Interactive comment on "CCN characteristics during the Indian Summer Monsoon over a rain-shadow region" by Venugopalan Nair Jayachandran et al.

We thank the Editor and appreciate the evaluation of our results by both the referees. We think that the recommendations lead to the overall improvement of the manuscript. We have carefully considered the comments and suggestions, and revised the paper accordingly. Our point-by-point responses to the comments, based on which the revisions are made, are given below. The review comments are given in italics, while the author responses are in bold font

Anonymous Referee #1

The manuscript "CCN characteristics during the Indian Summer Monsoon (ISM) over a rainshadow region" by Jayachandran et al. presents a comprehensive study of CCN characteristics of aerosol particles in the Indian sub-continent prior to, during and after the Indian Summer Monsoon. The paper discusses on-line particle measurements, including CCN number concentrations and aerosol particle size distributions, as well as the CCN parameters derived from these measurements. The aerosol data are supplemented by the aethalometer and meteorological data, as well as HYSPLIT trajectories.

At the moment, the presented study is of rather limited scientific relevance as the potential for CCNC in aerosol-cloud interactions studies has pretty much been exhausted. This is exemplified by the fact that the majority of referenced literature is at least a decade old or more. At this point, CCNC can no longer help us understand aerosol-cloud interactions, and the majority of presented outcomes are already known. This notion is not meant to change or take away from the paper; it is more meant as the direction of potential future work for the authors.

Having said that, the paper is of very good quality and written very well, and the authors do an excellent job at interpreting the data, presenting the results and discussing them in detail (sometimes too much detail). The objectives and conclusions are clearly stated, and the paper makes great use of existing literature and puts its results in perspective. The paper is well-structured and provides the interested reader with a lot of information about CCN characteristics in India during ISM. The authors make as much use of the data as I think is possible, which is definitely a benefit of the presented study. At some points throughout the paper authors make claims that are not supported by their observations, and the paper overall is quite long. However, I definitely recommend the manuscript to be published after the minor revisions suggested below are incorporated.

We thank the reviewer for the encouraging comments and fruitful suggestions.

General comments

1. Lines 40-41 and 594-595 – We know now that both of these statements are not true. CCN characteristics of aerosol particles (size, chemistry, etc.) have now been described in many locations

all over the world, yet challenges in understanding aerosol-cloud interactions (ACI) remain. The biggest challenges in ACI are understanding how CCN interact with water vapour in real atmosphere. We know very little about actual ambient supersaturation levels and the depletion of water vapour during CCN activation, something CCNC cannot help us with. Additionally, there is a very large disconnect between ambient CCN and cloud droplet number concentration (CDNC) (Moore et al., 2013), something we also fully don't understand. Please, rephrase or remove the statements.

Agreeing with the reviewer. Lines 40-41 is modified as follow, 'Characterization of the hygroscopic growth of AP, which is generally addressed by the Köhler theory (Köhler, 1936), is the most fundamental aspect in assessing the aerosol-cloud interactions (ACI) for reducing the uncertainties in indirect radiative forcing estimation'.

'In the real atmosphere, the supersaturation measurements are seldom possible and the large disagreements between the CCN and cloud droplet number concentration remains elusive (Moore et al., 2013).'

and lines 594-595 are removed.

2. In the overwhelming majority of CCN-related and referenced literature, k-value, or κ-value, is predominantly used to describe the hygroscopicity parameter kappa κ (Petters and Kreidenweis, 2007). In the current manuscript, it connotates an empirical fit value of the Twomey's fit. I found it very confusing while reading through the paper, especially since the magnitudes of both parameters are very similar. Is it possible to use any other connotation for the empirical fit value? I think it would make it clearer what parameter you are referring to, but I leave the decision at the discretion of the authors.

We understand the concern. Since all the papers using the Twomey's empirical fit have used the 'k' notation we are also following the same. Considering the reviewer's suggestion, we are clarifying this usage in the manuscript at Line 237 as,

'It should be noted that, the empirical fit parameter k is different from the effective hygroscopicity parameter-κ discussed by Petters and Kreidenweis (2007).'

3. There are three main periods discussed in this study – pre-Monsoon, Monsoon and post- Monsoon. Sometimes they are referred to as such. Sometimes they are referred to by the month. Sometimes they are referred to by continental and marine airmass. Sometimes they are referred to as dry and wet conditions. I found it confusing and I always had to go back and check which period is meant. I think it would make the paper a lot easier to read if the authors stick to one way of describing these periods.

All the analysis in the current manuscript are based on the continental and marine airmass trajectories which coincides with the dry and wet conditions over the region, respectively, within the Indian Summer Monsoon period (June to September) of 2018. Though months were mentioned, it represents the corresponding days only which

experienced the marine/continental air mass. However, to avoid the confusion the periods are uniformly referred as continental (1 and 2) and marine (1 and 2) conditions in the modified manuscript. Continental-1 and 2 comprise of 1-8 June and 15-30 September. While, the marine-1 and 2 comprises of 8-12, 15-31 of July and 1-27. 28-31 of August. These changes are implemented in the whole manuscript including figures.

These clarifications are mentioned in Lines 155-161

4. The paper should acknowledge more the fact that during July and August a lot of the aerosol particles and potentially good CCN are either washed out by wet scavenging or have already activated into cloud droplets, thereby in both cases being removed from the measured ambient aerosol population. In this sense, the aerosol properties measured in July and August represent a subset of APs that is already inherently CCN-inactive. The paper alludes to this on several occasions, but I think it should be present more throughout the paper. For example, lines 497- 498 – how do you know that low GMD and high dcri in August indicate the presence of freshly emitted water-inactive primary organic aerosols? Maybe your larger accumulation mode particles were simply removed by deposition/activation, leaving fine particles behind, skewing the NSD towards lower sizes and increasing your dcri.

We agree with the reviewer.

The missing of CCN active aerosol particles by wet scavenging or activation to cloud droplets leading to the low CCN activation fraction is now highlighted in the manuscript at line 705 as,

'In the presence of marine airmass trajectory (July and August), most of the fine AP which are potential CCN are either washed out by wet scavenging or already activated as cloud droplets. There is also possibility of less emissions due to wet conditions. Hence the measured AP are devoid of CCN active particles which are clearly seen from the aerosol NSD during the relevant periods. Thus, the low CCN activation fraction during the marine conditions is due to the missing of those CCN active particles near the surface.'

The drastic daytime increase in the absorption Angstrom exponent especially during marine conditions as seen in Fig 10(a) is considered as an indication for the local sources contributing to the primary organic aerosols. However, in the current paper those aspects cannot be proved and will be investigated in the ongoing work from the location. Hence, Lines 497-498 are removed.

5. Lines 499-524 – This whole section can be removed from the paper as it presents little to no new or exciting information. Of course, AF and dcri are anticorrelated. It makes perfect sense and wouldn't be any other way. Please, consider removing this section and starting a new section 3.6 at line 525 to describe the relationship between aerosol absorption and CCN properties.

As per suggestion, this part of the manuscript is removed in the revision.

6. Section 3.6 – In this section authors make an attempt to estimate the CCN concentration using several parameters, including a cut-off size, critical diameter or some predefined chemical information. It is immediately visible that setting a cut-off size alone does an excellent job in estimating CCN concentration. All R values are 0.95 or higher, which is amazing and unlikely to get any better. We already know well that size matters way more than chemistry (Dusek et al., 2006), and the fact that you have a >90% predicting capacity of CCN concentration simply by setting a lower size limit is very good and really all we need to know. Especially, since the correlations get even worse when you use dcri or chemical information. The discussion in section 3.6 needs to be reduced by quite much to highlight that particle size is more than enough to accurately estimate CCN concentration. There is absolutely no need to discuss and explain why R of 0.95 is worse than 0.97 because both of these values are very high, higher than in many other studies. The authors seem to be very perfectionist in this section and make statements that are not supported by observations. Chemistry does not play a crucial role in determining CCN efficiency, and there is no need to try and convince the reader that it does. The last bullet point in the Summary and Conclusions section should reflect this as well.

Thanks for the suggestions.

As the reviewer rightly pointed, the estimated CCN showed good correlation in all the three methods, indicating the prominence of aerosol size in CCN activation. Still, there is a significance difference in the activation efficiency of accumulation mode AP prior to and post the monsoon, indicating the difference in the composition. Similarly, the over-estimation of CCN concentration during marine condition for accumulation mode (2) and ammonium sulfate composition assumptions (3.38) shows the hygrophobic composition of those AP. The discussions in section 3.6 is modified as per the reviewer's suggestion and the conclusions are also modified.

7. Lines 703-714 – the discussion here needs to be changed to account for the facts that a) most of CCN-active particles during Monsoon are already removed by activation/deposition and what's left is inherently CCN-inactive, and b) size alone is sufficient to accurately estimate CCN concentration during all months and conditions of the performed study.

Complied with. The lines are modified as follows,

'The closure study indicates the size dependency of CCN activation especially during dry-continental conditions. Most of the CCN-active (fine) AP were removed from atmosphere by activation or wet removal and the remained particles were inherently CCN-inactive as seen in the aerosol NSD during the marine air mass.'

Minor comments

1. *Line 155 – please, define the observation period. It is seen in Table 1 and mentioned in the abstract, but I think it would be nice to include it in the main text as well as well.*

Complied with.

2. *Lines 162-164 – the sentence makes no sense. Please, rephrase.*

Lines 162-164 modified as,

'**From Figure 1(a), it can also be seen that the air mass history for the continental classification are mostly within 2 km above the surface, indicating the chances for the influence of local aerosol sources.**'

3. *Line 180 – winds were blowing from the north-east only during September. Please, state that.*

Figure 2(f) shows that continental air mass days in September experienced winds from both North-East and South-West. However, Line 180 is modified as,

'Westerly and South-Westerly winds were present during marine 1 and 2 conditions (Fig. 2d and e), while the continental-2 days (Fig. 2f) mostly North-Easterly winds were observed.'

4. *Lines 204-205 – "during (b) prior (June) and (c) after (September) monsoon" part makes no sense. Please, rephrase. Also, "…white star indicates…".*

The lines are modified as,

'**Aerosol optical depth (AOD) at 550 nm observed from the Moderate Resolution Imaging Spectroradiometer (MODIS) for the continental (a) 1 and (b) 2 conditions. The site-Solapur is indicated by the white star in the spatial AOD maps.'**

5. *Please, be consistent with units of measurement of BC concentration. Either ng or µg.*

Suggested changes are implanted in the revision. All the units are now represented in ng m^{-3}.

6. *Lines 231-232 – What is the reason for the difference in CCN concentration between continental and marine airmasses? Is it simply because the total number of all AP is different? Or because CCN are less hygroscopic during ISM?*

The study investigates this difference in the CCN population within the ISM period which coincide with different air mass history conditions. The analyses are meant to unravel the causes and found that, the aerosols present during the wet conditions are those CCN-inactive particles mainly due to the wet scavenging. The accumulation mode AP during the marine air mass was less hygroscopic. The distinct aerosol NSD during the marine and continental air mass conditions underline these findings.

7. *Line 247 – that is not really true. Jun, Jul and Sep values are basically the same, only Aug value is higher. But their variability (±) is high, and I would say there is no difference. This should be reflected in the discussion elsewhere, e.g. line 266.*

We agree with the reviewer. Line 247 is removed and the corresponding discussions at Line 266 is modified as,

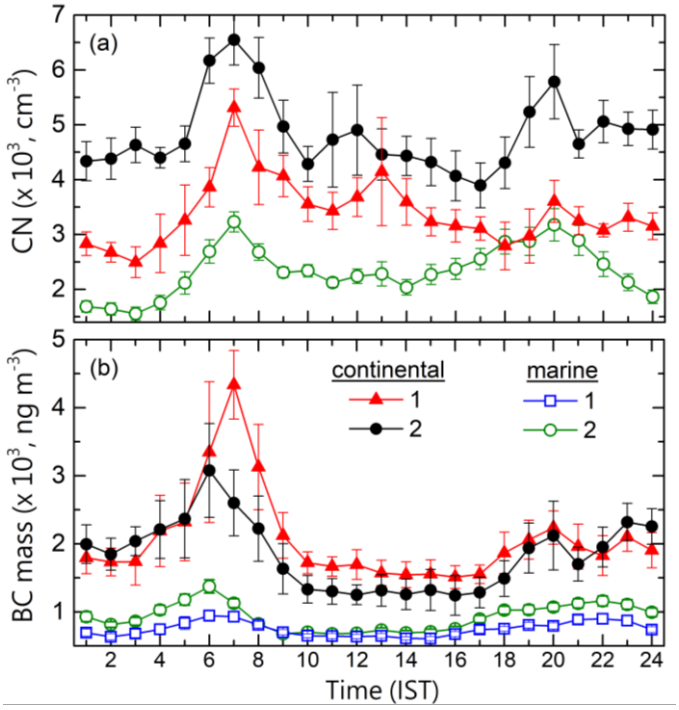
'Generally, an enhancement in k-values is observed during the monsoon period, which is seen only during the marine-2 conditions in this study. In all other cases, the k-values are comparable.'

8. *Line 281 – there is also an increase in CCN after sunrise in September (Fig. 5b). The increase is just not as dramatic as in other months.*

Thanks for the suggestion. This point is added in the manuscript at Line 281.

9. Figure 6 – have you tried combining panels a and b, and panels c and d? It would make comparing them much easier and different magnitudes of values would be easier to see. I have to carefully look at the y-axis values to see that the variation is less/more prominent.

Suggested changes are implanted in the revision. Figure 6 is modified as the following,

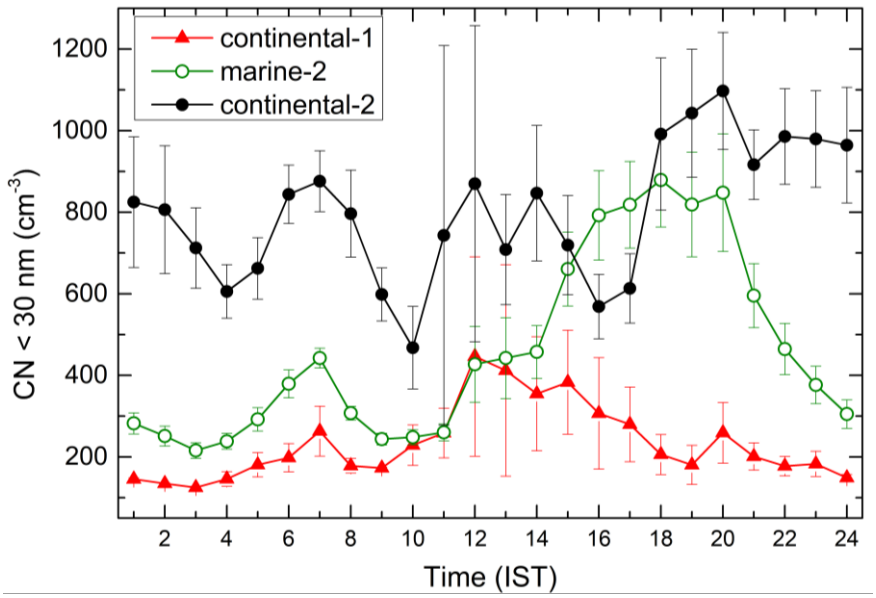


10. Line 288 – I would say that a small increase in CN is seen in the afternoon, instead of around mid-noon.

Suggested changes are implemented in the revision. 'Around mid-noon' is replaced with 'afternoon' in the manuscript

11. Figures 5 and 6 – so why is there a second peak in CN in the evening in all months, but there is no corresponding peak in CCN in the evening? What are those CCN inactive particles?

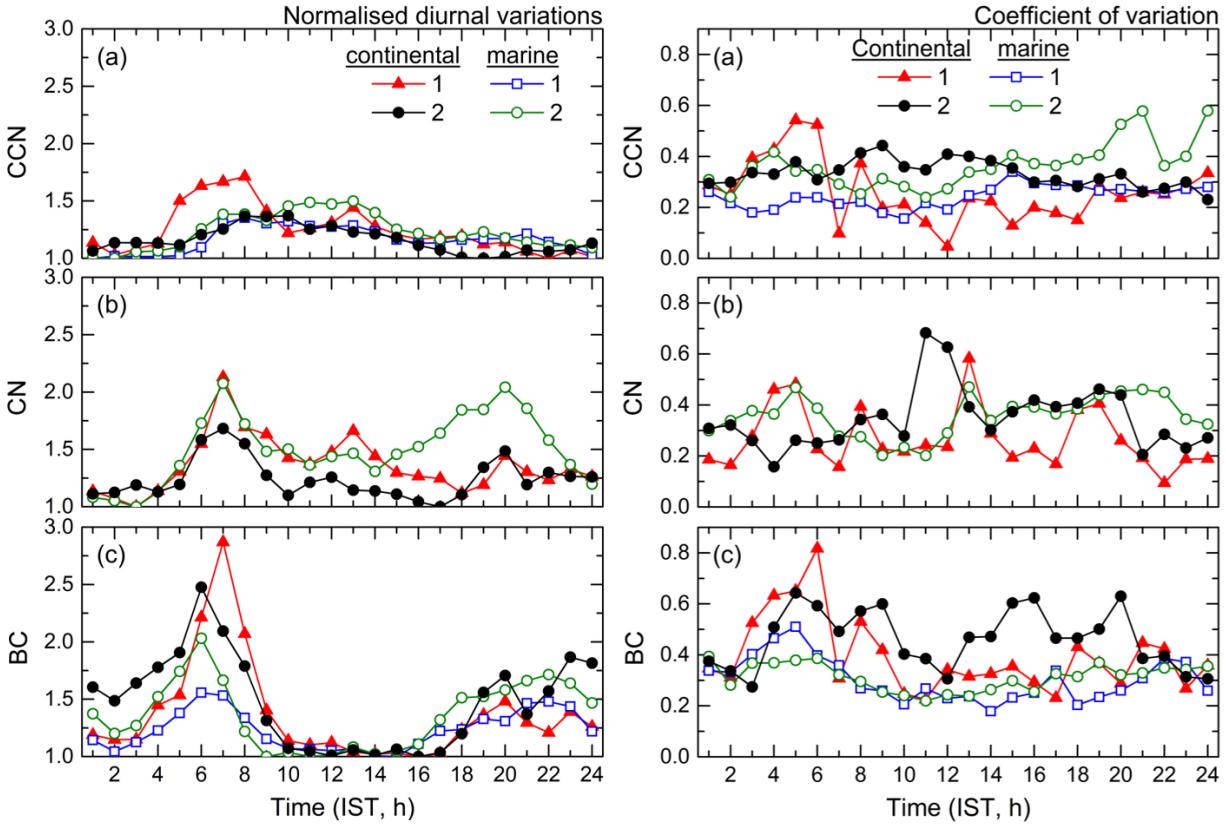
As the reviewer rightly pointed out, the evening peak observed in the CN concentration is not prominent in CCN diurnal variations, except during the marine air mass of July. Interestingly the CCN concentration peaked with sunrise and remained high during the daytime, especially during the marine airmass conditions. To investigate this pattern, the diurnal variation of nucleation mode particles is examined and is shown below, since the size mostly governs the CCN activation. From the figure, it is clear that the evening peak in CN concentration (August and September) is associated with an enhancement in the nucleation mode AP, which do not contribute to the CCN concentration.



12. Lines 312-313 – the statement is not true! For all three parameters (CCN, CN and BC) the diurnal variation during Jun and Sep is higher than in Jul and Aug.

We have examined the normalised diurnal variations (normalised with the least value in a day) and the coefficient of variance (CV) of CCN, CN and BC concentrations, and shown below. BC showed a clear diurnal variation under all the conditions. All the three parameters showed diurnal variations during the continental air mass (2) in September. Other than those, during the marine air mass (2) in August, both CCN and BC had a clear diurnal variation. But the CN concentration shows a prominent diurnal variation for the continental-1 also. Though the diurnal variations in BC was high during both continental conditions, the corresponding CV was also high, indicating the larger spread of the mean values. Thus, it is difficult to attribute the diurnal variations to marine or continental conditions. Hence, we are modifying the lines 312-313 as,

'The diurnal variations during the continental conditions indicate the consistently high AP background conditions. While the diurnal variations during the marine conditions indicate the significant presence of local AP sources.'



13. Lines 318-391 – neither are true. CCN is just a fraction of CN, so if CN increases, CCN is likely going to increase as well. Also, all three datasets in Fig. 7 are fitted well with a linear fit, with all R2 values above 0.8. Therefore, the dependence of CCN on total AP in your study is very much linear.

Suggested changes are implanted in the revision. The sentence is removed.

14. Line 334 – The CCN/CN relationship is August is not weak; at R2 of 0.82 it's quite strong actually. It's just weaker than in Jun and Sep.

Suggested changes are implanted in the revision. The sentence is modified as,

'The relationship between CCN and CN during marine-2 is weaker than the continental phases, and only a few AP are activating as CCN.'

15. Lines 341-342 – neither statements are true. The variation is linear, as mentioned above, and concentration is not 600 cm-3. In Fig. 7b most data points are below 500 cm-3, and Fig. 5a clearly shows CCN concentrations in August between 200 and 400 cm-3.

Agree. The variation is linear. The line is modified as,

'the CCN concentration at 0.3 % was not increasing beyond 600 cm^{-3}, despite of CN concentration increasing to ~7500 cm^{-3}.'

16. Lines 358-359 – the sentence is redundant. Basic physics tell us that is SS goes up, so will CCN and AF.

Suggested changes are implanted in the revision. The sentence is removed.

17. Lines 409-414 – biomass burning aerosol is not known to be particularly CCN-active, unless properly aged. The discussion here makes it sound as though high CCN AF is associated with biomass burning, which I don't think is true. Please, rephrase.

Suggested changes are implemented in the revision. Line 409-414 is modified as,

'The high CCN AF during the continental conditions at Solapur is similar to those reported during dry conditions in Nainital (Gogoi et al., 2015), where the high CCN AF was attributed to biomass burning.'

18. Lines 423-424 – please, indicate which reference you used for defining nucleation, Aitken and accumulation mode sizes.

Suggested changes are implanted in the revision. We followed the classifications from Ueda et al., (2016) and Willis et al., (2016). The references are added in the manuscript.

19. Lines 439 – instead of saying "in the mean picture", please, refer to the Figure and the panel in question.

Suggested changes are implanted in the revision. Line 439 is corrected as,

'Three modes are distinctly observed in Figure 11(c), with two peaks below 100 nm'

20. Line 457 – Figure 11a shows that 81% of particles are below 100 nm in diameter in August, not 60% as mentioned in the text.

For August, the fine particles (<100 nm) are 69%. It is corrected in the manuscript.

21. Lines 468-469 – I don't think all accumulation mode particles are always associated with biomass burning. Or maybe better to say that accumulation mode particles are associated either with biomass burning or with condensation and coagulation of smaller particles.

Suggested changes are implanted in the revision. The sentence is modified as,

The accumulation mode particles are associated with either aged biomass burning particles (Kalvitis et al., 2015) or condensation and coagulation of smaller secondary organics and inorganics particles (Seinfeld and Pandis, 2016)

22. Lines 473-476 – please, use punctuation in this sentence. It is currently not clear if oxygenated organic aerosol and sulfates are responsible for higher GMD or if long chain hydrocarbons are responsible for fine mode and oxygenated organic aerosol.

Suggested changes are implemented in the revision. The sentence corrected as,

'while oxygenated organic aerosols and sulfates are responsible for the higher GMD'

23. Lines 531-533 – there is no need to describe in the main text what should be and already is in the legend of the figure. All descriptions of the figures (symbols, lines, etc.) should be found in the legend and not in the main text. Please, correct this throughout the entire manuscript. This will also make the paper a bit shorter.

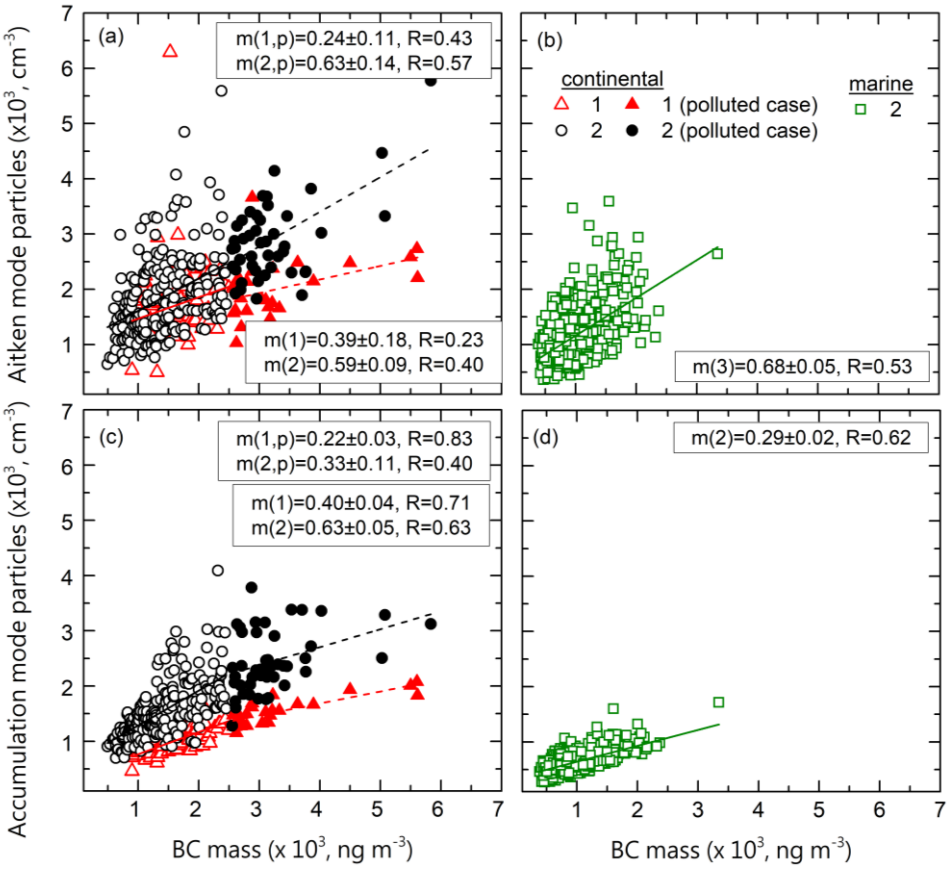
Suggested changes are implanted in the revision.

24. Line 534 – "comparatively high" should be replaced with "moderate".

Suggested changes are implanted in the revision.

25. Figure 14 – is impossible to read. First, what are the colours and symbols? The legend needs to be improved so it is clear what symbols and colours mean. Second, I would say that the BC mass is the independent variable and Aitken mode particles are the dependent variable, so the axes in the figure need to be switched.

Suggested changes are implemented in the revision. The figure is modified as below and the linear fits are carried out separately for BC concentrations 2500 ng m^{-3}, and above it which is considered as the polluted case. This classification is made since the BC mass during the wet periods was mostly (99%) below 2500 ng m^{-3}.



26. Line 676 – when is this true? During ISM?

The lowest values of CCN concentration is reported during the marine air mass conditions during the ISM period over Solapur.

Technical comments

There are several grammatical, punctuation and other errors, most of which will be corrected during the copy-editing stage of the manuscript. The errors indicated below stood out but are not exhaustive.

1. *Lines 73-77 – the sentence is missing a verb.*

Suggested changes are implanted in the revision. The following revision is done,

'Various studies have addressed the spatio-temporal distribution of AP (Padmakumari et al., 2013; Varghese et al., 2019), cloud microphysics (Prabha et al., 2011; 2012; Padmakumari et al., 2018), rainfall (Maheshkumar et al., 2014) properties, the relationship between cloud microphysics and thermodynamics (Bera et al., 2019), and ACI (Pandithurai et al., 2012; Prabha et al., 2012; Konwar et al., 2012; Gayatri et al., 2017; Patade et al., 2019) from the unique data obtained from the CAIPEEX.'

2. *Line 93 – "…data presented in this study are…". Data are plural and this should be reflected everywhere else in the paper.*

Suggested changes are implanted in the revision.

3. *Line 177 – remove the word "months" after "July and August"*

Suggested changes are implanted in the revision.

4. *Line 187 – "CCN characteristics at the site are…"*

Suggested changes are implanted in the revision.

5. *Line 266 – "An enhancement in k-values is…"*

Suggested changes are implanted in the revision.

6. *Line 356 – should say "inactive"*

Suggested changes are implanted in the revision.

7. *Line 436 – "…are also similar…"*

Suggested changes are implanted in the revision.

8. *Line 565 – my name is misspelled. Should be Paramonov* 😊

Sorry for the mistake. Corrected

References

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J.,Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., and Andreae, M. O.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, Science,312, 1375–1378, doi:10.1126/science.1125261, 2006.

Moore, R. H., Karydis, V. A., Capps, S. L., Lathem, T. L., and Nenes, A.: Droplet number uncertainties associated with CCN: an assessment using observations and a global model adjoint, Atmos. Chem. Phys., 13, 4235–4251, doi:10.5194/acp-13-4235-2013, 2013.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971, doi:10.5194/acp-7-1961-2007, 2007.

Ueda, S., Miura, K., Kawata, R., Furutani, H., Uematsu, M., Omori, Y. and Tanimoto, H., 2016. Number–size distribution of aerosol particles and new particle formation events in tropical and subtropical Pacific Oceans. *Atmospheric environment*, *142*, pp.324-339.

Willis, M. D., Burkart, J., Thomas, J. L., Köllner, F., Schneider, J., Bozem, H., Hoor, P. M., Aliabadi, A. A., Schulz, H., Herber, A. B., Leaitch, W. R., and Abbatt, J. P. D.: Growth of nucleation mode particles in the summertime Arctic: a case study, Atmos. Chem. Phys., 16, 7663–7679, doi:10.5194/acp-16-7663- 2016, 2016.

Thank you for an excellent paper and best of luck with the review process!

Anonymous Referee #2

General Comments:

The article provides a comprehensive account of cloud condensation nuclei characteristics over a rain shadow region in Western Ghats India. CCN study over Indian region, especially in the rain shadow regions are important in understanding aerosol-cloud interactions and their implications. The data collection and analysis are quite extensive and results are presented comprehensively. I recommend publishing this work after the comments are adequately addressed. One general lacuna is that the authors do not go beyond reporting the data and results of analysis, which though are good in themselves. A rigorous discussion in the light of the results on the CCN characteristics is needed to improve the scientific content in this work. In general, an improvement of the language would help to understand the importance of the finding better.

We thank the reviewer for the careful reading and fruitful suggestions.

Major Comments:

1. Entire analysis of this study is a comparison between CCN characteristics during continental air mass and marine air mass during 2018 monsoon over a rain shadow region in western ghat, India. However, it is not clear that how they delineated the continental air mass and marine air mass trajectories. It is important to make to clear whether the Hysplit model was ran for every 30 min (since the CCN and other data are available for 30 min interval) and the data are segregated accordingly for analysis for the entire study period or took a specific time and used that data only for further analysis. For a general reader it seems that entire June and September trajectories over study region are continental. But it is also mentioned that the monsoon onset is on 08 June.

From the continuous CCN and aerosol observations from 01 June to 30 September of 2018, we have classified days as continental and marine. For this we examined the air mass back trajectories for 5 days from Hysplit model, ran at 07 UTC (12:30 IST) every day. We checked the variability in air mass history reaching the site at different times in a day and found that there was no drastic change from continental to marine influence within a day. Thus the 12:30 IST was fixed as the standard representation for a day. Also, the different instruments had distinct sampling frequency and we have averaged the data from each instrument to hourly basis for the interpretations, which is mentioned in Line 150. As the summer monsoon onset was on 08 June, the air mass showed a clear marine influence from the date and continued the same up to 14 September. Prior to 08 June and from 15 September onwards, the air mass had predominant history over the land and was confined within two km above the surface. Thus, these days are categorised as continental which coincided with the dry conditions as revealed from the temperature

and relative humidity values measurements from the local AWS (Fig. 2 a and b). This way of classification is mentioned from lines 155 to 160.

2. Most of the analysis focuses on reporting of values and comparison with reported values from other sites. Authors should also try to add more science through discussion related to the implication of their observation. For instance, discussion on the implication of the role of carbonaceous aerosols in acting as CCN over the study region during dry conditions leading to semi-direct effect/rapid adjustment. Does dust aerosol have any role in modulating CCN properties over the region?

Suggested changes are implanted in the revision. In this regard we have modified Fig. 14 and more discussions as suggested is added in Line 585 as,

'The better association of CCN with absorption coefficient especially prior to the monsoon is interesting. The strong convective conditions existing during these conditions over the region can take the AP to high altitudes where it can absorb radiation and may lead to semi-direct effects. The association of BC with accumulation mode AP during continental conditions suggest that the carbonaceous AP existing in this size range can act as CCN. Hence, the role of carbonaceous AP in modulating both cloud microphysics and dynamics need to be investigated in detail. However, current investigation could not address these probable aspects.'

We didn't have enough evidence to show the role of dust aerosols influencing the CCN properties during the study period. As the reviewer mentioned in the 'minor comments', we have checked the possibility of dust coated with carbonaceous aerosols and found that possibility is absent. But we do agree that the point is valid over the region especially during the pre-monsoon to monsoon transition period and will be investigated using single particle and mixing state observations.

3. What hypothesis the authors put forward to explain the reduced activation ratio during monsoon/ marine airmass conditions, when normally the aerosols would be richer in hygroscopic species, that could be easily activated?

The reduced CCN activation ratio during the marine/wet conditions is due the removal of CCN active aerosol particles by wet scavenging and cloud activation. Similar observations of reduced CCN efficiency of AP due to the dominance of smaller AP are also observed over the Western Ghats during the monsoon (Jayachandran et al., 2018). This point is highlighted in the manuscript at Line 478 as,

'During the marine conditions, most of the bigger AP which are potential CCN are either washed out by wet scavenging or already activated as cloud droplets. Hence the measured AP are devoid of CCN active particles which are clearly seen from the aerosol NSD during the relevant periods (Fig. 11). Thus, the low CCN activation fraction during

the marine conditions is due to the missing of those CCN active particles near the surface.'

4. In CCN closure analysis, describe the methodology and assumptions used in estimating CCN. Why aerosol composition is assumed to be ammonium sulphate? In the analysis authors have explicitly tried to establish the influence of carbonaceous aerosols in acting as a CCN?

The details of CCN closure analysis are added in the Appendix.

Assumption of ammonium sulphate as the soluble fraction of the aerosol composition is the ideal case as sulfates are known to be the best CCN (Bigg, 1986; Covert et al., 1998). In the absence of concurrent aerosol composition measurements, we wanted to check the deviation of CCN activation from the ideal scenario. Hence ammonium sulfate was assumed as the aerosol composition following VanReken et al., (2003), Medina et al., (2007) etc. Though the study points to the pivotal role of carbonaceous AP toward CCN activation, we do not have quantitative information of aerosol composition in order to use in the CCN closure analysis.

Minor Comments

Line 32-40: Sentence is confusing and needs modification. It gives a feel that "Condensation nuclei" and "Cloud Condensation nuclei" are same. The sentence starting with "For a fixed liquid water content……." need to be revised.

Suggested changes are implanted in the revision. The Lines are modified as,

'Those AP or condensation nuclei (CN) which act as the Cloud Condensation Nuclei (CCN) at a specific supersaturation (SS) can indirectly affect the climate by altering the cloud micro-physical properties. In general, an increase in AP increases the cloud droplet concentration with smaller sizes (Twomey and Warner, 1967) for a fixed liquid water content, which in turn increases the cloud albedo (Twomey, 1977) and cloud lifetime (Albrecht. 1989).'

Line 40-45: "Characterization of CCN…… the physical and chemical characteristics of AP". These two sentences can be reframed to one as it tries to convey the same information.

Suggested changes are implanted in the revision. The lines are modified as,

'Characterization of the hygroscopic growth of AP, which is generally addressed by the Kohler theory (Kohler, 1936), is the most fundamental aspect in assessing the aerosol-cloud interactions (ACI) for reducing the uncertainties in indirect radiative forcing estimation.'

Line 52-55: "For a given particle……the accuracy of climate models to address the ACI (Fountakis and Nenes,2005)". The relevance of this sentence in the paragraph is not understood. In the second paragraph authors try to portray the heterogeneities of aerosol particles and CCN in the global scenario as well as in Indian context. Authors can discuss more on the role of organics as CCN as they can reveal the first indirect effect (Nenes et al., 2002) and as well as studies over organics in Indian context.

Suggested changes are implanted in the revision. More discussions as suggested are added in the modified manuscript.

Line 73-77: "Various studies…….. from the unique data obtained from the CAIPEEX". Rephrase the sentence.

Suggested changes are implanted in the revision. The lines are modified as,

'Various studies have addressed the spatio temporal distribution of AP (Padmakumari et al., 2013; Varghese et al., 2019), cloud microphysics (Prabha et al., 2011; 2012; Padmakumari et al., 2018), rainfall (Maheshkumar et al., 2014) properties, the relationship between cloud microphysics and thermodynamics (Bera et al., 2019), and ACI (Pandithurai et al., 2012; Prabha et al., 2012; Konwar et al., 2012; Gayatri et al., 2017; Patade et al., 2019) from the unique data obtained from the CAIPEEX.'

Line 80-90: Authors have mentioned that a few studies (Leena et al., 2016, Jayachandran et al., 2018) have already reported CCN characteristics over different locations of Western Ghats. If so, does this study address the same objectives with observations form a different site? Please bring more clarity to the objectives of this study.

The present study is the only study till date investigating the CCN properties at the leeward side of the Western Ghats, which is prone to drought conditions. Apart from the atmospheric dynamics, the role of aerosols in cloud nucleating over this region is still missing. Thus, the current study is the first attempt to understand the role of AP in cloud formation over the region. The main emphasis in the present study is the surface CCN observations during the Indian summer monsoon over the semi-arid rain shadow region, which has not been available from other observations

Line 110: Were the data corrected for the maximal activated fraction, which is of high importance, in particular for total CCN measurements (Paramonov et al., 2013; Rose et al., 2010)? Please give more information of reference data used in the köhler theory when performing the CCN calibration. This is very important because different parameterizations will retrieve different critical supersaturations (Rose et al.,2008; Wang et al., 2017). Also mention the uncertainty in measurements of different instruments.

CCN counter was calibrated with ammonium sulfate aerosol following Rose et al. (2008). During the calibration experiments CCN efficiency spectra were recorded for different CCN column ΔT values. The activation diameter corresponding to 50% CCN efficiency for each spectrum was taken as the critical dry diameter for the CCN activation of ammonium sulfate particles. The corresponding critical supersaturation was calculated with the activity parameterization Köhler model (AP3) mentioned in Rose et al., (2008). The calculated critical supersaturation was taken as the effective supersaturation at the given ΔT value.

The CCN counter used in the study was factory calibrated at DMT Inc prior to the experiment and all the instruments are periodically calibrated during the experiment. The uncertainty associated with all the measurements are less than 10%.

These details are added in the manuscript.

Line 162-165: Rewrite the captions specifically for Figure1. (a) & (b).

Suggested changes are implanted in the revision.

Line 186: Correct bullet numbering.

Corrected.

Line 190-220: There are several concerns in this analysis: (1) Does it mean that for all days in June and September, trajectories ending over study region were of continental origin? This is difficult to comprehend especially when authors have mentioned that monsoon onset over study region was on 08 June2018. (2) AOD retrieved from MODIS over land and especially during ISM is a matter of concern. (3) Authors also mentioned about MODIS retrieved fire count information, please do mention the confidence level used as well as its uncertainty.

All the analysis in the current manuscript are based on the continental and marine airmass trajectories which coincides with the dry and wet conditions over the region, respectively, within the Indian Summer Monsoon period (June to September) of 2018. Though months were mentioned, it represents the corresponding days only which experienced the marine/continental air mass. However, to avoid the confusion the periods are uniformly referred as continental (1 and 2) and marine (1 and 2) conditions in the modified manuscript. Continental-1 and 2 comprise of 1-8 June and 15-30 September. While, the marine-1 and 2 comprises of 8-12, 15-31 of July and 1-26. 28-31 of August. These are mentioned in Table 1. However, considering the reviewer's concern, the details of the observation periods are added in the text as well. Single terminology (continental and marine) is implemented in the whole manuscript including the figures.

The fire spots are detected on daily basis from the MODIS sensors aboard the Terra and Aqua platforms at 1 X 1 km^2 spatial resolution globally. The detection is performed based

on the algorithm (Giglio et al., 2003; 2016) that uses the strong emission of mid-infrared radiation from fires. In the present study, we have considered only the data having confidence value higher than 30, which comes under the classification of 'nominal' and 'high'. This information is added to the manuscript.

Line 225-270: From this study as well as those conducted over Mahabaleshwar and Amazon, an increase in ïA¸n´ values is reported during wet months. What is the scientific reason?

The increase in 'k' value during the wet months is associated with the enhancement in ultrafine particles which are CCN-inactive, in the total AP concentration. The increase of these small particles due to the wet scavenging necessitates higher supersaturation conditions for CCN activation. Hence, the CCN concentration increase drastically at high supersaturations resulting in a comparatively high 'k' value. The role of aerosol size in determining the k value is discussed in Nair et al., (2020).

*Line 315-350: Justify your arguments on the formation of NPF over the study region during wet conditions? Why the CN-CCN relationship weakens during September? CN-CCN relationship seems to hold strong when CN concentration is _3.7*10^3 particles/cm-3. Is it due to instrumental artefact? Or do you propose any process?*

The enhancement in nucleation mode particles (<30 nm) are seen mostly during the afternoon hours over the region. During the wet conditions, the AP concentration capable of providing deposition of pre-cursor gases are very less as seen in Figure 11, which can lead to the nucleation of AP. We have not investigated this aspect in detail in the current study. Hence new particle formation may be one of the reasons for the observed AP distribution having GMD less than 50 nm during the marine air mass.

There could be other reasons such as local emissions and turbulent mixing, which may be investigated later. Note that meteorology (PBL mixing) has a decisive role in the redistribution of particles of this size range.

From the closure studies, it can be seen that the accumulation mode particles (>100 nm) observed during continental-2 is less CCN active indicating the presence of hygophobic combination compared to continental-1. Also, from Fig. 11, it can be seen that the nucleation mode particles are more than twice during continental-2 than 1. Thus, both the presence of nucleation mode particles and lesser hygroscopic accumulation mode particles are responsible for the weak CN-CCN relationship during the September, compared to June.

Line 360: Figure 8 clearly shows that activation fraction (SS) is very low during wet months. What does it imply? Are similar observations are reported elsewhere? Discuss more on the implication and physical mechanisms?

Low CCN activation fraction during the wet periods are reported by Jayachandran et al., (2018) over the southern tip of peninsula. The concurrent AP number-size distribution (Fig. 11) points to the predominance of smaller particles due to wet scavenging during these conditions and are not getting replenished. The CCN-inactive particles are responsible for the low CCN activation fraction. This has a major impact on cloud microphysics which is a challenging task to quantify. The enhanced ultrafine AP concentration during the large supersaturation conditions can intensify the convective strength (Fan et al., 2018) and is an active area is further research.

Line 365-370: Why the diurnal variation in Twomey's empirical fit parameter –ïA¸n´ and activation fraction is not showing (revere) relationship that is obviously seen in other months. During September, diurnal variation in GMD showed morning high values, which is also reflected in AF but not in Twomey's empirical fit parameter –ïA¸n´ . Similarly, in June, GMD showed low values during morning hours without any significant change in AF and Twomey's empirical fit parameter –ïA¸n´. Discuss the scientific implications of these

As the reviewer have rightly pointed out, the clear diurnal variations observed in GMD and activation fraction is not seen in the Twomey's empirical fit parameter-k, during September. The inverse relationship between CCN activation fraction and Twomey's empirical fit parameter- k fails during the presence of very small AP (Nair et al., 2020). There is a predominant nucleation mode AP during September (Fig. 11). The aerosol composition prior to and post the monsoon rainfall is a major missing factor in this analysis. These inferences point to the fact that the Twomey's empirical fit- k is not a perfect parameter to assess the CCN activation of AP. From Fig. 10, it can be seen that the diurnal variation in GMD during June is negligible (within the error bars) even though there is a dip during 10-16 hrs local time. Correspondingly there is no diurnal variation in CCN activation fraction and the 'k' value.

Line 395-400: Authors have tried to associate high Twomey's empirical fit parameter –ïA¸n´ values observed during morning hours of wet months to organic aerosol mostly produced by biomass burning. However, the MODIS fire count map during August shows very less fire count over the study region. Justify the statement?

As the reviewer rightly pointed out, the biomass burning indicated by the MODIS fire count is less over the study region during the wet period. Since the sampling site is on a rural setting, the use of solid fuels is dominant for domestic purposes which can contribute to the organic aerosols. Studies (Zhang et al., 2013) have shown that secondary organic carbons have high absorption Angstrom exponent.

Line 400-414: "Thus, the aerosol composition especially the organic aerosols: : :.." Please justify. In literature AAE greater than 2 is usually inferred as biomass source and AAE between 1 and 2 is usually

considered as mixture of BC and OC (Bergstorm et al., 2007). Authors should present independent observations or data to attribute this to organic aerosols.

Agree with the reviewer. AAE between 1 and 2 is due to the mixture of BC and OC and higher the AAE value point to the dominance of OC. The references cited in the corresponding discussion in the manuscript also indicate the same. Thus, in the present study, the sudden increase in 'k' values was associated with a high AAE, which indicate the predominance of OC. Due to the absence of aerosol composition measurements in the current study, it is difficult to attribute the exact type of organic aerosols causing this enhancement in both AAE and k values.

Line 515: Are the estimated 'a' and 'b' value are site and season specific. ?

The values are specific to the range of critical diameters which are season specific. However, the plot and the associated discussions are removed from the manuscript as per the other reviewer 1 suggestions.

Line 555: What about the role of dust aerosol (local/transported) acting as CCN? As can be seen in Figure, there are a few points where the AAE is less than 1. Some studies have attributed such points to dust coated with BC. Do back-trajectories in these case support dust transport?

We have looked the air mass trajectory for those cases. As it can be seen from the air mass back trajectories none of them are originating from the north-west or west part, from where dust transport is expected. To confirm the absence of dust over the region in this study, we have examined the aerosol NSD also for AAE<1 instance. Both the air mass back trajectories and NSD show negligible role of dust AP.

Line 590: Figure 13 and Figure 14 clearly indicates the role of carbonaceous aerosols acting as CCN over the study region during dry conditions (June and September). Is there any specific reason for the better correlation observed in June?

The co-occurrence of carbonaceous AP and CCN and associated relationship as seen in Fig. 13 and the weakening of the same during the September is noted. AP size plays a major role in this difference. A seen in Fig. 11, the NSD during the continental conditions in June has negligible nucleation mode particles (7%), compared to that of September (15%). This aspect is brought out more clearly by the modified Figure 14. The accumulation mode AP have a better correlation with the absorption coefficient during the dry condition prior to the monsoon. The atmospheric conditions will be favouring the aging of carbonaceous AP leading to the CCN activation. Detailed studies regarding the mixing state and size distribution of AP over the region is needed to confirm.

Straighten up the formatting errors in reference list. Check line 855 for example.

Suggested changes are implanted in the revision. All the references are now modified according to the journal norms

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CCN characteristics during the Indian Summer Monsoon over a rain-shadow region

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Abstract. Continuous aerosol and Cloud Condensation Nuclei (CCN) measurements carried out at the ground observational facility situated in the rain-shadow region of the Indian sub-continent are illustrated. These observations were part of the Cloud-Aerosol Interaction Precipitation Enhancement EXperiment (CAIPEEX) during the Indian Summer Monsoon season (June to September) of 2018. Observations are classified as dry-continental (monsoon break) and wet-marine (monsoon active) according to air mass history. CCN concentrations measured for a range of supersaturations (0.2-1.2 %) are parameterized using Twomey's empirical relationship. CCN concentrations ~~even~~ at low (0.2 %) supersaturation (SS) were high (>1,000 cm^{-3}) during continental conditions ~~associated~~ and observed together with high black carbon (BC~2,000 ng m^{-3}) and columnar aerosol loading. During the marine air mass conditions, CCN concentrations diminished to ~350 cm^{-3} at 0.3 % SS and low aerosol loading persisted (BC~900 ng m^{-3}). High CCN activation fraction (AF) of ~~≅~~0.55 (at 0.3 % SS) were observed before the monsoon rainfall, which reduced to ~~≅~~0.15 during the marine air mass~~onsoon~~ and enhanced to ~~≅~~0.32 after that. Mostly mono-modal aerosol number-size distribution (NSD) with a mean geometric mean diameter (GMD) of ~~≅~~85 nm, with least (~~≅~~9 %) contribution from nucleation mode (<30 nm) particles persisted before monsoon, while multi-mode NSD with ~~≅~~19 % of nucleation mode particles were found during the marine air mass~~onsoon~~. Critical activation diameters (d_{cri}) for 0.3 % SS were found to be about 72, 169, and 121 nm prior, during and, after the marine conditions~~onsoon~~, respectively. ~~The estimated d_{cri} were inversely correlated to the AF, linearly up to 100 nm and non-linearly beyond that.~~ The better association of CCN with aerosol absorption, and the concurrent accumulation mode particles during continental conditions, point to the possibility of aged (oxygenated) carbonaceous aerosols enhancing the CCN activity prior to the monsoon. ~~The~~ An enhancement in CCN concentration and k-values during daytime along with ~~the increase in~~ absorption Angstrom exponent~~, indicate the freshly emitted local anthropogenic aerosols dominated by organics reducing the CCN AF~~was observed during the ~~monsoon~~marine conditions. Best closure obtained using measured critical diameter, and ammonium sulfate composition during continental conditions emphasize the role of aged aerosols contributing to the accumulation mode, enhancing the CCN efficiency. T~~But t~~he over-estimation of CCN and less hygroscopicity of accumulation mode aerosols during the marine air mass~~onsoon point to the significant~~indicate the role of size-dependent aerosol composition in CCN activity during the period.

1 Introduction

Atmospheric aerosol particles (AP) emitted from both natural and anthropogenic sources, affect the radiation budget as well as the hydrological cycle of Earth, mainly through its direct and indirect effects. Those AP or condensation nuclei (CN) which act as the Cloud Condensation Nuclei (CCN) at a specific supersaturation (SS) can indirectly affect the climate by altering the cloud micro-physical properties. In general, an increase in AP increases the cloud droplet number concentration and decreases the size of droplets (Twomey and Warner, 1967) for a fixed liquid water content, which in turn increases the cloud albedo (Twomey, 1977) and cloud lifetime (Albrecht. 1989). In the real atmosphere, the superstaturation measurements are seldom possible and the large disagreements between the CCN and cloud droplet number concentration remains elusive (Moore et al., 2013).~~Cloud Condensation Nuclei (CCN) the sub-set of AP or Condensation Nuclei (CN), that can be activated at a specific water vapor supersaturation (SS), and indirectly affect the climate by altering cloud micro-physical properties. For a fixed liquid water content, an increase in CCN concentration increases the cloud droplet number concentration (Twomey and Warner, 1967), while reducing the cloud droplet size (Twomey, 1974), and thereby altering the cloud albedo (Twomey, 1977) and lifetime (Albrecht, 1989).~~ All these effects eventually modify the precipitation pattern (Lohmann and Feichter, 2005; Rosenfeld et al., 2008). Some of these aerosol indirect effects are moderately understood, while others are not, which contribute to significant uncertainty among all the climate forcing mechanisms (IPCC, 2013). Characterization of the hygroscopic growth of AP, which is generally addressed by the Köhler theory (Köhler, 1936), is the most fundamental aspect in assessing the aerosol-cloud interactions (ACI) for reducing the uncertainties in indirect radiative forcing estimation.~~Characterization of CCN is the most fundamental challenge in assessing the aerosol-cloud interactions (ACI) and reducing the associated uncertainties in indirect radiative forcing assessment. In this regard, the primary aspect is to characterize the hygroscopic growth of AP with respect to relative humidity, which is generally addressed by the Köhler theory (Köhler, 1936) and direct bearing from the physical and chemical characteristics of AP.~~ However, Köhler theory is modified to accommodate the real atmospheric conditions and applied for both laboratory and field measurements, as well as in the climate models (Shulman et al., 1996; Laaksonen et al., 1998; Raymond and Pandis, 2003; McFiggans et al., 2006; Petters and Kreidenweis, 2007; Rose et al., 2008; Mikhailov et al., 2009). For a given particle, the size and composition determine its CCN activity at a specific SS, while the CCN spectrum (CCN at different SS) depends on the median diameter and standard deviation, number concentration, and the mixing state of the aerosol system (Quinn et al., 2008). In this regard, closure studies are necessary to understand the role of each parameter in the activation of AP as CCN, which may improve the accuracy of climate models to address the ACI (Fountakis and Nenes, 2005).

Large spatial and temporal heterogeneities are found in AP as well as CCN properties and thus, the regional characterization of CCN in different meteorological settings are imminent. Temporal and spatial heterogeneities of CCN and different mechanisms affecting CCN are investigated in several studies (Hoppel et al., 1973, Hudson and Xie, 1999, Paramonov et al., 2015, Schmale et al., 2018; Nair et al., 2020) over both continental and marine environments. Over the land mass, significant variability in CCN activation properties are reported due to urban and industrial influences (Sotiropoulou et al.,

2007; Asa-Awuku et al., 2011). Carbonaceous combustion AP produced mostly from urban and industrial activities, contribute more than half of the global CCN concentration (Spracklen et al., 2011). Though nascent black carbon (BC) AP are insoluble (Weingartner et al., 1997), coating, condensation and coagulation of organic and inorganic AP can increase its hygroscopicity, thereby acting as CCN (Liu et al., 2013). ~~For a given particle, the size and composition determine its CCN activity at a specific SS, while the CCN spectrum (CCN at different SS) depends on the median diameter and standard deviation, number concentration, and the mixing state of the aerosol system (Quinn et al., 2008). In this regard, closure studies are necessary to understand the role of each parameter in the activation of AP as CCN, which may improve the accuracy of climate models to address the ACI (Fountakis and Nenes, 2005).~~ However, the role of organics, mostly from carbonaceous combustion sources, in determining the CCN activity is still uncertain. Ervens et al., (2005) have reported a broad range (-86 % to 110 %) of changes in cloud droplet number concentration due to the organics. The reduction in surface tension by organic AP can even perturb the first indirect (Twomey) effect (Nenes et al., 2002). The presence of water soluble organic carbon can increase the CCN concentration, especially in rural and urban settings (Mircea et al., 2002). Recent studies (Singla et al., 2017; Nair et al., 2020) highlight the dominance of organic AP and its significant role in CCN activation over the Indian subcontinent. The increasing trend in aerosol loading (Babu et al., 2013) and the significant contribution of carbonaceous aerosols from both fossil fuel and biomass burning over the Indian subcontinent (Nair et al., 2007) highlight the necessity of the characterization of CCN and the role of carbonaceous AP over distinct environments in India ~~and the role of carbonaceous AP~~.

Even though the aerosol properties such as aerosol optical depth (Babu et al., 2013), black carbon (BC) mass concentration (Manoj et al., 2019) have been studied across the Indian sub-continent through a network of observatories (Moorthy et al., 2013) for decades, only a few CCN studies are availbale since last few years over specific regions. Year-round CCN measurements are reported from the high altitude observatory over the Western Ghats (Leena et al., 2016), Indo-Gangetic Plain (IGP, Patidar et al., 2012), Central Himalayas (Gogoi et al., 2015), and Eastern Himalayas (Roy et al., 2017). CCN characteristics for a specific season, including closure analysis were reported by Jayachandran et al., (2017; 2018) at peninsular India, and by Bhattu and Tripathi, (2015) at IGP. Apart from these studies, Indian Ocean EXperiment (INDOEX, Ramanathan et al., 2001), Cloud-Aerosol Interaction Precipitation Enhancement EXperiment (CAIPEEX, Kulkarni et al., 2012), and South-West Asian Aerosol-Monsoon Interaction - Regional Aerosol Warming EXperiment (SWAAMI-RAWEX, Jayachandran et al., 2020~~19~~), Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB-2018, Nair et al., 2020~~19~~) are other major multi-platform campaigns carried out over the sub-continent and nearby marine environment to study the regional ACI. CAIPEEX conducted both aircraft and ground-based observations of aerosols, clouds, and planetary boundary layer (PBL) since 2009, in a phased manner. Details of the CAIPEEX are available in Prabha et al., (2011) and Kulkarni et al., (2012). Various studies have addressed~~ing~~ the spatio-~~-~~temporal distribution of AP (Padmakumari et al., 2013; Varghese et al., 2019), cloud microphysics (Prabha et al., 2011; 2012; Padmakumari et al., 2018), rainfall (Maheshkumar et al., 2014) properties, the relationship between cloud microphysics and thermodynamics (Bera et al., 2019), and ACI (Pandithurai et al., 2012; Prabha et al., 2012; Konwar et al., 2012; Gayatri et al., 2017; Patade et al., 2019) from the unique data obtained from the CAIPEEX.

Varghese et al., (2016) investigated the linkages of surface and cloud base CCN spectral characteristics over the rain shadow region.

The assessment of the effects of AP on clouds and precipitation due to the changes in the atmospheric composition by anthropogenic activities is very significant over India as the agriculture and economy of the region mostly depend on the Indian Summer Monsoon (ISM) rainfall. The west coast of India, which is the gateway of the ISM, receives almost 2.5 times the long-term monsoon mean rainfall observed all over India (Parthasarathy et al., 1995). The mountain ranges along the western coast of India known as the Western Ghats (WG) mountains, play a pivotal role in ISM rainfall due to orography (Grossman and Duran, 1984; Sijikumar et al., 2013). ~~WG mountain range is oriented in the north-south direction and extends about 1,600 km and has an average altitude of about 1.2 km.~~ A few studies have been carried out till date to understand the aerosol loading (Udayasoorian et al., 2014), CCN characteristics (Leena et al., 2016; Jayachandran et al., 2018) and its influence on the aerosol indirect effects (Anil Kumar et al., 2016) from different locations in the WG. However, the rain shadow region (leeward side) is prone to drought conditions with predominant continental effects, and relevant studies are sparse~~It is observed that the orography and the associated atmospheric dynamics cause heavy rainfall (~500 cm yr^{-1}) over the windward side of the WG, while the rain shadow region (leeward side) is prone to drought conditions~~.

CAIPEEX observations were conducted over the rain shadow region to understand the cloud and precipitation microphysics and AP properties to derive guidelines for the precipitation enhancement over the region. CAIPEEX Phase IV was designed to address the major objectives for the science of weather modification. The background observations of CCN were trivial for the design and validation of the experiment and the data presented in this study are~~is~~ aimed at understanding the aerosols and its cloud activation properties near the surface.~~-~~ The present study addresses the first reporting of CCN and its characteristics under different air mass and meteorological conditions throughout the ISM season (June to September) of 2018 over this region. The study focuses on the variations in CCN characteristics within the ISM season, and the possible factors are investigated using the concurrent and collocated aerosol size distribution and BC measurements. Another focus of the study is the CCN closure analysis to assess the role of size and composition at different atmospheric conditions.

2 Experiment details

2.1 Location, measurements, and database

As part of the ground segment of CAIPPEX IV campaign, aerosol and PBL measurements have been going on since May 2017 from Sinhgad College of Engineering at Solapur (17.70° N, 75.85° E, ~490 m a.m.s.l), which is at the cente~~ore~~ of the rain shadow region. The location is marked as a circle in Figure 1 and is a semi-arid region. The college site is ~~at the~~12 km away from Solapur city~~ outskirts (~12 km away)~~ and the aerosol sampling lab is on the third floor of the building, away from all local activities in the rural setting. Even though the sampling site is well isolated from the urban contamination, Solapur consists of numerous sugar and textile industries emissions~~that emit smoke~~, apart from the seasonal emissions from agricultural activities.

Details of instrumentation and data used for the present study are~~is~~ illustrated in Table 1. Aerosol sampling was carried out through separate PM 2.5 inlets from about 2 m above the rooftop connected with conductive tubing. CCN concentrations were measured at every second using a continuous flow streamwise temperature gradient CCN counter (CCN-100, DMT Inc; Roberts and Nenes, 2005). Initially (June 2018) CCN were measured at five SS (0.2, 0.4, 0.6, 0.8 and 1.0 %) and in July CCN counter was calibrated again and the SS was set at 0.3, 0.5, 0.8 and 1.2 %. Calibrations were carried out both before and after the experiments using ammonium sulfate AP following Rose et al., (2008). Instrument was also factory calibrated before the campaign. During the calibration experiments, CCN efficiency spectra were recorded for different ΔT values. The activation diameter corresponding to 50 % CCN efficiency for each spectrum was taken as the critical dry diameter for the CCN activation of ammonium sulfate particles. The corresponding critical supersaturation was calculated with the activity parameterization Köhler model (AP3) mentioned in Rose et al., (2008). The calculated critical supersaturation was taken as the effective supersaturation at the given ΔT value.

CCN counter uses the fundamental principle of the difference in the diffusion rate of heat and water vapor. A fixed temperature gradient is maintained along the walls of the wetted cylindrical column inside the instrument in which the desired SS is generated depending on the temperature gradient and the flow rate. The aerosols are fed at a constant sheath to sample flow of (10:1) along the center-line of the column and the total flow rate was maintained at 500 Vccm. The details of the working principle of the instrument are available in Roberts and Nenes, (2005) and Lance et al., (2006). During June, each SS was maintained for five minutes, except for 0.2 % which was for 10 minutes. About two minutes of data during the SS transition were discarded to avoid the uncertainty in establishing the required SS during the transition. At 0.2 % SS (lowest set-SS), about four minutes of initial data were discarded. Except June, all the SS were set for seven minutes each, except for 0.3 % SS, which was maintained for nine minutes. Here also, the initial 3-4 minutes data were discarded to ensure the set SS conditions. Thus, one cycle of the complete set of SS took 30 minutes, and the cycle was repeated. AP were continuously exposed to the SS inside the column, and those having their critical SS less than that of the set-SS inside the column, activated as liquid droplets and counted by the optical particle counter operated by a laser diode at 660 nm at the exit of the column. Since the CCN concentration was always less than 6,000 cm^{-3}, correction for water vapor depletion inside the column as suggested by Lathem and Nenes, (2011) was not applied.

Size segregated aerosol number concentration (NSD) from about 15 nm to about 685 nm, distributed among 107 size bins, was measured every three minutes using a scanning mobility particle sizer (SMPS, TSI model 3082). The set up consists of an electrostatic classifier, including a long Differential Mobility Analyser (LDMA, TSI model 3081), and a butanol based-Condensation Particle Counter (CPC, TSI model 3772). Before entering the LDMA the AP are charged to a known charge distribution by a bipolar charger in the electrostatic classifier, which were size segregated according to their electrical mobility (Wiedensohler, 1988; Wang and Flagan, 1990) in the DMA. The AP classified according to their sizes were counted by the CPC. The sheath and sample flow were maintained at 0.3 and 3 L min^{-1}, respectively. Multiple charge correction and diffusion charge correction were applied to the aerosol NSD data during the data inversion. AP were passed through a diffusion dryer before the classifier to prevent the high humidity conditions.

Radiation absorption properties of AP at different wavelengths were measured using a dual spot Aethalometer (AE 33, make: Magee Scientific) at every minute. Aethalometer operated at a flow rate of 2 LPM measured the attenuation of light due to the AP deposited on a filter tape (Hansen et al., 1984) at seven different wavelengths - 370, 470, 520, 590, 660, 880 and 950 nm. From this, the absorption coefficient (σ_{abs}) is estimated from the rate of attenuation, filter spot area, and the flow rate (Weingartener et al., 2003). The new-generation AE33 compensates the loading effect and multiple scattering effects (Arnott et al., 2005) associated with the filter-based optical attenuation techniques (Drinovec et al., 2015).

The wavelength dependence of absorption coefficient of aerosols is parameterised using the equation

$$\sigma_{abs}(\lambda) = \beta \times \lambda^{-\alpha_{abs}} \quad (1)$$

where, β is a constant and α_{abs} is the absorption Angstrom exponent. The nature of the carbonaceous sources can be inferred from the value of α_{abs}. Humic-like substance (HULIS) and brown carbon produced from biomass burning have higher absorption at lower wave length (ultra violet and blue) regions (Gelencser et al., 2003). Hence, α_{abs} will be higher (~2) for biomass dominant sources, while fossil fuel dominant sources will have α_{abs} close to unity (Kirchstetter et al., 2004)

Ambient weather parameters such as temperature, pressure, wind speed, wind direction, relative humidity and rainfall were also used in the present study from the Automatic Weather Station (AWS) measurements located at the site. All the instruments operated during CAIPEEX were calibrated periodically, especially before and after the experiments. The uncertainty associated with all the measurement techniques used in the present study is <10 %. All the measurements having different sampling frequencies were averaged to hourly intervals for analysis and interpretations. Air mass pathways were investigated using Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) (Draxler and Rolph, 2014) available from NOAA ARL READY Website.

2.2 Meteorology

The air mass back trajectories for five days reaching 50 m above the site were examined using back-trajectory analysis, and found that two distinct air masses reached the site during the observation period (01 June to 30 September, 2018). These are classified as (a) continental (dry) and (b) marine (wet), and shown in Figure 1. ~~Continental classification is carried out for t~~Those days in which air masses ~~which~~ were over the landmass and within 1 km a.g.l for minimum 3 days before reaching the site, and hence having a significant continental influence are segregated as continental. ~~All other trajectories, which were~~While, those from the nearby marine atmosphere are classified as the marine and the ~~observation~~ corresponding period include~~s~~ the monsoon rainfall period over the site. The ~~C~~continental air mass consistently prevailed over the site during the first week of June (denoted as continental-1) and from September 15 to 30 (denoted as continental-2) of 2018. ~~The ISM onset over Solapur was on 8 June 2018. Monsoonal circulation consistently prevailed over the site during July and August months of 2018~~Marine air mass days consist of 08-12, 15-31 July (denoted as marine-1) and 1-26, 28-31 August (denoted as marine-2). Thus, the observations and findings throughout this manuscript are examined on the basis of this classification. From Figure 1(a), it can also be seen that the air mass history for the continental classification are mostly within 1 km above the surface, indicating the chances for the influence of local aerosol sources~~It can be seen from Figure 1(a) that most of the~~

~~cases, air masses were confined within lower 2 km, while typical monsoon circulation was reaching the site during the July and August months (Fig. 1b)~~.

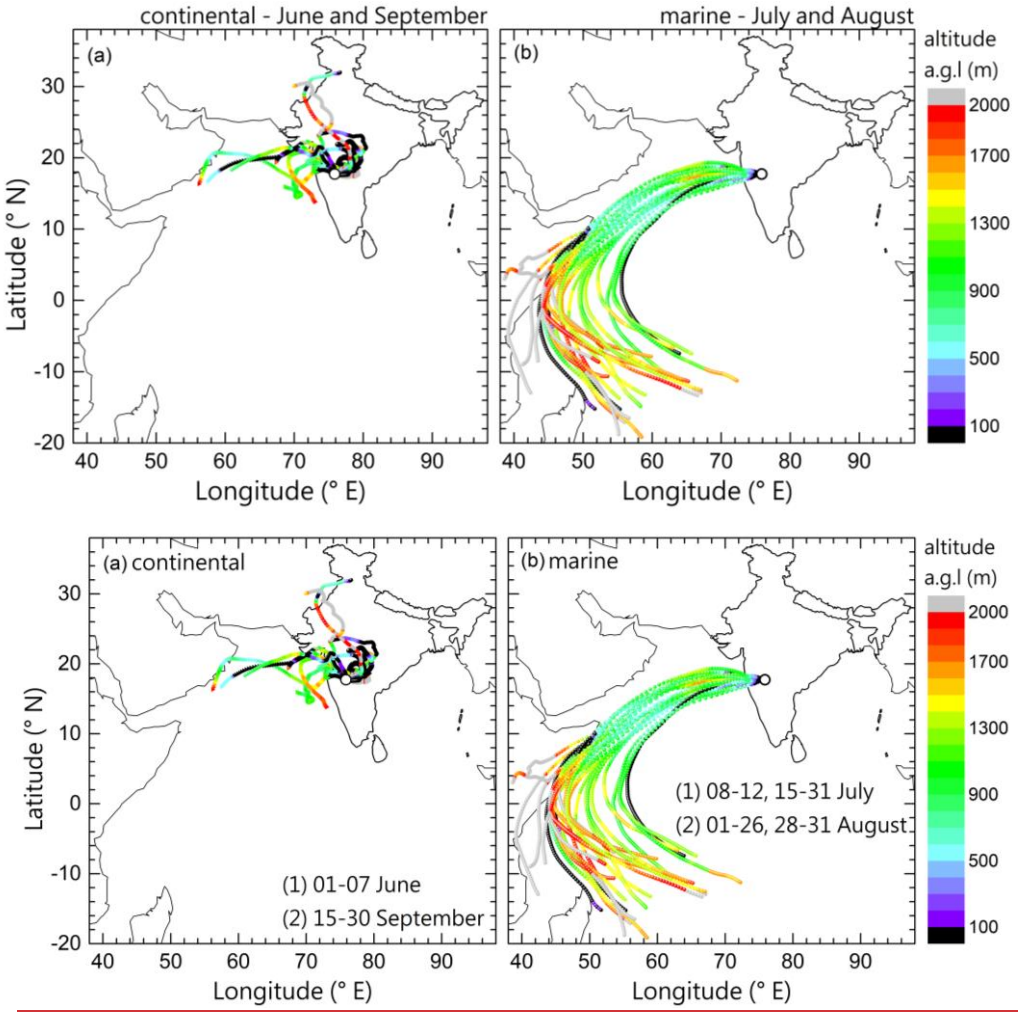
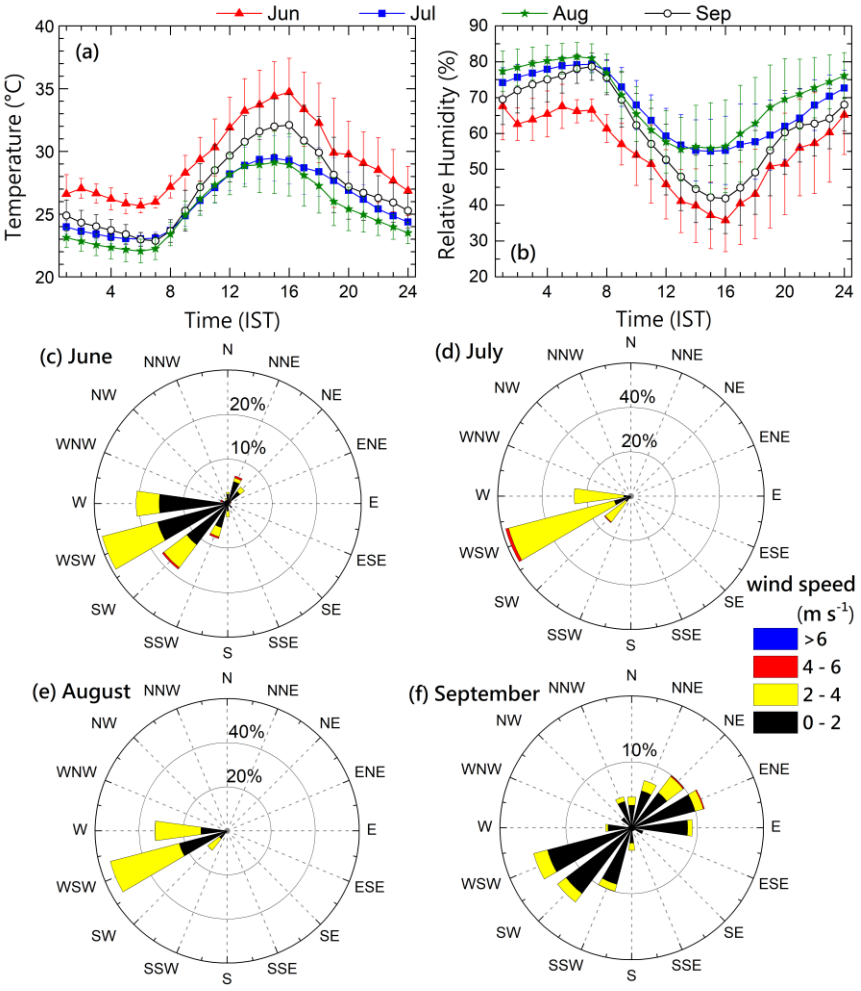


Figure 1. Air mass back trajectories with 5 days duration reaching 50 m above the site segregated to (a) continental and (b) marine. The color of the trajectories indicates the altitude of the air mass above the surface.

The meteorological parameters observed at the site from the AWS measurements during these periods are shown in Figure 2. Diurnal variation of temperature and relative humidity (RH) are shown in Figure 2(a) and (b), respectively. The temperature and RH values are distinctively different during continental-1~~prior to the monsoon (June)~~, compared to other periods. Marine-1 and 2 periods~~onsoon (July and August) months~~ experienced low temperatures and high RH throughout the day, while continental-2~~September~~ had higher temperature and lower RH during noon and afternoon hours. The monthly mean temperature during the study periods of continental-1~~June~~, marine-1~~July~~, marine-2~~August~~, and continental-2~~September~~ were 29.5±3.6ºC, 25.9±2.6ºC, 25.4±2.8ºC, and 27.1±3.5ºC; respectively. A dry spell existed during the ~~campaign~~ continental-1 days

~~of June~~ when the maximum hourly mean temperature recorded was ~38 °C. The maximum temperature at all the periods~~months~~ was observed at the 1500 and 1600 hours (IST), and the lowest temperatures were observed before sunrise of the day. Intermittent rainfall happened during July and August ~~months~~ and a few heavy rainfall events occurred during these months. The aerosol/CCN measurements during heavy rainfall are not included in the analysis for interpretations (missing days in Table 1). From the wind rose diagram (Fig. 2c), it can be noted that the strong winds were blowing mostly from the west and south-west part of the site during continental-1~~June~~ and in few cases, winds were blowing from the north-east direction. Westerly and South-Westerly winds were present during marine 1 and 2 conditions (Fig. 2d and e), while the continental-2 days (Fig. 2f) mostly North-Easterly winds were observed~~Winds were reaching the site from west and south-west during July and August months (Fig. 2 d and e), while September had winds mostly from North-East and South-West with significantly weak wind conditions~~.


Jun
Jul
Aug
Sep
(a)
(b)
Temperature (°C)
Relative Humidity (%)
Time (IST)
Time (IST)
(c) June
(d) July
(e) August
(f) September
N
NNE
NE
ENE
E
ESE
SE
SSE
S
SSW
SW
WSW
W
WNW
NW
NNW
20%
10%
40%
20%
40%
10%
wind speed
(m s-1)
>6
4 - 6
2 - 4
0 - 2

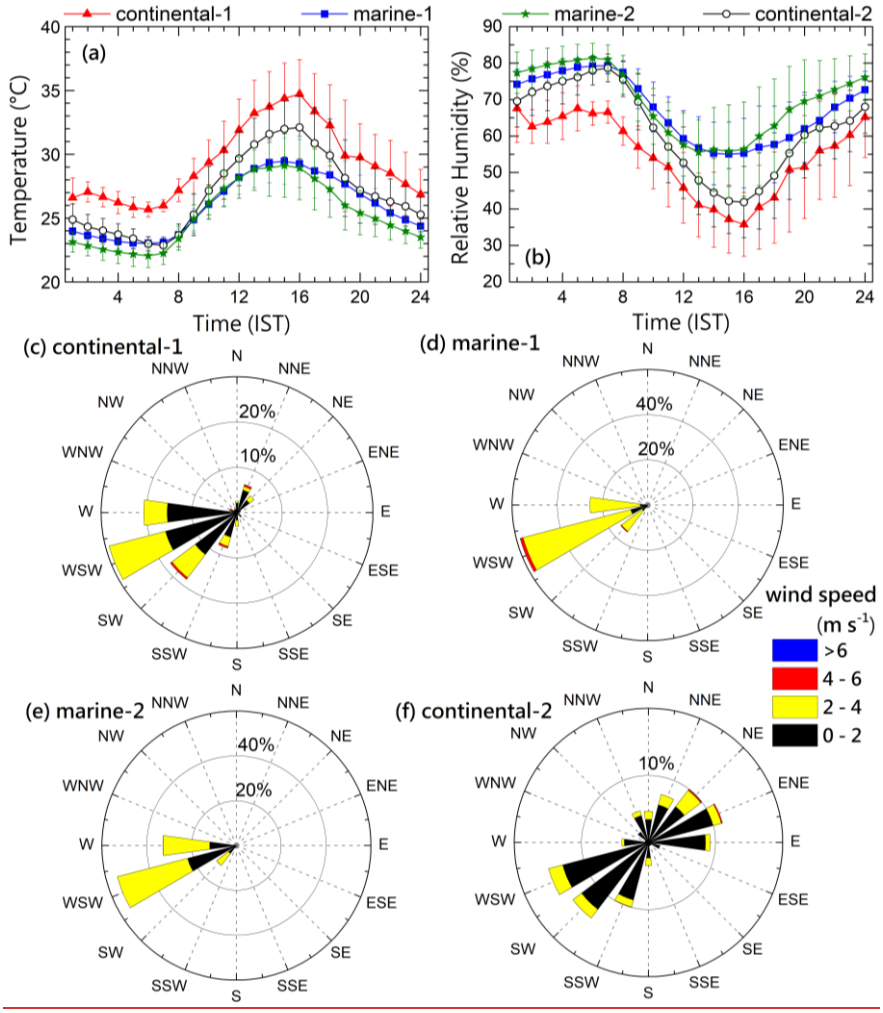


Figure 2. Diurnal variation of (a) temperature, (b) relative humidity along with the standard deviation of the mean values. The wind rose diagrams from the co-located AWS measurements for ~~the months of~~ (c) continental-1~~June~~, (d) marine-1~~July~~, (e) marine-2~~August~~, and (f) continental-2 conditions~~September~~.

3 Results and Discussions

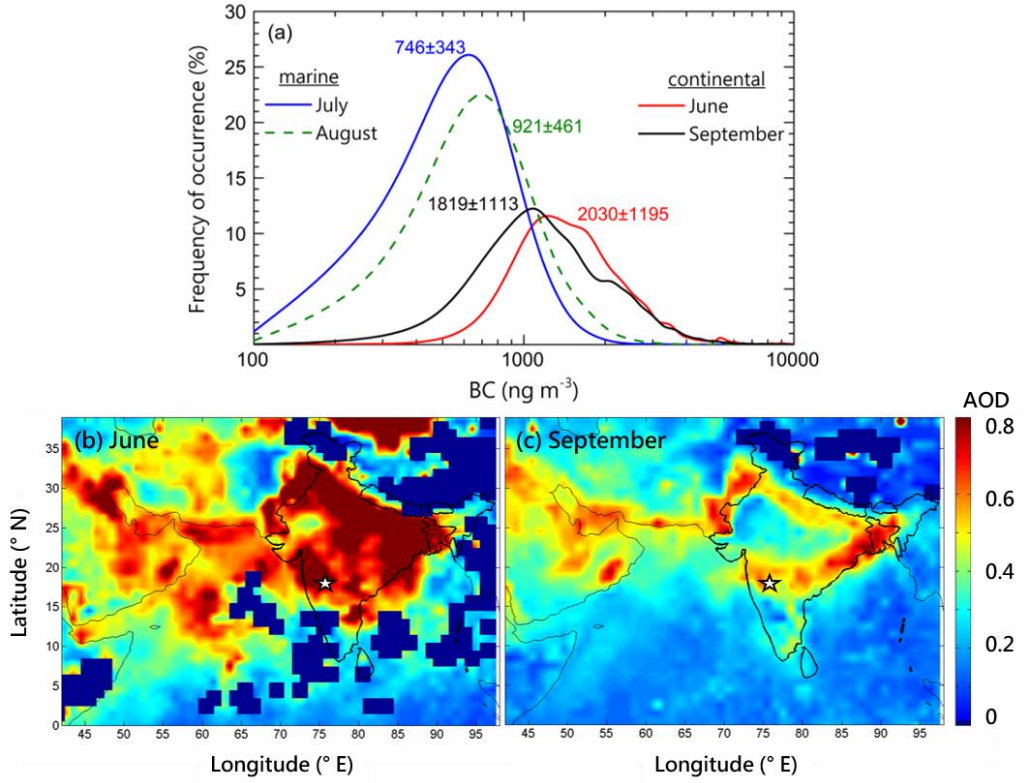
CCN characteristics at the site are~~is~~ investigated with aerosol size distribution and BC measurements**.**

3.1 Overview of aerosol loading

The frequency distribution of BC mass loading and the mean values (and its standard deviation) during the observation days are shown in Figure 3(a) with an aim to understand aerosol loading and the influence from anthropogenic activities. The distinct atmospheric conditions and the air mass history are evident in the BC mass loading at the site. Before the onset of

monsoon, under the influence of continental air mass, the mean BC values were above 2,000 ng m^{-3}, which reduced to very low values (~746 ng m^{-3}) during the marine~~wet~~ conditions. BC concentration was even higher than 4,000 ng m^{-3} during the continental air mass, while in many cases values were almost 100 ng m^{-3} under marine~~onsoon~~ conditions.

Apart from the near-surface measurements, the columnar aerosol optical depth (AOD) is examined using the Moderate Resolution Imaging Spectroradiometer (MODIS) - Aqua at 550 nm. The AOD observed from MODIS ~~before the monsoon onset and~~ during the continental air mass conditions along with the site (white star) are shown in panels (b) and (c) of Figure 3. It can be seen that heavy aerosol loading (AOD>0.5) persisted around the rain shadow regions and the Mumbai coast (northwest of the site) in addition to the high loading over the IGP. After the monsoon rainfall, the aerosol loading has reduced all over India as seen in Figure 3(c). Still, high aerosol loading (AOD>0.4) was observed around the observation site, IGP and the northern part of the east coast.


(a)
746±343
921±461
1819±1113
2030±1195
marine
July
August
continental
June
September
Frequency of occurrence (%)
BC (ng m-3)
(b) June
(c) September
Latitude (° N)
Longitude (° E)
Longitude (° E)
AOD
0.8
0.6
0.4
0.2
0

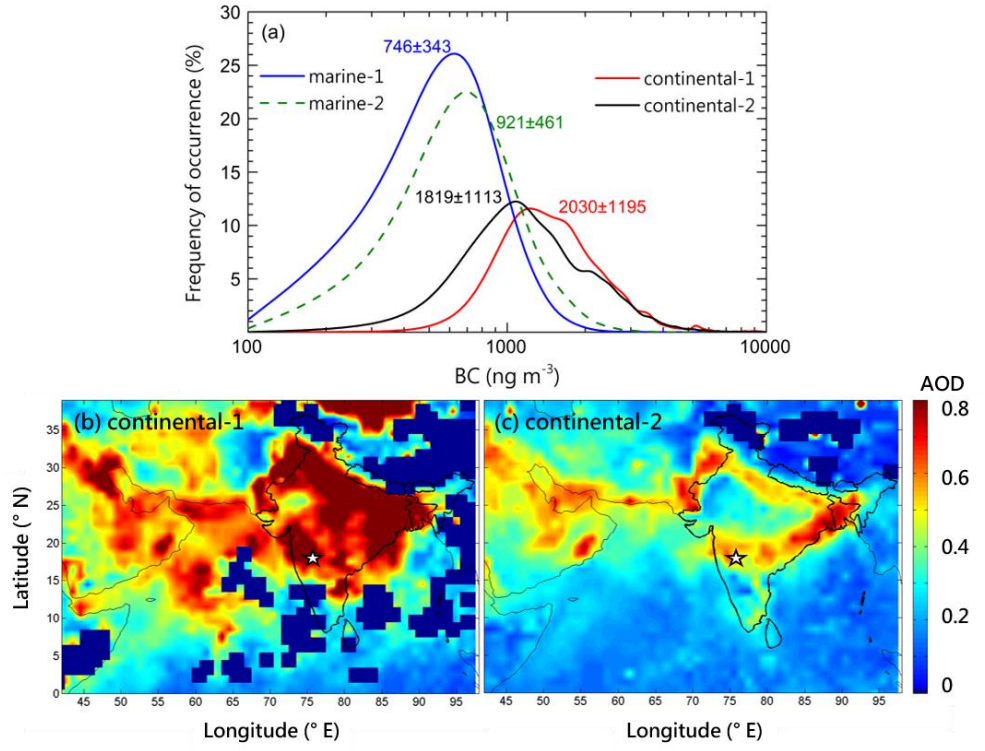


Figure 3. (a) Frequency distribution of BC mass concentration and its mean values for marine and continental conditions. Aerosol optical depth (AOD) at 550 nm observed from the Moderate Resolution Imaging Spectroradiometer (MODIS) for the continental (b) 1 and (c) 2 conditions. The Solapur site is indicated by the star in the spatial AOD maps~~Aerosol optical depth (AOD) at 550 nm observed from Moderate Resolution Imaging Spectroradiometer (MODIS) during (b) prior to (June) and (c) after (September) monsoon, and the white star indicate Solapur~~.

BC can be consider~~r~~ed as a proxy for the anthropogenic activities (Myhre et al., 2013; Lelieveld et al., 2019), and the BC loading over Solapur indicates that the anthropogenic influence is predominant during the continental air mass conditions. This observation is in line with the columnar aerosol observation from MODIS. Apart from fossil fuel combustion, biomass burning may also contributes to the carbonaceous aerosols that prevailed over the site. The fire counts observed from the MODIS (collection 6 product obtained from https://earthdata.nasa.gov/firms) may support this inference, which is given in the Appendix (Figure A1). The high aerosol loading locations in Figure 3 are associated with the numerous fire events which can be seen in Figure A1. From another site in the rain shadow region closer to the central part of India - Nagpur, Kompalli et al., (2014) have reported BC mass of ~2,000 ng m^{-3} before the monsoon, which is similar to the present study. From the long-term observations of BC from the north-west part (Pune) of the current study, Safai et al., (2013) have reported a mean BC mass of ~1,200 ng m^{-3} during the monsoon period. Both the high surface BC and total column aerosol loading observed before the monsoon, indicate the significant anthropogenic influence on the total aerosol loading. The low BC values (<1000 n~~n~~g m^{-3})

during the marine conditions~~wet-monsoon months~~ at Solapur represent a cleaner atmosphere, while ~1~~.~~500 n~~μ~~g m^{-3} BC was reported from a coastal location in peninsular India (Babu and Moorthy, 2002) during monsoon. About 50 % reduction (~10000 to 6000 ~~μ~~ng m^{-3}) in BC mass associated with the dominance of fossil fuel source replacing the biomass, during monsoon compared to the pre-monsoon values, was reported by Vaishya et al., (2017) from a heavily polluted IGP site. The low BC loading during the marine conditions~~onsoon months~~ over Solapur is due to the wet scavenging of aerosols and the distinct air mass reaching the site as well as due to ~~less~~ reduced local burning during the active monsoon conditions. The high AOD and BC observations identify Solapur as a polluted-continental environment, which is cleaner during active monsoon compared to the other periods~~.~~.

3.2 CCN number concentrations and its variations

The mean CCN concentration at different SS, known as the CCN SS spectra, segregated according to the air mass conditions are shown in Figure 4. It can be seen that the CCN concentrations at all SS are higher during continental~~June and September~~, compared to marine conditions~~onsoon months (July and August)~~. The highest CCN concentration is observed during continental-1~~June~~ which is as per the surface BC loading and the total columnar aerosol loading. CCN spectra are similar for the marine conditions~~onsoon months~~, except the slight difference at the lowest SS. CCN concentration before the monsoon ranged from ~1,600 to 3,600 cm^{-3} for 0.2 to 1.0 % SS. Meanwhile, the CCN concentration was only ~900 cm^{-3} during marine air mass~~July and August~~, even at 1.2 % SS. Thus, a clear distinction is seen in the CCN concentration between the marine~~wet~~ and continental air mass~~dry~~ conditions within the same ISM period.

The CCN concentration varies with SS, and its parameterization is very important for its applicability in climate models (Khvorostyanov and Curry, 2006). The measured CCN spectra are parameterized by the Twomey's empirical fit relationship (Twomey, 1959; 1977), which is widely used due to its simplicity (Cohard et al., 1998) and given as,

$$CCN(ss) = C \times SS^{k} \quad (2)$$

where C and k are the empirical fit parameters characterizing the spectra. It should be noted that, the empirical fit parameter k is different from the effective hygroscopicity parameter-κ discussed by Petters and Kreidenweis (2007).

More than 90 % of cases of the current observations show a high correlation coefficient (R>0.95) with the Twomey's empirical fit, except during continental-2~~September~~, during which about 65 % of the cases only had high (> 0.95) correlation coefficient with the Twomey's fit. The spectra having a correlation coefficient of more than 0.95 with the empirical fit are only considered in the present study.

Hygroscopic or bigger particles have flat CCN spectra and low k values, while hydrophobic and ultrafine (UF) mode (<100 nm) AP will have steep CCN spectra and high k values (Hegg et al., 1991; Jefferson, 2010) as those particles need higher SS to activate as CCN. Thus, the empirical fit parameter 'k' indicates the nature of the aerosol system towards CCN activation and 'C' indicates the CCN concentrations at 1.0 % SS. Generally, h~~H~~igh C and k values are reported for the anthropogenic, while low values are reported for the natural/marine AP (Seinfeld and Pandis, 2016; Andreae, 2009). ~~From Figure 4, it can be seen that the mean k values are higher during the monsoon conditions than the continental conditions.~~ The

highest k value (~0.67) is observed during marine-2~~August~~ and the minimum (~0.52) during continental-1~~June~~. As may be noted, bigger or hygroscopic (or both) particles which are CCN active were abundant during continental-1~~June~~ compared to the marine conditions~~onsoon months~~, when fine or hydrophobic (or both) particles were predominant.

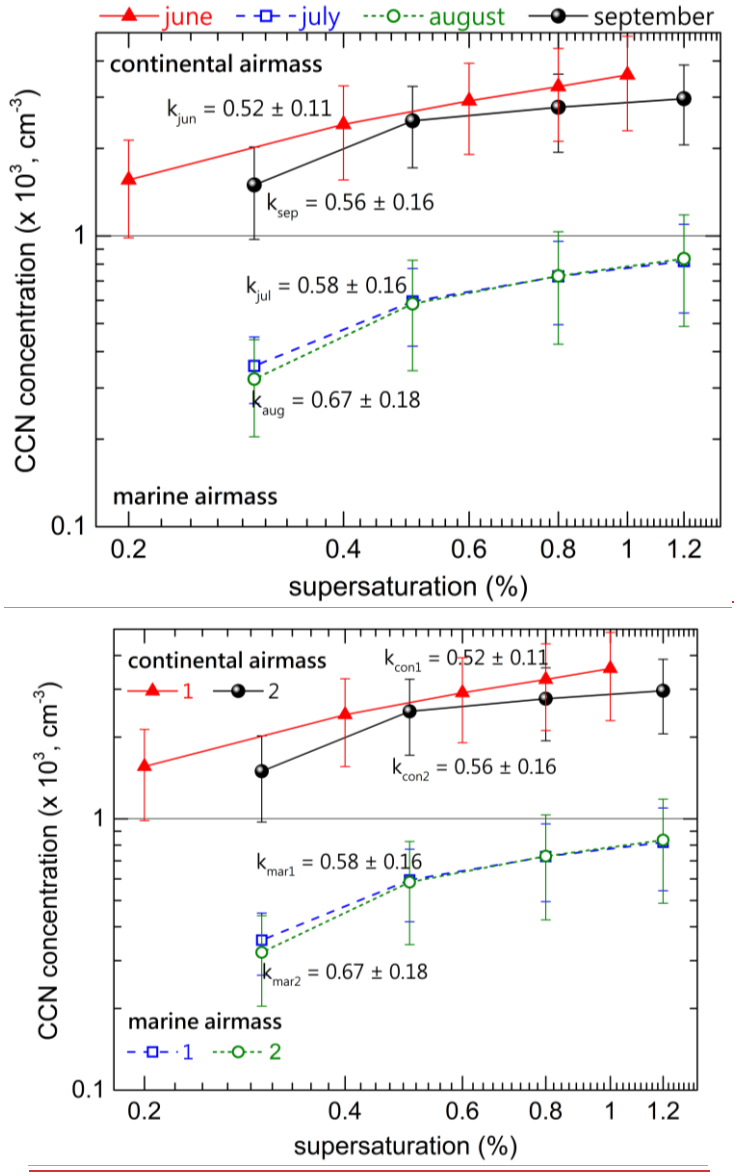


Figure 4. Mean CCN concentrations (± standard deviation) for different SS during continental ~~(June and September)~~ and marine ~~(July and August)~~ conditions. The power-law fit k-value of each spectrum is also given.

CCN concentrations and the k values reported during the current study, along with a few other studies are given in Table 2. Generally, most of the aerosol abundance measurements such as BC mass (Kompalli et al., 2014), and aerosol number

concentration (Babu et al., 2016) showed the lowest seasonal mean value during ISM over the Indian region, mainly due to the wet removal of AP. The CCN concentration at semi-arid Solapur before the onset of ISM is comparable to the values (~2,000 cm^{-3}) observed over the arid north-west region of India reported by Jayachandran et al., (202019). Interestingly, the CCN concentrations at Solapur during active monsoon periodthe marine conditions is the lowest among the values reported over the Indian sub-continent. The current values (~350 cm^{-3}) are comparable to those reported from Ponmudi (~400 cm^{-3}) at the southern part of the WG, and another site at WG - Mahabaleshwar (~500 cm^{-3}) at 0.2 % SS. From Table 2, very high values of CCN concentrations are reported from polluted urban environments. Very low CCN concentrations (<300 cm^{-3} at 1 % SS) are also reported from pristine environments like Amazon (Pohlker et al., 2016; 2018), and Alps (Juranyi et al., 2011). The mean CCN values observed at Solapur during ISM are comparable to those classified as 'polluted-marine' by Andreae, (2009).

Generally, an enhancement in k-values is observed during the monsoon period, which is seen only during the marine-2 conditions in this study. In all other cases, the k-values are comparable.An enhancement in k-values are observed during the monsoon period. Jayachandran et al., (2017; 2018) reported similar results for the monsoon period both at a coastal site, and at a hill station in the WG. From the southern tip of India, Jayachandran et al., (2017) have shown that the enhancement of k values (~0.7) associated with wet scavenging and lower k values (~0.55) during no rainfall conditions, within the same ISM period. The enhancement in k values (two-times) associated with the monsoon rainfall can be seen from Mahabaleshwar also (Table 2). Thus, the CCN concentrations at different SS at Solapur during ISM are low compared to those reported from other environments in India, while the CCN spectra show the common characteristics to those values reported from WG and peninsular India.

Significant diurnal variations are seen in the PBL AP properties over the Indian sub-continent (Nair et al., 2007). Daytime high and nighttime low aerosol abundance characterized by anomalous high values just after sunrise is known as the fumigation peak (Prakash et al., 1992), is generally observed. This diurnal pattern is mostly due to the evolution of the PBL and due to local emissions (Nair et al., 2007; Kumar et al., 2015). As the CCN activation and its properties are highly heterogeneous, it is very important to know its variation in a day. The diurnal variation of CCN concentration at 0.3 % SS, segregated to air mass wet and dry conditions are shown in Figure 5(a) and 5(b), respectively. The CCN variations in a day are similar during the marine conditionswet months (Fig. 5a), while it differs before and after the monsoon rainfall. In general, CCN concentrations show a slight enhancement (more prominent during clean background-marine air mass) during daytime due to the anthropogenic activities. A rapid increase is seen just after the sunrise in all monthsthe conditions, except though weak in continental-2September, which is due to the mixing of the nocturnal residual layer with the evolving PBL (fumigation peak). There is no vivid diurnal variation in CCN during continental-2September. The diurnal variations of CN and BC concentrations for different periodsmeteorological conditions are shown in Figure 6. The diurnal variation of the total APCN concentration and BC mass concentration is more vivid than that of the CCN concentration. A clear bi-modal variation is seen in both CN and BC diurnal variations during marine conditionsJuly an August. A sharp peak is seen in both CN and BC after the sunrise (0600-0700 IST) and the next peak starts increasing from 1500 IST maximum is at around 2000 IST. The diurnal variations in CN and BC are less prominent during the marinewet months, compared to that during the continental

conditions~~dry months~~. In both~~,~~ the conditions a small increase is seen in the CN concentration around ~~mid-~~afternoon. The fumigation peak seen in BC ~~in the early monring was of~~during the continental~~dry~~ conditions ~~such and~~ was~~ere~~ more than twice the corresponding daytime average values. Another important observation is that the CN values were consistently higher during continental-2~~September~~ throughout the day than continental-1~~June,~~ while the daytime BC mass was higher during continental-1~~June~~ than 2~~September~~. In contrast to the CCN, the BC had well defined multiple peaks (morning and evening), indicating the contrasting aerosol source characteristics during the diurnal cycle. The well-mixed conditions~~ are reached~~ during the late afternoon hours and the PBL evolution~~mixing~~ has a well defined role in the reduction of concentration during the daytime, until new sources of aerosol are injected in to the atmosphere in the evening hours. The nighttime increase in BC could be due to~~attributing~~ the stable conditions and less vertical mixing. The role of the PBL in modulating regional aerosol characteristics will be dealt in a separate study.

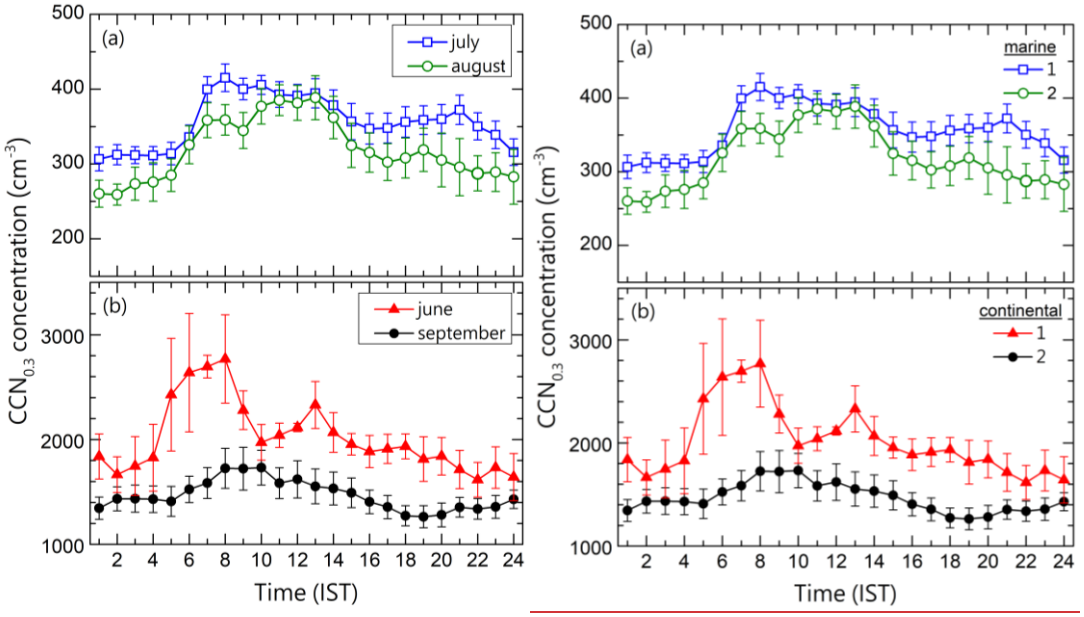


Figure 5. Diurnal variation of CCN concentration at 0.3 % SS during (a) marine~~wet~~ and (b) continental~~dry~~ conditions.

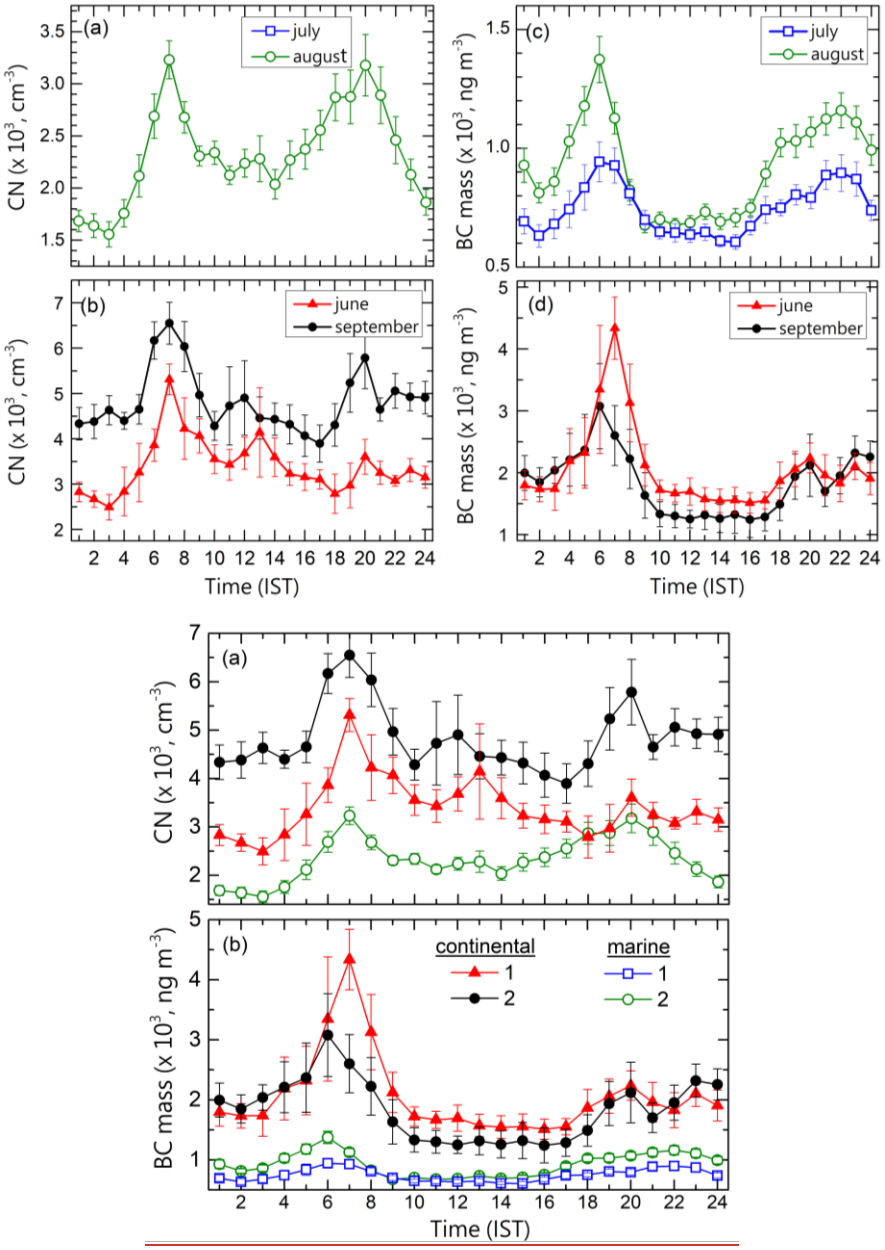


Figure 6. Diurnal variations of (a) CN and (b) BC~~during (a) wet and (b) dry conditions, and BC mass~~ concentrations during ~~(c) wet and (d) dry~~ marine and continental conditions.

A few studies report~~ed~~ the diurnal variations in CCN and its properties; at coastal (Jayachandran et al., 2017), W~~estern~~ G~~hats~~ (Leena et al., 2016; Jayachandran et al., 2018~~; Leena et al., 2016~~), rain-shadow (Varghese et al., 2016), IGP (Patidar et al., 2012), central Himalayan (Gogoi et al., 2015), and Eastern Himalayan (Roy et al., 2017) environments of the Indian sub-

continent. Weak diurnal variations in CCN concentrations during ISM, similar to the present study but with opposite patterns were reported from the southern coast by Jayachandran et al., (2017) and from WG by Leena et al., (2016). Day-night variations in the CCN concentration can be due to the changes in aerosol sources, PBL dynamics, or both. Since the sky is generally overcast during the ISM and hence a shallow moist PBL (Sandeep et al., 2014) prevails, the observed diurnal variations during marine conditions are mainly due to the diurnal variations in the source and sink processes. The bi-modal diurnal pattern seen in BC mass concentration at Solapur is seen similar to the observations reported by Safai et al., (2007) over Pune. Apart from the fumigation process happening during the sunrise, vehicular and biomass emissions also have a role in the peaks observed in a day. Thus, both local emissions and PBL dynamics contributed to the diurnal variations observed in the AP characteristics. The diurnal variations during the continental conditions indicate the consistently high AP background conditions. While the diurnal variations during the marine conditions indicate the significant presence of local AP sources.~~The less diurnal variations during the dry conditions indicate the consistently high AP background conditions over Solapur, while the evident diurnal variations during the wet conditions indicates the presence of significant local aerosol sources~~

3.3 CCN-CN association

The association of CCN concentration at 0.3 % SS with the concurrent total AP number concentration, CN (~15-685 nm) is investigated separately for different conditions and is shown in Figure 7. CCN concentration at 0.3 % ~~for~~ during continental-1 ~~the month of June~~ is estimated from the measured CCN spectra. ~~Though CCN forms the sub-set of the total AP system, the highly uncertain and non-linear dependence of CCN on the total AP is worth investigating.~~ The role of the aerosol NSD is revealed through the color of the scatter which represents the Geometrical Mean Diameter (GMD) of the corresponding AP system. It can be inferred from the Figure that the relationship between CCN and CN is different for different conditions. A least-square linear fit forced through origin (as there is no CCN in the absence of CN) is made through the scatter. ~~and the corresponding fit parameters and the linear fit line is also shown in the Figure.~~

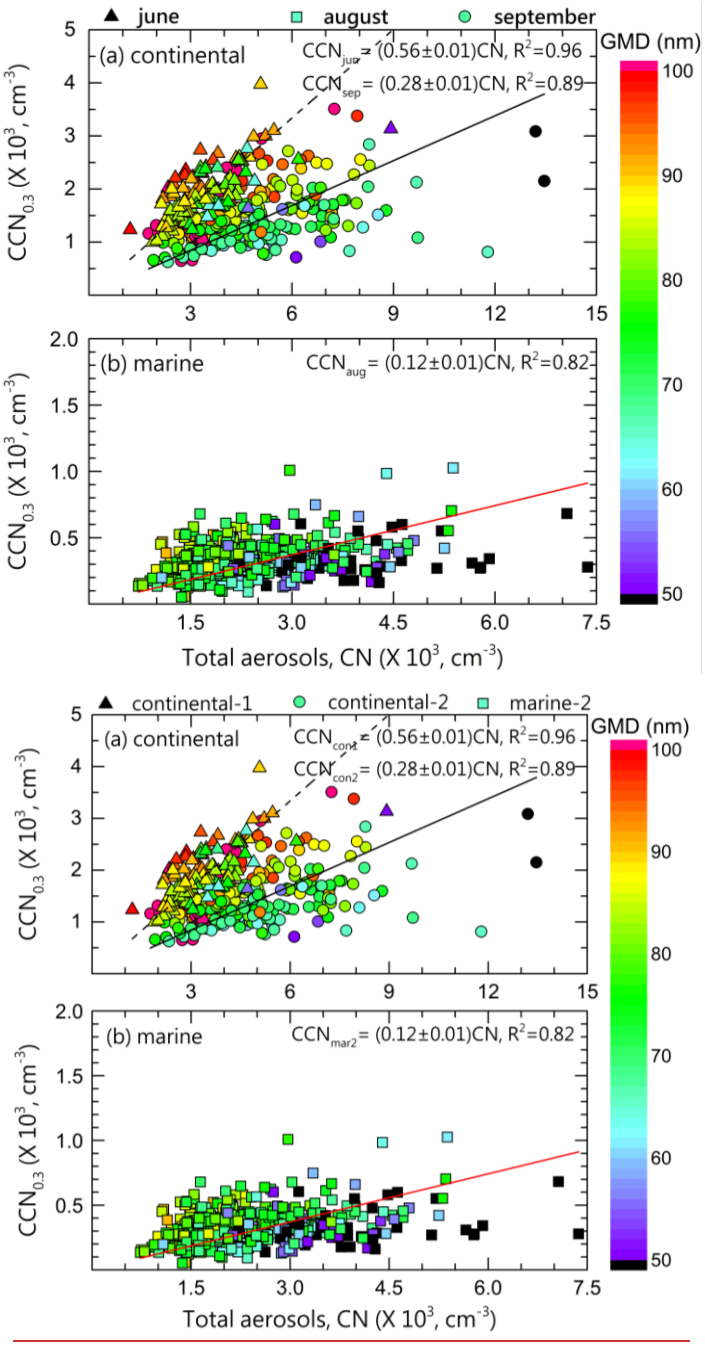


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Figure 7. Association between CCN (at 0.3 % SS) and CN concentrations segregated to (a) continental and (b) marine conditions. The color of the scatter indicates the concurrent geometrical mean diameter of the aerosol system. The least-square linear fit is also shown along with the fit parameters.

The best linear relationship between the CCN at 0.3 % and CN concentrations is seen during continental-1~~June~~, and the corresponding slope and correlation coefficient (R) of the fit are ~0.56 and 0.98, respectively. The linear association weakens during the marine~~onsoon~~ condition when the slope and correlation coefficient values of the fit reduce to ~0.12 and 0.90, respectively. Even though under continental conditions, the slope of the linear fit during continental-2~~September~~ (~0.28) reduces to the half of that measured during continental-1~~June~~, and the correlation coefficient value (R=0.94) also reduces. The relationship between CCN and CN during marine-2 is weaker than the continental, and only a few AP are activating as CCN~~The relationship between CCN and CN is weak during August and only a few AP are activating as CCN~~. It can be seen that most of the scatter points which lies below the linear fit line and corresponding to the higher (than the monthly mean) CN values are having GMD less than 50 nm. Even though the number of cases is less, similar observations can be seen during the continental case also. The two points (black-circle) corresponding to CN concentrations higher than 13,000 cm^{-3} are having GMD less than 50 nm. These cases which reduce the CCN activation indicate the presence of an UF mode, probably due to the new particle formation (NPF) events. However, the presence of UF particles is not the only cause for less activation of CN as CCN in marine conditions~~onsoon months~~ as the scatter and the linear fit excluding the UF particles are also having low correlation and slope values.

During ~~August~~ marine-2 (Fig. 7b), the CCN concentration at 0.3 % was not increasing beyond ~~seems to be not varying linearly with the CN and is nearly constant at ~~600 cm^{-3}, despite of CN concentration increasing to ~7,500 cm^{-3}. This is indicative of a significant number of UF~~ or Aitken mode particles~~ that require high SS for activation. Similar behavior of AP system towards CCN activation is observed at Eastern Himalayas (Roy et al., 2017). The drastic difference in CCN-CN association, similar to the present study, is also reported by Asmi et al., (2012) between winter and summer months at a high-altitude site in France. They have attributed the predominance of accumulation mode particles and fine mode particles during winter and summer months, respectively. From the Central Himalayas, Dumka et al., (2015) have shown an~~the~~ increase in CCN-CN scatter during ISM due to the change in the aerosol physico-chemical properties. The spread of the scatter between CN and CCN increased~~s~~ for polluted conditions (Jayachandran et al., 2020~~19~~), which was attributed~~is mainly due~~ to the associated complex aerosol size distribution and mixing state. Thus, the CCN dependence on CN population during the ISM shows a complex dependence on the aerosol size and mixing state.

3.4 CCN Activation Fraction

The fraction of AP acting as CCN at a given SS is known as the CCN activation fraction/ratio (AF) and is an important parameter to characterize the CCN activity (Dusek et al., 2006; Andreae, 2009; Deng et al., 2013). The CCN AF values for all the SS segregated to different air mass history is shown in Figure 8. During continental-1~~June~~, more than 40 % of the AP are getting activated as CCN at 0.2 % SS. Nevertheless, during marine-2~~August~~, about 40 % of the particles only are activating as CCN even at 1.2 % SS, revealing the highly CCN in~~-~~active nature of the aerosols. During continental-2~~September~~, the CCN AF values at all SS are between those of continental-1~~June~~ and marine-2~~August~~. ~~The CCN AF values at all SS show a similar feature in CCN-CN relationship at 0.3 % SS (Fig. 7).~~

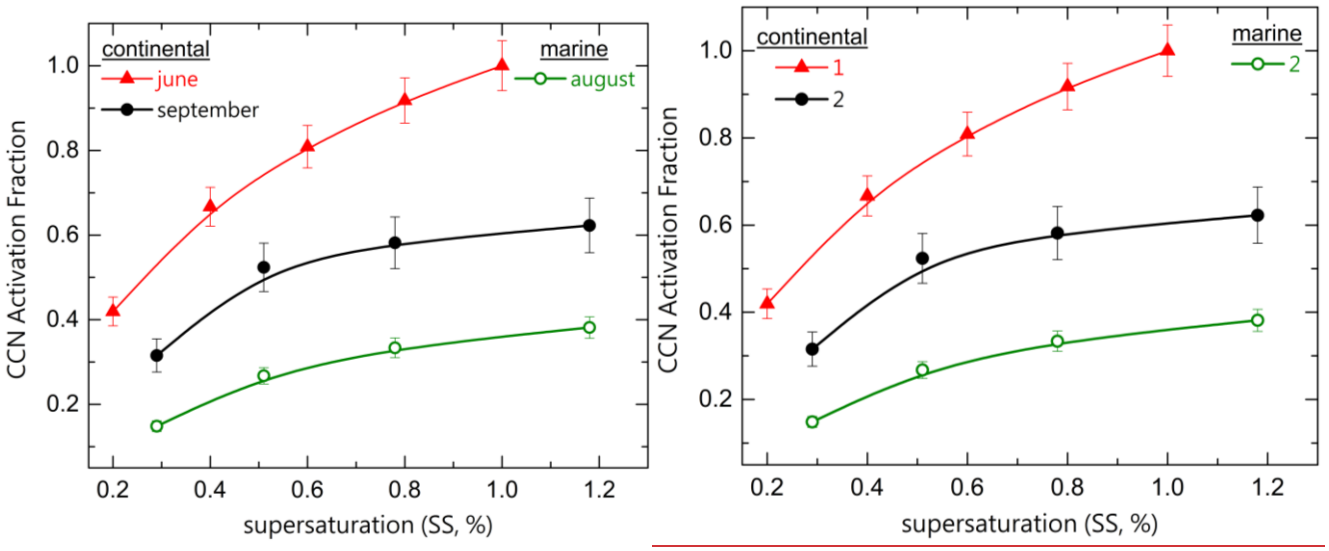

Figure 8. Variation of CCN activation fraction at different SSs during continental ~~(June and September)~~ and marine (~~August)~~ conditions. Vertical error bars indicate the standard error.

The diurnal variations of k and AF (0.3 % SS) values ~~segregated for the wet and dry conditions~~ are shown in Figure 9. Unlike the continental~~dry~~ conditions, the k values show a clear diurnal pattern during the marine~~wet~~ conditions, similar as in the case of CN and BC concentrations. The k values increase almost twice after the sunrise and decrease thereafter reaching the nighttime values by evening hours (1600 IST) during marine conditions~~July and August~~. Again, the k values peak at around 2100 hours IST. The enhancement in CCN and k values during daytime in marine airmass conditions, indicate the influence of local anthropogenic aerosol sources in determining the CCN activation. As discussed in Figure 8, the CCN AF values are very low throughout the day, with a slight increase during noon hours in marine-2~~August~~, when the k-values are low. Contrastingly, the AF values are consistently high throughout the day during continental-1~~June~~. During continental-2~~September~~, an increase in CCN AF (from ~0.3 to 0.4) can be seen during the daytime.

It is well understood that the CCN characteristics are a function of aerosol size and composition. Hence it will be interesting to know the diurnal variations of the concurrent aerosol size and composition. In the absence of continuous aerosol composition measurements, the absorption Angstrom exponent (α_{abs}), which is a proxy to identify the nature of the carbonaceous aerosols is estimated, and its diurnal variation for different conditions are shown in Figure 10 (a and b). The diurnal variation of the GMD for the corresponding periods are shown in Figure 10 (c and d). The diurnal variations of α_{abs} is similar for the marine conditions~~July and August months~~ with the values peaking by sunrise (0600-0900 IST) and late evening hours (1800-2000 IST). Almost the same pattern is seen during continental-2~~September month~~ also, but of different magnitudes. Meanwhile there is no clear diurnal variations in α_{abs} during continental-1~~the June~~ observations similar to the diurnal variations of ~~'~~k~~'~~ and AF.

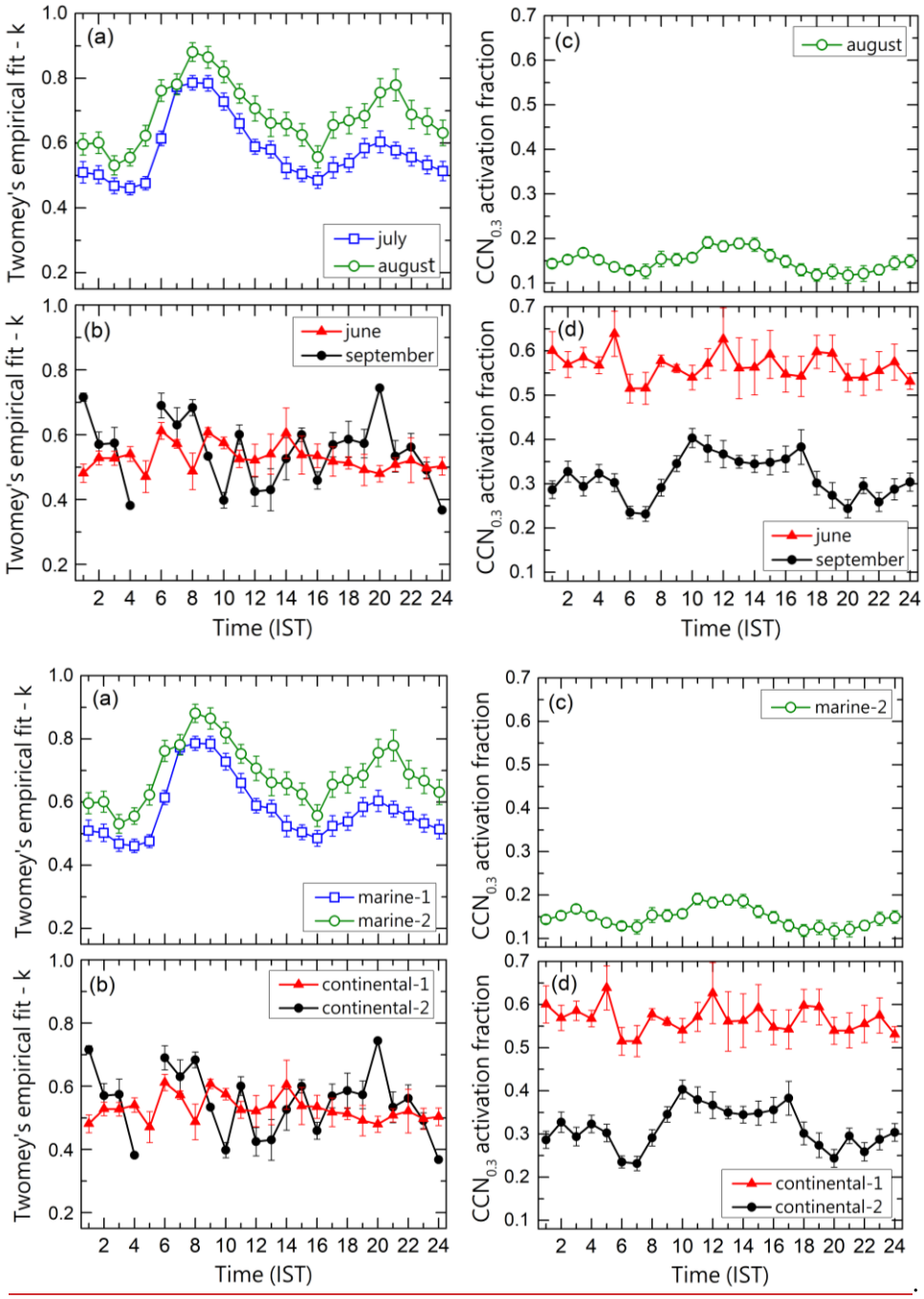

Figure 9. Diurnal variations of k-values (a and b), and CCN activation fraction (c and d), during marine and continental conditions. Vertical bars indicate the standard error values.

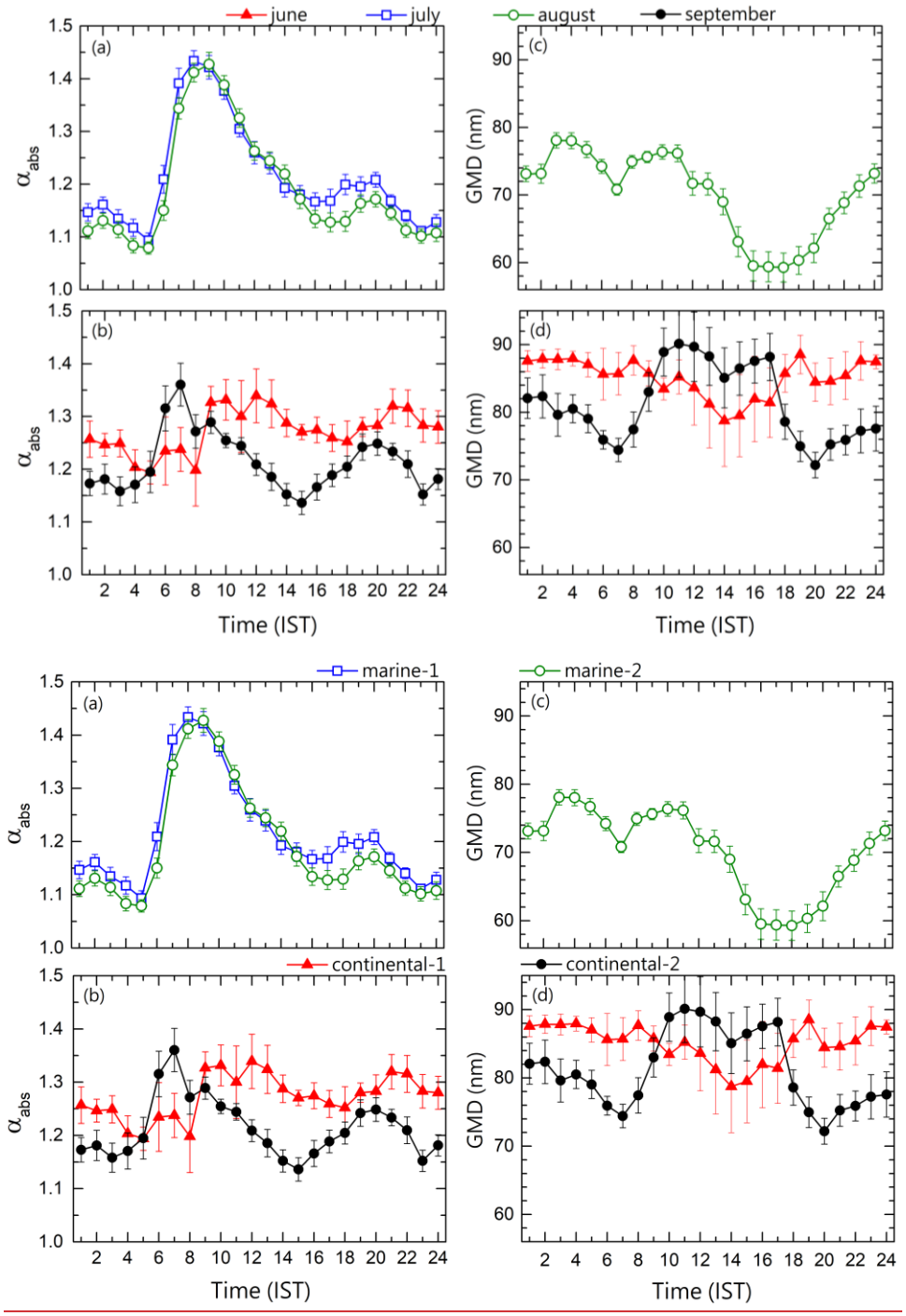

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Figure 10. Diurnal variation of absorption Angstrom exponent (a and b) and geometrical mean diameter (c and d) during continental and marine conditions. Error bars indicate the standard error associated with the mean values.

The mean GMD values during the marine conditions decrease from 1200 IST onwards and reaches the minimum value (<60 nm) from 1600 to 1800 IST, which increases back to ~72 nm by midnight hours. Interestingly, a small dip in the GMD is observed during the fumigation peak (~0700 IST), associated with the sharp increase in α_{abs} values. The GMD values were consistently higher throughout the day during continental-1~~June~~, such that the lowest mean value (~79 nm) observed at the 1400 IST is higher than the maximum mean GMD (~78 nm) observed in a day (0300 and 0400 IST) during marine-2~~August~~. GMD during continental-2~~September~~ depicts a clear diurnal variation which is opposite to that observed during continental-1~~June~~, with distinct high values during daytime. Similar to marine-2~~August~~, GMD decreases during the fumigation peak associated with the sharp increase in the α_{abs} values.

Even though the mean α_{abs} values are almost similar in all periods~~the months~~ with comparatively higher (1.29 ± 0.09) during continental-1~~June~~ and lower during marine-2~~the August~~ (1.19 ± 0.14), the values show diurnal variations systematic with aerosol abundance diurnal variations. The sudden sharp increase in the k values (Fig. 9a) during sunrise hours of marine-2~~August~~ is associated with a similar enhancement in the α_{abs} values. The high k values (>0.8) during these hours is due to the organic aerosols, inferred from α_{abs} values. Chung et al., (2012) have reported α_{abs} values above 1.6 for organic aerosols while, Gyawali et al., (2009) have reported α_{abs} values above 1.4 for biomass smoke. The daytime enhancement (~2 times) in CCN AF during ~~September~~ continental-2 is exactly according to the daytime enhancement seen in the aerosol GMD. Jayachandran et al., (2017) have reported the similar association between CCN AF and aerosol GMD diurnal pattern during ISM from a coastal site in southern peninsular India. Interestingly, similar association is not seen in other periods~~months~~. Thus, the aerosol composition especially the organic aerosols inferred from the high α_{abs} values is playing a ~~major~~ role in determining the CCN activation during the marine~~onsoon~~ conditions, while the aerosol size is determining the CCN activation during the continental conditions.

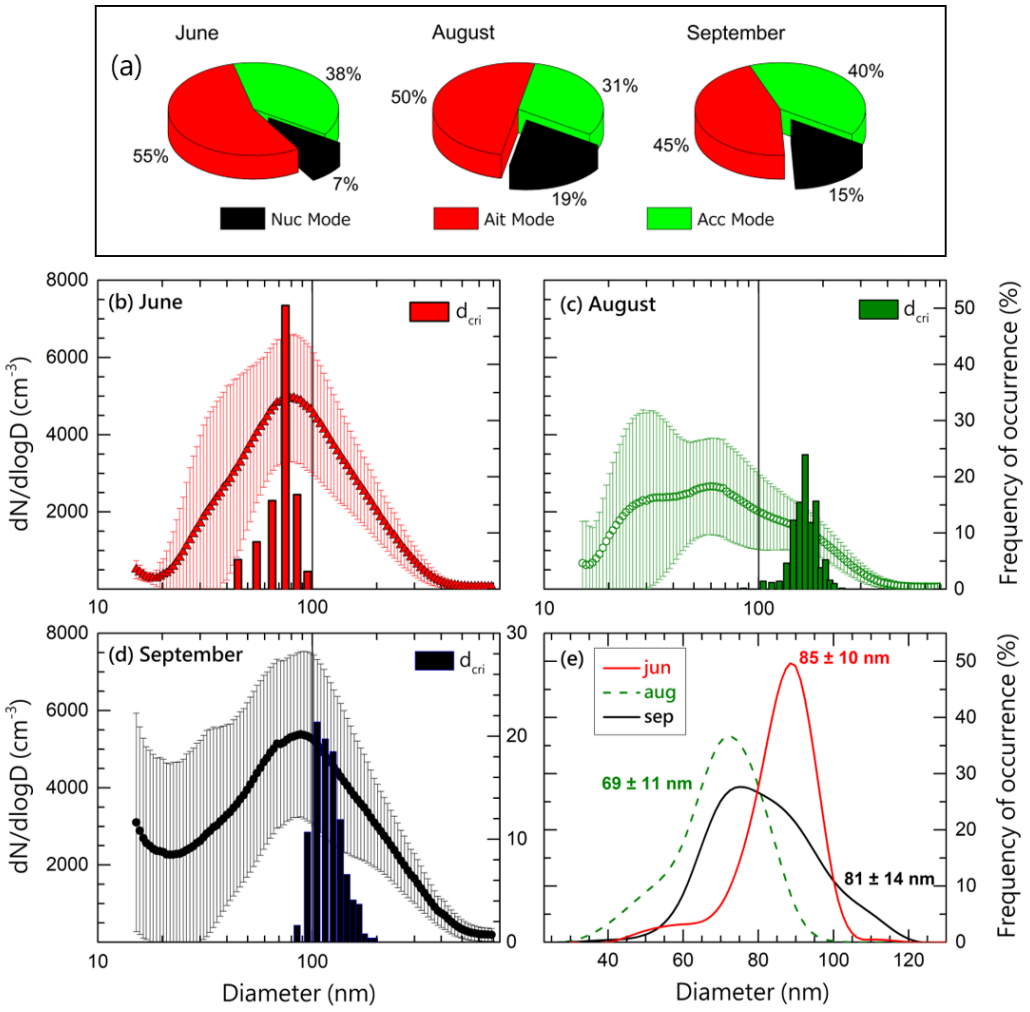
In general, high AF is found for aged background aerosols, while freshly emitted polluted urban aerosols have low CCN efficiency (Andreae and Rosenfeld, 2008). CCN AF values reported from India and some relevant studies reported across the globe are mentioned in Table 2. The similarity seen in CCN concentration and k values are seen in CCN AF also between Solapur and Ponmudi during the ISM. At both the places, only a small fraction (15-20 %) of the ambient AP is activating as CCN at 0.3 % SS. As seen in the Table, high AF values are reported from the coastal location and central Himalayas. The high CCN AF during the continental conditions at Solapur is similar to those reported during dry conditions in Nainital (Gogoi et al., 2015), where the high CCN AF was attributed to ~~possibly associated with~~ biomass burning. The low CCN AF observed at Solapur during ~~monsoon rainfall, possibly~~marine conditions resulting from the wet scavenging, is consistent with the values reported over the sub-continent during similar conditions, while the high CCN AF before and after monsoon rainfall is observed by several studies, resembling a biomass burning dominant polluted environment (Andreae, 2009).

As mentioned earlier and reported by several studies (Dusek et al., 2006; McFiggans et al., 2006), aerosol size plays a major role in determining the CCN activation ability of aerosols. It has been found that the UF particles were present during the monsoon conditions, when CCN AF was very low (Fig. 8). Meanwhile, the presence of bigger particles is enhancing the

CCN activation in other cases. To investigate the role of aerosol size in the observed CCN activity, the aerosol NSD during each condition is examined in detail.

3.5 Aerosol size distribution and critical activation diameter

The simultaneous and co-located aerosol size distribution observations and critical diameter are examined. The fraction of particles in the nucleation mode, Aitken mode, and accumulation mode is estimated and shown (in %) in Figure 11(a). Nucleation mode particles are those observed below 30 nm, Aitken mode particles are those from 30 nm to 100 nm and accumulation mode particles are those beyond 100 nm (Ueda et al., 2016; Willis et al., 2016). The corresponding mean NSD of AP along with the standard deviations for the study period are shown in panels (b), (c), (d) of Figure 11. The frequency of occurrence of the GMD for each period~~month~~ along with the mean GMD values are shown in Figure 11(e). As seen in the CCN characteristics, aerosol NSD also depicts distinct features prior to the marine~~onsoon~~ (Fig. 11b), during marine~~onsoon~~ (Fig. 11c) and after the marine air mass ~~onsoon~~ (Fig. 11d).


(a)
June
August
September
38%
55%
7%
50%
31%
19%
40%
45%
15%
Nuc Mode
Ait Mode
Acc Mode
(b) June
(c) August
(d) September
(e)
d cri
dN/dlogD (cm-3)
Frequency of occurrence (%)
Diameter (nm)
jun
aug
sep
85 ± 10 nm
69 ± 11 nm
81 ± 14 nm

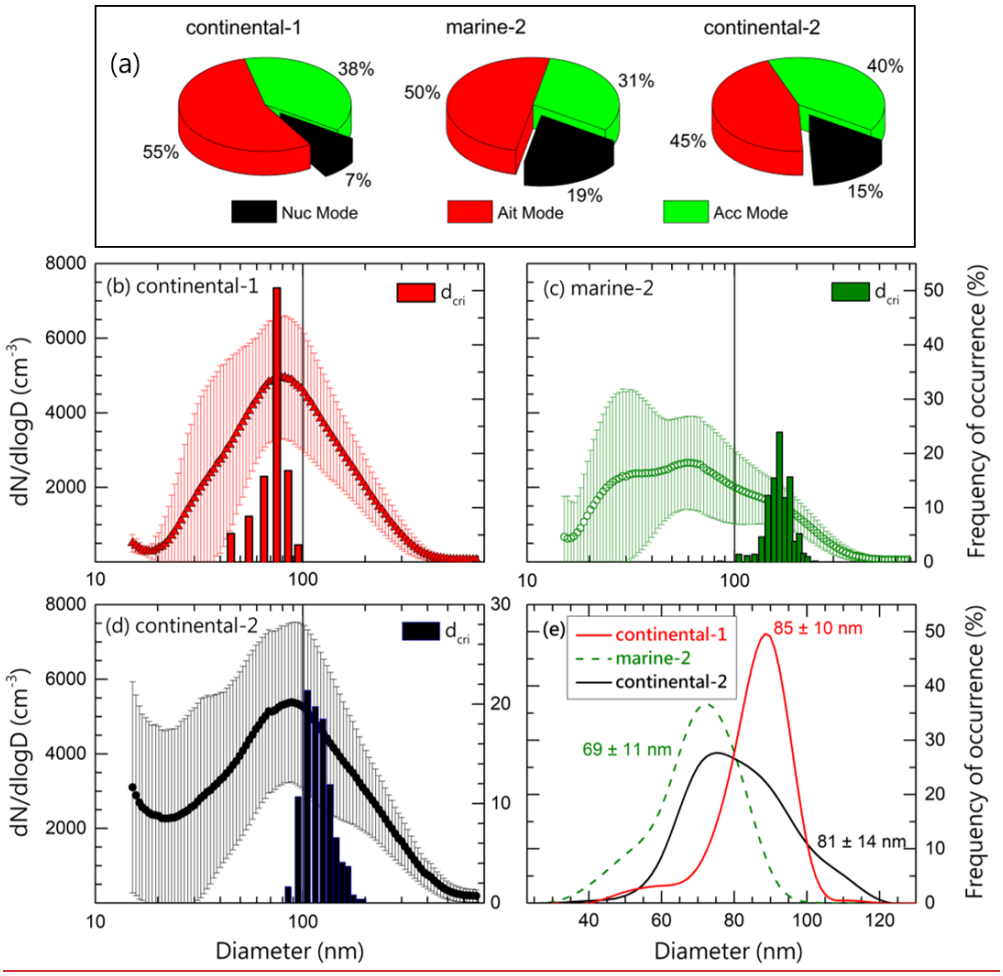


Figure 11. (a) Fraction (in %) of nucleation mode, Aitken mode, and accumulation mode particles during the observation periods. Aerosol mean number-size distribution (with standard deviations) during (b) continental-1~~June~~, (c) marine-2~~August~~, and (d) continental-2 condition~~September month~~s. The bars in the same plots indicate the frequency distribution of critical activation diameters at 0.3 % SS of the corresponding condition~~month~~s (right axis). (e) Frequency distribution of the geometric mean diameter of the aerosol system during the observation periods.

During continental-1~~June~~, most of the distributions are mono-modal, peaking around 80 nm and the mean GMD during this period is ~85 nm. In this period, the majority (~55 %) of the AP were present in the Aitken mode and least (~7 %) in the nucleation mode. During ~~similar~~ the continental-2 air mass conditions~~, but after the monsoon rainfall, the aerosol size distributions during the September is also similar. But~~ a prominent presence of nucleation mode particles (15 %) ~~i~~was also seen ~~(in the~~ Figure 11~~(~~d). The consistent presence of such particles is seen as the spread of the distribution of the GMD in Figure 11(e). Unlike the continental air mass conditions, the aerosol size distributions are entirely different during the marine air mass under the monsoon rainfall ~~conditions~~ as seen in Figure 11(c). Three modes are distinctly observed in ~~the mean~~

~~picture~~Figure 11(c), with two peaks below 100 nm. The mean GMD during this period is ~69 nm with the frequency distribution spreading towards the lower size range. About 19 % of the total aerosols were found in the nucleation mode (<30 nm) during marine-2~~August~~ and this feature continues in continental-2~~September~~ also, even though the air mass history changes. The accumulation mode (30-100 nm) AP concentration diminished (only 31 %) during the marine air mass ~~August~~.

For a given aerosol NSD, the critical activation diameter (d_{cri}) serves as an important parameter for representing the CCN activity, along with the CCN activation fraction and the empirical fit parameter – k values. Assuming homogeneous composition, d_{cri} for a specific SS can be estimated by integrating the aerosol NSD from the higher to lower size, until the integration becomes equal to the measured CCN number concentration at that SS (Furutani et al., 2008; Kammermann et al., 2010; Deng et al., 2011; ~~Varghese et al., 2016;~~ Fang et al., 2016). The lower limit of the integration can be considered as the 'apparent' critical activation diameter, as the ambient aerosol system can have both internal and external mixing state, and size-dependent composition. The frequency distribution of d_{cri} estimated for 0.3 % SS is also shown as the bars (right axis) in Figure 11(a-c). Comparing the d_{cri} for different conditions, the values were always less than 100 nm during continental-1~~June~~, contrastingly the values were always greater than 100 nm during marine-2~~August~~. The d_{cri} values were around 100 nm during ~~the September month~~continental-2. The mean (± standard deviation) values of d_{cri} were 72 ± 12 nm, 169 ± 38 nm and 121 ± 20 nm for continental-1~~June~~, marine-2~~August~~ and continental-2 condition~~September month~~s, respectively.

Different factors such as heterogeneous sources (Kim et al., 2002; Morawska, 2002), local meteorology (Wehner and Wiedensohler, 2003; Du et al., 2018), long-range transport (Birmili et al., 2001), and cloud processing (Noble and Hudson, 2019) can influence and modify the NSD. The predominant (69~~0~~ %) fine particles (<100 nm) size distribution (bi-modal) during ~~monsoon~~ marine air mass is similar to the two modes observed in the fine size range during monsoon at the urban site, Kanpur (Bhattu and Tripathy, 2014) during the ISM. The GMD values and the corresponding CCN properties from the present study and relevant other studies are listed in Table 2. Less AP concentration with low GMD (74 nm) was observed during monsoon at an urban site by Kanawade et al., (2014) and at a background Himalayan site (86 nm) by Kompulla et al., (2009) over the Indian sub-continent. Similar to the present study, GMD was high~~er~~ before and after the monsoon period, in both studies. From a high-altitude site in WG, Leena et al., (2016) have reported the lowest seasonal mean GMD of ~77 nm during ISM. The enhancement of the smaller particles in the total aerosol system, causing the reduction of the GMD especially during monsoon as seen in the present study is consistent with the previous studies (Babu et al., 2016). The mean GMD value observed over Solapur during ISM is the lowest (69 nm) reported value during the similar period over the Indian region.

Similar to the present observations, comparable accumulation and Aitken modes, and a dominant accumulation mode was reported over Amazon for wet and dry months, respectively by Pohlker et al., (2016). The accumulation mode particles are ~~usually aged,~~ associated with either aged biomass burning particles (Kalvitis et al., 2015)~~, and mostly resulting from the~~or condensation or coagulation of secondary ~~in~~organics and inorganics~~, and coagulation of smaller~~ particles (Seinfeld and Pandis, 2016). Interestingly, low~~smaller~~ aerosol GMD values (<70 nm), similar to the present observations during marine air mass~~August~~, were consistently observed near anthropogenic sources by Quinn et al., (2008). In the same study, they found bigger AP (GMD>70 nm) for observations carried out away from anthropogenic sources, which is similar to the present

observations during June and continental conitionsSeptember. From the concurrent Aerosol Mass Spectrometer measurements, they found that hydrocarbon-like organic aerosols (HOA) having mass spectrum characteristic of long chain hydrocarbons from fresh diesel exhausts were responsible for the fine mode, while and oxygenated organic aerosols and sulfates are responsible for the higher GMD. Hence the presence of freshly produced local fossil fuel combustion aerosols in the UF mode can be athe reason for the low CCN activity during marine-2August in the present study. During the marine condition, most of the bigger AP which are potential CCN are either washed out by wet scavenging or already activated as cloud droplets. Hence the measured AP are devoid of CCN active particles which are clearly seen from the aerosol NSD during the relevant period (Fig. 11). Thus, the low CCN activation fraction during the marine conditions is due to the missing of those CCN active particles near the surface. The low CCN activity in August could also be due to the wet scavenging conditions that were prevailing over the site, which indeed contributes to significant washout of bigger and hydrophilic AP.

Since the CCN activity depends mainly on the aerosol size and chemical composition (Dusek et al., 2006; McFiggans et al., 2006), d_{cri} estimated from concurrent aerosol NSD and CCN measurements can be considered as a proxy for the variations in the chemical composition of the aerosol system. As the aerosol size distribution and chemical composition are intrinsically associated with each other, any shift in the physical size distribution is mostly associated with the change in the aerosol composition, arising mainly due to the change in the sources or due to different processes such as ageing, coating or scavenging, except for externally mixed systems (Crosbie et al., 2015). Quinn et al., (2008) have correlated the d_{cri} with the HOA mass, and found that HOA can explain about 40 % of the variance in the d_{cri}. They have reported 70-90 nm and higher values (>90 nm) as the d_{cri} for marine and inland regions, respectively, at 0.44 % SS. For anthropogenic and marine environments, Furutani et al., (2008) have reported d_{cri} values of 70-110 nm and 50-60 nm, respectively, at 0.6 % SS. The d_{cri} values observed during continental conditions at Solapur is similar to the values observed at a tropical monsoon climate region by Fang et al., (2016), under urban influence. The d_{cri} values during marine-2August are higher than the corresponding values reported from polluted North-China plain by Deng et al., (2011). From an urban site, Burkart et al., (2011) have reported an average value of ~169 nm for d_{cri} at 0.5 % SS. Freshly emitted carbonaceous combustion particles can have large d_{cri} values up to ~350 nm, even at a high SS (0.7 %) (Hitzenberger et al., 2003). The d_{cri} can exhibit very low values also, than theoretically estimated ones in the presence of partially or fully soluble particles, as their slight presence can largely enhance the CCN activity of insoluble particles such as BC and dust (Dusek et al., 2006; Begue et al., 2015). The sharp distinction in the d_{cri} values and aerosol NSD between different atmospheric/air mass conditions within the same season in the present study indicate the difference in the aerosol composition. The low GMD and high d_{cri} during the August indicate the presence of freshly emitted water inactive primary organic aerosols.**Relationship between CCN activation fraction and critical activation diameter**

The association of CCN activation fraction with d_{cri} is examined and shown in Figure 12. The different months are indicated by the shape of the scatter, while the color indicates the concurrent UF fraction (CN<30 nm in %). In general, the CCN AF and d_{cri} are anti correlated. From the Figure, it can be seen that the points corresponding to June (triangles) and August (stars) months lie at opposite corners of the scatter, while those during September (circles) are between the other two.

As seen in the aerosol NSD discussions, nucleation mode AP are almost absent during June, while it is more common during August and a few cases are observed during September. But there are cases of having low CCN AF (<0.1), but not associated with UF events, but associated with higher BC mass concentrations. Thus, the presence of UF particles cannot be pointed out as the only cause for the lower CCN efficiency of the aerosols, but the composition is also having a role in determining the CCN efficiency.

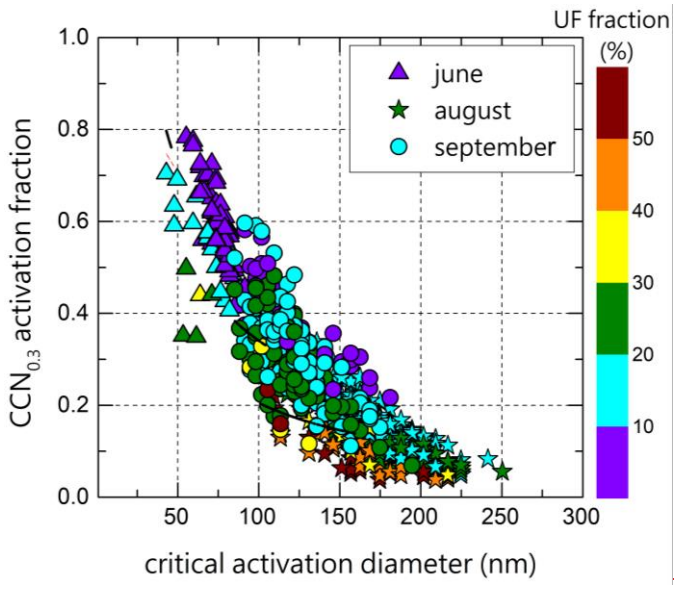


Figure 12. Scatter between CCN activation fraction at 0.3 % SS and the critical activation diameter. The color of the scatter indicates the nucleation mode (UF) fraction.

From Figure 12, it can be seen that the CCN AF is not varying linearly with the d_{cri}. Except for the month of June, the CCN AF exhibited a power law dependence on d_{cri}. Hence a non-linear fit was applied to the scatter as

$$CCN_{AF} = a \times d_{cri}^{-b} \quad (3)$$

'a' is a constant as the CCN AF is assumed to be one when the d_{cri} reaches zero, in all the cases. The value of 'b' and the corresponding correlation coefficient (inside bracket) of the fit was estimated as 0.67 (0.97), 0.73 (0.97), and 0.84 (0.98) for June, September, and August months, respectively. During June, the CCN AF (in %) decreases linearly with d_{cri} with a negative slope value of 0.59 with a better correlation coefficient of 0.99, than that obtained using the non-linear fit.

The inverse linear relationship between the CCN AF and d_{cri} during June is similar to that reported by Furutani et al., (2008). In their study the critical diameter varied within a range (30-130 nm) similar to that of the present observation during June (40-90 nm), where the linear relationship exists. As the d_{cri} increases beyond 100 nm, linear relationship switches to non-linear, and the order of the relationship increases as d_{cri} increases, which is illustrated by September (b = 0.73) and August (b = 0.84) months values.

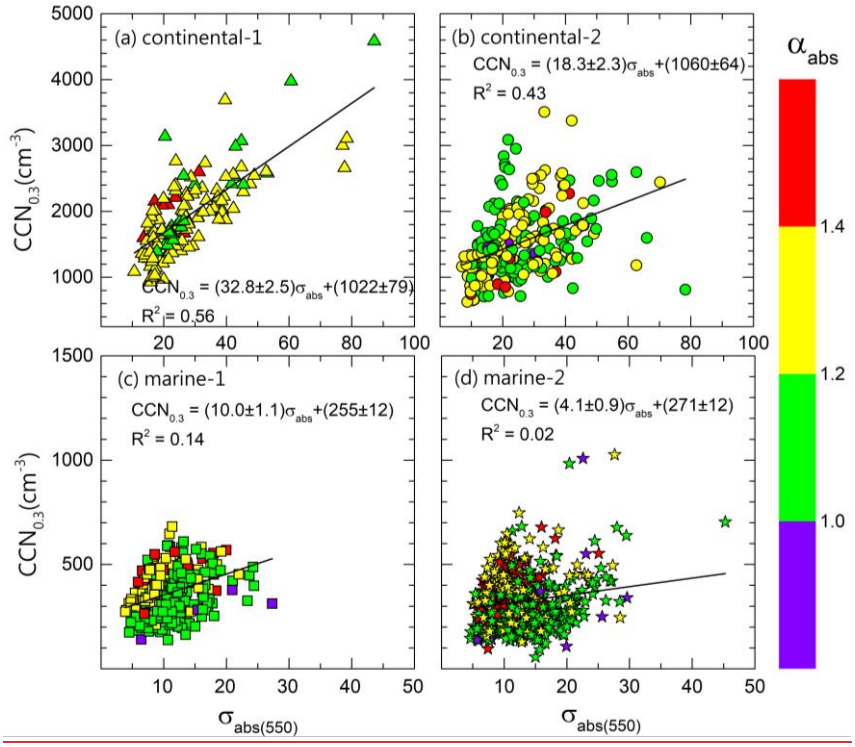
3.6 Relationship between ~~CCN activation~~ aerosol absorption and CCN properties

The aerosol composition plays an important role associated with the changes in the aerosol NSD due to the meteorological processes and active source and sink mechanisms prevailing during the monsoon conditions. The significant influence of aerosol composition in determining the CCN activity at lower SS which is more probable in real atmosphere, is demonstrated in many studies (Cubison et al., 2008; Kammermann et al., 2010; Bhattu et al., 2015; Jayachandran et al., 2017). In this aspect, the role of carbonaceous aerosols in determining the CCN activation is investigated.

The association of CCN concentration at 0.3 % SS and absorption coefficient at 550 nm, segregated for different periods are shown in Figure 12~~3~~. ~~Each point in the Figure indicates the CCN at 0.3 % SS corresponding to the aerosol absorption coefficient given in the X-axis. The color of the scatter indicates the α_{abs} values. A least square linear fit of the scatter is also made and shown along with the fit parameters.~~ The association between aerosol absorption and CCN concentration is generally low, but ~~comparatively high~~moderate during the continental air mass and very weak during the marine air mas~~onsoon condition~~s. The better association between the CCN concentration and absorption properties may be due to (i) absorbing AP itself acting as CCN or (ii) aerosol species co-emitted with the absorbing AP activating as CCN. The higher slope and better association observed during continental-1~~June~~ indicate that the low k, high AF values and the high association of between CCN and CN during the period are due to the major contribution of carbonaceous aerosols towards CCN activation. It can be either due to the co-emitted organics enhancing the CCN efficiency of the aerosol system or due to the aged carbonaceous aerosols itself activating as CCN or due to the combination of both. The enhancement in the accumulation mode aerosols supports this observation as oxygenated organic aerosols and sulfates are found in the accumulation mode (O'Dowd et al., 1997; Quinn et al., 2008). During marine air mass ~~(wet conditions)~~ there is no clear association between aerosol absorption and CCN, and less AP are activated as CCN. It indicates that there is a change in the source/sink and nature of CCN during marine/wet and c~~C~~ontinental/dry conditions.

Carbonaceous aerosols form a major source of CCN concentration and thereby contribute to indirect effect of aerosols (Novakov and Penner, 1993). Anthropogenic carbonaceous aerosols cause an indirect effect of -0.9 W m^{-2}, while sulfates cause only -0.4 W m^{-2} (Lohmann et al., 2000). Spracklen et al., (2011) have shown through simulations that about 60 % of the global CCN concentration is from carbonaceous sources. Various atmospheric processes such as ageing, coating and mixing can enhance the water activity of BC (Lammel and Novakov, 1995; Kuwata et al., 2009), which is hydrophobic when freshly emitted. Mixing with hydrophilic substances like inorganic salts can also enhance the CCN activity of carbonaceous aerosols (Dusek et al., 2006). Thus, the better correlation observed between CCN and aerosol absorption, and the associated high CCN efficiency during the continental conditions indicate the significant role of carbonaceous aerosols in CCN activation at Solapur. Jayachandran et al., (2018) illustrate~~d~~ the close association of CCN with aerosol absorption properties (better than present study) from WG and the lack of association between the parameters at a coastal site during the monsoon conditions. Enhancement in CCN concentration along with increase in the aerosol absorption coefficient was observed at central

Himalayas (Gogoi et al., 2015). In general, carbonaceous aerosols have a significant role in CCN concentration during continental conditions.



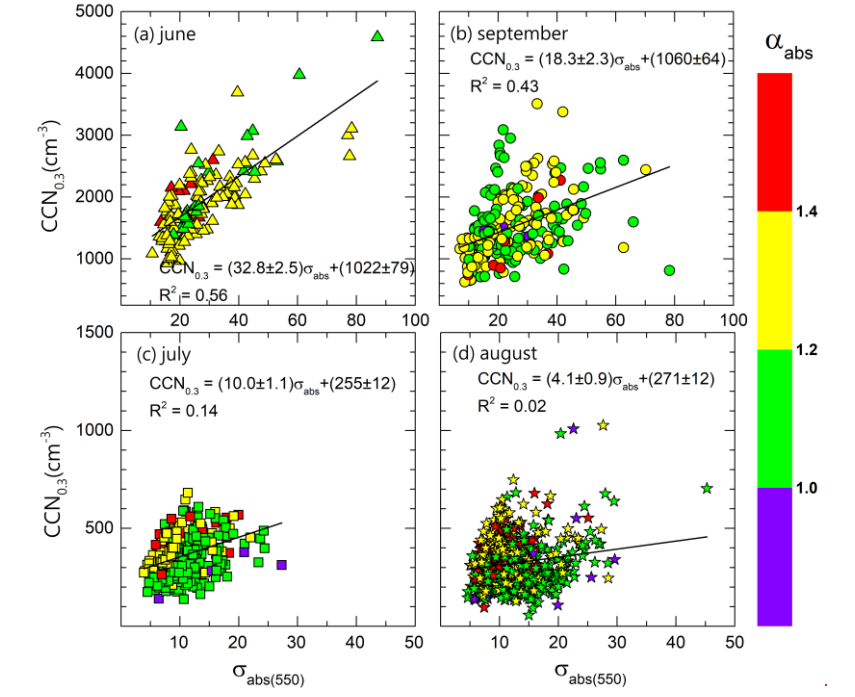


Figure 12~~3~~. Association between CCN (at 0.3 % SS) and absorption coefficient (at 550 nm) for (a) continental-1~~June,~~ (b) marine-1~~July~~, (c) marine-2~~August~~, and (d) continental-2 condition~~September month~~s. The color of the scatter indicates the concurrent absorption angstrom exponent values. The least square linear fit is also shown along with the fit parameters.

As the very low AF is observed along with the enhancement in the nucleation mode particles and depletion of Aitken particles after the onset of ISM over the study region, the chemical characteristics of Aitken, as well as accumulation mode particles, are investigated. Most of the studies, from different parts of the globe, have reported about the high hygroscopicity of accumulation mode particles and less hygroscopicity for the Aitken mode particles (Paramo~~a~~nov et al., 2013; 2015). From the long-term observations from Amazon, Pohlker et al., (2016) have concluded that organics predominantly present in the Aitken mode reduces the hygroscopicity, while the dominance of inorganics in the accumulation mode enhances the aerosol hygroscopicity which was in-line with other studies (Gunthe et al., 2009; C. Pohlker et al., 2012).

The CCN concentration ~~found to have~~exhibited an association with the absorption coefficient during the continental air mass compared with the marine air mass conditions. The reduced CCN efficiency due to the presence of Aitken or UF mode is already discussed. To ascertain the contribution of the carbonaceous AP to the NSD, the association of BC with Aitken mode particles and accumulation mode particles are examined under high and low aerosl loading conditions, and shown in Figure 13~~4~~. Since the BC loading was almost (>99 % cases) less than 2500 ng m^{-3} during marine-2, cases above this value is taken as polluted (p, high loading) in continental air mass. ~~–~~A least square linear fit is also made and the corresponding parameters are shown in Table 3. Under similar BC loading (low), Aitken mode AP are better associated with BC with higher slope (m=0.68) during marine-2. While, the accumulation mode AP showed similar correlation with BC during low loading and the lowest slope during marine-2. The Aitken mode AP showed better association with BC during continental-2 under high BC loading case. Except during continental-2, the accumulation mode AP were more associated with BC compared to Aitken mode AP. These all indicate that BC or the co-emitted AP from carbonaceous combustion sourecs were predominant in Aitken mode during marine-2 and polluted continental-2 conditions. In all other cases the carbonaceous combustion AP contributed to the accumulation mode. ~~The BC mass concentration is better associated with the accumulation mode AP during June and less associated during August. The BC mass concentration is not associated with the Aitken particles during June, while comparatively better associated during August and September. The C values (Y-intercept) of the linear fit show that the BC mass is present even in the absence of Aitken particles in all the conditions and the highest during the June (C~1224 ng m^{-3}). Interestingly, the negative C values indicate, BC mass is absent while accumulation mode particles reduce to zero, except during monsoon conditions. This shows the BC or the co-emitted AP are present in the accumulation mode only during the continental (dry) air mass.~~

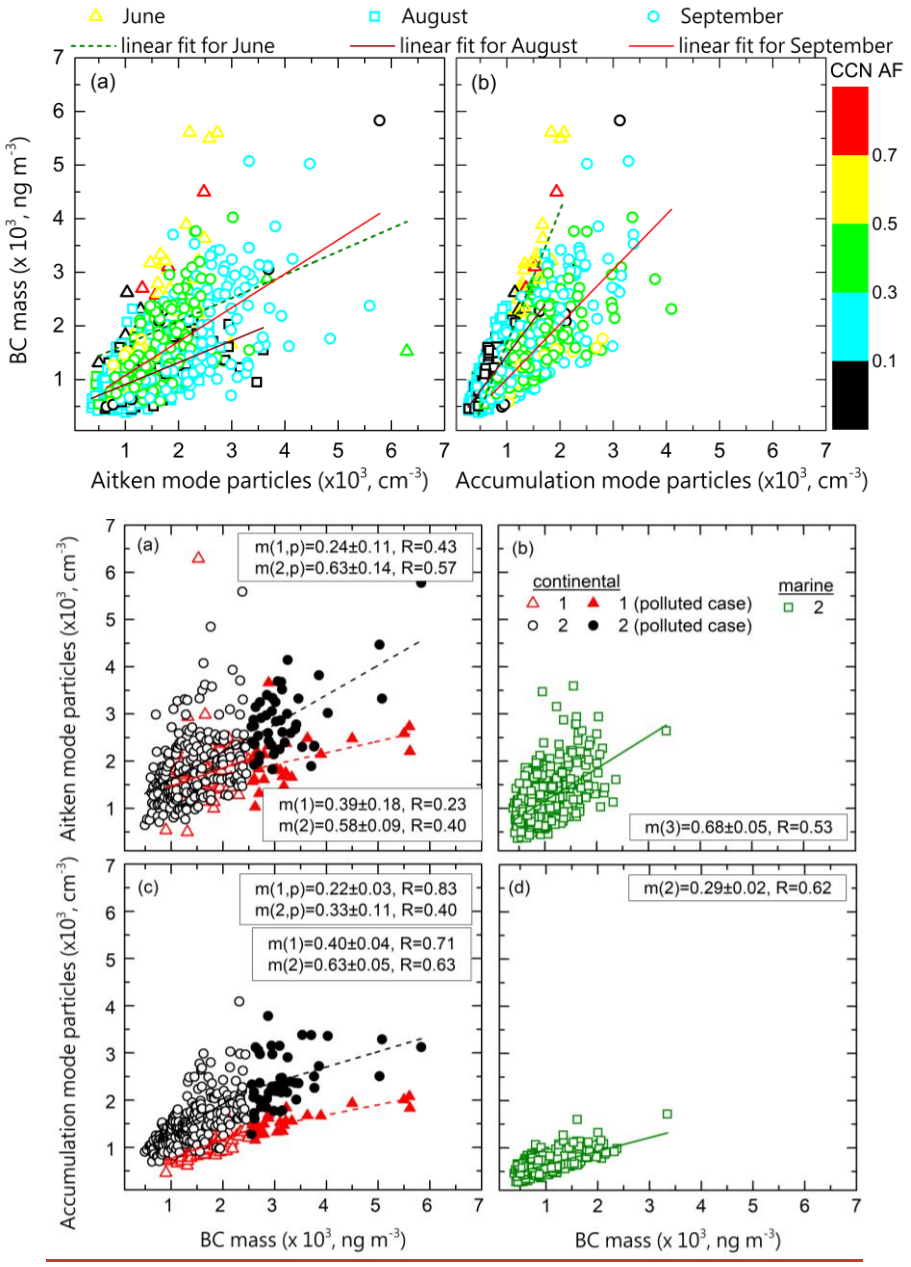


Figure 1~~3~~4. Association of BC mass concentration with (a) Aitken and (b) accumulation mode particles, segregated for different air mass conditions. The least square linear fit lines are also shown. The color of the scatter indicates the corresponding CCN AF values.

The association of BC with the accumulation mode particles and association of CCN with absorption coefficient during ~~June~~continental-1 along with high CCN AF indicate~~s~~ the aged, large size hygroscopic particles from carbonaceous combustion sources present prior to the monsoon at the location. The strong convective conditions existing during these

conditions over the region can take the AP to high altitudes where it can absorb radiation and may lead to semi-direct effects. The association of BC with accumulation mode AP during continental conditions suggest that the carbonaceous AP existing in this size range can act as CCN. From the eastern coast of India, Kompalli et al., (20~~19~~20*19*) have reported highly coated larger BC particles (>110 nm) in dry conditions under the continental influence, while nascent BC particles (~80 nm) with less coating were found during ISM due to wet scavenging. This finding is in-line with the current marine-2 observations. The enhanced fine particle concentration having better association with BC mass concentration ~~after the onset of ISM~~during continental-2 underlines the possibility of freshly emitted carbonaceous aerosols reducing the CCN AF. The association of BC mass and accumulation mode aerosol number concentration also points to the possibility of inorganic aerosols like sulfate, co-emitted along BC from carbonaceous combustion sources, enhancing the CCN activity of AP in ~~dry,~~ continental conditions~~.~~. Hence, the role of carbonaceous AP in modulating both cloud microphysics and dynamics need to be investigated in detail. However, current investigation could not address these probable aspects.

3.~~6~~7 CCN closure

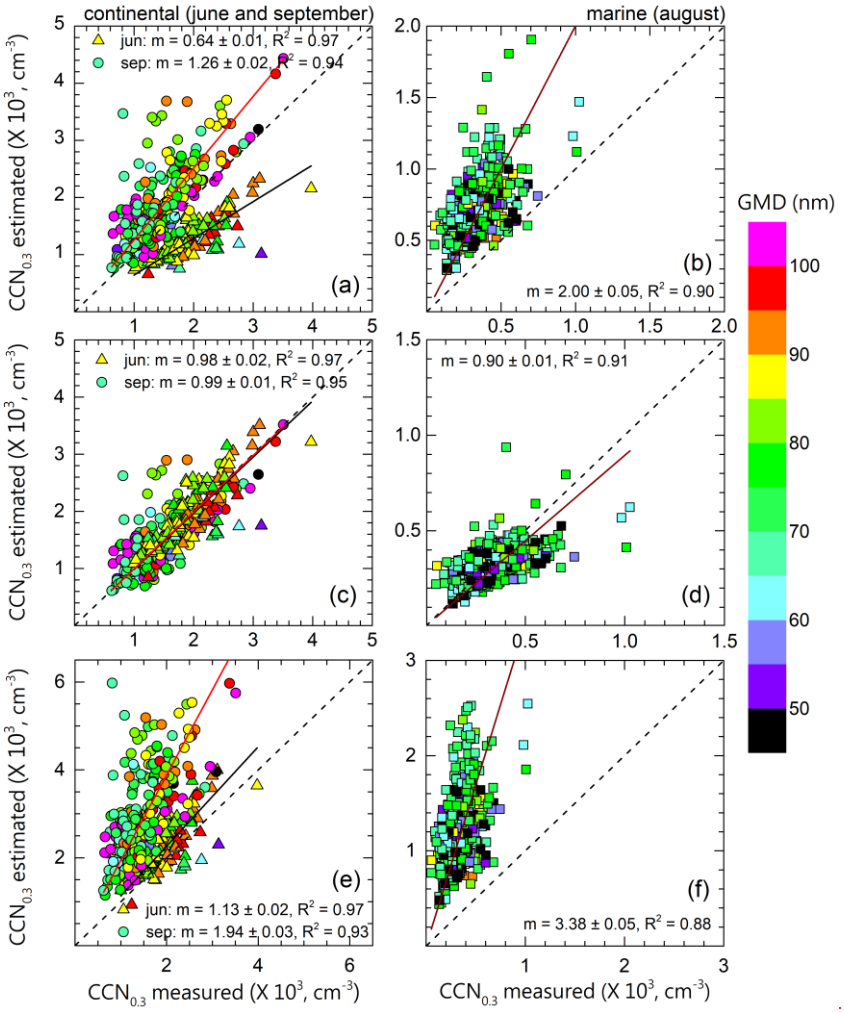
~~The biggest uncertainty in ACI regarding the CCN concentration and characteristics is due to the lack of proper understanding on the dependence of CCN on aerosol size and composition.~~ To understand the role of size and composition in CCN activity ~~minimize this uncertainty~~, CCN closure studies have been carried out by many investigators (Brokehuizen et al., 2006; Lance et al., 2009; Juranyi et al., 2011; Bhattu and Tripathy ~~et al.~~, 2015; Crosbie et al., 2015; Jayachandran et al., 2017) at different environments, leading to the better understanding on the CCN activation from aerosols. CCN concentration at the rain shadow region under different air mass conditions are estimated and validated with the measured CCN concentrations. The CCN concentrations are estimated by (i) assuming accumulation mode aerosols activating as CCN (ii) applying the mean 'apparent' critical diameter and (iii) assuming the aerosol composition as ammonium sulfate ($(NH_4)_2SO_4$), and compared with observations. The scatter between the CCN concentration at 0.3 % SS estimated and the corresponding measured CCN concentrations segregated according to marine and continental conditions are shown in Figure 1~~4~~5. ~~The color of the scatter plot represents the concurrent GMD values of the aerosol system. Least square linear fit is applied to the scatter and the corresponding fitting parameters are also mentioned in the Figure. The dash line indicates the unit slope (m = 1) line.~~

CCN are generally approximated as AP above 100 nm (accumulation mode) in many studies when there are no concurrent CCN measurements (eg. Andreae, ~~2005~~2009). Still, it is a rough approximation due to the non-linear dependence of CCN activation of AP, and this assumption is examined in Figure 1~~4~~5 (a and b). The variations in accumulation mode ~~aerosols~~ AP are correlated well (R>0.95) with the CCN concentration in ~~continental~~ all the conditions ~~(R = 0.98 and 0.97), compred to the marine conditions (R = 0.95)~~. Interestingly under the same conditions (continental), the accumulation mode ~~aerosols~~ AP have different activation efficiency as CCN and is under-estimated (m = 0.64) during ~~June~~ continental-1 and over-estimated (m = 1.26) during ~~September~~continental-2. But during ~~August~~ marine-2 the linear fit of the scatter ~~indicates~~ shows

that the estimated CCN concentration is almost twice as that of the measured concentration, when the accumulation mode particles are considered to be CCN active.

Rather than taking an assumed value as the critical activation diameter (100 nm), the mean of the measured critical diameter (d_{cri}) is used to estimate the CCN concentration and shown in panels (c) and (d) of Figure 1~~5~~. The mean d_{cri} for 0.3% SS of ~70, ~165 and ~120 nm are used for estimating CCN during ~~June~~continental-1, ~~August~~ marine-2 and ~~September month~~continental-2 conditions, respectively. The estimated and the measured CCN concentration correlate well during the continental conditions with high correlation coefficient (R = 0.97) and almost unit slope. From the Figure 1~~5~~(c), it can be seen that most of the points lie along the diagonal 1:1 line, irrespective of the number concentrations and GMD values. ~~Compared to these, the CCN concentration is under estimated (m = 0.90) and correlation coefficient is less (R = 0.95) during August. The comparatively low correlation and slope of the linear fit indicate the absence of a sharp size cut in the activation of CCN. This may points to the role of chemical composition of small (<100 nm) particles and the mixing state of the aerosol system in CCN activation.~~

The CCN concentration at 0.3 % SS at different conditions are estimated (Fig. 1~~5~~ e and f) by assuming an inorganic composition of $(NH_4)_2SO_4$ as an ideal scenario. The methodology used for the estimation is given in Appendix. In all the conditions, CCN concentrations were over-estimated by this approach, indicating that the ambient aerosol system has a lesser hygroscopicity (κ) than that of $(NH_4)_2SO_4$. The highest over-estimation (m = 3.38) is observed during ~~August~~ marine-2~~and the correlation of the linear fit of the scatter is also comparatively weak (R = 0.94)~~. During ~~June~~continental-1, the maximum correlation coefficient (R = 0.98) is obtained and the slope is also nearer to the unity (m = 1.13). However, ~~even~~ for the continental-2 conditions, the CCN concentrations are over-estimated ~~(m = 1.94)~~(almost twice) ~~during September~~ when $(NH_4)_2SO_4$ composition is assumed. These observations confirm that the nearly mono-modal aerosol NSD observed during continental-1~~June~~ is more similar to an aged continental aerosol system having similar hygroscopicity of sulfate aerosols. This observation has to be considered along with the association of BC with accumulation mode aerosols (Fig. 1~~3c4b~~). The AP system observed during marine conditions are of very less hygroscopic and the multiple size modes observed in the smaller size range indicate a heterogeneous composition in a complex mixing state, during the wet conditions.


continental (june and september)
marine (august)
jun: m = 0.64 ± 0.01, R² = 0.97
sep: m = 1.26 ± 0.02, R² = 0.94
(a)
m = 2.00 ± 0.05, R² = 0.90
(b)
jun: m = 0.98 ± 0.02, R² = 0.97
sep: m = 0.99 ± 0.01, R² = 0.95
(c)
m = 0.90 ± 0.01, R² = 0.91
(d)
jun: m = 1.13 ± 0.02, R² = 0.97
sep: m = 1.94 ± 0.03, R² = 0.93
(e)
m = 3.38 ± 0.05, R² = 0.88
(f)
GMD (nm)
100
90
80
70
60
50
CCN0.3 estimated (X 10³, cm⁻³)
CCN0.3 measured (X 10³, cm⁻³)

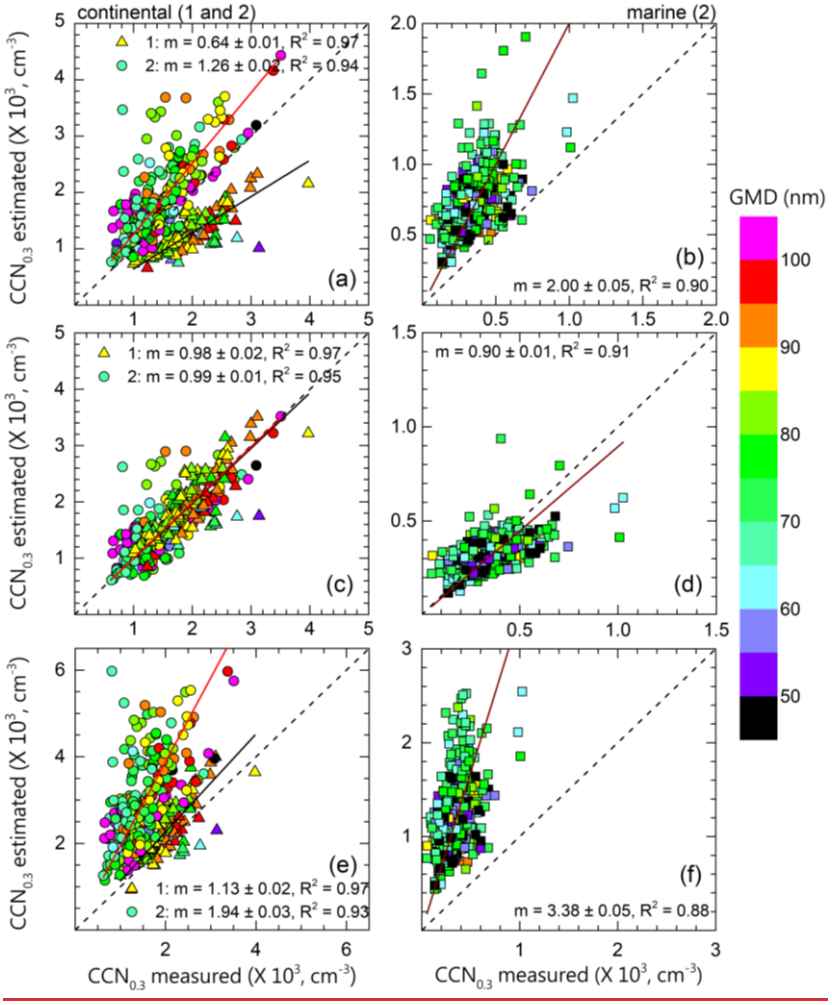


Figure 1~~4~~5. Scatter between estimated and measured CCN concentration at 0.3 % SS for continental and marine conditions with the color indicating the GMD. CCN estimated (a and b) as particles above 100 nm, (c and d) from critical activation diameter, and (e and f) using aerosol NSD and ammonium sulfate composition. Linear fit and the parameters are also shown. The dash lines indicate the unit slope (m=1) line.

Comparing the three approaches used to estimate the CCN concentration, using a sharp size cut for estimating CCN concentration suits well for ~~the continental aerosol systems~~all the cases. ~~Perfect closure is obtained using the measured critical diameter indicating the size-dependency of CCN activation during these conditions. Still~~However, there is a variation in the AP composition in the continental air mass before the monsoon rainfall and after that, reflecting in the different CCN efficiency of the accumulation mode during these periods. The aged aerosol system prior to the monsoon resembles a sulfate aerosol composition with a very high CCN activation efficiency and low k values. ~~The CCN AF during this period inversely varies in a linear manner with the d_{crit}.~~ During continental-2~~September~~ all the accumulation mode aerosols are not participating in the

CCN activation (Fig. 145a) and the assumption of $(NH_4)_2SO_4$ composition nearly doubles the estimated CCN concentrations. ~~Thus, fresh anthropogenic fine aerosols mostly from carbonaceous combustion sources, during this period (Fig. 14a) are inactive towards the CCN activation.~~ Comparing the aerosol NSD in continental conditions, there is a depletion of Aitken mode particles and an enhancement in nucleation mode particles during continental-2~~September~~. Similar to the different AF between ~~June and September~~continental 1 and 2, Pohlker et al., (2016) have showed high CCN AF in the absence of nucleation mode.

This study ~~underlines the inability to make use of either aerosol size distribution or uniform (internally mixed) composition to explain the CCN activation during the monsoon (wet) conditions. This finding~~ is similar to the ~~weak~~ CCN closure reported by Crosbie et al., (2015) for North-American monsoon conditions. The complex meteorological pattern including the monsoon showers and regional aerosol production (both primary and secondary) causes large variability in the aerosol NSD as seen in Figure 11. The ~~very~~ less hygroscopicity of the accumulation mode aerosols during the monsoon is revealed in Figure 145(b). ~~Even the measured mean d_{cri} cannot predict the CCN concentration from the measured NSD accurately.~~ The least closure is obtained while assuming a uniform internal mixture of hygroscopic inorganic composition. These all points to the highly complex mixture of the size-dependent composition of the prevailing aerosol system during monsoon. Studies (Cubison et al., 2008; Ervens et al., 2010) have highlighted the need for size-resolved composition information for estimating the CCN concentration for freshly emitted AP, near to the sources. ~~It can also be noted that the GMD may not have much role in deviating the closure during active monsoon, which is similar to the inference from Figure 7.~~ Thus, even though the nucleation mode AP present during the period hinder the CCN activity, the presence of bigger particles in the same period is not supporting the CCN activation. It indicates that apart from the size of the aerosols, the composition/mixing state of the aerosol system during marine-2~~August is~~ also ~~playing a crucial role in determining~~influence the CCN efficiency. From the aerosol optical properties (Fig. 10a), it is seen that the low CCN AF and high k value is associated with the enhancement of the organics at the site. These organics observed after sunrise hours during marine~~onsoon~~ conditions ~~are~~ may be limiting the CCN activation of the aerosols. The quantification and classification of these species are essential to address the effect of aerosols on clouds in the~~a~~ rain shadow region, especially during the monsoon ~~conditions~~.

4 Summary and Conclusions

CCN characteristics at a rain-shadow region during the Indian Summer Monsoon (ISM) are studied with respect to the different air mass and meteorological conditions that prevailed over the region. It is found that the polluted-continental conditions transform into a polluted marine condition by the onset of ISM with a significant change in aerosol size distribution and composition affecting the cloud nucleating properties. The important findings are listed below.

- Comparatively high BC (~2,000 ng m^{-3}) loading and AOD (> 0.5) prevailed over Solapur before and after the marine air mass~~onsoon~~, which reduced to very low values (BC~800 ng m^{-3}) during the monsoon-clean background conditions.

- The lowest CCN concentrations at any SS (~900 cm^{-3} at 1.2 % SS) is observed at Solapur, compared to the values reported during ISM over the Indian sub-continent. However, the k values (~0.6) during ISM are high and similar to those reported over Western Ghats (WG) and peninsular India under similar conditions.
- A slight daytime enhancement in CCN is seen due to the influence of anthropogenic activities, while a significant enhancement in k values (2 times) was observed during the daytime of monsoon ~~associated~~ with ~~organic aerosols, inferred from the~~ concurrent high absorption Angstrom exponent values.
- Significant diurnal variations in CN, BC concentrations and properties like 'k', CCN AF and α_{abs} during the marine~~wet conditions~~ air mass indicate the dominant presence of local aerosol sources, while the weak diurnal variations of the same parameters during the continental~~dry~~ air mass~~conditions~~ indicate the consistent polluted background conditions at Solapur.
- The aerosol system prior to the onset of ISM having a mono-modal number-size distribution (NSD) with a geometrical mean diameter (GMD) of 85 ± 10 nm depicted high CCN activation fraction (AF) of ~55 % at 0.3 % SS. During the ISM, multiple modes were observed in aerosol NSD with a predominant nucleation mode fraction (~19 %) resulting in the lowest CCN AF of ~15 %. Just after the monsoon rainfall, aerosols were significantly present both in nucleation (~15 %) and accumulation mode (~40 %) and the CCN AF enhanced to ~32 % only even though the corresponding aerosols GMD was 81 ± 14 nm.
- The mean critical activation diameters (d_{cri}) estimated for 0.3 % SS from concurrent CCN and aerosol NSD measurements were highest during the monsoon (~165 nm) and lowest just prior to the monsoon (~70 nm), and ~120 nm just after the monsoon for 0.3 % SS. ~~The CCN AF decreases with an increase in d_{cri}, linearly up to ~100 nm (prior to the monsoon) and beyond which non-linearly (during and after monsoon) decreases.~~
- Better association of absorbing type aerosols with CCN and accumulation mode aerosols during continental air mass conditions indicate the aged, bigger sized particles from carbonaceous combustion sources possibly enhancing the CCN activity prior ~~and after~~ to the marine air mass~~onsoon~~. ~~CCN closure analysis revealed that the CCN population during continental air mass (before the monsoon) is more of sulfate type.~~ While, aerosol absorption correlated well with Aitken mode particles during and after the marine air mass resulting in low CCN activation.
- The closure study indicates ~~that~~ the size dependency of CCN activation especially during dry-continental conditions. Most of the CCN-active AP were removed from atmosphere by activation or wet removal and the remained particles were inherently CCN-inactive as seen in the aerosol NSD during the marine air mass. ~~weakens during wet monsoon conditions. CCN estimation using measured aerosol NSD and sulfate composition assumption is not valid during monsoon.~~ However, the CCN activation efficiency of the accumulation mode ~~The role of Aitken mode composition and mixing state is very significant in CCN activation~~particles reduced during and after the ~~wet monsoon~~marine air mass~~conditions~~.

~~Even though the aerosol-CCN conditions correspond to a 'polluted-marine' conditions over the rain-shadow region,~~ ~~t~~The very low aerosol loading (towards an aerosol limited regime) with low CCN efficiency during the ISM rainfall, adds the significance of CCN in cloud droplet concentrations. The regional CCN concentration can be estimated from the aerosol size distribution alone, indicating the size dependency of CCN activation during the continental ~~(dry)~~ airmass conditions. But the distinct aerosol NSD and CCN properties during the monsoon due to the change in the aerosol source and sink mechanisms ~~lead to the stronger~~suggest the dependence of CCN activation on the composition of Aitken mode aerosols and its mixing state~~. Aerosols similar to the composition of sulfates existing in accumulation mode enhances the CCN activation during continental air mass, while the accumulation mode aerosols during the monsoon have low hygroscopicity.~~. However, ~~T~~the predominance of ultrafine particles in the boundary layer and the corresponding very low CCN efficiency ~~after the onset of ISM~~ demand further studies using the simultaneous cloud base observations to understand the ACI affecting the precipitation pattern over the rain shadow region against the backdrop of cold phase invigoration (Rosenfeld et al., 2008; Gayatri et al., 2017) and condensational heating (~~Khain et al., 2012;~~ Fan et al., 2018) mechanisms of tropical convective clouds.

Appendix

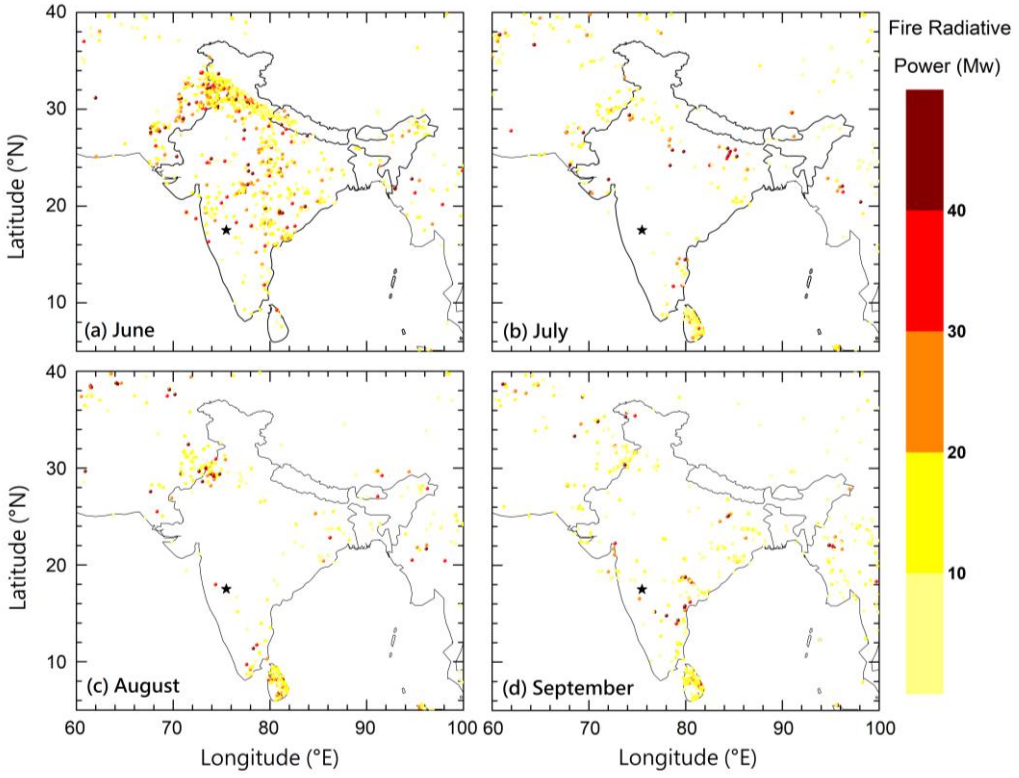


Figure A1. Spatial distribution of the Moderate Resolution Imaging Spectroradiometer (MODIS) fire radiative power (Collection 6 product obtained from https://earthdata.nasa.gov/firms) for the measurement periods, along with the observation site marked as a black star. Data points having confidence value higher than 30, which comes under the classification of 'nominal' and 'high' are only used.

Estimation of CCN concentration

The saturation ratio, $s = a_w \, exp\left(\frac{A}{D_p}\right)$ (A1)

and $a_w = \left(\frac{4\sigma M_w}{RT\rho_w}\right)$ (A2)

where a_w is the water cativity of the solution droplet, σ is the surface tension of the solution. M_w and ρ_w are the molecular mass and density of water. R is the universal gas constant and D_p is the size (Seinfeld and Pandis, 1998). The critical diameter for a given aerosol system can be estimated from the Köhler theory based on its size distribution, chemical composition, hygroscopic growth information. The critical diameter derived from the Köhler equation is as follows (Lance et al., 2009)

$$d_{cri} = \left[\frac{27}{4}\left(\log\left\{\frac{SS}{100}+1\right\}\right)^2\left(\frac{\rho_w RT}{4\sigma M_w}\right)^3\left(\frac{M_w}{\rho_w}\right)\frac{\rho_s \vartheta \in}{M_w}\right]^{-1/3} \quad \text{(A3)}$$

where, ρ_s, M_s, and $\in$ denote density, molecular mass and volume fraction of the solute, respectively. ϑ is the effective Van'thoff factor.

Assuming a pure internally mixed aerosol system with uniform composition, CCN concentration can be predicted using the following equation based on the measured aerosol NSD and estimated critical diameter (Juranyi et al., 2011).

$$N_{CCN}(SS_{set}) = -\int_{D_{max}}^{d_{cri}} \frac{dN}{d\log D}(D)\, d\log D \quad \text{(A4)}$$

In the present study, the aerosol composition is assumed to be of ammonium sulfate as an ideal case (Covert et al., 1998) in order to examine the deviation of CCN activation from the ideal scenario. Hence the insoluble fraction was taken as zero. Density of $(NH_4)_2SO_4$ is taken as 1.76 g cm^{-3} (Hinds, 1999).

Data availability

Data used in the present study can be obtained by making a request through http://www.tropmet.res.in/~caipeex/ registration form.php or contacting thara@tropmet.res.in.

Competing interests

The authors declare that they have no conflict of interest.

Author contributions

TVP conceptualized the experiment. TVP and VJ designed the study. TVP, PM, KST, SPB, GD, NM, JR, MK, SD, MV and PDS were responsible for conducting the campaign and data collection. VJ carried out the scientific analysis of the data and drafted the manuscript. TVP carried out the review and editing of the manuscript.

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Table 1: Details of aerosol measurements used in the current study

Sl. No.	Measurements	Instrument	Period (2018)	Reference
1	CCN concentrations at different SS	CCN counter (CCN-100, make: DMT)	June 1-8	Roberts and Nenes, (2005)
2	Aerosol NSD from ~15-655 nm	SMPS (LDMA + CPC, make: TSI)	July 8-12, 15-31 August 1-267, 28-31	Wiedensohler, (1988)
3	Aerosol absorption properties at 7 wavelengths	AE33 (make: Magee Scientific)	September 15-30	Drinovec et al., (2015)

Table 2: CCN and aerosol characteristics reported over various locations along with the present results (at 0.3 % SS). CCN reported for (@ 0.4%, # 0.36 SS

Location (coordinates; a.m.s.l)	Type/Condition	Period	CN (cm^{-3})	CCN (cm^{-3})	k	AF (%)	GMD (nm)	Reference
Solapur (17.65° N, 65.9° E, ~480 m)	Continental Monsoon Monsoon Continental	Jun 2018 Jul 2018 Aug 2018 Sep 2018	3427±1064 - 2356±984 4381±1824	1946±594 357±92 322±118 1497±524	0.52±0.11 0.58±0.16 0.67±0.18 0.56±0.16	0.55±0.09 - 0.15±0.06 0.32±0.10	85 ± 10 - 69 ± 11 81 ± 14	Present Study
Mahabubnagar 17°N, 78°E	Continental (Polluted)	Oct 2011	-	~5400 at 1 % SS	~0.45	~0.9	-	Varghese et al., (2016)
Ponmudi (8.8°N, 77.1°E; ~960 m)	Western Ghats Monsoon	Jul-Sep, 2016	~2000	~400	0.65±0.28	~0.20	-	Jayachandran et al., (2018)
Mahabaleshwar (17.56°N, 73.4°E; 1348 m)	Western Ghats/ Pre-monsoon Monsoon	Mar-May Jun-Aug, 2012	~3100 ~3200	~1200 ~500 at 0.2 % SS	~0.5 ~1	~0.35 ~0.35	~90 ~77	Leena et al., (2016)
Thumba[@] (8.5°N, 76.9°E; 3 m)	Coastal Monsoon	Aug-Sep, 2013	~4900	2096 ± 834	0.54 ± 0.21	0.46 ± 0.15	~103	Jayachandran et al., (2017)
Nainital (29.2°N, 79.3°E; 1960 m)	Central Himalayas	Jun 2011 July 2011 Aug 2011 Sep 2011	2425±1112 1874±776 1606±453 2304±904	925±601 881±500 684±396 1233±677	0.57±0.11 0.45±0.08 0.45±0.04 0.39±0.03	0.38±0.11 0.47±0.11 0.42±0.18 0.54±0.12	-	Dumka et al., (2015)
Darjeeling (27.02°N, 88.25°E; 2200 m)	Eastern Himalayas	Mar-May, 2016	7220 ± 1988	~1600	0.38 ± 0.05	~0.25	-	Roy et al., (2017)
Kanpur (26.5°N, 80.3°E; 142 m)	Urban/Polluted	May-Jun (dry) Aug (wet)	~7110 ~6450	~4570 ~2360	-	~0.64 ~0.36	-	Bhattu and Tripathy, 2014

Korea (37.6 ºN, 127.04° E)	Urban/Polluted	May-Jun, 2016	10825 ± 4863	3105 ± 1521	-	-	44 ± 14	Kim et al., (2018)
Guangzhou, (23.07°N, 113.21°E)	Clean Polluted	Summer	8246±3595 7193±3775	3017±1450 2883±1158	-	0.39±0.12 0.45±0.13	-	Duan et al., (2017)
Colarado[#] (38.64° N, 105.11° W, 2300 m)	Forest	Jun Jul Aug	~1400 ~1800 ~1250	~500	-	~0.30	~68 ~80 ~90	Levin et al., (2012)
Amazon 2.13° S, 59° W; 130 m	Forest Dry Wet	Aug-Nov Feb-May	1520±780 330±130	~1469 ~289 at 1% SS	0.36±0.06 0.57±0.03	-	-	Pohlker et al., (2016; 2018)

Table 3: The linear fit parameters between BC mass and Aitken/accumulation mode particle concentration under different loading and air mass conditions

Period	loading	BC – Aitken particles			BC – Accumulation particles		
		m	C	R^2	m	C	R^2
Continental-1June	low	0.3943 ± 0.183	1082224 ± 302241	0.2309	0.40 ± 0.042.48 ± 0.14	364 ± 72-804 ± 165	0.710.74
	high	0.24 ± 0.11	1194 ± 385	0.43	0.22 ± 0.03	815 ± 109	0.83
Marine-2August	low	0.6841 ± 0.053	477503 ± 4935	0.5328	0.291.32 ± 0.027	356133 ± 1648	0.6238
	high	-	-	-	-	-	-
Continental-2September	low	0.5863 ± 0.095	1025446 ± 14114	0.4037	0.63 ± 0.051.02 ± 0.06	657 ± 82-9 ± 111	0.630.53
	high	0.63 ± 0.14	889± 444	0.57	0.33 ± 0.11	1398 ± 375	0.40