Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2020-1241-RC2, 2021

Comment on acp-2020-1241

Anonymous Referee #2

Referee comment on "The Asian tropopause aerosol layer within the 2017 monsoon anticyclone: microphysical properties derived from aircraft-borne in situ measurements" by Christoph Mahnke et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1241-RC2, 2021

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:

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.

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.

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.

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.

- a) Line 9: Please don't use "density" when you mean "concentration" here, and elsewhere in the manuscript.
- b) Line 10: What is "NIXE"?

- c) Line 41: Does deep convection really provide "efficient" transport of aerosol particles and precursors (this implies low losses)? Or do you mean "rapid"?
- d) Line 51: Replace "production" with "emission".
- e) Line 55: Nitric acid is needed as well as ammonia.
- 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.
- g) Line 61: "In this paper we examine the vertical distribution. . . ."
- h) Line 65: Change "calculated" to "calculate".
- i) Line 70: The title of the field campaign should be capitalized, even if it doesn't match the acronym.
- j) Line 70: Throughout this paragraph, please use past tense verbs when discussing StratoClim.
- k) Line 78: Is there a reference for the Geophysica and the basic payload?
- l) Line 80: Remove "flight paths see" when referring to Fig. 1.
- m) Line 82: Change "were headed to India" to "were over northeastern India".
- n) Line 100: Change "wing-sonde" to "underwing".
- o) Line 105: Change to ". . . version of the UHSAS-A were necessary: integrating a. . . ."
- p) Line 106: Change to ". . . of the UHSAS-A and installing a new pump system. . . ."
- 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?
- r) Line 109: Remove "Also, ".
- s) Line 112. Change to "characterized as a function of pressure."
- t) Line 115: Change "has been" to "was" and Polystyrol Latex spheres" to "polystyrene latex spheres." (note use of lower case)
- u) Line 117: Add ". . . to remove doublets and contamination particles." (Why else use a DMA?)
- v) Line 118: Add ". . . without the DMA". I assume these sizing checks were performed without the DMA.
- w) Line 119: What was the reference standard from which you determined 10% uncertainty in counting efficiency. A CPC?
- 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.
- 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. . . ."
- z) Line 135: "NIXE" again.
- aa) Line 140: Change to "et al. (2017). More detailed descriptions. . . ."
- bb) Line 141: Remove "have been used to"
- cc) Line 147: Change "close to" to "from".
- dd) Line 148: Change to "which translates into a horizontal resolution of 1-2 km at the M55. . . ."
- ee) Line 150: Please provide the detection limit in m^-1 sr^-1, since you are examining a small signal.
- ff) Line 158. Also provide the detection limit for the MAL.
- gg) Line 160: "carbon monoxide" is not capitalized.
- hh) Line 165: Change to "and updated electronics".
- jj) Line 166: Do not capitalize "tunable diode laser spectroscopy". It's a method, not a product name.
- kk) Line 171: Same for "new particle formation".
- ll) Line 185: Change "altitude" to "theta" (the Greek character).

- mm) Line 187: Change "inclines" to "increases". An "incline" is an upward slope from horizontal.
- nn) Line 193: "NPF" is already defined.
- oo) Line 193: I would imagine that convective outflow in laminae is also a major source of variability at this altitude.
- pp) Line 203: Change "begins to abate" to "decreases with increasing (theta symbol)."
- qq) Line 210: Change "weak" to "poor".
- rr) Line 211: Remove the unnecessary sentence, "However, due to the high number. . . ."
- 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.
- tt) Line 217: Change "read out of" to "digitized from".
- uu) Line 226: Change "that was also" to "was".
- 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 . . . ."
- ww) Line 233: Change "densities" to "concentrations".
- 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?
- 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.
- zz) Line 253: I don't know that a change from 470 to 170 per mg is "subtle".

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

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

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

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

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.

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

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

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.

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

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.

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.

a12) Line 373. Please provide a reference for the Junge layer.

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

a14) Line 407. Change to "The ATAL's relation".

a15) Line 416. Change "lagging" to "lagged".

a16) Line 417: Change "correlation" to "relationship".

a17) Line 449: Change "in situ measured" to "measured in situ"

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

a19) Line 457: Change "ATALs" to "ATAL".

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

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.

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.

a23) Please place latitude and longitude markers on the axes of Fig. 1.

a24) Fig. 2 is very nice!

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.

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.

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.

a28) Fig. 6. Where are the 3 COPAS channels?

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.

a30) Fig. 8. A theta axis would be helpful here, as well.

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