

## ***Interactive comment on “New Particle Formation at a High Altitude Site in India: Impact of Fresh Emissions and Long Range Transport” by Vyoma Singla et al.***

### **Anonymous Referee #2**

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In their work, Singla et al., have studied new particle formation events that have been observed at “high altitude” in India. During the measurement time they observed 47 NPF events. The data have been recorded using a ToF-ACSM, a WRAS and a CCNC, actually not the best set of instruments to study those type of processes. The authors calculated formation rate and growth rates for the most important events (10 events) and found quite high value. Despite that the conclusions of the paper and the scientific message is not very clear neither new.

After a careful read of the manuscript I would not recommend this work to be published in ACP at this current form. I would also add that there is no new information on what we

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know already about NPF. Especially considering the fact that most of the conclusions don't have evidence in the manuscript.

Below I reported few major concerns.

In the title, the authors mention new particle formation at high altitude. After that, there is basically no reference anymore on the height of the sampling site. What does it mean high altitude? Are the measurements done in the free troposphere (FT) or in the planetary boundary layer (PBL)? This needs to be explained in the manuscript. It is fundamental to understand where are all these particles going after been formed. The lifetime in the free troposphere is larger than in the PBL making their climate impact very different. Without this information is actually useless to know the altitude especially because around the globe high and low altitude can mean totally different scenario. See for example the study conducted from Martine Collaud Coen et al., (2017) (<https://www.atmos-chem-phys-discuss.net/acp-2017-692/>). I would recommend the authors to look deeply in the newest literature focusing on new particle formation / nucleation at high altitude / free troposphere. Here just few papers (Rose et al., ACP 2017, Garcia et al., ACP 2015, Tröstl et al., JGR 2016, Venzac et al., PNAS, 2008)

In the abstract, but in general in the whole study, the authors' just report a series of numbers and characteristics of the most prominent NPF events. However there is no a clear message on the mechanism behind the NPF events or newer information on this process. Additionally, in the manuscript, there are basically no indications that support their conclusion. For example in the abstract they reported: “. . .New particle formation (NPF) events were observed on 47 days and mainly associated with these north-easterly air masses and high SO<sub>2</sub> emissions and biomass burning activities, while weaker or non-NPF days were associated with westerly air masses and relatively higher influence of local air pollution. . .” This is not confirmed by any of the data shown here.

Additionally, in the abstract, they reported:”. . . A closer examination of strong NPF

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events showed that low relative humidity and solar radiation favored new particle formation". It is very common to see NPF correlating with radiation therefore nothing new but here as well there are no evidence that the Relative humidity is affecting NPF. Changed in RH could just reflect a change in air mass.

As mentioned already, the authors have a good set of instruments but unfortunately it is not the proper one to study NPF. They would need a chemical ionization mass spectrometer (CIMS) in order to measure sulfuric acid concentration or organic precursor concentration. Additionally, Since they based the conclusion on the SO<sub>2</sub>, would be important to have a monitor to measure this gas as well. Then it is extremely important to measure also physical properties of the aerosol smaller than 5 nm, the most critical size when study NPF. Obviously it is difficult to measure all these parameters however would be good if the author would mention that in the text. For this kind of studies I believe that most of the conclusions can't be driven with the instruments used in this study. ToF-ACSM is a very good instrument but the cut off of the instrument is far too big to say something about nucleation. Here I would recommend the author to look for the latest study on NPF that have been conducted at the CLOUD chamber in CERN and their instrumentation.

Additional major comments:

Formation rates: when comparing no NPF day with NPF day would be important to calculate J<sub>5</sub> for the non NPF days as well.

Growth rates: In order to explain a GR of 2.5 nm/h or more at such size, you would need a lot of H<sub>2</sub>SO<sub>4</sub>. This is probably not possible if you are not in a plume of an air mass with very high H<sub>2</sub>SO<sub>4</sub> concentration. More considerations need to be done here as well and probably organics would play a major role as well.

Lines 268 to 275: the statements reported in these lines are not confirmed by the data. There is no evidence that the neutralized nature of aerosol favored nucleation. Additionally, the study mentions that NH<sub>3</sub> plays a major role in those events. They

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based this conclusion on the fact that during the non-events, ammonia is taken up by the acidic aerosol. Although the ammonia scavenging by the acidic aerosol might be real, there are no evidence the NH<sub>3</sub> is driving NPF.

Additionally because of the nature of the ACSM the paragraph is 4.2 is not very useful in order to understand NPF. Also the diameter has to be defined. Is it Electrical mobility or aerodynamic diameter? If Electrical the cut off is around 40-50 nm otherwise around ~75 nm. Additionally, if the aerodynamic diameter is converted the authors need to mention the density approximation that they used to make such conversion. Summarizing the ACSM can't say that the aerosol measured comes from NPF or not. As far as it is shown in this data, it can come from any sources. Therefore once more there are no evidences of the statements at the end of the paragraph (from line 286 – 292).

4.4 Cluster analysis The paragraph is quite long, vague and here as well the statements are not supported by the data. It is very difficult to believe that NPF is triggered by biomass burning without further evidence, chemical information and source appointments.

Finally, as the reviewer 1 mentioned already, most of the conclusions reported here are not supported by the data.

Minor Issue:

Paragraph 2.1 Measurement site. Here would be very helpful to explain what are the major sources around the site and if the site is situated in the planetary boundary layer, free troposphere or some layer in between. I also assume that this depends on the season and on the time of the day.

Line 230: Usually cold temperatures favour nucleation not hot. (Kirkby et al., Nature, 2011)

Line 233: How can be that a particle that is 5 nm big and grow at 2.5 nm/h reach 40

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nm in 6 hours?

Line 255 :” The CS on NPF days was much lower than on non-NPF days ( $\sim 4.2 - 4.4 \cdot 10^{-2} \text{ s}^{-1}$ ). “ The difference in CS between event and no event doesn’t seem to be much lower. They are actually comparable. I would consider using more non-event days in the data analysis. One option would be to take an average of many them.

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