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Atmospheric Chemistry and Physics Discussions

Interactive comment on "In situ observations of new particle formation in the tropical upper troposphere: the role of clouds and the nucleation mechanism" *by* R. Weigel et al.

Anonymous Referee #2

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Weigel et al. compare measurements of the concentrations of particles greater than 4/6 and 10/15 nm using condensation particle counters with different size sensitivities. Regions where concentrations at the small and large sizes differ by more than 100 cm-3 are identified as regions of new particle formation (NPF). These regions are then characterized in terms of altitude, frequency, relationship to clouds, spatial scale (to be added), and compared with model predictions of so2 concentrations and particle nucleation from h2o-h2so4. The results are quite interesting and add to our understanding of the importance of the tropical tropopause layer as a source region for stratospheric constituents, particularly so2 and particles. The paper is well organized and well written, except for a few small problems mentioned below, and should be published when

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these are corrected.

The one somewhat major comment has to do with Fig. 4 and the FLEXPART modeling. What is the grid scale for the FLEXPART model and how does it compare to the "spatial scale" in Fig. 4. Although the abscissa of Fig. 4 is time it represents a horizontal distance traveled by the a/c particularly for the South America flight. Since the air mass trajectories are so long for the NPF events, what leads to the high temporal/spatial variability of the so2 plumes?

A more difficult and more important question is, what fraction of the time were so2 plumes observed corresponding to flight legs when there was no NPF observed? Would the so2 concentrations on other flight legs have a similar appearance as the ones associated with the two flight legs shown, even when no NPF events were observed? This question may exceed the work envisioned for this paper, but such a nice correspondence here, naturally raises the question as to how often such a correspondence is observed. NPF events were observed on half of the South American flights and all African flights, so if the modeling is not too hard, a statistical answer would strongly reinforce the importance of so2 to the NPF events observed.

It would be nice to provide a rationale for the selection of case studies since there were many NPF events observed. Why for example was a ferry flight used for the South American case rather than a targeted scientific flight?

Detailed comments:

9255.3-5: It is not obvious how "mixing of two subsaturated air masses can result in supersaturation". If they are both subsaturated, mixing them will not change that.

9257.20: . . . described the transport . . .

9257.24 . . . predominantly originates from . . .?

9266.20: 350-370 K is a little restrictive for the high concentration layers for SCOUT-AMMA, where the layers extend to 340 K.

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9267.6-20: Suggest replacing all of this text, which is quite confusing, with the following. For the COPAS measurements analysis a conservative definition of an NPF event is used, which includes the measurement uncertainty of 15% for each COPAS channel. Thus, a positive difference between N6 and N15 is only considered to be an NPF event if 0.85*N6 – 1.15*N15 > 100 particles per cm3. At background conditions, without NPF, the subtraction of number concentrations that are measured by two independently operated CPCs lead to positive and negative values of Nuf that statistically vary around zero. Intentionally a conservative approach was adopted here. Application of other NPF criteria, as suggested by Lee et al. (2003), on our measurements would lead to more frequent observation of NPF events with longer duration of each event.

9267.11: I do not understand the point of the following sentence, "According to a measurement uncertainty of 10% an NPF criterion, equivalent to the one used for the COPAS data, was applied on the measurement data from the DLR CPC system." Was a different uncertainty applied to the DLR CPC system? What is meant with this sentence? Is this necessary to include?

9268.6: . . . at corresponding altitude levels . . .?

9269.12: Here and elsewhere (9276.8, 9284.5, . . .) this is not the proper use of according, in English. Replace it with corresponding.

9269.6-16: The times of various NPF events and speeds of the various a/c are not all that interesting unless the picture is completed by providing the spatial scale of the events. This is what is interesting, not the time over which the events are observed in a speeding a/c. In fact, skip the detail about a/c speeds and times of observation, and just say that, based on the time over which the events were observed, and the relative a/c speeds, the spatial scale of the NPF events ranged from x – y km. Same for Table 1. Replace the last three columns with the distances representing the NPF events.

9271.13: . . . over Brazil . . .

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9273.20-23: Why mention 4-7 days? This represents less than 50% of the so2 in South America and well less than 50% in Africa. An equivalent or larger fraction of so2 comes from 15-20 days which implies synoptic scale lifting. This raises the question as to why the great temporal dependence of the so2, see the major question above?

9281.9-11: The standards for agreement seem to have relaxed somewhat here. The measurements fall within the four order of magnitude range covered by the model results, so yes it should be easy to find a combination of so2 concentrations and surface area which match the measurements; however, all would require low values of so2 and surface areas between 2 and 1 cm-3.

9282.19-25: It is not clear where the CO mixing ratio numbers come from. For example in the CO range from 67-82, Nuf ranges from 4-4000 cm-3. But also in this Nuf range CO ranges from 67 to over 90. So where does 82 come from? I assume it is the highest CO value when Nuf < 100 cm-3, but even this isn't quite true as Nuf is < 100 for CO up to 85.

9283.24: Do not use a colon here. Make it two sentences to not confuse the reader.

9284.10: What is meant by, "(see . . . black curve in Fig. 10)? That curve is Nuf and not related to backscatter or depolarization.

9284.11-12: . . . up to and 4000 ???

9286.9-11: Change to, "New particle formation, which was observed on half of all flights over South America and during each local flight over West Africa, was confined . . .",

9286.14: Suggest, . . . were to large extent volatile . . ., but it should be quantified with a per cent or fraction.

9287.10-12: The conclusion from this statement, " Nucleation mode particles were detected not only in clear air but also within thin cirrus cloud layers, indicating that new particle formation occurred in both, clear air, as well as in clouds.", does not follow from the modeling shown here. The observation, as the modeling shows, is not made at the

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time of the new particle formation, but measures the residue of such an event. Thus, high concentrations of Nuf in a cloud does not imply NPF in the cloud, merely that the resulting Nuf were still present even in cloudy air. The authors do qualify this statement in the following text, but not before this bold statement is made. The qualifications should rule and the bold statement eliminated or strictly qualified.

9287.19-22: These statements are again not strictly supported by the modeling or observations. In all case studies the times of nucleation in the NPF events always occurs 1-2 days prior to the observation. This is not consistent with nucleation occurring in thunderstorm anvils which are only a few hours old. Thus this has to be qualified as a pathway far upstream of the observations for providing source material to the TTL.

Fig1: How are the N4 particles shown? There are no symbols included in the N4 legend. What do the percentile gray scales refer to, the N6 or N4 particles? Where are the "red dots"

Fig2: The caption is confused. The description for b) is for c), and c) for d), and there is no description for b). Where is the black scatter-line plot?

Fig. 4: Include on the figure, with another scale, the particle number concentrations as in Fig. 3, so the correspondence between so2 and NPF can be seen clearly.

Fig. 6: There are no cyan circles on the plot.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 9249, 2011.