spheres. To go back to total light scattering, you should just integrate the amount of scattering each bin represents--which is the scattering by a PSL sphere. While backscattering is not the same as the side scattering measured by the UHSAS, it's probably more accurate to assume the PSL refractive index that was originally used to establish the bin sizes for the instrument.

[Authors]: We agree that this was an inconsistence in our method. We decided for the aerosol backscatter calculation to recalibrate the UHSAS-A bin-limits from the calibration with PSL standards (refractive index m = 1.59) to m = 1.5 and m = 1.45 (Appendix A4 in the appendix), before recalculating the aerosol backscatter coefficients and BR using the respective refractive indices. Additionally, we added Appendix B to the appendix as a sensitivity study to show how the aerosol backscatter coefficient calculation is affected by using different refractive indices and shifting the bin-limits of the size distribution.

[Rev2]: a11) Section 6.2. Can you estimate the hygroscopicity of the aerosol and the ambient size they would have? This might substantially affect the backscatter comparison with the remotely sensed measurements, which are at ambient RH.

[Authors]: A reliable estimation for the influence of the hygroscopicity of the aerosol on the backscatter comparison would require particle size resolved

knowledge about the aerosol chemical composition along the flight tracks. This information is not available.
The sentence: “Also the effect of the particles hygroscopicity on the measured particle sizes and the resulting calculated backscatter compared with the remotely sensed backscatter measurements, which are at ambient relative humidity, can not be ruled out.” was added to Section 6.1.

[Rev2]: a12) Line 373. Please provide a reference for the Junge layer.

[Authors]: One general reference for the Junge layer and one showing it together with the ATAL (observed in a different year than our observation) were added.

[Rev2]: a13) Line 397: Change to, "there are fewer cloud free flight segments at altitudes of up to ~15 km."

[Authors]: Changed as suggested.

[Rev2]: a14) Line 407. Change to "The ATAL's relation".

[Authors]: Changed as suggested.

[Rev2]: a15) Line 416. Change "lagging" to "lagged".

[Authors]: Changed as suggested.

[Rev2]: a16) Line 417: Change "correlation" to "relationship".

[Authors]: Changed as suggested.

[Rev2]: a17) Line 449: Change "in situ measured" to "measured in situ"

[Authors]: Changed as suggested.

[Rev2]: a18) Line 452: Change "ATALs" to "ATAL". You've already defined "SR" (although it should be backscatter ratio).

[Authors]: Changed as suggested. SR was changed to BR throughout the manuscript.

[Rev2]: a19) Line 457: Change "ATALs" to "ATAL".

[Authors]: Changed as suggested.

[Rev2]: a20) Line 470: Where did "probably spiraling" come from? Over what time/spatial scales? Why do you think this? Is this relevant?

[Authors]: We agree that this is not relevant here and removed "probably spiraling".

[Rev2]: a21) Line 482: I don't understand what this sentence is saying. Are you saying that coagulation is insufficient to quickly reduce the observed concentrations of small particles, therefore NPF must be ongoing? Please clarify. Again, this is an opportunity to use the sizing afforded by the COPAS channels and examine the concentration of 6-10 nm particles in the smallest channel for evidence of very recent NPF.

> [Authors]: What we want to say is, that the coagulation of the nucleation mode particle happens so fast, that the still frequent NPF encounters during StratoClim 2017 detected by COPAS indicate the prevalence of such events within the ASM region. The sentence was rephrased to make this clear. The occurrence of recent NPF during StratoClim 2017 was already discussed based on the COPAS measurements in detail in the cited publications from Weigel et al. (2021a) and Weigel et al. (2021b) (Weigel et al. (2020a) and Weigel et al. (2020b) in the preprint), to which this sentence refers to.

[Rev2]: a22) Please go over the references thoroughly an ensure compliance with ACP formatting guidelines. There are many obvious discrepancies--some paper titles are capitalized, some are not; some journals are abbreviated, some are not, etc. Please do not rely on reference manager software--it always does a poor job of formatting and this causes a lot of work for Copernicus technical editors.

> [Authors]: The references were updated according to the ACP guidelines.

[Rev2]: a23) Please place latitude and longitude markers on the axes of Fig. 1.

> [Authors]: Done as suggested.

[Rev2]: a24) Fig. 2 is very nice!

> [Authors]: We thank Rev2 and note that the median profile shown here was recalculated based on the 0.1 Hz resampled data for Theta levels > 420 K. This is noted in the Figure legend, caption, and the discussion within the manuscript.

[Rev2]: a25) Fig. 3. Please mark the region of the ATAL, between ~370 and 410K, and also the approximate latitude range of the TTL. Fig. 3b relies on color vision to discriminate the lines; please add some symbols or different line types.

> [Authors]: We changed Fig. 3 as suggested with different line types and symbols. Because Fig. 3 (a) and (b) show data measured in different years and global regions, we decided not to mark the ATAL and TTL in these figures, as these altitudes/theta-levels vary for different years and regions, what could result in misinterpretations of the figures.

[Rev2]: a26) Fig. 4. Where are the 3 COPAS channels? The agreement between UHSAS and the UCSE data looks quite poor. Please quantify the level of agreement in the text.

> [Authors]: Concerning the 3 COPAS channels, please see authors response to [Rev2]: Major comment 2). The discussion of the comparison between the

balloon data and the UHSAS-A data was extended. Additionally, the overlapping size-bin of the NIXE-CAS measurement was added.

[Rev2]: a27) Fig. 5. Also a very nice figure. I'd like to see the ATAL marked, and a second plot showing dV/dlogDp, which should very clearly show the ATAL.

[Authors]: Fig. 5. was changed to a three-panel figure, showing the size distributions for number, surface area, and volume concentration. The ATAL was marked as suggested.

[Rev2]: a28) Fig. 6. Where are the 3 COPAS channels?

[Authors]: See authors response to [Rev2]: Major comment 2).

[Rev2]: a29) Fig. 7. Please mark the ATAL. Could you put a potential temperature axis, using climatological values, on the right axis? All the other plots are in theta-space, so this is confusing and hard to compare to other figures.

[Authors]: We marked the ATAL in this figure as suggested. We understand the wish for a Theta axis. Due to the wide range of observed Theta values at a specific altitude (or vice versa) over the campaign period (i.e. the 360 K surface covers a vertical range of ~4 km, while the 370 K surface covers a vertical range of ~1.5 km), a direct translation between altitude and Theta over this regional and temporal range is very ambiguous and misleading (especially in the altitude/Theta region of the ATAL). This is the case for each of the individual data sets shown here, but especially for the combination of the in-situ and the remote data. To avoid misinterpretations, we decided not to add a potential temperature axis.

[Rev2]: a30) Fig. 8. A theta axis would be helpful here, as well.

[Authors]: See authors response to previous comment for Fig. 7.

[Rev2]: a31) Fig. 9. These plots to not show "correlations"; they show scatterplots of y vs x.

[Authors]: We agree and changed the text and the figure caption accordingly.

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