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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurement report: Altitudinal variation of CCN activation across the Indo-Gangetic Plains prior to monsoon onset and during peak monsoon periods: Results from the SWAAMI field campaign" *by* Mohanan R. Manoj et al.

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At the outset, we thank the reviewer for the meticulous review, constructive comments and the overall appreciation of the work. We have considered each of the comments carefully and revised the manuscript. Our responses to the comments, which formed the basis for the revision, are given below along with the page and line numbers in the

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revised manuscript, where the revisions are incorporated.

Reply to comments of Anonymous Reviewer #2 (RC2)

General comments:

1) A more thorough review of relevant literature in the area and on the subject should be presented. Many recent studies are not mentioned, and what the recent study offers in comparison to others is not clear.

Yes, we agree. We have added a thorough review of the related literature in the revised manuscript. The page and line numbers are mentioned along with the reply to specific comments. There have been several works with CCN in India mostly ground-based focusing on case studies or long-term measurements on seasonality. However, we focus on the vertical structure of CCN characteristics within the ABL and in the free troposphere and the changes that occur in these properties as the season changes from just prior to the onset of Indian monsoon to its active phase. This urged us to restrict our literature review to those investigating the vertical variations in CCN in the previous version of the manuscript.

2) There is a complete lack of mentioning operational supersaturation levels, which is crucial for a notion of particle activation size. Without this information all discussion falls short. Also other sampling information, such as drying of the aerosol prior to CCN and CN measurement should be mentioned.

We thank the reviewer for pointing this out. We have made the necessary changes in the revised manuscript. The page and line numbers are given along with the reply to the specific comments. We have also added a section in the supplementary material explaining these in detail.

Specific comments:

1) Introduction: As the manuscript refers to CCN and hygroscopicity, the importance of chemical composition should be also discussed and a more excessive review of the

literature in the area should mentioned. To my knowledge, there are at least two recent studies focusing on CCN in the area, also taking into account chemical composition and number size distribution. Shika et al. (2020) focus on aerosol properties also during pre-monsoon and during monsoon season and implications on cloud droplet formation. Furthermore, Arub et al. (2020) characterize chemical composition and size distributions in the area of Delhi based on air masses origin and their impact on hygroscopicity and CCN formation. Singla et al. (2017) study the role of organics in CCN activation in Western Ghats, India. Also another study part of the same experiment (CAIPEEX) by Jayachandran et al. (2020a) although mentioned in the discussion section (4.1.1) general outcomes are not mentioned in the introduction, in order to put into context the present study. Finally, Jayachandran et al. (2020b) also report airborne CCN measurements across the Indo-Gangetic Plain which also are mentioned in the discussion section (4.1.1) but not mentioned in the introduction. Overall, the introduction section needs to be enriched with other relevant studies in the area so that the current study is put into context.

Complied with in the revised manuscript (page 2, lines 48-49; page 2-3, lines 56-79; page 3, lines 87-88 and pages 3-4, lines 97-100).

2) P3, L80-86: A map with the locations of the focus areas would be helpful for the reader to get an idea of the topography and type of environment and possible aerosol sources which can impact aerosol size and chemical composition.

We have modified figure 1 in the manuscript to clearly show all the important locations. We believe that after the modification, the location of the important areas along with the direction of the wind would provide a fair idea of the environment and the possible sources.

3a) P4, L100-108: Is there a drier at the inlet?

Yes, the CCN and CPC flown during SWAAMI sampled through a Nafion dryer (Permapure MD-110-12S). This Nafion dryer was operated in the atmospheric vacuum set up.

The drier was installed to stop relatively warm, wet ambient air from condensing in the cool conditioned cabin sample lines. It was not used to force dry the sample to some set standard. Internally a bleed air is removed from the sample line and trickled over a Rotronic HC2-IExxx Screw-in Relative Humidity (RH) and temperature sensor. This provides the sample RH and temperature measurements used here. Changes made in the revised manuscript (page 5, lines 136-145).

3b) What is the RH of the sampled aerosol which enters inside the CCN counter and the CPC?

The mean RH for the pre-monsoon portion of the campaign are 49.7% and 28.7 % for the ambient and CCN sample lines respectively. This shows a big reduction in RH, which is due to both the nafion and the equilibrium of the ambient air to the temperature conditions of the in-cabin sample line. These two effects, can be untangled to some extent by using the Magnus equation to solve the RH for ambient dew point and cabin influenced sample line temperatures. The mean RH in the sample line would be 36.1%, as shown by 'Theoretical' in Figure R1 if the nafion is removed. However, the big difference can be seen in the interdecile upper range where the values are close to that of condensation, the environment the nafion dryers were used to eliminate. As expected, the RH values were much higher and the effect of the drying less pronounced during monsoon with mean values of 54.4, 73.5 and 57.6 % for sample, ambient and theoretical respectively (Figure R2). Changes made in the revised manuscript (page 5, lines 136-145).

3c) Also how was the CCN instrument operated? Was it on scanning flow analysis (Moore and Nenes, 2009; Moore et al., 2012; Lathem et al., 2013)? To my knowledge, this is the most appropriate analysis for airborne measurements as it ensures the correct supersaturation spectra over very limited timescales. If not, the CCN analysis by staying at a constant supersaturation for a given time allows for a complete CCN spectrum every, say, hour, during which obviously the aircraft has moved on to other areas, with other aerosol characteristics and sources. Even in Trembath (2013) it is not clearly

stated how the CCN instrument supersaturation varied: “Each column supersaturation was set using the proprietary dual column CCN software (DMT inc, Boulder); the set point ranged between 0.1 and 0.5 % across all flights.” p.122 Or was the CCN instrument operated in a constant supersaturation? A few details on the operating mode should be included, and how the time at each supersaturation compares in terms of aircraft velocity and distance covered.

The CCN instrument used on the BAe-146 is a dual column instrument. During the SWAAMI campaign the instrument was operated behind a constant pressure inlet set to 400 hPa. This allows a constant supersaturation to be maintained between sea level and approximately 7000 m. All the supersaturation data and concentration data are corrected for this constant pressure using Roberts et al (2010) for the former and a simple density correction for the latter. To gain as much information as possible from a flight the instrument is set up to have one column scanning three different supersaturations (here 0.12%, 0.23% and 0.34%) and the other stable (0.1% SS). This returns CCN concentrations for four different supersaturations every 15 mins and keeps one channel as a reference over the entire period. More details on the operation and relative humidity variations of the sample are provided in the supplementary material. Changes made in the revised manuscript (page 5, lines 136-145).

4) P5, L136-148: Figures 3 & 4: Are all provided CCN and AFs at 0.1% supersaturation? If yes, it should be clear both in the figures and the text. There is no mention whatsoever of the instrument supersaturation in the text, not at what particle sizes this supersaturation corresponds to.

and

5) P6, L168-174: Once more, no mention of instrument supersaturation. Was it constant? Was it the same during all flights for which the ARs are compared between sites?

Figures 3 and 4 represent the CCN counts at 0.1% supersaturation. The mean su-

persaturation was 0.099 $\pm$0.005 (mean $\pm$ standard deviation), with 95 % of the values within the interval 0.098 to 0.100, and with minimum and maximum values of 0.07 and 0.11 respectively. The figures and text are modified in the revised manuscript and the supersaturation is clearly mentioned (page 6, lines 171-176; page 24, Figure 3 and lines 714-716). The value of supersaturation is now included in figures 5 & 6 as well (pages 27-28). We do not have size resolved CCN measurements and the Rosemount inlet limits the maximum size of particles measures to $\sim$3 μm. The CCN values reported in our study represent the size-integrated CCN concentration for input particles below 3 μm, but practically the size of the particles entering the CCN counter was less than 600 nm. Passive Cavity Aerosol Spectrometer Probe (PCASP) measurements (not included in this study) showed that the optical diameters of the particles rarely crossed 600 nm and the numbers of particles above this size was negligibly small. Including the size distribution from PCASP measurements will not address the changes happening below 100 nm (particle diameter), relevant to CCN activation, hence not attempted here. The same supersaturation settings were used throughout the campaign.

6) P6, L181-184: When evoking the anthropogenic impact, anthropogenically impacted emissions are mostly in the lower particle sizes, which means that particles indeed activate in lower ranges of supersaturation. Once more the instrument supersaturation and respective particle size range should be clearly stated.

CCN counts at 0.1% supersaturation are reported here and the corresponding particle size ranges are lower than 600 nm. In general particles are activated if they have sizes larger than the critical diameter required for activation at a given supersaturation. Anthropogenic particles are smaller in size and are activated even at lower ranges of supersaturation, depending on the particle size. If they are big enough they activate at a relatively lower supersaturation. The supersaturation is mentioned in page 6, lines 171-173 and page 8, line 226.

7) P7, L203-214: All this discussion should be put in context also with particle size. Sulfate is mostly found in particle sizes larger than organics.

Complied with in the revised manuscript. The CCN activation shows strong dependence on the size of the particles. Deng et al 2011 showed that the activation curves for ambient aerosols vary significantly from those for ammonium sulfate particles. Following this study, in the range of supersaturation used in our study (0.1%) the particle diameter varies between 75 and 125 nm. If ammonium sulphate and adipic acid with size of 100 nm are considered the former can be activated at 0.15% SS compared to and 0.27% SS for the latter (Hings et al., 2008). Changes made in the revised manuscript (page 8, lines 249-251).

8) P7, L215-225: Therefore the current study offers insight of what happens above the boundary layer? This is the difference between the other studies (Brooks et al., 2019a; Jayachandran et al., 2020a)? This should be clarified, even in the introduction section.

Complied with in the revised manuscript (page 3-4, lines 97-100). The current study provides height resolved information of CCN activation up to 6 km whereas Jayachandran et al, (2020a) made measurements only up to 3.5 km. We observed the lowering of activation ratios due to the presence of BC above the boundary layer, similar to what Jayachandran et al, (2020a) observed close to the surface. Our study explores the impact of the changes in chemistry reported by Brooks et al, (2020a) on the CCN activation across the IGP. These are explicitly mentioned in the revised introduction.

9) P10, L294-195: Operational mode and settings should be comparable to those during the prior monsoon period, correct? Otherwise no comparison is possible.

and

10) Figures 8 & 9: It is clear from these figures that the CCN instrument was operated in different supersaturation levels, therefore it becomes even more imperative that the whole discussion on ARs is clarified, as well as operational conditions between premonsoon and monsoon flights. Also the scatter in these figures is sometimes so high, which raises confidence issues concerning the fitting (e.g. Fig. 8 d & e, 9c)

Yes, the same operational mode was followed throughout the SWAAMI campaign. The operational mode and the supersaturation has been clearly mentioned in the revised manuscript (page 5, lines 136-142). We agree that figures 8 and 9 have high scatter. Our main objective here is to show the relative changes in the properties of CCN across the IGP and not to quantify these changes in terms of their absolute magnitudes. We feel that the figures hold good in this respect. The scatter can be reduced to a certain extent by re-plotting (Figure R3) the mean CCN count against the respective supersaturation bins after omitting a few outliers. This leaves us with limited number of data points as the range of supersaturation was restricted from 0.1 to 0.4, which enabled high resolution measurements even at higher altitudes. The general observations do not change after re-plotting but the robustness of the fit is still questionable considering the limited number of points especially in the case of figure 9. The modified figure 8 is shown below (figure R3). The general pattern remains almost the same except in panels a & f having limited number of points and large spread in the data respectively. Hence, after considering the different options we decided to retain the original figures 8 and 9.

Technical corrections P7, L218: precursor gases (one word) Corrected in the revised manuscript (page 9, line 265)

We thank the reviewer for the detailed comments.

References

Deng, Z. Z., Zhao, C. S., Ma, N., Liu, P. F., Ran, L., Xu, W. Y., Chen, J., Liang, Z., Liang, S., and Huang, M. Y.: Size-resolved and bulk activation properties of aerosols in the North China Plain, Atmospheric Chemistry and Physics, 11, 3835-3846, 2011. Hings, S. S., Wrobel, W. C., Cross, E. S., Worsnop, D. R., Davidovits, P., and Onasch, T. B.: CCN activation experiments with adipic acid: effect of particle phase and adipic acid coatings on soluble and insoluble particles, Atmospheric Chemistry and Physics, 8, 3735-3748, 2008. Jayachandran, V. N., Suresh Babu, S. N., Vaishya, A., Gogoi,

M. M., Nair, V. S., Satheesh, S. K., and Krishna Moorthy, K.: Altitude profiles of cloud condensation nuclei characteristics across the Indo-Gangetic Plain prior to the onset of the Indian summer monsoon, Atmospheric Chemistry & Physics, 20, 2020.
Roberts, G. C., Day, D. A., Russell, L. M., Dunlea, E. J., Jimenez, J. L., Tomlinson, J. M., Collins, D. R., Shinozuka, Y., and Clarke, A. D.: Characterization of particle cloud droplet activity and composition in the free troposphere and the boundary layer during INTEX-B, Atmospheric Chemistry and Physics, 10, 6627-6644, 2010.

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2020-1233/acp-2020-1233-AC2-supplement.pdf

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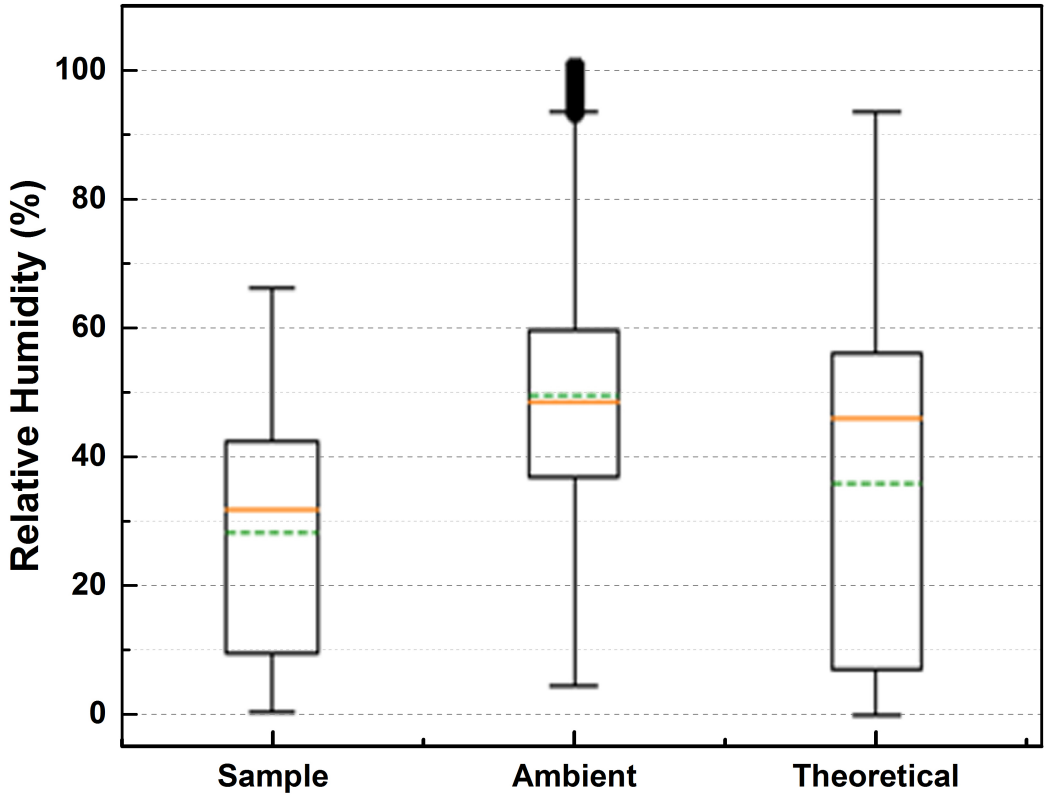

Fig. 1. Figure R1. Boxplots showing, mean (dashed green), median (solid orange) interquartile (box) and interdecile (caps) for the RH data from the three pre-monsoon flights (B956, B957 and B958).

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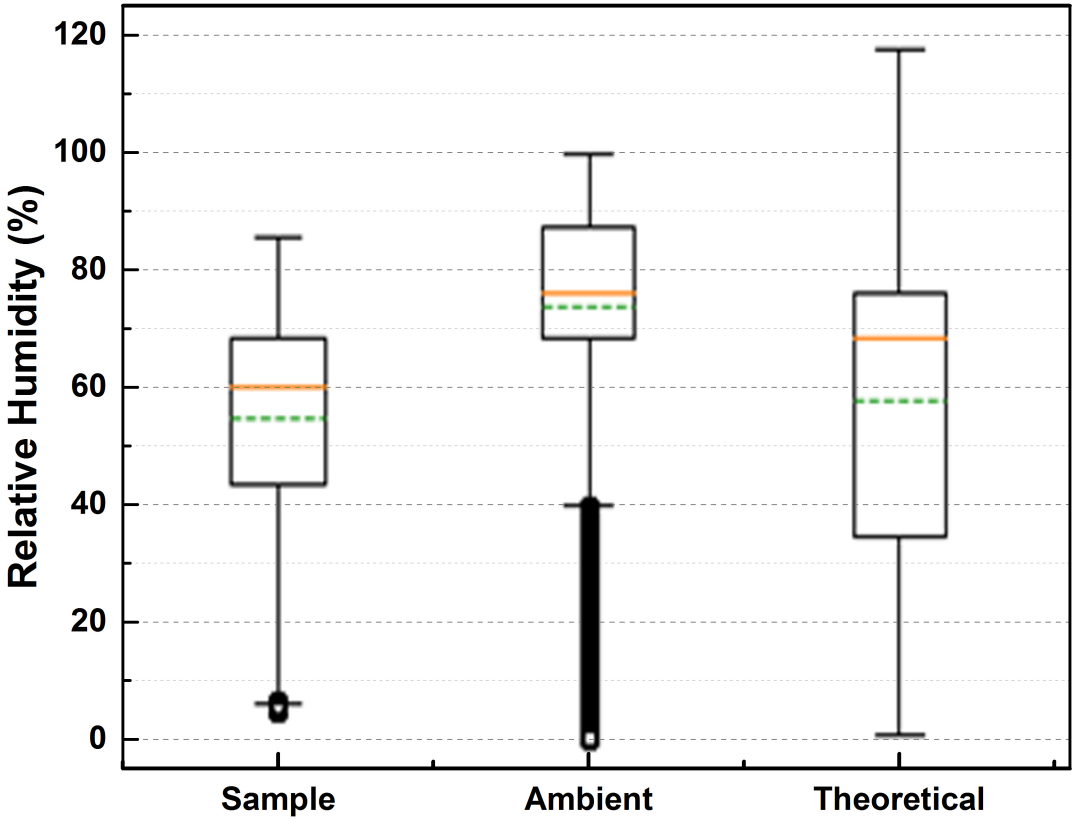

Fig. 2. Figure R2. Boxplots showing mean (dashed green), median (solid orange), interquartile (box) and interdecile (caps) for the RH data from the six monsoon flights (B969 to B974).

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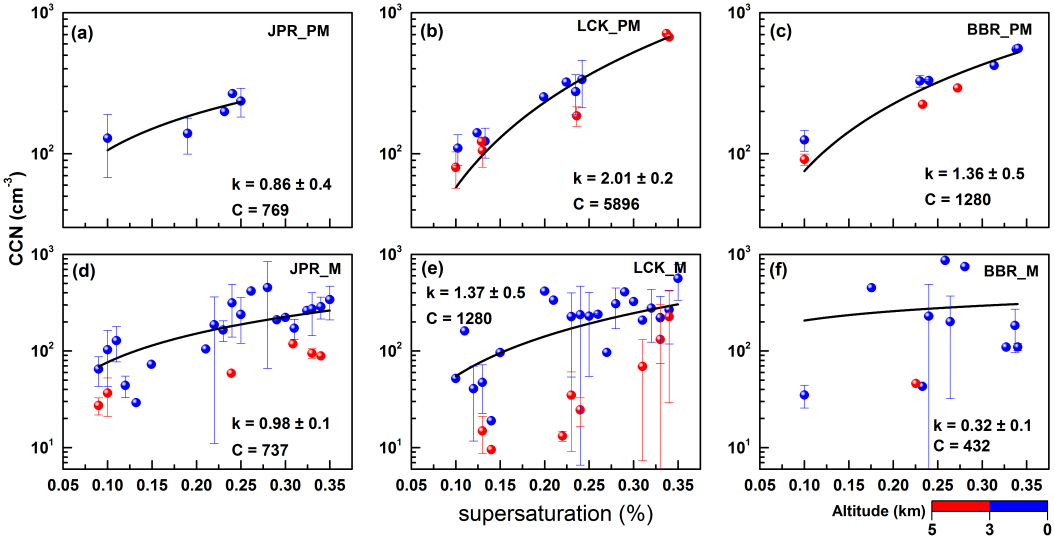

Fig. 3. Figure R3. Figure 8 in the manuscript has been re-plotted taking mean CCN versus supersaturation.

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