

## ***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 #1**

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Review of the ACP manuscript acp-2011-191 “In situ observations of new particle formation in the tropical upper troposphere: The role of clouds and the nucleation mechanism” by Weigel et al.

General Comments:

The manuscript “In situ observations of new particle formation in the tropical upper troposphere: The role of clouds and the nucleation mechanism” by Weigel et al. deals with in situ measurements of aerosol particles in the tropical upper troposphere and tropopause layer carried out with the Russian Geophysica and the German Falcon aircraft. The focus on the study is on new particle formation events, which are interpreted

C2171

by using measurement and model results. As these particles are supposed to maintain the stratospheric aerosol layer, the subject is of high interest to ACP and I suggest to accept the manuscript, however, with some changes as described below. In particular I find it important to emphasize what is new/different compared to the paper by Borrmann et al., ACP 10, 5573–5592, 2010, who used the “same” data (cf. e.g., Fig. 1 in this study and Fig. 6 in Borrmann et al.) from the same campaigns and came to similar conclusions e.g., the broad particle concentration maximum in the 340 K to 390 K altitude range and its role in maintaining the stratospheric aerosol layer. There are new/different data/findings in the manuscript, but this should be emphasized.

Generally, in some parts the paper is too lengthy. For instance the “Introduction” should be shortened (this is not a review paper on the source of stratospheric aerosol particles), rearranged, and focused on the for this study essential issues. For instance, on page 9252 the discussion on SO<sub>2</sub> as particle precursor gas is started but not completed and is restarted again on page 9254.

Furthermore the purpose of the microphysical box modeling is not clear to me. When you look into the literature you will find that parametrizations of particle formation vary over a wide range in the relevant parameter space (cf. e.g. Zhang et al. JGR, 2010). Hence, as long as there is no progress on understanding the relevant mechanisms, one will always find a nucleation parametrization which reproduces the observed concentrations. But this is no proof. A similar approach as chosen by the authors here, trajectories and a microphysical box model to explain observations, was already made by others, e.g., Kanawade and Tripathi, JGR, 2006 (by the way, this reference is missing in the manuscript). So please convince me and make it clear to the reader why the MAIA simulations are helpful and what do we learn from it what we didn’t already know.

On the other hand, the paper addresses one important question in UT/LS aerosol science: Does particle formation take place inside cirrus clouds and if yes, under which conditions? The authors should strengthen this part (maybe there are even more cases

C2172

available for analysis). Likewise, the information that particle formation takes place mainly in the lower part of the TTL is important the question is why there and not above.

Specific Comments:

- p. 9251 l. 9, The term “ultrafine particles” is nowadays used for particles smaller than about 100 nm. The particle size range the authors refer to, 6-15 nm, is better described by “nucleation mode particles”. Please change accordingly in the whole manuscript.
- p. 9252 l. 17, For the ultrafine particles (< 100 nm) sedimentation is not important, even at the low pressures at the tropical tropopause. Please remove “sedimentation” from the list of microphysical processes.
- p. 9252 l. 24, Remove “lower”.
- p. 9252 l. 26, The authors describe the stratospheric aerosol life cycle, but if you do so, please include all important processes, e.g., the sink of stratospheric aerosol particles is the transport into the troposphere, either by sedimentation, or tropopause folds, or downward transport in the polar vortex.
- p. 9253 l. 29, The non-volatile components, please specify which inorganic salts are meant. And concerning the mineral dust, I doubt that it reaches altitudes of 11 km or above. In the literature one can find individual cases where this was the case, but mineral dust is likely not a component of the majority of aerosol particles in the UT/LS. If the authors are aware of references with opposite statements, please provide them to the reader.
- p. 9254 l. 20, Barth et al., 2001 is only one reference. There are other references which state that only a smaller fraction of the SO<sub>2</sub> survives the transport inside deep convective clouds, and not the “major fraction” (e.g., 30% given by Ekman et al., 2006, J. Atmos. Sci.). In my point of view the fraction of SO<sub>2</sub> reaching the upper troposphere is one of the important open questions with respect to UT/LS particle formation.

C2173

- p. 9261 l. 13, Where there no trace gas measurements on the Falcon or are they not used?
- p. 9262 l. 10, Where are the 31 additional level for simulating the SCOUT-AMMA flight located, were they just added above or was a higher vertical resolution used? Why are the numbers of vertical levels different for the two campaigns?
- p. 9264 l. 29, The estimation of the pre-existing particle surface area, where does it come from, which numbers or references are used?
- p. 9265 l. 20, “The MAIA simulations of this work serve as sensitivity study” for what? Considering all the uncertainties likely only an estimate on which of the input parameters is the most critical one can be obtained, but probably not a quantitative result.
- p. 9268 l. 6, I don’t see that the Falcon measurements exceed the Geophysica measurements. For an easier comparison, a log-scale on the x-axis of Fig 2.a would help.
- p. 9269 l. 1, Remove the sentence “It can be speculated ...”, it is of no help to understand the data.
- p. 9269 l. 11, What is the reason to provide the aircraft speed here. In my point of view the sentence can be removed. (And by the way, the True Air Speed, TAS, and not the ground speed would be relevant to derive spatial scales from measurement periods.)
- p. 9269 l. 16, Reading the description of Fig. 2 in the above paragraph is a little bit exhausting, wouldn’t it be enough to describe the shape of the curves and to discuss the statistical numbers provided in Tab. 1?
- p. 9273 l. 22, As you are making case studies I would say that it is not totally impossible to distinguish between the different kinds of vertical transport. You could take respective weather maps and satellite pictures together with your FLEXTRA trajectory and see which kind of cloud (convective, frontal) is prevailing in the region with the

C2174

strongest SO<sub>2</sub> source contribution. This would be quite a simple exercise.

- p. 9273 l. 17, In order to estimate how reliable the SO<sub>2</sub> loads to the air masses are (calculated with FLEXPART), please plot the whole flight track together with the respective SO<sub>2</sub> loads in Fig. 4. In this way it would be possible to see, if the high SO<sub>2</sub> loads at the time of particle formation are just coincidence.

- p. 9276 l. 15, Where do the numbers for the particle surface area come from? Please give a reference or describe why you have chosen these values.

- p. 9277 l. 7, The calculations of the new particle formation shown in Fig. 6 partly range back 6 days. I can not imagine that the respective air masses/trajectories didn't have any cloud contact within this period, even considering the high altitude of the events. Most of the trajectory path lies in the tropics! And on p. 9280 you even state that "cloud processing along the trajectory" is likely. However, if a cloud contact occurred particle concentrations will be different (scavenging, particle formation). Hence how realistic/reliable is this modeling?

Technical Comments:

- p. 9252 l. 18, The sentence starting with "Strong new ..." prolongs over six lines. Please shorten or separate into two or three sentences.

- p. 9253 l. 25, Make the description of the aerosol particle composition a new paragraph.

- p. 9257 l. 12, I'm not a native speaker, but "beside others" seems not to be the right expression to me.

- p. 9257 l. 14, Please use "LMS" for lowermost stratosphere, because the lower stratosphere (LS) is a different region.

- p. 9257 l. 20, "describe" instead of "described"

- p. 9257 l. 20, Remove "at".

C2175

- p. 9257 l. 20, What is meant with "transformation of H<sub>2</sub>O"?

- p. 9257 l. 21, The "impact" of what "on the stratospheric water budget"?

- p. 9258 l. 3, Remove "Fierli et al." reference, it is already provided in the previous sentence.

- p. 9258 l. 21, Exchange "has been" with "was".

- p. 9258 l. 27, Exchange "size" with "particle".

- p. 9259 l. 2, Exchange "These" with "Generally, CPC".

- p. 9259 l. 3, Insert a comma before "which".

- p. 9260 l. 2, Is "automation" correct or shouldn't it be "automatic"?

- p. 9260 l. 8, "tropics of the Americas" should rather be "tropics of Central and South America" as I don't see any tropics in North America.

- p. 9260 l. 11, Remove the "of"s before the years.

- p. 9260 l. 20, Remove "size".

- p. 9260 l. 21, Exchange "size" with "diameter".

- p. 9261 l. 2, Please specify what "in the vicinity of the aircraft" means, 10 m, 100 m, 1000 m?

- p. 9263 l. 22, Please insert "freshly formed" before "aerosol particles".

- p. 9264 l. 3, Remove "(decay probability)", this was already introduced as synonym for stability the sentence before.

- p. 9264 l. 5, The sentence starting with "The thermodynamical data ..." prolongs over six lines. Please separate into two sentences.

- p. 9264 l. 10, A trifle, but sometimes "see" and sometime "cf." is used to refer to a

C2176

paper or figure. Please harmonize in the whole manuscript.

- p. 9265 l. 8/9, Please use either "particle size range" or "diameter range" but not "diameter size range".
- p. 9266 l. 15, "data set (...) is a summarized result", please change wording.
- p. 9266 l. 20, Change to either "is between" or "ranges from ... to".
- p. 9267 l. 13, Change "applied on" to "applied to".
- p. 9267 l. 14, The sentence "Intentionally a conservative ...", please rephrase this sentence.
- p. 9267 l. 18, Add an "s" to "lead".
- p. 9268 l. 6, Replace "according" with "respective", similar p. 9269, l. 12.
- p. 9270 l. 2, Shouldn't it be "Fig. 2c and d"?
- p. 9271 l. 26, Replace "by" with "according to".
- p. 9273 l. 16, Remove the heading "Results of ...", it has no numbering and is actually not needed.
- p. 9274 l. 9, The reference "Kazil and Lovejoy, 2007" is missing in the reference list. Or it must be "Kazil et al., 2007" or "Kazil and Lovejoy, 2004" here.
- p. 9282 l. 21, Remove "(Nuf)", you use this abbreviation already before.
- p. 9283 l. 15, Please use past tense when describing measurements.
- p. 9283 l. 18, Change "in average" to "on average".
- p. 9283 l. 21, Must be "Table 3", do. p. 9284 l. 10.
- p. 9284 l. 12, Remove "and".
- p. 9286 l. 2, Change "aircraft borne" to "aircraft-borne".

C2177

- p. 9286 l. 7, Please separate the sentence, the second sentence starting with "One NPF event ...".
- Tab 3: Please make the diameter ranges for the surface area a new line.
- Fig. 1: c) There are not "red circles" as mentioned in the figure caption.
- Fig. 5: Caption: l. 3 Exchange "or" with "and".
- Fig. 6: Please, make the figure larger. Caption: l. 8 There are no "cyan circles" in the figure, but "green bars", please correct.

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

C2178