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Comment on acp-2020-1285

Anonymous Referee #1

Referee comment on "In situ observation of new particle formation (NPF) in the tropical tropopause layer of the 2017 Asian monsoon anticyclone – Part 2: NPF inside ice clouds" by Ralf Weigel et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1285-RC1>, 2021

Review

The authors present in-situ observations with a high altitude aircraft of size-resolved ultrafine aerosol particle concentrations, ice water content, ice particle size, carbon monoxide mixing ratio, and meteorological quantities in the tropical tropopause layer over Nepal, India, and Bangladesh, during the Asian Monsoon season in 2017.

During the eight flights, recent nucleation of ultrafine aerosol from the gas phase (new particle formation, NPF) was identified from the size-resolved ultrafine aerosol particle concentrations. NPF events that produced large amounts of new particles in the altitude range 11-16 km were identified both in clear air and cloudy air, with cloud ice reaching number concentrations as high as 3 cm^{-3} . While earlier in-situ observations have identified NPF inside cirrus clouds, and numerical modeling showed that NPF in cirrus clouds is possible based on known mechanisms, such systematic occurrence of NPF in cloudy air in the tropical tropopause layer, producing large numbers of new particles, is remarkable. The observations presented by the authors, a result of their significant scientific, technical, and organizational ability, are a major contribution to a better understanding of the upper troposphere. In this respect, the manuscript is of high quality. The analysis and discussion of the observed ultrafine aerosol concentrations is very good with respect to the structure of the upper troposphere and carbon monoxide. In particular, the discussion of the role of surface air and pollutants for the new particle formation is very interesting: The moderately elevated carbon monoxide values in the majority of observed NPF case show that lofting of surface air contributes the observed NPF events, but the lofted air is almost always diluted when NPF has been observed. The findings that in-cloud NPF is strongly suppressed in the presence of predominantly liquid-origin ice particles (as opposed to in-situ cirrus ice particles), and that the observed NPF is largely independent of the carbon monoxide content (which indicates time since surface contact and NPF precursor load) are important. The statistical analysis of NPF events in the presence of ice particles is a very useful quantitative analysis of the results.

Where the manuscript falls short is the valiant, but lengthy, complex, and mostly inconclusive attempt to produce quantitative relationships between ice cloud properties and ultrafine aerosol concentrations / NPF strength (Sec. 5). Relationships that are identified are weak and only applicable to subsets of the collected data, producing much

hypothesizing and conditional statements that prevent strong conclusions. The main insight is that below the integrated radius (IR) threshold of $\sim 1 \mu\text{m cm}^{-3}$, the observed ultrafine aerosol concentrations are independent of IR, while above it, the maximum concentration of ultrafine aerosol falls linearly with increasing IR. This insight may hold in general, but it also could be limited to this data set. The reason why it is so difficult to construct quantitative relationships between the observed quantities is, in this reviewer's opinion, the complexity and nonlinearity of the processes (transport, precursors, scavenging, mixing, chemical conversion, time since NPF, etc.) that shape the observations, rather than any failure on the part of the authors.

This reviewer's recommendation is to focus on the very significant results and insights in this work, of which there are plenty, and drop the complex, hypothetical, conditional, and inconclusive elements of the analysis that weigh the manuscript down and compromise the overall quality of the work. The manuscript would also benefit from proofreading for English; some expressions are used in an unusual way ("constrained" is a better expression than "confined" in many places, etc.), and the language could be simplified for clarity and ease of reading. A major revision is recommended to provide sufficient time for any necessary changes.

Specifics

Line 58: "From the CLOUD experiments, which were performed under a variety of controlled conditions, it can be deduced that the intensity of NPF (the formation rate of new particles per air volume and per time unit) depends on the concentration of the NPF precursors."

Hasn't this been known long before the CLOUD experiments - e.g., <https://doi.org/10.1029/2003JD004460>, and others?

Line 65: ... indeterminable, because ... (expand a little bit on this for the reader's sake)

Line 76: "however" may not be necessary here - could be dropped for simplicity.

Line 77: ". Under real conditions in the atmosphere, however, the concentration of precursor material is spatially and temporally highly variable."

Please provide one or two references.

Line 93: "Investigations concerning the occurrence of NPF within clouds, or in their immediate vicinity, are sparse ..."

This statement would appear incompatible with the work of Clarke and Kapustin (2006), who report decade of data on particle production, transport, evolution, and mixing in the troposphere, much, if not most of it, near clouds.

Line 95: "... possible reasons for this are discussed by Wehner et al. (2015)."

Please briefly give some of these reasons - this will be illuminating to the reader.

Line 229 "... principally based on the difference of both quantities (cf. Weigel et al. (2011)). "

This could be removed.

Line 247: "coincidentally"

replace with "coincide"

Line 280: "computations"

replace with "computational"

Line 375: "The encountered in-cloud NPF events at altitudes between approximately 11 km and 16.5 km ($\sim 355 - 385$ K) had a mean event duration of 14.5 seconds (ranging from one second to a maximum of about 300 seconds)."

"event duration" means "flight time spent in air with in-cloud NPF", is that correct? If yes, please make sure that this is clear, because the reader might otherwise assume that this refers to the time period during which NPF took place.

Line 648: " It is not likely that a high number of interstitial, non-activated aerosol is accountable for the abundance of submicrometre-sized particles."

Please substantiate that "it is not likely", or if substantiation is not possible, remove the passage.

Line 666: "... likely suffice ..."

Please substantiate this, or if substantiation is not possible, remove the passage.

Line 912: "The IR turned out as appropriate cloud ice related parameter to juxtapose with NPF data."

This is a very confident statement given the very limited explanatory power of the IR.