Authors' replies to the reviews 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"

Please note the changed title that now reads as: In-Situ observation of New Particle Formation (NPF) in the tropical tropopause layer of the 2017 Asian Monsoon Anticyclone - Part I: summary of StratoClim results

On behalf of all authors, I would like to express my gratitude and appreciation to the two reviewers for their valuable suggestions and constructive critiques, which contributed decisively to optimising and completing the presented study. We hope to have adequately addressed all comments and objections and hereby submit a revised version of the article for re-evaluation and thank the reviewers in advance for their renewed efforts.

Anonymous Reviewer #1 (Rev1)

…

[Rev1]: The manuscript is well organized, but in many sections too lengthy.

> [Authors]: As both reviewers agree that the overall length of the paper should be reduced, much of the original text has been removed and some new aspects added, not least through the valuable suggestions of the two reviewers.

[Rev1]: 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.

> [Authors]: The section is significantly shortened to a useful level of information for the following discussion.

[Rev1]: 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.

> [Authors]: Please refer to the extensively revised manuscript version. The text has been changed by shortening unnecessary sections, removing speculation where it existed and rewording sections to better clarify the intent behind the statements made.

Specific remarks of the first Reviewer:

[Rev1]: As above, the introduction is quite long an 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.

> [Authors]: Since the two reviewers agree also on this point and consider the compilation of the state of knowledge and the background to be too detailed, the introduction as well as all parts of the rest of the article were significantly shortened and reduced to the essentials.

[Rev1]: Moreover, Sec. 1.2 has a misleading headline, this sections describes the Asian Monsoon Anticyclone and ATAL, but not the StratoClim campaign, please change.

[Authors]: corrected as suggested

[Rev1]: 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.

[Authors]: The list is reworded, please refer to the corresponding sentence in the revised paper version.

[Rev1]: p. 3 1. 89: Please cite Brock et al. 1995 as reference for the statement of the first sentence.

[Authors]: corrected as suggested

[Rev1]: p. 3 1. 101: Why do you give a 40% - 90% range for the SO_2 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.

[Authors]: The targeted text areas have been rephrased and rearranged according to the recommendation.

[Rev1]: p. 4 1. 114 ff.: In situ measurements of SO_2:
Rollins et al, GRL, 2017 and the discussion of their results is missing here!!!

[Authors]: The reference to Rollins et al 2015 is integrated in a new sentence at this point.

[Rev1]: p. 4 1. 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.

[Authors]: We agree with the suggestion and throughout the manuscript the term "ultrafine" has been replaced by "nucleation-mode". Also all axes labels in the figures were modified accordingly.

[Rev1]: p. 6 1. 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.

[Authors]: To better clarify the content of this article and to emphasise the differences between this article and the parallel-paper, the paragraph has been rephrased and restructured.

[Rev1]: p. 9 1. 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?

[Authors]: The expression denuder was replaced as suggested. In addition, the reviewer raises another important point, which is addressed and explained by an addendum in the same section of this article. However, the reviewer's question about the quantification of the re-condensate cannot be answered for any of the differently used evaporator systems under real and variable conditions (outside the very controlled laboratory conditions), as the effectiveness of the re-condensation depends on the device geometry, the atmospheric conditions, the chemical composition of the vapour or condensate, and the interactions between these factors. Please, refer to the revised paper version for clarification. Moreover, in Borrmann et al. 2010 measurements of a CPC with a real denuder on board the DLR Falcon were shown together with data from the COPAS setup. In the upper troposphere over Burkina Faso both showed a distinct feature in the vertical "nv" profile with numbers that were close to each other.

[Rev1]: p. 9 1. 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?

[Authors]: We agree with the reviewer that there has been an increasing focus on the study of organic aerosol components, particularly in the context of NPF. Certainly, organic aerosol components may have been detected up to altitudes of 8 km above the tropopause, although freshly formed nucleation-mode particles associated with these organics have not yet been detected in situ at these altitudes to our knowledge. Whether these organics are involved in NPF itself is just as possible as the option that organic vapours condense on particles that had formed by NPF (with reference to the CLOUD experiments at CERN). However, these possibilities are also discussed in the present article.

[Rev1]: 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.

[Autors]: At this point in the text, references and a short sentence were added. However, as the used vaporiser device in combination with the CPC does not provide any information on the oxidation state of organic aerosol species any statement on that based on COPAS measurements would be highly speculative. A different manuscript is in preparation, which discusses the oxidation state based on AMS mass spectrometric measurements. These, however, only include particles larger than ~70 nm.

[Rev1]: p. 10 1. 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 $0.85 \times N6 - 1.15 \times N15 > 0$? Where do the 0.8 and the 1.2 emerge from?

[Authors]: The corresponding text has been rephrased and expanded to make this point clearer. Please refer to the revised paper version.

[Rev1]: p. 11 l. 329 ff.: The discussion on the different NPF event terminology is confusing, please simplify.

[Authors]: The specified text has been revised and rearranged, please refer to the revised paper version.

[Rev1]: 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?

[Authors]: corrected as suggested

[Rev1]: p. 16 l. 470 ff.: What are the implications of the “However, ..” sentence for you simulation? Did you exclude such conditions from your simulations?

[Authors]: To clarify the statement, the sentence was rephrased - in fact, variable conditions, in particular on very short time scales, cannot be taken into account in the simulation.

[Rev1]: p. 16 l. 473: What is meant with “sample” here?
Can you please provide a time resolution, e.g. every minute or so?

[Authors]: Each sampling position along *Geophysica*’s flight track in 1-Hz resolution was meant. The revised paper version includes a rephrased sentence at this point.

[Rev1]: 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?

[Authors]: The statement does not raise doubts but explains why both data sets are used in parallel (for very different applications). ERA-5 offers new products with higher resolution than ERA-interim provided - yet ERA-interim is useful until it is not completely replaced by ERA-5. Until that time, when ERA-5 products fully replace ERA-interim, the latter still represents the state of the art.

[Rev1]: 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?

[Authors]: Basically, the sentence was an expendable rudiment. After a new review and a corresponding demand for general cuts in the article, this sentence was deleted without replacement in the revised version of the paper. Furthermore, in our opinion it is quite reasonable to choose exactly the same temporal (spatial) resolution for the trajectory analysis as is given for the detection of NPF events. Any higher resolution would further complicate the analysis, a lower resolution of the trajectory analysis might not allow for investigating the fine structure in the variable NPF rate of the observed events, i.e. particularly intensive NPF events would then not be investigated with the coarse resolution trajectory analysis.

[Rev1]: p. 18 1. 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?

> [Authors]: The idea was to use two different approaches in the analysis of the air mass history in order to find a mutual support/confirmation of our findings. The different resolutions result from the requirements, which were defined by the operators of respective tool and are deliberately not adapted to each other. Furthermore, the two analyses were deliberately kept as independent as possible so that the resulting conclusions of both methods can be compared without bias. The model set-ups of TRACZILLA and CLaMS are both very different. The TRACZILLA trajectory calculations include higher statistics and account for vertical diffusion. Vertical velocities are calculated differently in the two approaches. CLaMS uses the total diabatic warming rate from the reanalysis forecast, while TRACZILLA uses only the radiative contribution (e.g. Bucci et al., 2020). Moreover, TRACZILLA bases its convective identification on satellite observations of cloud tops. Detailed comparisons between both approaches and further insights will be provided in a campaign overview paper which is currently in preparation.

[Rev1]: p. 18 1. 528-530: There are previous publications who have used this approach, e.g. Weigelt et al., 2009, JGR. Why do you cite these two?

> [Authors]: The approach of Weigelt et al. may have been similar. The citations in this article refer to this approach using TRACZILLA. In the revises paper version, the reference to Weigelt et al. was included and the use of TRACZILLA was specified more concretely.

[Rev1]: p. 19 1. 548: What do you mean with “certain” distance?

> [Authors]: The concretisation "at a linear distance of more than 500 km" was inserted at this point.

[Rev1]: p. 19 1. 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?

> [Authors]: Here, also, the corresponding text passage was concretised in order to eliminate misleading formulations, please refer to the revised paper version.

[Rev1]: p. 20 1. 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.

> [Authors]: We disagree with the reviewer on this point, as in Fig 2 and section 4.1 the median number concentrations (ambient) are compared with the percentile series of the different sites to illustrate the height levels of the main NPF activity observed at the different sites. In contrast, in Fig3a and connected text sections, the mixing ratios of the median distributions are compared - this has a different objective and relates to other

units of particle density. One should also keep in mind, that the two figures are particularly valuable in the light of the observation by Williamson et al., 2019. They provided tropical ATOM results albeit only for altitudes below 10 km. The Figures 2 and 3 highlight the (very few) tropical locations on 3 continents for much higher altitudes. Therefore, Fig3a should be seen as a transitional element rather than a repetition. To clarify the intentions, short motivation statements for the two representations have been added to the text to emphasise the differences in the content of the images. Nevertheless, unnecessary text has been eliminated in the revised paper version.

[Rev1]: p. 20 1. 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.

[Authors]: Basically, the measurements are from different altitudes without overlap. This is mentioned in the submitted paper version: "Panels a and b) exhibit merged data of two independent CN-detectors with individual dp50 (i.e. N6 for $\theta > 350$ K and N4 for $\theta < 350$ K), which were deployed on individual aircraft, the M-55 Geophysica and the DLR Falcon-20 (cf. Borrmann et al. (2010) and Weigel et al. (2011))." The different cut-offs were not treated. The main point of this figure is to illustrate the layers of NPF occurrence at different locations.

[Rev1]: 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.

[Authors]: done as suggested

[Rev1]: 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.

[Authors]: We agree with the reviewer that there was a lack of clarity in the structuring described. The revised paper version corrects the structure.

[Rev1]: p. 22 1. 644: Fig 3b: Why is $N_{6\text{-}15}$ 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.

[Authors]: Details on that were provided in the Figure's caption and now are also added in the text with a reference to the paragraph about the definition of n_{nm}.

[Rev1]: 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.

[Authors]: We agree with the reviewer's opinion that we have not made clear enough why the representation in Figure 3 c and d is exactly the way it is. We agree with the reviewer's suggestion and we have made additions both in the caption and in the text.

[Rev1]: 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 N6 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.

> [Authors]: We follow the reviewer's opinion and we have shortened the section considerably.

[Rev1]: 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?

> [Authors]: Also because Reviewer #2 has raised concerns at this point, we have changed Figure 4 to increase the information content of the figure. We have also removed a large part of the descriptive text. However, in the course of the image renewal and to answer the (very important) questions posed here, we had to add another part of the text. Please see the revised paper version.

[Rev1]: 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)

> [Authors]: We have revised the section, reduced the interpretations and we have emphasised that the observations are based only on the limited StratoClim dataset, whereas previous measurements in the same region bear much better statistics for detecting the diurnal dependence of NPF. Please, refer to the revised paper version.

[Rev1]: p. 26 1. 773: Sorry, I did not get it, why are the short NPF events excluded from further analysis?

> [Authors]: In our opinion, the evaluation of events with too short duration creates a falsifying weighting in the event-wise analysis due to uncertain data points. Above all, this method filters out the singular 1-2 second features, which can also be noise events. For the evaluation of NPF events, arithmetic averages of concentrations or mixing ratios over less than five data points is not very robust under highly varying conditions. Finally, the accuracy of the duration for very short NPF events is comparatively low because the running mean of the raw signal smooths the temporal discrimination of short events. Please refer to the revised paper version at this point.

[Rev1]: p. 27 1. 775: whole Sec. 4.4: again very descriptive. What do we learn from these distributions about the underlying mechanisms?

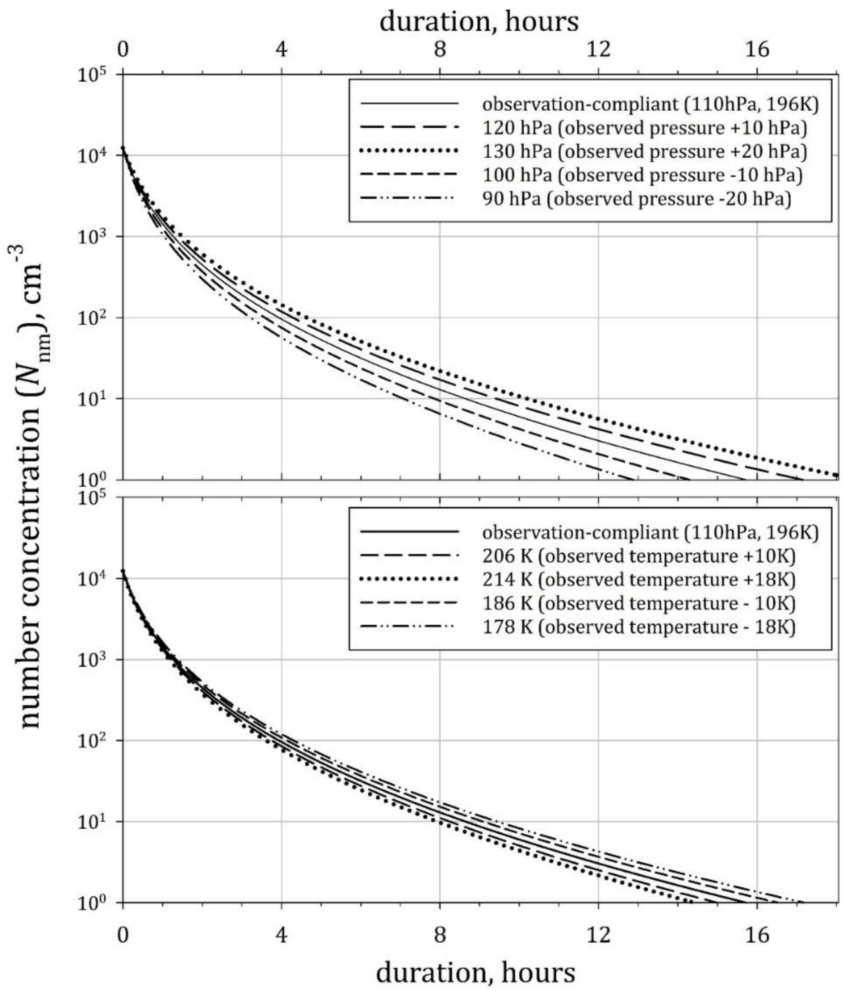
[Authors]: In correspondence to the reviewer's suggestions, we have shortened and streamlined the text. However, we would like to emphasise at this point that in Section 4 we are thematically in the descriptions of observation. Here we will be concerned with the classification of events in the metrics of time and location and their integration into different frames of reference. Nothing else is done in section 4.4. The NPF events related to the lapse-rate tropopause are a basis for the discussion (location within the TTL, range of action of convection, etc.) in subsequent sections. A new variable is the equivalent latitude. The relation of NPF events to the centre of the anticyclone is provided for the first time in this article. Nevertheless, the text in section 4.4 has been revised to make these aspects clearer to the reader. Please refer to the revised version of section 4.4.

[Rev1]: 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.

[Authors]: In fact, a misleading formulation had been chosen here. In the revised paper version this sentence has been rephrased. Please, refer to the new text at this point.

[Rev1]: 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.

[Authors]: Here the reviewer made a particularly important point. The simulations were repeated with stepwise deviations of pressure and temperature from the conditions encountered in the measurements (110 hPa, 196K) (see results in the two figures below). There is no significant dependence of the simulations observable unless pressure and temperature deviations are exaggerated. - additions to the text were made accordingly at the appropriate point. Regarding dilution, we would like to point out to the reviewer that the simulations made based on one tenth of the aerosol concentration should correspond to a dilution, and the simulation does not result in increased persistence of the nucleation-mode particles at this point. However, the reviewer is correct that dilution lowers the coagulation rate ($\Delta N/\Delta t$), but the duration for which an NPF event is detectable is insignificantly prolonged, as shown in the simulation (within the paper) for the case of the initial particle concentration multiplied by 0.1. Further dilutions would result in parallel-shifts of the curve towards lower particle number concentrations.



[Rev1]: p. 30 1. 878: Sec. 5.1: section is too lengthy, please shorten.

[Authors]: The section is significantly shortened in the revised paper version.

[Rev1]: p. 35 1. 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 ascents 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.

[Authors]: We gratefully accept the reviewer's advice and have thoroughly revised the text for this illustration (former Fig 11 now Fig 12). In this process, some of the descriptive text has been reduced to a level that is at least necessary to explain the figure and convey the intended message. The link between convective transport and NPF is not as systematic and universal as assumed. Individual observations point to a limitation of this general view. Taken on their own, Fig11 (now Fig 12) and the explanation are of course only a weak indication. However, the article combines very different aspects in a chain of indications that supports that the connection between convective transport and NPF is less direct than generally assumed.

[Rev1]: p. 36 1. 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.

> [Authors]: 1) The section is reduced to the text on the observation made and freed from speculation. 2) Regarding the second point, we would like to refer to the wording of the submitted paper at this point, where it says "The presence of ammonium species (Höpfner et al., 2019) or organics could then promote the NPF of H2SO4 in the TTL even at low supersaturations (Metzger et al. (2010); Kerminen et al. (2010); Kirkby et al. (2011); Kürten (2019)).". The role of organic substances was therefore not disregarded in any way and the four references listed on this point at the end of the sentence all lead to the CERN experiments. The ERICA instruments detects only particles larger than ~70 nm, which is too far from the nucleation mode. For this reason it is not possible to state for the detected organics whether they were part of the nucleation process itself or simply later condensed onto the freshly made particles.

[Rev1]: p. 39 1. 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.

> [Authors]: We agree with the reviewer that the time series shown is only a shallow indication of the contribution of a wave-like temperature fluctuation. In the revised version, a detailed analysis of the fluctuation was carried out taking into account the stated accuracy (not uncertainty) of the temperature measurement. By applying a low-pass filter to the data, the noise of the temperature measurement could be extracted. Within two time windows comprising the two NPF events and further time intervals of the measurement, the noise level remains almost constant with a maximum scatter of +/-0.75K. A fit of the filtered data was achieved by an overlaid wave fit. With this approximation, a wave character could be more clearly indicated for the temperature fluctuation, which remained after noise filtering during the NPF events.
>
> The described analysis is included in the text below the previously shown Figure 12 (with discussion) as a new Figure 13 (with explanation). The detailed qualitative description of the analysis procedure as well as the effects of small temperature variations on the potential NPF rate have been added in appendices in the revised version.

[Rev1]: p. 40 1. 1173: The “Summary and Conclusions” section is in in some parts too speculative, please focus on your results and your line of evidence.

> [Authors]: The vast majority of this section summarises the observations made, and the conclusions in this section are freed from unproven conjecture.

Figures:

[Rev1]: 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.

> [Authors]: We understand the point the reviewer is trying to make here, but plotting the flight paths with a colour code of altitude was not going to provide any meaningful information in the overlay of different flights with the same pattern over Nepal. Instead, we think that the new version of Figure 4 and the numerous other vertical profiles provide the desired and essential information.

[Rev1]: 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)

> [Authors]: The fact of a faulty image export was overlooked. The corresponding figure in the revised version contains the missing information.

[Rev1]: 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.

> [Authors]: The fraction *f* was defined in section 2.1.1 which has moved in the revised version to a new Appendix. The figure's caption specify the fraction *f*, with equation and explanation. Nevertheless, for clarity we added a remark in the image.

[Rev1]: 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.

> [Authors]: We understand the point the reviewer is making here. In the revised version we have split the images and presented them in two separate figures. For each of the new figures, two levels of zoom-ins have been made. With each new zoom level, the data points are moved apart and the overlap of data points is clearly reduced as much as possible. The data points are displayed semi-transparently. All of this should avoid the

unnoticed obscuring of data points by overlaps. The text and image numbering as well as the captions have been adapted accordingly.

[Rev1]: In addition, the information “position” and “fasted transport region” are not visible in the plot, the figure is hard to understand without this information.

[Authors]: Please refer to the answer to the previous question and the revision of the article. We are convinced that the identified limitations of recognisability could be eliminated with the re-edited paper version.

Technical corrections:

[Rev1]: p. 3 1. 90-94: Please split this lengthy sentence into two.

[Authors]: done as suggested.

[Rev1]: p. 3 1. 92: Please add the chemical formula for dimethyl sulphide as well and start with a lowercase letter i.e. “dimethyl”.

[Authors]: done as suggested.

[Rev1]: 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 …

[Authors]: The remark was added (slightly modified).

[Rev1]: p. 8 1. 225: Please replace “aerosol” with “particle”.

[Authors]: done as suggested.

[Rev1]: 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.

[Authors]:corrected.

[Rev1]: p. 9 1. 252: Not totally sure, but shouldn´t it be “larger then” instead of “greater than”?

[Authors]: The validity of the expression used has been confirmed by a native English speaker.

[Rev1]: p. 12 1. 352: The chemical names of the salts should start with lowercase letters.

[Authors]: corrected.

[Rev1]: p. 32 1. 955: Please replace “vertically variable”. You mean that the uncertainties in the altitude coordinate in the reanalysis data is high, right?

[Authors]: At this point, the entire sentence and the preceding one could be deleted for the purpose of shortening, as the uncertainties are dealt with in section 3.3 anyway.

[Rev1]: p. 39 1. 1135: What do you mean by “clear positive signal”? Number concentrations can only be positive.

[Authors]: The misleading formulation has been replaced. Please refer to the revised paper version.

[Rev1]: p. 41 1. 1202: Please replace “amounts” by “concentrations”.

[Authors]: The expression "amounts" was replaced by "numbers".

Anonymous Referee #2 (Rev2)

GENERAL REMARKS

[…] The manuscript is well organized and presents interesting and relevant scientific findings, however, it lacks a precise and compact representation. The far too detailed description of the instrumentation and methods (18 manuscript pages including the introduction) combined with complex and difficult-to-understand figures gives the reader a hard time to follow the red line of arguments.

[Authors]: As both reviewers agree that the overall length of the paper should be reduced, much of the original text has been removed and some new aspects added, not least through the valuable suggestions of the two reviewers.

In the present version of the manuscript, many of the excellent findings may get lost because the key results are hidden in the details of the extensive descriptions and discussions. In addition, arguments are repeated, and the manuscript contains a significant amount of textbook knowledge and repetition. […]

[Authors]: Please refer to the new manuscript version. The text has been extensively revised by shortening unnecessary sections, removing speculation where it existed and rewording sections to better clarify the intent behind the statements made.

SPECIFIC COMMENTS

[Rev2]: 1. Introduction: In the first section of the introduction, the role of SO2 and sulphuric acid in particle nucleation are widely discussed, but immediately afterwards a dedicated section on new particle formation follows which extends over more than two manuscript pages. I suggest merging the sections and reducing the length significantly by focussing on new particle formation.

[Authors]: We agree with the reviewer's suggestion to shorten the text to the bare essentials, which has been done in large parts of the introduction, but we would also like to keep the two sections separate, as the subjects of the two paragraphs point in very different directions, although they are both part of the of the NPF's theme.

[Rev2]: 2. Instrumentation and Methodology: In the field study, a set of established and well characterised instruments has been deployed. However, the instrumentation section stretches over six manuscript pages. There is certain danger that readers will drop out here and miss the interesting parts. The authors should please focus here on the relevant information, omit textbook knowledge, and refer to published instrument papers instead. This section can also be shortened significantly.

[Authors]: The sections on Instrumentation and Methodology were also significantly shortened or simplified in the revised version of the article to strengthen the essential content of this section, as reviewer #2 rightly notes here. For example, a subsection on the aerosol evaporator has been moved to a new appendix so that the reader has the necessary technical details at hand without them interrupting the flow through the main text.

[Rev2]: 3. Observations and Results: The discussion and interpretation of figures appears to some extend speculative which is indicated by the frequent use of terms like “the impression could arise“, “may also indicate”, “may primarily”, etc. See as examples the paragraph from line 749 to 766, or the paragraph from line 924 to 938. Please also check the frequent use of the term “however”. I suggest focusing on the description of the excellent observations and well-founded explanations, while avoiding extensive descriptions of the figures. By doing this, a significant reduction of the text can be achieved.

[Authors]: We have revised large parts of the Observations and Results - also because Reviewer#1 expressed concerns here - and in particular reconsidered the frequent use of certain phrases as well as shortened text wherever the given information was considered dispensable. Please refer to the revised paper version.

[Rev2]: 4. Summary and Conclusions: This chapter is far too long and repeats a significant amount of content discussed before. The summary and conclusions chapter should deliver the key messages of the study without repeating the details. An adequate way may be using bullet points. In the current form of the manuscript, the key messages get lost and the scientific impact of the study is diminished.

[Authors]: We agree with the reviewer that the summary section needed to be revised. In the revised version we have better focused the main findings as well as the conclusions and removed dispensable repetitions.

[Rev2]: 5. Figure 4: The complexity of the figure can be reduced significantly by switching to a log-scale representation of the occurrence frequency. Then the inserted figures are no longer needed, and the message comes across smoothly.

[Authors]: We have - also because Reviewer#1 has raised concerns at this point - changed Figure 4 to increase the information content of the figure. Please see the revised paper version.

[Rev2]: 6. Figure 6: This figure is difficult to digest. Panels a and b show the data coloured by flight date which is not telling much. Panels c and d show the same data but coloured by CO mixing ratio as an indicator for boundary layer influence. In fact, only panels d and e are needed since they transport the key message that observations associated with low CO mixing ratios are located above the thermal tropopause whereas high CO mixing ratios are located well below the thermal tropopause.

[Authors]: We do not fully agree with the referee’s opinion as the flight-wise coloured profiles illustrate two things: A) they represent how the observed phenomena distribute over the campaign period (with Fig 3), over the time of day (with Fig. 5), and in relation to the flight patterns (with Fig. 1).

B) through this illustration it becomes clear that some of observations were made consistently over several or all flights.

Nevertheless, the text has been shortened. Moreover, the revised version of Figure 6 is reduced to four panels. The revised text is freed of statements that relate to the earlier Figure 6 and the message of the new figure is more focused. Please refer to the revised paper version.

[Rev2]: 7. Figure 9: I question the value of this figure and the transported message is not clear to me. In case the authors want to establish a link between the number density of ultrafine particles and the transport time from the boundary layer, another form of illustration is needed. Potentially, this figure can be moved to the supplement since Figure 10 conveys the message right away.

[Authors]: Reviewer#1 has also expressed concerns (albeit different ones) about Fig. 9 - therefore, and due to the reservation mentioned here, the former Fig. 9 has been revised and split into two figures (9 and 10) in the revised article version. However, the statement made here does not correspond to what can be seen in the former Fig. 10 (now Fig. 11). The new Fig. 11 shows in the target height (theta) under NPF how fast the previous transport was. In contrast, Fig. 9 (now Fig. 10) shows the geo-position of the fastest ascent (which does not necessarily correspond to the location and altitude where the NPF event was detected).

MINOR ISSUES

For the current version of the manuscript, I refrain from discussing minor issues but offer a review of the revised and shortened – best to about half of the current size – manuscript with a stronger focus on the key results and findings.