Response to Reviewer #2 comments:

The study by Sebastian et al. (2021) reports asynchronous measurements of particle number size distributions (PNSD) from six stations located in contrasting environments in India. The shape of the PNSDs is first discussed, with a specific focus on the concentrations in the Aitken and accumulation modes, and the occurrence of new particle formation (NPF) in investigated in a second step. The contribution of NPF to the formation of potential cloud condensation nuclei (CCN) is finally analysed.
Although the objectives associated with this study are very interesting, I find that the methodology employed is not necessarily adequate, and, in my view, the analysis of the results could have in addition been enriched on some aspects. Therefore, I do not recommend the publication of this study in its current form. The most decisive points in my opinion are listed below.

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 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 the 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. The first point concerns the selection of the datasets. Data availability is considered "adequate" (>60%, on what criteria?) at some sites (RNC, MUK, MBL, HYD) and limited for the others (<50% at TVM and DEL). Data availability is, in particular, very limited at TVM (34%) and one can question the relevance of the statistics that are reported for this station. Further reason to this question comes from the recent study by Rose et al (2021), who investigate the impact of reduced data availability on seasonal and annual statistics of the particle number concentration, and suggest that 50% and 60% of the data should be available to derive relevant statistics at the seasonal and annual scale, respectively.

   In addition, the data from the different sites correspond to periods that are sometimes relatively distant (between 2011 and 2020). A clear decreasing trend of the particle concentration was reported by Asmi et al. (2013) for the majority of the sites they consider in their study (located in Europe, North America, Antarctica and on Pacific Ocean islands), I therefore question the relevance of the comparative study that is made here, and which does not alert on these aspects. Further, to my knowledge, there is no study that has looked at the evolution of NPF on a global scale, and even if Nieminen et al. (2014) show that there is no clear/homogeneous trend at the boreal site of Hyytiälä over the period 1996-2012, I do not think that the authors should ignore this possibility here. To sum up, I do not think that the authors can exclude the fact that the differences observed between the sites may also be related to the selected periods, in addition to the signature of their environments. If this is not enough to completely question this study, this aspect should at least be discussed, and the following points also considered.

Response:

In the originally submitted manuscript, we have clearly stated that the data availability is adequate at four sites while it is limited at two sites (TVM and DEL) for readers to relate the statistics derived from these sites. Figure R1 shows the bar plot of seasonal data availability for all the sites. DEL has lower data availability during winter, while TVM has lower data availability during pre-monsoon, monsoon and post-monsoon seasons. Rose et al. (2021) used data availability of >50% so that more data can be used for the analysis as opposed to Laj et al. (2020), which used data availability of >75%. (Rose et al., 2021) indicated that the criterion of >50% is not estimated on strict statistical analysis. Though the data availability is lower for TVM and DEL, these measurements are very useful to understand the general features in the data and can be used with caution, especially the regions for which such analyses are rarely reported.

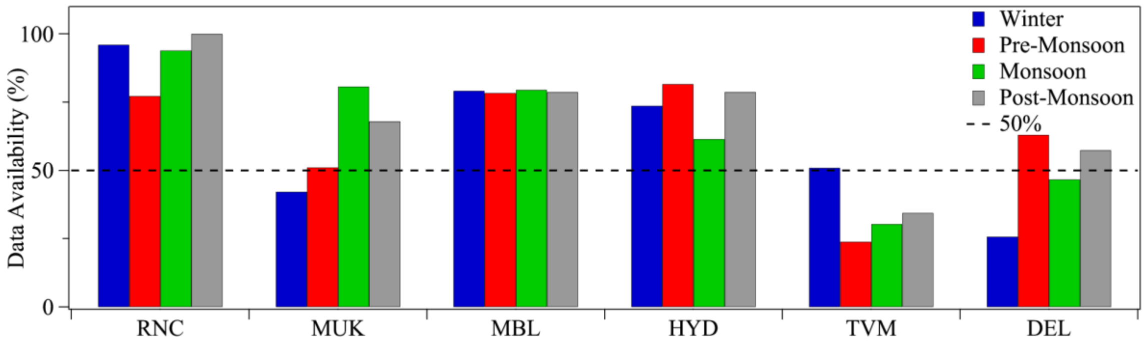

Figure R1. Seasonal data availability for all the sites.

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 them even though they have been plotted on the same figure (Figure 3, 6 and 7). In effect, we have restructured 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 R2). 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 R3). 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.

In the originally submitted manuscript, Figure 9 shows a **s**catter plot of the particle formation rate and the growth rate as a function of condensation sink for each site and by no means compared between the sites. Overall, each site shows a positive correlation between the formation rate and growth rate.

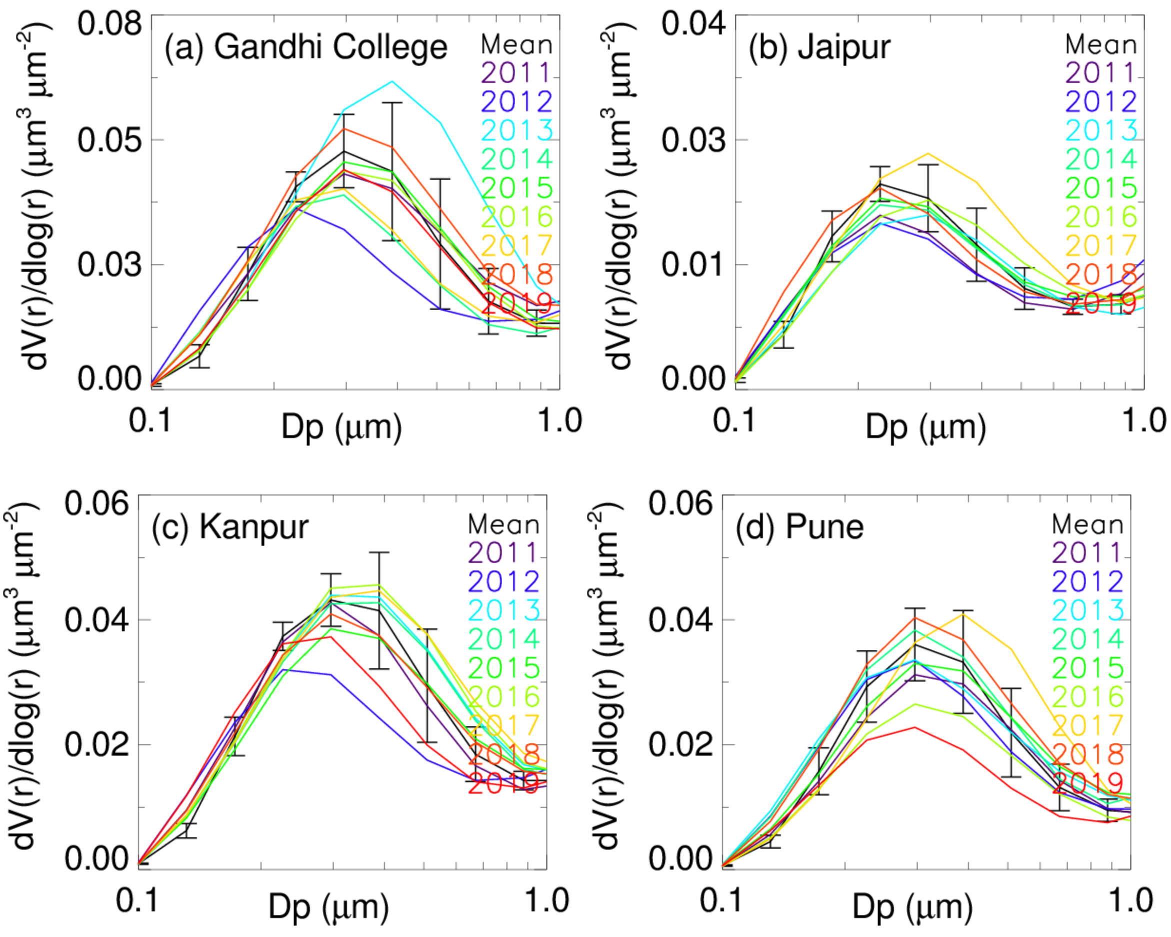

Figure R2. Yearly averaged particle volume size distribution in the size range from 0.1 to 1 μm during 2011-2019 (colored lines) and the mean particle volume size distribution with standard deviation (black line) based on AERONET at (a) Gandhi College, (b) Jaipur, (c) Kanpur, and (d) Pune

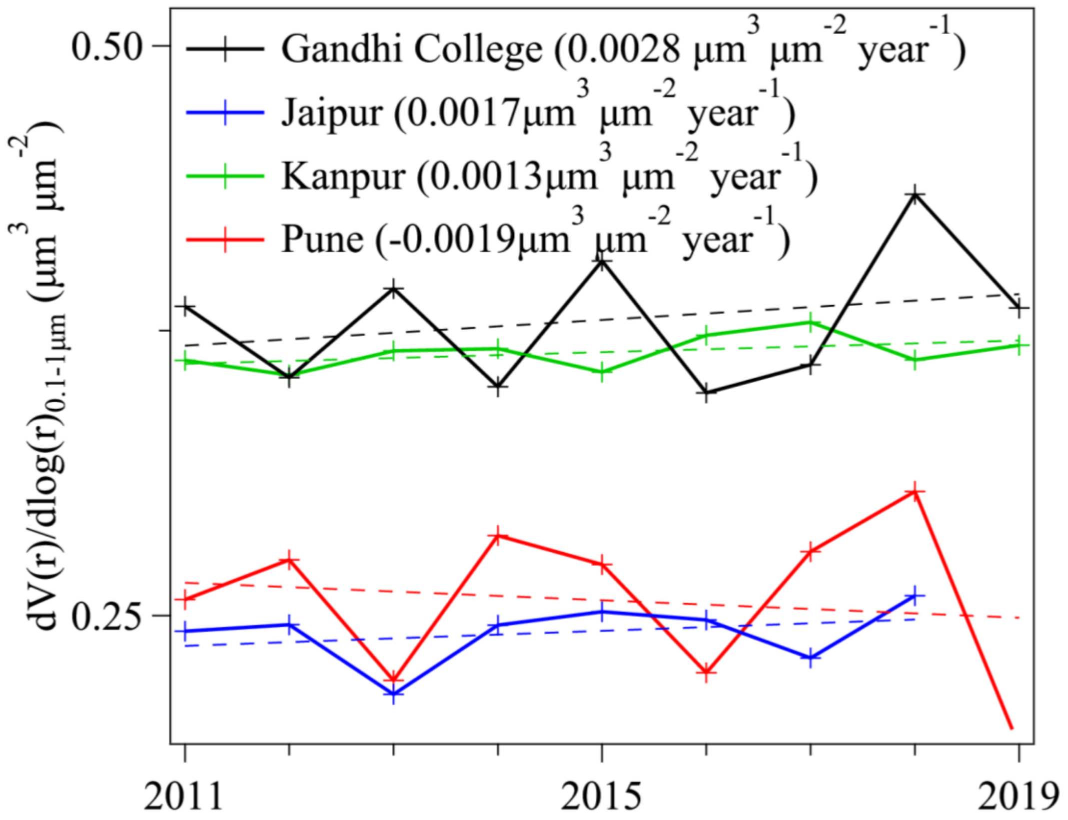

Figure R3. The trend in yearly averaged total volume in the size range of 0.1-1.0 µm during 2011-2019 at Gandhi College, Jaipur, Kanpur and Pune. The dotted line shows the linear fit line, and the slope of the linear fit is given in the legend.

In my opinion, one of the interests of a multi-site study such as this one is to be able to highlight observations common to sites with similar characteristics, or to highlight particularities, and discuss as well what explains (or may explain) the observed differences. I think that in its current form, the manuscript does not sufficiently address this last aspect. For example, the discussion at L362-368 should in my view be developed. More broadly, Section 3.1, is for me too descriptive and I find it difficult to extract a message from it. On the other hand, some additional information useful to the modelling community could easily be extracted from this analysis, such as the parameters (N, σ, d) of the representative modes of the distributions presented in Fig. 12 (similar to Asmi et al. 2011 or Rose et al. 2021); such numbers would also benefit the discussion reported at L272-312.
Concerning the analysis of J and GR, the calculation of J10 (with the exception of TVM, but the coverage at this site may on the other hand be too limited for such study, see previous point) and a GR on a fixed range common to all stations would have allowed a comparison of the sites between them and with the literature. Again, I find it difficult to extract a message from this analysis in its current form.

Response:

We agree with the Reviewer's point of view on a multi-site study to be able to highlight observations common to sites with similar characteristics, but we refrained from comparing observations from similar sites as the period of study is different for different sites. The discussions at L362-368 point to the possible reasons for the seasonality in particle number concentrations. Major findings from other studies related to seasonality in particle number concentrations across these sites are discussed in the manuscript.

Section 3.1 mostly deals with explaining the size distribution characteristics across all the sites. We believe that the section adequately discusses the seasonality in particle number size distributions and number concentrations in size ranges common to all sites (Aitken and Accumulation mode) for all the sites.

As suggested by the Reviewer, we have calculated N, σ and Dp values for the particle number size distributions for all the sites on a seasonal basis. The values are tabulated in Table R1. The table is included in the supplementary information as Table S1.

Table R1. Parameters of the modes identified for the description of median particle number size distributions from all six sites shown in Figure 3. N, Dp and σ are the number concentration, geometric mean diameter and the standard deviation of the distribution.

	Unimodal			Bimodal					
Site	N	Dp	σ	N_1	Dp_1	σ_1	N_2	Dp_2	σ_2
Annual									
RNC	2555	87.5	2.0	591	49.3	1.9	1963	101.8	1.9
DEL	9670	50.9	2.2	8237	44.9	2.0	1465	121.3	1.9
HYD	6401	63.4	2.5	2097	27.3	1.8	4186	90.0	1.9
MBL	3166	74.2	2.3	3104	72.9	2.2	48	197.3	1.2
MUK	2573	85.5	1.9	301	65.1	1.5	2276	90.5	1.9
TVM	3463	111.2	1.8	3379	109.6	1.8	85	330.8	1.3
Winter									
RNC	3205	94.6	1.9	876	53.0	1.9	2357	109.2	1.8
DEL	13555	68.7	2.3	12878	65.9	2.1	678	298.0	1.2
HYD	7314	61.1	2.3	3165	33.9	1.8	3990	95.2	1.8
MBL	3817	84.4	2.3	4877	100.2	2.6	789	319.4	0.6
MUK	3374	86.0	1.9	3344	85.5	1.9	28	256.0	1.2
TVM	4437	113.2	1.8	4266	110.6	1.8	169	320.0	1.3
Pre-Monsoon									
RNC	4012	81.2	2.0	2721	64.6	1.9	1280	118.7	1.7
DEL	7708	49.8	2.3	4622	35.8	1.9	3093	96.0	2.1
HYD	7726	82.0	2.2	1858	24.5	1.8	6007	98.7	1.8
MBL	3702	78.6	2.1	5034	100.5	2.4	1342	228.3	0.5
MUK	6488	91.1	1.8	1748	62.5	1.9	4760	101.4	1.7
TVM	3241	122.4	1.8	2933	115.3	1.7	282	313.0	1.3
Monsoon									
RNC	1774	78.4	2.0	85	58.3	1.3	1693	81.1	2.0
DEL	9336	40.2	2.2	5059	27.4	1.9	4194	66.8	1.9
HYD	3141	49.2	2.8	2844	45.4	2.5	210	196.9	1.4
MBL	2187	50.3	2.1	1960	47.8	2.3	255	58.9	1.4
MUK	1984	79.4	1.9	1765	73.9	1.7	223	199.5	1.5
TVM	2603	103.4	1.8	1565	93.5	2.1	1109	110.1	1.6
Post-Monsoon									

RNC	2072	102.0	2.0	441	52.0	1.8	1629	118.7	1.9
DEL	12152	60.6	2.2	11881	59.5	2.1	286	263.9	1.1
HYD	9949	58.7	2.5	9335	57.5	2.5	123	157.5	1.3
MBL	3277	88.5	2.4	2937	79.4	2.2	289	237.6	1.3
MUK	1782	93.7	1.9	1743	93.3	2.0	50	99.0	1.2
TVM	3176	117.5	1.8	3099	116.2	1.7	86	360.7	1.3

We now use J_{LDS} and $GR_{LDS\text{-}25nm}$ to define the formation of the lowest detectable size and particle growth rate between the LDS and 25 nm for respective sites. $GR_{LDS\text{-}25nm}$ and J_{LDS} for each site have been plotted in Figure 9 for visualizing the overall association between $GR_{LDS\text{-}25nm}$ and J_{LDS} when considering all sites. We understand that the GR and J values cannot be made for a fixed range as the LDS is different for each site.

2. To conclude with science, the section dedicated to the contribution of NPF to the formation of CCN also has some gaps in my opinion. I think the authors should have:

- first recalled the main assumption that is made in this approach: particle size is considered to play a more determining role than chemical composition.
- been clearer in the explanation of the method: for example, it is indicated "We calculated the seasonally averaged change in CCN-active particles on non-event days over the same time of day as the NPF events". What does this mean given that each event is characterized by its own start / end times? Are average start / end times considered?
- finally, provided all the elements allowing to really evaluate the importance of NPF with respect to the (potential) CCN population at these sites: all the events certainly do not present a growth of the particles beyond 50 nm (it is at least indicated for HYD), therefore it would be interesting to know the percentage of events during which the formed particles reach a priori sizes of climatic importance, and only consider these events in the statistics reported in Fig. 10. It would also be interesting, especially for high altitude sites, to indicate the "concentration increase" observed on non-event days over the time period of interest, in order to really be able to measure the importance of NPF compared to other sources of potential CCN.

Responses:

- We have edited the statement as "In typical ambient in-cloud supersaturations, the total number of particles from 50 nm to >100 nm can be considered as a proxy for CCN concentrations assuming fixed chemical composition."
- We have addded statements in the revised mansucript as "The start of the NPF event is the time when nucleation mode particle number concentrations increase rapidly during an NPF event." and "For non-event days, the seasonally averaged start of the NPF event time was chosen to calculate $N_{CCNprior}$. N_{CCNmax} on non-event days was taken similar to NPF event days, as a maximum one-hour average concentration of particles larger than 50 nm (and 100 nm)."
- All identified NPF events have particle mode diameter growing beyond 50 nm. Only these events are used for calculating the values plotted in Figure 10. The time evolutions of seasonally averaged diurnal particle number size distributions (Figure 4) show that particles grow beyond 100 nm at all sites, with the exception of monsoon season at some sites. The red open circles show an absolute increase in CCN concentrations for 50 nm and

100 nm on non-event days (second term in Eq. 2) in the Fig. R4.

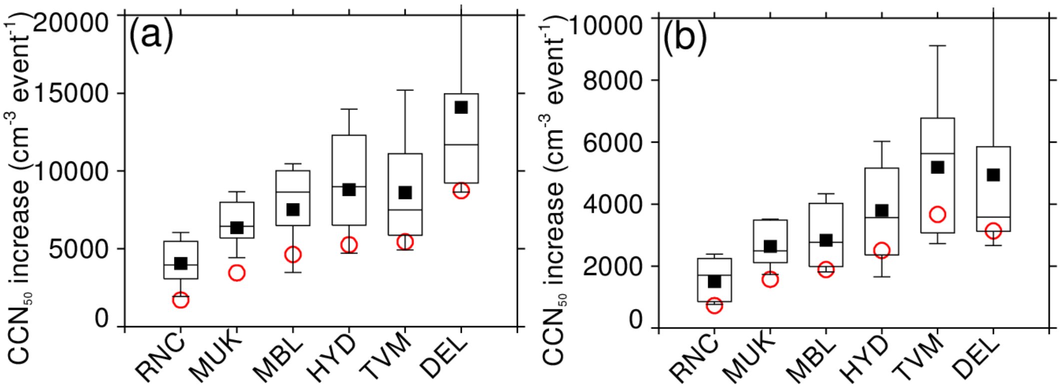

Figure R4. Box-whisker plot of absolute increase in CCN concentrations for (a) 50 nm and (b) 100 nm particles on NPF event days (First team in Eq. 2). The filled square box indicates the mean, the horizontal line indicates the median, the top and bottom of the box indicate 25th and 75th percentiles values and the top and bottom of the whiskers indicate the 10th and 90th percentile values. The red open circles show the mean CCN concentrations for (a) 50 nm and (b) 100 nm on non-event days (Second term in Eq. 2).

3. Finally, this paper could in my view be improved in its form. For example, some lists of numbers could be replaced by tables (e.g L421-430, L495-503). Some sentences are also confusing, or have a structure that could be revised (e.g. L547-549, L575-577). Concerning Section 2.1, in particular, the information given, especially about the cities near the stations, should be homogenized (number of inhabitants missing for some). Furthermore, the reader would appreciate guidance on the impact that can be expected from these urban areas on the observations (air mass back trajectory analysis?). More generally, a selection/reorganization of the information would often benefit the clarity of the messages.

Response

Agree, and we have included a table summarising the frequency of occurrence of NPF events and non-NPF events, $GR_{LDS\text{-}25nm}$ and J_{LDS} in Supplement as Table S2 as shown below in Table R2.

Table R2. Number of valid observation days, number of NPF days (percentage), number of non-NPF days (percentage), mean formation rates and mean growth of particles at all six sites of study.

Site code	valid observation days	NPF days	Non-event days	J_{LDS} ($cm^{-1}s^{-1}$)	$GR_{LDS\text{-}25\ nm}$ ($nm\ h^{-1}$)
RNC	586	21 (3.9%)	493 (83.7%)	0.11±0.05	6.3±2.4
MUK	440	13 (2.9%)	321 (73.1%)	0.04±0.02	2.5±1.6
MBL	281	16 (5.9%)	188 (66.1%)	0.04±0.02	4.7±3.0
HYD	270	38 (16.3%)	124 (44.8%)	0.13±0.11	5.7±3.6
TVM	133	23 (16.6%)	55 (41.4%)	0.007±0.005	1.1±1.1
DEL	139	39 (28.1%)	30 (21.1%)	0.13±0.10	3.7±2.1

Lines 547-549 is rephrased as

"High background number concentrations of CCN_{50} and CCN_{100} in Delhi resulted in a smaller relative increase of CCN from NPF, during post-monsoon and winter seasons when compared to the other sites."

Lines 575-577 is rephrased as

"The high pre-existing particle concentration is also an indication of precursor-laden air. But when the condensation sink gets very high, it inhibits aerosol nucleation."

The airmass history is analysed using HYSPLIT transport model (Fig. R5). The seasonal trajectory density plots for all six sites are added to the supplementary as Figure S1.

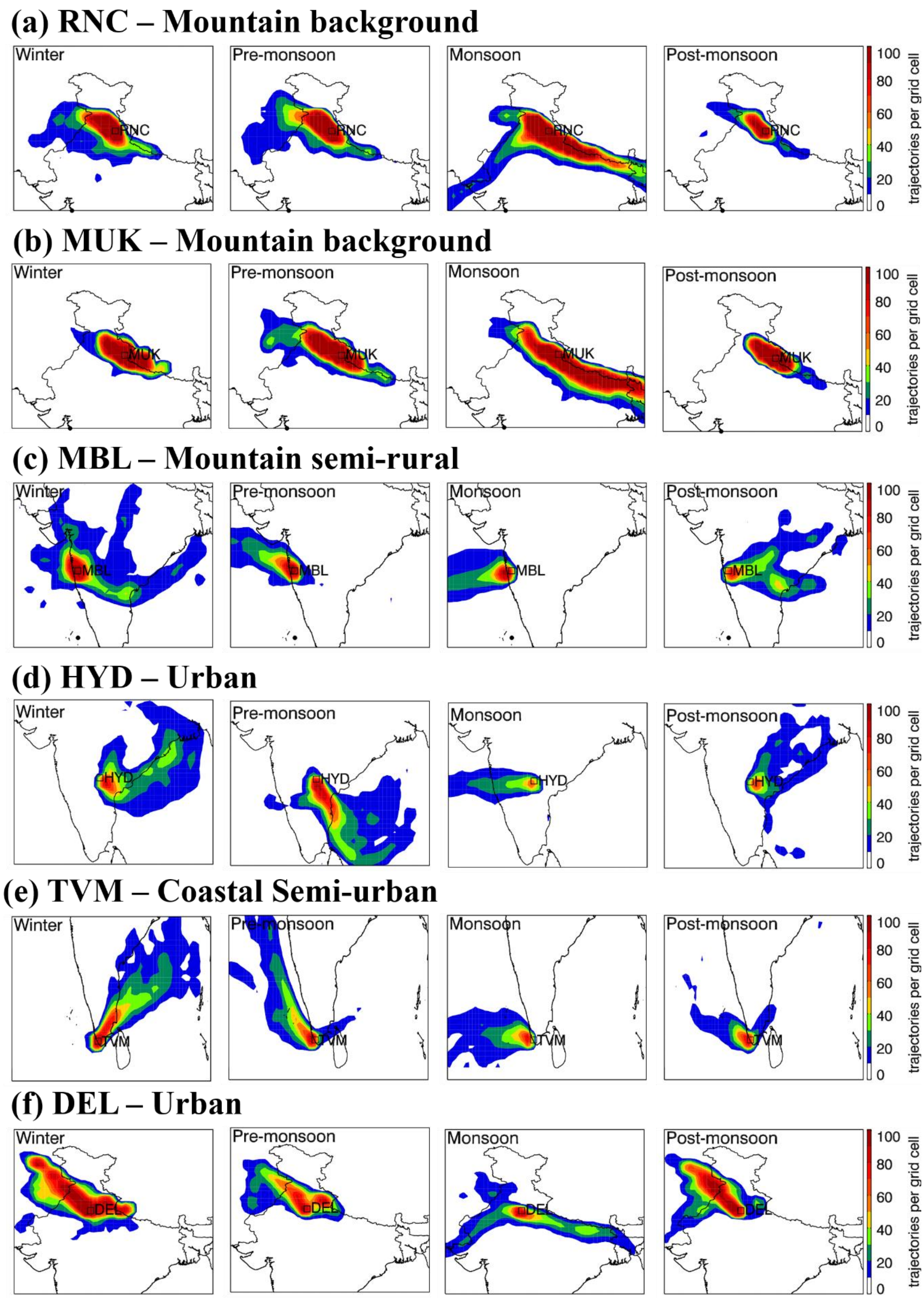

Figure R5. HYSPLIT modelled 72-hour backward air mass back trajectory density starting at 500 m for (a) Ranichauri, (b) Mukteshwar, (c) Mahabaleshwar, (d) Hyderabad, (e) Thiruvananthapuram and (f) Delhi.

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