

Review of the ACP manuscript acp-2020-1158

“In-Situ observation of New Particle Formation in the upper troposphere /lower stratosphere of the Asian Monsoon Anticyclone”

by R. Weigel et al., 2020

The above manuscript deals with in situ measurements of aerosol particles, in particular ultrafine ones, in the tropopause region of Nepal. Knowing the complexity of such measurements, both on the technical as well as on the organisational level, the measurements are highly valuable.

The manuscript is well organized, but in many sections too lengthy.

For instance the section on carbon monoxide, 2.3 could be shortened, it is not necessary to give text book or review article type knowledge here. Second paragraph in this section is fine, but the first one is not needed in this detail. Other examples are too many references for individual statements.

Besides the length of the manuscript, my major criticism is that in some parts there is too much interpretation of or speculation on the data set (e.g. wrt. Fig 5a).

So, please shorten the manuscript and try to keep a line of evidence, and do not speculate so much.

Specific remarks:

- As above, the introduction is quite long and reads more like an introduction to a thesis. This is also reflected by 11 pages of literature, this is not a review article, or? Could you try to shorten and focus it? For instance, much of Sec. 1.1 is not needed here to get introduced into the topic of your paper. Moreover, Sec. 1.2 has a misleading headline, this section describes the Asian Monsoon Anticyclone and ATAL, but not the StratoClim campaign, please change.

- p.2, l. 64/65: The list of the most likely aerosol particle constituents here mixes chemical composition (e. g. soot or inorganic salts) with source processes (e. g., biomass burning or meteoritic ablation). I suggest to harmonize the list.

- p. 3 l. 89: Please cite Brock et al. 1995 as reference for the statement of the first sentence.

- p. 3 l. 101: Why do you give a 40% - 90% range for the SO₂ transport in DCCs as calculated by CRMs, but in line 104 you cite a study where a CRM gives only 30%? Change the range and merge the sentences.

- p. 4 l. 114 ff.: In situ measurements of SO₂:

Rollins et al, GRL, 2017 and the discussion of their results is missing here!!!

- p. 4 l. 130: Please delete “(ultrafine)” as usually this term refers to all particles smaller than 100 nm diameter, but here you mean really the nucleation mode particles.

- p. 6 l. 160-165: You state here that you discuss here all NPF events during StratoClim 2017, but in the next sentence you say that you discuss the NPF events related to thin cirrus clouds in another paper. How does this fit together? Please clarify in the text.

- p. 9 l. 259: The “denuder” device, is it really a denuder? It is a vaporizer, as called in line 273. But the vaporized gases are not removed from the sampling air, right? How much of the vaporized gas re-condenses later on the non-volatile particle cores, not changing the number, but the size?

- p. 9 l. 272: The picture of the stratospheric aerosol changed in recent years. A significant fraction of organic material is found up to altitude of 8 km above the tropopause. How does this influence your interpretation of the measurements downstream the vaporizer? Could it be that a larger fraction of the non-volatile material is of highly oxidised organic nature? This question is more relevant for the results section.

- p. 10 l. 298: If you state in line 302 that you took the 15% uncertainty of the COPAS instruments into account to receive the criterion in equation (1), should the equation then not look like $0.85 \times N_6 - 1.15 \times N_{15} > 0$? Where do the 0.8 and the 1.2 emerge from?

- p. 11 l. 329 ff.: The discussion on the different NPF event terminology is confusing, please simplify.

- p. 15 l. 447 ff.: Not sure what is meant with “this case study” please specify more precisely, do you mean a coagulation simulation later on?

- p. 16 l. 470 ff.: What are the implications of the “However, ..” sentence for you simulation? Did you exclude such conditions from your simulations?

- p. 16 l. 473: What is meant with “sample” here?
Can you please provide a time resolution, e.g. every minute or so?

- p. 17 l. 485: The “However, sentence ...”. In this paragraph you emphasize the value of the new ERA data, but in the last sentence you raise doubts. Why?

- p. 17 l. 505/506: I do not understand the approach of including the trajectory uncertainty. I would assume that you use neighbouring trajectories as well and calculate an average over all these trajectories. And actually, does it really make sense to have trajectories every 154 m?

- p. 18 l. 515: The use of another back-trajectory model is, in the first instance, astonishing. Why do you calculate one back-trajectory with ClAMS every second, when you get 1000 using the other model?

- p. 18 l. 528-530: There are previous publications who have used this approach, e.g. Weigelt et al., 2009, JGR. Why do you cite these two?

- p. 19 l. 548: What do you mean with “certain” distance?

- p. 19 l. 553: The conclusion of the “Hence, ...” sentence is not clear to me. How did get to it? Based on 2 Weeks of measurement only. Is this valid?

- p. 20 l. 567: I’m not sure about section 4.1. Why is it needed in this manuscript (I don’t think so)? In particular as you do the comparison with previous campaigns in Fig 3a again.

- p. 20 l. 572: How was this merging accomplished, i.e. how did you account for the different lower threshold diameters? Please give a statement on that in the text.

- p. 21 1. 619: Please delete “which may also be a result of recent NPF”, this is speculation here, at this point you describe the scatter plot shape only.
- p. 21 1. 620: 3rd part of the vertical profile, is there really a difference between 400-415 K and above? If yes, please prove this by providing numbers, e.g. the variance in the different sections. By eye it does not look so different.
- p. 22 1. 644: Fig 3b: Why is N_{6-15} only provided in a smaller altitude range? Or do you show only data which fulfil your “event” criteria? This should be clarified in the text and the legend.
- p. 22 1. 650: Fig. 3c and d are a little bit misleading. In 3c you compare N_6 with N_{10nv} , and in 3d N_{10} with N_{10nv} . Hence the difference between the curves in 3c is much larger than indicated in 3d. This is of course correct, but misleading. Clarify this in the legend of Fig. 3d.
- p. 23 1. 657 - 672: This paragraph is a good example of too much text in the manuscript. It would be sufficient to describe a) the low fraction of N_{nv} in the lowest part of the profile, b) the local minimum coinciding with the maximum in N_6 and c) the nearly constant ratio above? This information could be provided in three sentences. In particular as you repeat this information in the following lines 675 to 690. Please shorten the paragraph.
- p. 24 1. 711 - 728: This paragraph and Fig 4 is very descriptive. What do we learn from this exercise? Could short NPF events be caused by other aircraft? Any indication of NPF event duration is shorter at lower altitudes (higher probability to encounter aircraft)? And long events by DCC outflow?
- p. 25 1. 739 ff.: This paragraph tries to interpret Fig. 5a, but, honestly, I believe you have a statistical problem, too small number of flights and hence too small number of different measurement conditions. Hence it is not worth analysing the plot in that detail. Other studies, with better statistics, have shown for the same region that there is a diurnal cycle in NPF, albeit at slightly lower altitudes (Hermann et al., 2003, Fig. 9)
- p. 26 1. 773: Sorry, I did not get it, why are the short NPF events excluded from further analysis?
- p. 27 1. 775: whole Sec. 4.4: again very descriptive. What do we learn from these distributions about the underlying mechanisms?
- p. 28 1. 824: The diffusive properties of a particle does not depend on the number concentration, only on individual particle and gas properties. What you mean is that coagulation losses depend on number concentration.
- p. 29 1. 852: Initial UFP number is just one parameter of the simulation. How is the sensitivity of your simulation to the ambient conditions, i.e. pressure and temperature? And what is the influence of dilution on your simulation? Thinking about NPF in the outflow region of DCCs I would assume that the UFP concentrations will be diluted and hence the lifetime be extended.
- p. 30 1. 878: Sec. 5.1: section is too lengthy, please shorten.

- p. 35 l. 1022: Fig. 11 is nice to see, but again to not put in too much of interpretation on what you see. The figure shows nicely what is known from other studies namely that the main outflow region of DCCs is well below the tropical tropopause (i.e. at 350 – 370 K) and that above the air still ascends but with much lower vertical speed. Moreover clouds play a dominant role as above 380 K (i.e. “no” clouds) NPF is quite limited. That’s it. Do not make a too detailed description.

- p. 36 l. 1066 ff.: The role of different particle materials in NPF, this paragraph is a gain speculative. Either you provide more evidence from the ERICA measurements here, or you should delete this paragraph. The focus are gravity wave here and not the precursor gases. Anyhow, what would be missing again is the potential role of organics in the nucleation process as well as in the initial growth process. There are several publications on this, e.g. from measurements at CERN.

- p. 39 l. 1149: Similar to the authors, I believe that gravity waves play a role for the UT/LS aerosol. But unfortunately, the discussion about Fig. 13 is again pure speculation. Taking temperature fluctuations of +/- 1 K as indication for GWs, based on measurements that have +/- 2 K uncertainty is not valid. This is measurement noise. Please provide evidence that your data really show GWs, e.g. by providing the variance of the temperature measurements in this period compared to measurements in the same region and at similar altitude. Only if the variance for that period would be higher your discussion would be based on evidence.

- p. 40 l. 1173: The “Summary and Conclusions” section is in in some parts too speculative, please focus on your results and your line of evidence.

Figures:

Fig. 1 b): It would be valuable to indicate with a three colour code for the flight tracks which sections of the flight were clearly in the troposphere, which ones in the tropopause region and which ones above the local tropopause. This might already partly explain the observed UFP pattern.

Fig 2: a) and b): The legends are missing, i.e. “N4/N6”. Colour code legend as well. Please indicate the break level between the two data set by a line.

Is the concentration given at ambient conditions (as indicated) or at STP conditions?

Why is there no tropopause region bar for Fig 2 d)

Fig. 3: d): please add “nv” to “fraction” in the legend, otherwise the reader would have to guess which fraction is meant. Same in the text on p. 23, l. 655.

Fig. 9: I have problems with the representation in Fig 9. The plots contain so many data points that they overlap! This could, in practice depending on the used plot program, mean that for instance the blue area in Fig. 9 c represents 10% of the data in that region and the other 90% of the data in the same region could be yellow or red and be hidden below the blue ones. This could also be the reason why “no systematic structure is visible”, it might be hidden.

Please think about a different way of showing the data, e.g. to split each plot into four plots by limiting the colour code range for each of the for subplots or separating according to the measurement altitude.

In addition, the information “position” and “fasted transport region” are not visible in the plot, the figure is hard to understand without this information.

Technical corrections:

- p. 3 1. 90-94: Please split this lengthy sentence into two.
- p. 3 1. 92: Please add the chemical formula for dimethyl sulphide as well and start with a lowercase letter i.e. “dimethyl”.
- p. 3 1. 95: ... but also medium volcanic eruptions with VEIs of 4 not reaching the stratosphere directly, but indirectly via the TTL can play a role ...
- p. 8 1. 225: Please replace “aerosol” with “particle”.
- p. 8 1. 246: Please replace “size mode” with “size range”, as the ultrafine particles contain particles from two modes, nucleation mode and Aitken mode. Same in line 450.
- p. 9 1. 252: Not totally sure, but shouldn't it be “larger then” instead of “greater than”?
- p. 12 1. 352: The chemical names of the salts should start with lowercase letters.
- p. 32 1. 955: Please replace “vertically variable”. You mean that the uncertainties in the altitude coordinate in the reanalysis data is high, right?
- p. 39 1. 1135: What do you mean by “clear positive signal”? Number concentrations can only be positive.
- p. 41 1. 1202: Please replace “amounts” by “concentrations”.