Response to Reviewer #1 comments:

This manuscript presents a comparison of at least 1 year of asynchronous particle size distribution (PSD) data from 6 sites across India. These include two mountain background sites (Ranichauri: Dec 2016 to Sep 2018; Mukteshwar: Jan 2012 to Dec 2013), one mountain semi-rural site (Mahabaleshwar: Mar 2015 to Mar 2016), one urban site (Hyderabad: Apr 2019 to Mar 2020), one urban coastal site (Thiruvananthapuram: Jan 2013 to Jan 2014), and one megacity (Delhi: Nov 2011 to Jan 2013). Sebastian et al. use the PSD data from the sites to compare number concentrations (Aitken mode, accumulation mode, and total number concentrations), frequency of new particle formation (NPF), and contribution of NPF to cloud condensation nuclei (CCN) concentrations among the sites. The study provides an important analysis spanning multiple sites (and all seasons) across India with implications for understanding NPF in the context of both air pollution and cloud properties. When observed, NPF contributed to higher fraction of CCN concentrations at urban sites compared to mountain (rural) sites. Overall, NPF played an important role in driving particle concentrations and size distribution.

Response:
We are thankful to the Reviewer for his/her suggestions and comments on our manuscript. Below, we provide a point-by-point response to comments and suggestions in the BLUE colour text. The associated modifications are shown in a red colour in the revised manuscript.

The following major changes were made to the revised manuscript.

- Figure 5 in the originally submitted manuscript was revised to reflect seasonal changes in size-segregated particle number concentrations.
- A percentage increase in CCN_{50} and CCN_{100} is included in Figure 10 (c and d).
- Parameters (N, σ, d) of the representative modes of the log-normal distributions are calculated and presented in the Supplement Table S1.
- The mean particle formation rate for TVM site in the originally submitted manuscript was incorrectly stated in the text as 0.07 cm^{-3} s^{-1}, which is corrected to 0.007 cm^{-3} s^{-1} in revised manuscript supplement Table S2.
- Airmass trajectory analysis is presented for each site and season in the Supplement and briefly discussed in the revised manuscript Section 2.1.
- A histogram of the relative occurrence of total particles is also presented in the Supplement Figure S2.

1. Since there are relatively few PSD based studies from South Asia, this manuscript is timely and important. I have some comments which may help improve the manuscript: I think the main weakness of the analysis in its current form is that the authors compare across sites which do not have the same observed size ranges, especially in the context of the comparison of growth rates and formation rates across sites. The authors themselves write (page-10 line-241) "A direct comparison of GR and J between all of the sites is not possible because of the different size ranges covered by the instruments.". Yet, this manuscript is essentially a comparison of PNSD, GR_{Nuc}, and J_{Nuc} between the six sites. Interestingly, the authors define J_{LDS} (formation rate at the lowest detectable size) in the Methods section (Section 2.2) and never refer to it again, switching to GR_{Nuc} and J_{Nuc} in the Results and discussion (Section 3.2). One suggestion to provide consistent comparison across sites is to fit a multi-lognormal distribution for the particle size distributions (e.g., Hussein et al., 2005) and extrapolate for the same size range (e.g., 5–1000 nm) for each site. Then these reconstructed size

distributions can be used to compare number concentrations (then the authors can even include nucleation mode in addition to the Aitken and accumulation modes that are included in the analysis) and subsequent analysis (J, GR, etc.). [Page-9 line-225 suggests that the authors may have a mode-fitting analysis already set up.

Response:

Thank you for noting it. We now use J_{LDS} and $GR_{LDS\text{-}25nm}$ to define the formation of the lowest detectable size (LDS) and particle growth rate between the LDS and 25 nm throughout the revised manuscript. Figure 9 shows a scatter plot of the particle formation rate (J_{LDS}) and the growth rate ($GR_{LDS\text{-}25nm}$) as a function of condensation sink for each site and by no means compared between the sites. Overall, each individual site shows a positive correlation between particle formation rate and growth rate.

We completely agree with the reviewers' point of view that a direct comparison between different sites is not viable because (i) the data from different sites is asynchronous, and (ii) the size distributions are not uniform. We are comparing particle data between seasons at respective sites and do not intend to compare particle data between sites even though they have been plotted on the same figure (Figure 3, 6 and 7). In effect, we have restructured/removed some sentences in the results and discussion section.

This study essentially covers the time period from 2011 to 2019 when considering all sites. We have calculated yearly averaged particle volume size distributions in the size range from 0.1 to 1.0 μm for four sites in India where more than five years of AERONET data is available (Gandhi College, Jaipur, Kanpur and Pune) (Figure R1). Gandhi College is a typical semi-urban type, Pune and Kanpur are typically urban, and Jaipur is a mixed urban semi-arid environment. We avoided the year 2020 due to nationwide lockdown owing to COVID-19, which reduced primary anthropogenic emissions. There is no clear linear increasing trend in particle volume size distributions in the size range from 0.1 to 1.0 μm, while several studies found a significant rise in anthropogenic aerosol loading over India (Dey and Di Girolamo, 2011; Krishna Moorthy et al., 2013; Ramachandran et al., 2012; Thomas et al., 2019). The averaged particle volume size distribution over the entire period show large variability for particles larger than 0.3 μm. The calculated trend in total volume concentration in the size range from 0.1 to 1 μm also shows an insignificant increasing trend at all sites over the time period from 2011 to 2019 (Figure R2). Considering all the sites, the total volume concentration changed from -6% to 14%. From this analysis, it can be concluded that particle volume size distribution properties in the size range from 0.1 to 1 μm may not have changed drastically over the study time period. Similar trends and variability can be applied to sites considered in this study. It may also be noted that a variety of factors can influence trends in aerosols like urbanization, meteorology and regional climate. Nevertheless, we do not exclude the rising trends in aerosols over India. Still, we have refrained from comparing NPF characteristics between the sites.

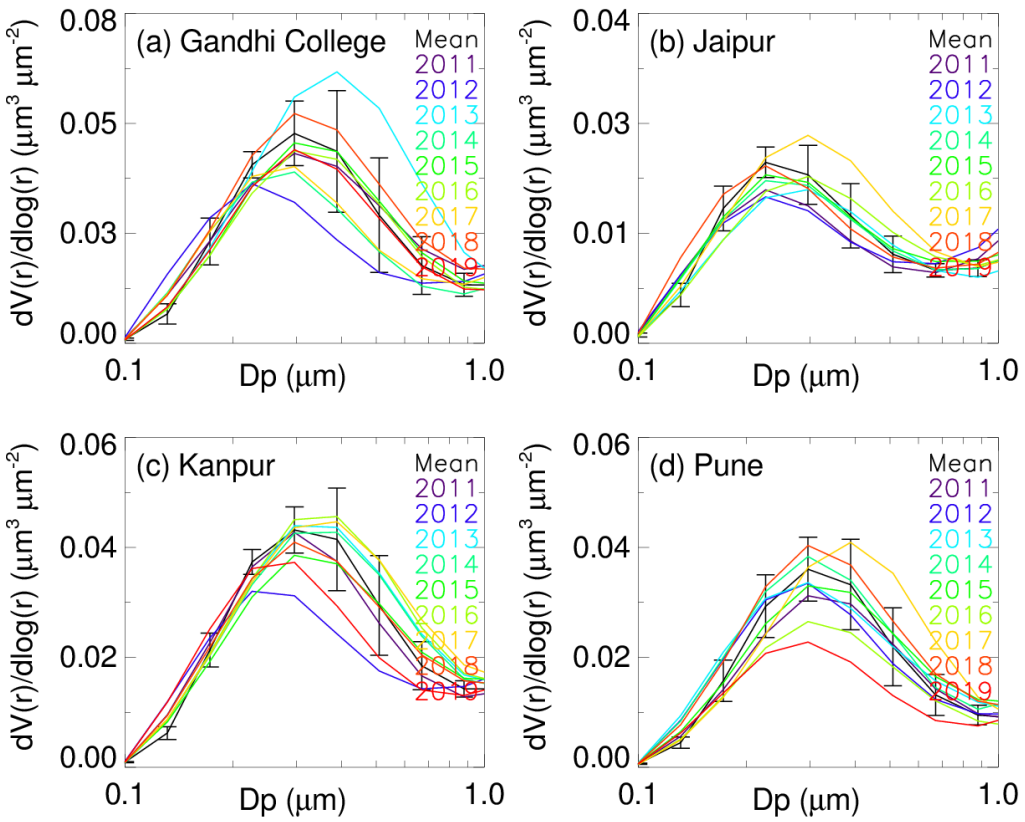

Figure R1. Yearly averaged particle volume size distribution in the size range from 0.1 to 1 μm during 2011-2019 (colored lines) and the mean particle volume size distribution with standard deviation (black line) based on AERONET observations at (a) Gandhi College, (b) Jaipur, (c) Kanpur, and (d) Pune

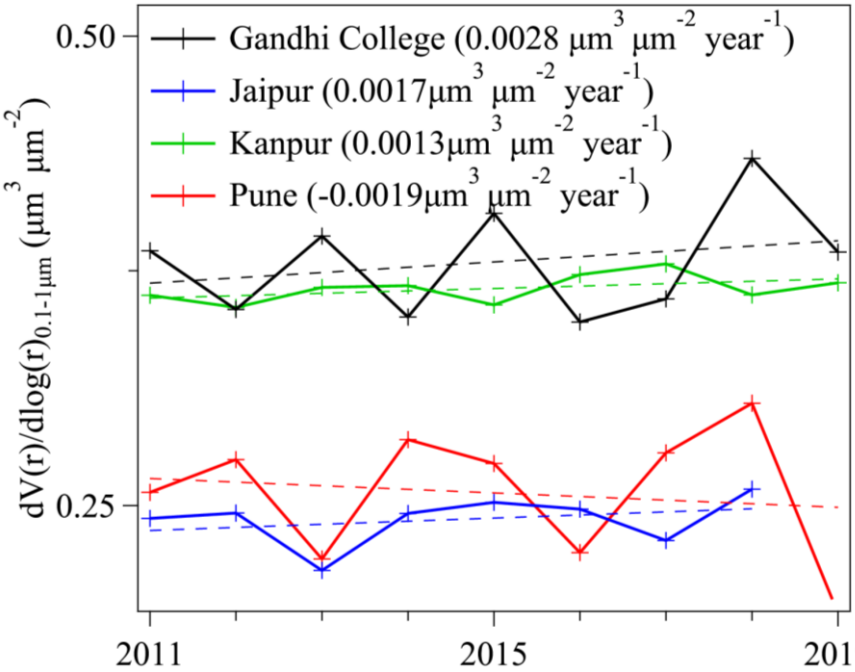

Figure R2. The trend in yearly averaged total volume concentration in the size range of 0.1-1.0 μm during 2011-2019 at Gandhi College, Jaipur, Kanpur and Pune. The dotted line shows the linear fit line, and the slope of the linear fit is given in the legend.

As suggested by the Reviewer, we have used (Hussein et al., 2005) to extrapolate the PNSDs down to 5 nm to have uniform particle number size distribution data across all the sites. The multi log-normal distribution was fitted through measured particle number size distributions to extrapolate the data down to 5 nm. However, we show that multi log-normal fitted PNSD down to 5 nm deviates from the actual value (Fig. R3). We used measurements down to 5.6 nm from two sites, Delhi and Mahabaleshwar. For these sites, the extrapolated PNSD using a multi log-normal fitting largely deviates from the actual value during a typical NPF event observed on 22 June 2012, while the extrapolated PNSD fitted well to the measured PNSD during a typical non-event observed on 24 June 2012. The same exercise was performed for the Mahabaleshwar site for typical NPF and non-event days. It can be concluded from this analysis that a multi log-normal distribution fitting fails to capture/identify/detect the particle modes below ~15 nm on NPF event days. The NPF characteristics calculation is subjective, and estimation based on fitted PNSDs will further augment errors in the computation of NPF characteristics e.g., nucleation mode number concentration, particle formation rate and growth rate. Therefore, we abstain from extrapolating the PNSDs for the size range for which it was not measured.

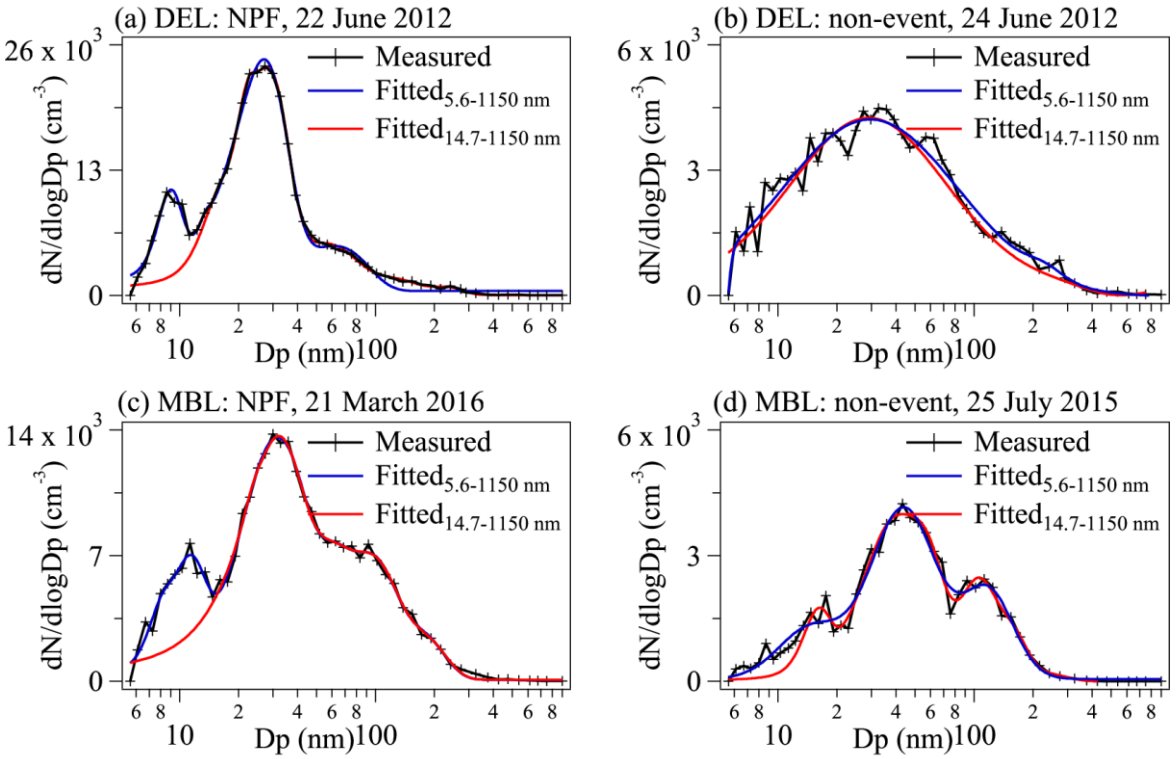

Figure R3. Measured PNSD in the size range 5.6-1050 nm (black line connected by a plus sign), fitted multi log-normal distribution in the same size range (blue line), and fitted multi log-normal distribution in the size range 14.7-1150 nm (red line) during an (a) NPF event on 22 June 2012, (b) non-event on 24 June 2012, at Delhi (c) NPF event on 21 March 2016 and (d) non-event on 25 July 2015, at Mahabaleshwar.

2. The manuscript should include instrumentation setup details (including inlet and sampling tubing information) for each site or refer to previous articles from these sites which contain this information.

Response:

We have included instrumentation setup details for each site where applicable or cited relevant references which contain instrumentation setup details as indicated below.

For MUK, the reference has been cited as "More details of the site and aerosol sampling can be found in Hyvärinen et al. (2009)".

For Delhi, the following details included - "The WRAS system uses a stainless-steel inlet tube with an integrated Nafion drier to dry the aerosol sample. A detailed description of the site and aerosol sampling is given elsewhere (Jose et al., 2021)."

For MBL, the following discussion is added, and the paper has already been cited. "The WRAS has a stainless-steel inlet tube with a an integrated Nafion dryer to reduce the relative humidity to ~40%."

For TVM, the following details are included in the revised manuscript: "The ambient air was sampled from a height of 3 m above ground level through a manifold inlet fitted with PM_{10} size cut impactor at 16.67 LPM flow rate. Subsequently, the flow was distributed among various aerosol instruments connected with electrically conductive tubing. To restrict high relative humidity conditions, a diffusion dryer (Make: TSI, Model: 3062) employing silica gel was used."

3. Since the datasets range across a decade (Delhi 2011-2012 to Hyderabad 2019-2020), it may be helpful to present the dates in the figures (or caption) where the comparison across sites is presented. This will help put the comparison in the context of not only the different sites, but also different years as presumably most of these sites have become more polluted over the last decade. Furthermore, a brief discussion on the possible implications of the changes in particle size distribution over the past decade will be helpful.
Response:
Table 1 summarizes measurement details, including time periods, instrument, particle size range and time resolution. To help the reader, we have added a statement in each figure caption. "Note that measurements are from different time periods for each site (refer to Table 1)"

4. The definition of seasons (Table 2) warrants some discussion. For example, why use "pre-monsoon" and not "spring" and "summer". Furthermore, "monsoon" spans across four months for all sites. What are the implications of the season definitions to the summary results (when averaging using these periods) given the differences in climatology for each site? To be clear, I am not asking the authors to necessarily change the season definitions, just to justify and discuss their implications on the results.
Response:
The classification is based on India Meteorological Department (IMD). December, January and February are the coldest months in a year at all these sites (Winter). March through May are the warmest months at all the sites (summer or pre-monsoon). We referred to it as pre-monsoon as per the IMD definition. The onset of monsoon happens on the southern tip of India in early June and encompasses the entire country by mid-August, and the retreat phase lasts till the end of September. Therefore four months are considered as the monsoon season as per IMD definition. October and November months consist of the post-monsoon season. Also, Table 2 clearly identifies the meteorological characteristics of each season.

5. The discussion on precursors in this manuscript seems to be primarily based on existing literature. Is it possible to include some approximate quantitative comparison of precursor concentrations across the six sites (and by season), perhaps using SO_2 data (if available) to

calculate H_2SO_4 proxy (Dada et al., 2020)?

Response:

Unfortunately, we do not have measurements of precursor gases (such as SO_2, organics etc.) for these sites except Hyderabad. Our recent study calculated sulfuric acid proxy based on SO_2 concentrations in Hyderabad (Sebastian et al., 2021).

Since "primary" and "secondary" is now routinely used in the context of mass-spectra derived source apportionment, the authors should be intentional and clear while using the terms "primary" and "secondary" in the context of the PSD-based NPF analysis presented throughout the manuscript (e.g., on page 10, lines 255).

Response:

The "primary" and "secondary" spectra are used to separate between primary and secondary mass (regardless of whether secondary mass condenses to a particle that came from primary emission). We do not classify the aerosols based on mass concentration. Here, the likely "primary' and "secondary" sources of particle number concentrations are indicated.

6. The analysis on the "relative occurrence of Aitken mode and accumulation mode" (pages 16-19, including Figures 6 and 7) is not clear to me. To my knowledge this is not a standard analysis and requires more/clearer context and guidance for the reader to understand the results and their interpretations. For example, in the context of Figure 6 (x-axis: Aitken mode concentration; y-axis: "relative occurrence"), the authors write "a reasonably log-normal shape…" (page-16 line 351). Perhaps I am missing something, but I am unable to understand this discussion.

   Response:

   Relative occurrence explains how frequently is a particular number concentration occurs for a particle mode. The relative occurrence of size-segregated particle number concentrations is presented to find the maximum relative occurrence of a particular particle size range (type) in different seasons to infer possible causes of variability on relative occurrence. For instance, higher Aitken mode particle number concentrations in pre-monsoon (March through May) than other seasons indicate the potential contribution from NPF processes and are in line with the highest NPF frequency in the pre-monsoon season. Delhi has the highest occurrence of Aitken mode particles during the winter season, indicating the dominance of anthropogenic sources and conducive meteorological conditions (Kanawade et al., 2020). The term 'reasonably log-normal distribution' is removed to avoid confusion for the reader.

7. (Page 25 line 506) "Expectedly, the condensation sink at the start of the NPF event is higher at urban sites than the mountain sites. The mean condensation sink at urban sites (16.1×10^{-3} s-) was twice as compared to mountain sites (7.9×10^{-3} s^{-1})." What do "start of NPF event" condensation sink values mean? Are they averaged over a few minutes or hours? In the second sentence, what is the averaging period for the "mean condensation sink"?

   Response:

   The condensation sink at the start of the event is taken as one-hour average CS just before the start of the NPF event. It has been clearly stated in the Figure 9 caption.

8. When presenting CCN increase, the authors should consider also including the fraction increase (%) over the "baseline" in addition to the magnitude increase (cm^{-3}) which the authors have done (in abstract and conclusions as well).

   Response:

   We have included percentage change in CCN_{50} and CCN_{100} increase in the revised manuscript as suggested by the Reviewer as shown below Figure R4.

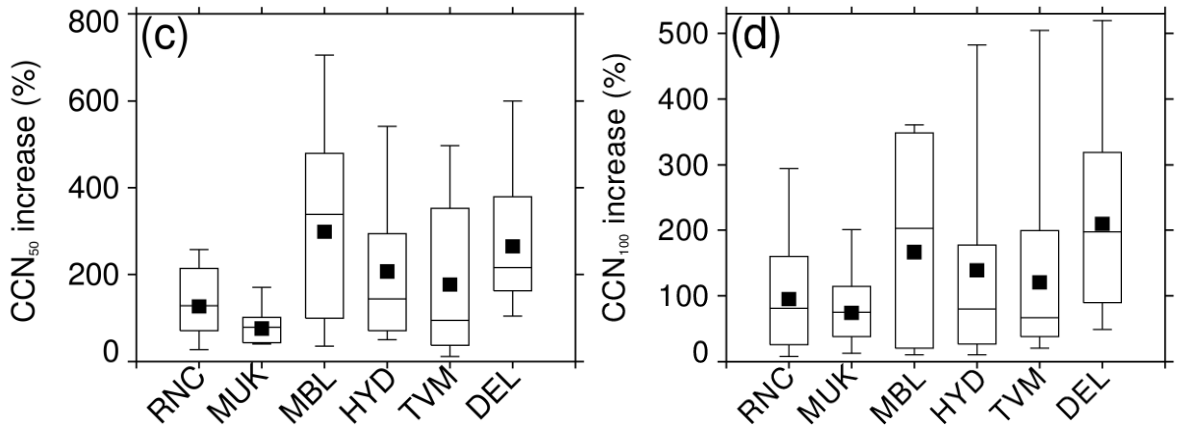

Figure R4. Box-whisker plot of percentage increase in CCN concentrations for (c) 50 nm and (d) 100 nm particles at all the sites based on the observed NPF and non-event events.

9. Please use full caption in Figure 7 (should be able to stand alone).
   Response:
   Included the full caption in Figure 7.

10. Updated ACP/Copernicus guidelines state "it is important that the colour schemes used in your maps and charts allow readers with colour vision deficiencies to correctly interpret your findings." This means that jet/rainbow color scales need to be changed to other appropriate color scales. More here: https://www.atmospheric-chemistry-and-physics.net/submission.html#figurestables
    Response: We have checked figures for colour vision deficiencies, and it seems to be appropriately reflected.

References:

Dada, L., Ylivinkka, I., Baalbaki, R., Li, C., Guo, Y., Yan, C., Yao, L., Sarnela, N., Jokinen, T., Daellenbach, K. R., Yin, R., Deng, C., Chu, B., Nieminen, T., Wang, Y., Lin, Z., Thakur, R. C., Kontkanen, J., Stolzenburg, D., Sipilä, M., Hussein, T., Paasonen, P., Bianchi, F., Salma, I., Weidinger, T., Pikridas, M., Sciare, J., Jiang, J., Liu, Y., Petäjä, T., Kerminen, V.-M., and Kulmala, M.: Sources and sinks driving sulfuric acid concentrations in contrasting environments: implications on proxy calculations, 20, 11747–11766, https://doi.org/10.5194/acp-20-11747-2020, 2020.

Hussein, T., Dal Maso, M., Petäjä, T., Koponen, I., Paatero, P., Aalto, P., Hämeri, K., and Kulmala, M.: Evaluation of an automatic algorithm for fitting the particle number size distribution, Boreal Environment Research, 10, 337–355, 2005.
Citation: https://doi.org/10.5194/acp-2021-804-RC1