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Comment on acp-2021-804

Anonymous Referee #1

Referee comment on "Observations of particle number size distributions and new particle formation in six Indian locations " by Mathew Sebastian et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-804-RC1, 2021

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.

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.]

- 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.
- 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 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.
- 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.
- 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 $H2_SO_4$ proxy (Dada et al., 2020)?
- 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).
- 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.
- (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"?
- 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).
- Please use full caption in Figure 7 (should be able to standalone).
- 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

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.

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