Response to Reviewer #1

[Reviewer Comment; RC1] This manuscript presents measurements from a 5 month study of fluorescing biological aerosol particles (FBAP) at an elevated, moderately remote site in southern India. The data quality appears to be very good and the scientific significance is potentially high. The problem is that the paper is probably twice as long as it needs to be and the conclusions drawn are much too speculative with not enough concrete statistics to support the hypotheses put forth regarding either the source of the FBAPs or the physical mechanisms underlying the observed trends. When discussing the results, the objective is to highlight the main points without trying to describe each and every peak and wiggle. Let the Tables list all the statistical details and focus on the most important variations that will then be used to drive the discussion. At the moment there are probably twice as many pages of results and twice as many figures as needed.

[Author Response; AR1] We would like to thank Dr. Baumgartner for his positive evaluation of our results mentioning, "The data quality appears to be very good and the scientific significance is potentially high". The comments provided by the reviewer and corrections suggested in the manuscript have greatly helped us to improve the quality of the manuscript. The suggestions have been meticulously implemented wherever appropriate and we believe changes will be acceptable. As suggested by the Reviewer manuscript is shortened by cutting short the description on of monthly plots and supplementary figures and also by rearranging the text wherever appropriate. We have also revised the figures by combining few of them and moving one to the supplement. The description on size distribution is also shortened and modified accordingly, to highlight the main points.

What is missing:

[RC2]A topographical map of the research site and surrounding area, preferably something like a Google Earth rendition that would show the surrounding vegetation as well as areas with no vegetation such as is referred to in the text. Given the very low wind speeds, it is likely that most of the trends that are seen can be linked to more local sources.

[AR2] We have now provided a land-use map of the southern India in the supplement clearly indicating the significance of the chosen site. As for the topography, note that Fig. 1 in the main text is scaled by the altitude. Relation between low wind speed and local source is explained in the manuscript (L726 – L730)

[RC3] A more in depth analysis of the periods with rain, taking a much closer look at the properties of the FBAP just before, during and after each event.

[AR3] We have performed the in-depth analysis exploring the relation between rainfall and FBAP number concentration just before, during, and after rainfall and the result indicated no significant effect on FBAP number concentration.

Hence, it was very difficult to make any sound and conclusive statement pertaining to effect of rainfall on FBAP number concentration. One reason, as pointed out by Reviewer#3, could be the persistent and heavy rainfall. However, during the second part of the campaign carried out during winter season, we did observe impact of rainfall on the FBAP number concentrations. These results will be discussed in detail in follow up study. In addition we have given a hypothesis explaining the absence of effect of rainfall on FBAP number concentration at this particular location (L850 – L863).

[RC4] A cross-correlational analysis between the FPAB properties and the meteorological conditions. One would not necessarily expect a one to one correlation between the meteorology and the FPAB concentrations, particularly if the spore production is local. Only for advected aerosols would they be linked one to one.

[AR4] We agree with Reviewer that one cannot expect one to one correlation between meteorology and FBAP concentration, however, it may not be true in all the cases and not necessary be always linked for advected bioaerosols. For example, FBAP concentration, depending up on the type of spore, is known to have varied response/relationship with relative humidity (RH). The emissions of few specific types of spores emitted locally are positively related with relative humidity whereas some have shown decrease in concentration with increasing RH (Oliveira et al., 2010; Herrero et al., 1996; Kurkela, 1997; Oh et al., 1998; Healy et al., 2014., Quintero et al., 2010; Hjelmroos, 1993; Ho et al., 2005., Sabariego et al., 2000., Calderon et al., 1995). This has been clearly mentioned in the manuscript (L764 – L774). The relation of wind speed and direction may be useful to further study the impact of advected bioaerosols, which has been detailed in the manuscript (L731 – L738; Hameed et al., 2012; Almaguer et al., 2013; Lyon et al., 1984; Quintero et al., 2010; Huffman et al., 2012; Jones and Harrison, 2004; Troutt and Levetin, 2001; Kurkela, 1997).

[RC5] It is impossible to compare the shapes of size distributions when they are on different figures. Since the concentrations are quite different, normalize them to a unit area then put only the means or medians on the same figure so that any differences in the shapes can be clearly seen. These differences can then be evaluated quantitatively and linked to the hypothesis.

[AR5] Based on Reviewer's suggestion we attempted to prepare a figure comparing the mean FBAP number and mass size distribution for each focus period in single panel. We have replaced Fig. 10 and 11 (original manuscript) with the new figure (Fig.8, revised manuscript). We have also added the prepared figure below for Reviewer's kind read-through.

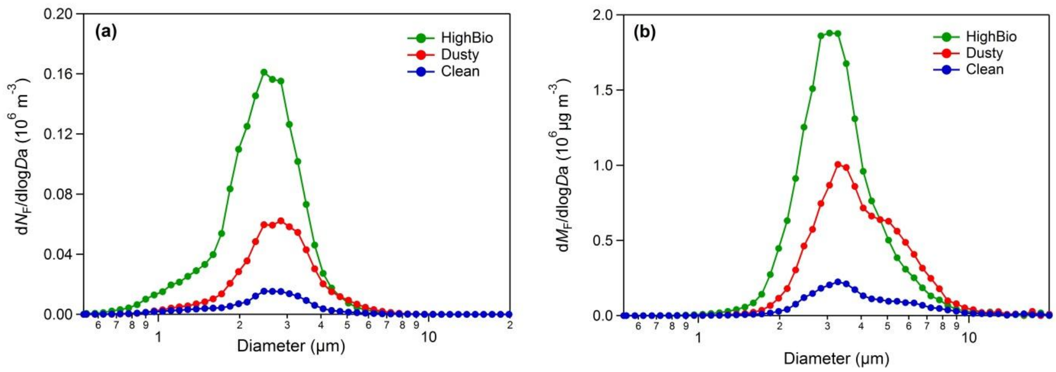

Figure R1.1: *FBAP number size distributions (dN_F/dlog D_a) and mass size distribution (dM_F/dlog D_a) averaged over each distinct focus periods during the measurement campaign carried out at Munnar.*

[RC6] The back trajectory analysis is useless as currently given. There needs to be an analysis of not only where the air came from horizontally, but vertically as well. Questions to be asked: a) How long had the air been close to the surface before reaching the site? b) What were the meteorological conditions along the trajectory, i.e. precipitation and humidity and c) How long had the air been close to the site? These all will impact the history of the particle as well as the air and whether particles had been removed after leaving their source.

[AR6] Indeed, the back trajectory plots shown in Figure 1 are color-scaled by pressure (altitude) to highlight the vertical component of the air motion. We chose not to discuss this in great detail in the paper, because it would make the already long manuscript even longer and we felt this analysis would be beyond the scope of the manuscript. We also felt that it would not be helpful in improving the primary scientific conclusion specifically that during the monsoon season over Indian region, and could lead to further confusion. Further, our sole aim with this figure is to show the back trajectory is only to provide an orientation to reader that this site experiences very contrasting winds during contrasting seasons (monsoon vs. winter), and a broader picture of air mass origin with rough estimates about air mass composition (Vinoj and Satheesh, 2003; Li and Ramanathan, 2002; Satheesh and Srinivasan, 2002; Vinoj et al., 2010, 2014; Prospero, 1979; Moorthy et al, 1991.). To investigate the relation of impact of wind direction and speed separate meteorological measurement in high time resolution were performed and were reported. In any case, we believe that Referee has overlooked the fact that back trajectories are already color scaled by pressure.

[RC7] An error analysis of the measurements. What is the expected error in size and concentration based on error propagation that no doubt has already be detailed in earlier publications. Nothing is said about FBAPs that are not complete particles but are fragments. There needs to be an estimate of the size dependent losses in the sampling system. Even with no bends, there will be diffusion and turbulence losses that can easily be calculated with Aerocalc (Baron and Willeke).

[AR7] Reviewer may be aware of the fact that UV-APS cannot differentiate the particle type; instead it only measures the aerodynamic diameter and spectrally integrated fluorescence. Under this scenario, a plant/animal fragment sampled will be treated as a complete particle where instrument will assign the equivalent aerodynamic diameter. We have calculated the equivalent aerodynamic diameter of a non-spherical particle. For example an ellipsoid particle with length of 4 μm long and width of 2 μm will be treated as particle with aerodynamic diameter of 3.6 μm. As suggested by Reviewer a sentence (average penetration efficiency of 99.8% at 290K and 840 hPa) discussing about the size dependent losses in the sampling system is now added based on Aerocalc/previous literature.

I have attached an annotaed version of the paper that includes many more questions and comments, The paper is relatively readable, given that the first author is not a native English; however, I am annoyed whenever I read a paper written by a non-English speaker but who has co-authors that are but who obviously have not read the paper, otherwise the numerous grammatical errors would have been corrected.

The suggestions provided in the annotated version of the manuscript have been implemented wherever appropriate and we thank Reviewer for his valuable edits.

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Response to Reviewer #2

[Reviewer Comment; RC1] The manuscript presents measurements of Fluorescent Biological Aerosol Particles (FBAP) made using an Ultraviolet Aerodynamic Particle Sizer (UV-APS) at a high-altitude tropical site in southern India over a period covering the southwest monsoon season. A thorough background research, set of measurements and description of these has been presented and is potentially worthy of publication. However, I do feel some improvements could be made to the current manuscript as detailed below. Although I'm sure any new field measurements using a UVAPS or similar qualify as valid research, it is difficult to see the real contribution of this work to the field, other than to validate previous similar measurements at a new location. I believe the information is there, but is swamped in meticulous depiction and description of all measurements. Perhaps the manuscript could be streamlined and given more structure to emphasize this. The properties of the new site and results specific to this should be highlighted, with an eye on how it will be useful for future research and how the presented measurements will support that research. The technical content of the paper appears good and accurate. Figures have been chosen well to depict and compare the measurements (haven't seen the supplementary figures, which do not appear to be with the manuscript). Work in the field I was aware of, and much more besides has been appropriately sited.

[AR1] We would like to thank Reviewer for positively evaluating our manuscript stating following important points:

- "A thorough background research, set of measurements and description of these has been presented and is potentially worthy of publication"
- "The technical content of the paper appears good and accurate"
- "Figures have been chosen well to depict and compare the measurements"
- "Work in the field I was aware of, and much more besides has been appropriately sited"

The reviews provided here have been very helpful for us in improving the overall quality of the manuscript. We have also tried to fine-tune the key messages highlighting the findings of this study and how it would be useful in future in conclusion section. In addition to reduce the length of the manuscript we have now moved few figures to supplementary material and merged Fig. 3 and Fig. 5.

Some suggested alterations below.

[RC2] Pg 10: The inlet system is described here. Could the effects of the inlet system be assessed by a comparison of measurements of a range of particle sizes under controlled conditions both with and without the inlet tubing in place?

[AR2] Following the experiment, we performed a test with room air to estimate particle losses in the inlet tube. Observing particle distributions with and without tubing showed that no noticeable difference in the particle number concentration or size distribution. We also performed theoretical calculations to calculate the diffusion losses during the entire sampling for all the size-ranges sampled and found that average penetration was 99.8% at 290K and 849 hPa.

[RC3] Line 445: Should read "Basically the UV-APS measures the particle number and aerodynamic size; the average mass of size-resolved particles can" Lines 445-447: Can this statement be backed-up by a reference?

[AR2] We have now added an appropriate reference for this statement.

[RC3] Line 457 and few lines preceding: Were all the compared measurements made using the same density value?

[AR3] As far as we could found out from the literature all the measurements were carried out using same density values (1 g cm^{-3}).

[RC4] Lines 709-716: Description is confusing; maybe a mistake in the period references in the figure has been made? Here and everywhere else the three named focus periods are mentioned, the period names should perhaps be written within inverted commas or in italics to make reading easier.

[AR4] We thank Reviewer for pointing this out. The *focus* periods are now written in italics throughout manuscript.

[RC5] Figure 2: The shadowed blocks representing the focus periods don't seem to be visible on my copy of the manuscript.

[AR5] This mistake has been corrected. We have now added the shadowed blocks in Fig. 2.

[RC6] Finally grammar details. While the document is largely well written, there are numerous grammar mistakes that, at times, make it quite difficult to read. These often consist of a missing 'a' or 'the'. Although too numerous for individual attention, I have indicated some instances below and suggest that one of the authors go through the whole manuscript again and tidy this up.

[AR6] We thank Reviewer for pointing this out. Once we receive the final acceptance from The Editor before the final publication we intend to get our manuscript English edited from a professional service.

[RC6.1] Line 27: Missing 'a' between 'constitute' and 'large'.

[AR6.1] Done

[RC6.2] Line 55: Should read "selected areas. These measurement results confirm the fact that the fraction of PBAP to TAP is".

[AR6.2] Done

[RC6.3] Line 56: Missing ’a’ between ’constitute’ and ’significant’.

[AR6.3] Done

[RC 6.4] Line 172: Missing ’the’ between ’including’ and ’Arabian’.

[AR6.4] Done

[RC6.5] Line 173: Should read, "movement of the ITCZ reaching up to the equator is associated with the NE monsoon, which is also marked".

[AR6.5] Done

Response to Reviewer #3

General

[Reviewer Comment; RC1] This paper reports results of a three month measurement campaign of fluorescent biological aerosol particles (FBAP) at a high altitude tropical site in southern India. There are some unique aspects on the data. First, the marine air masses can be compared to local FBAP sources. Secondly, the campaign included long periods of heavy and persistent rain. Consequently, the authors observed a lack of correlation of FBAP with precipitation, contrary to several recent studies from other areas. The data has been presented in diverse ways that are also comparable to earlier studies. The material seems to merit publication. However, there are several issues that should be treated.

[Authors Response; AR1] We would like to thank reviewer for his thoughtful and detailed comments, which have helped us in improving the quality of the manuscript. We also thank reviewer for making following positive observations about our work.

- "There are some unique aspects on the data"
- "First, the marine air masses can be compared to local FBAP sources. Secondly, the campaign included long periods of heavy and persistent rain"
- "The data has been presented in diverse ways that are also comparable to earlier studies"

[RC2] Most importantly, the paper is unnecessarily long. The authors should concentrate on the findings that are unique to this study. Although it is good to treat many facets of the data, I think the paper would benefit of focusing also in this respect. The authors have divided the three month period first into months and later to three focus periods that they call dusty, clean, and high bio. I find the latter division much more useful. I recommend keeping it and getting rid of the monthly results. Because the data is treated from many angles, many of the explaining factors and arguments are presented several times. An example is the effect of the clean SW winds. It would be good to try to collect the findings first and then treat them at once.

[AR2] We thank Reviewer for pointing this out and we have now implemented Reviewer's suggestions. Accordingly, we have reduced the length of manuscript by moving figures related to monthly division of the observed data to the supplement and corresponding description has also been reduced substantially.

Specific

[RC3] In subsection 3.5 on SEM images the authors state that "these images are not being presented here for any quantitative purpose and to draw any specific scientific conclusions". Indeed, there are only a few particles shown. However, the authors use the images to support their hypotheses on the particle species. I

propose either analyzing a large number of samples and particles to corroborate the hypotheses or moving the subsection to the supplement and being cautious on using them as evidence.
[AR3] We understand the reviewer's concern. What we meant was not to draw any scientific conclusions using the SEM images regarding neither the variabilities in bioaerosol number/mass concentration nor the type of bioaerosols. Our sole intention was to orient the reader to the dominant particle types in an air mass during three distinct focus periods. Note that we have investigated more than 100 individual particles randomly collected on five ocassions and have shown few as exemplary images. This has now been clarified in the revised manuscript text (L637 – L639).

[RC4] The measurements have been done with the UV-APS. Regarding the data interpretation, it would be good to acknowledge and point out that the detection efficiency for fluorescent particles of the UV-APS is low especially below approximately 2 microns (e.g. Healy, at al., ACP2014, Saari et al., AST 2014). This mostly affects the reported fluorescent particle size distributions. Further, it would be good to state whether a zero check cycle for the instrument was used.

[AR4] As pointed out by the Reviewer few studies have reported that counting efficiency of UV-APS decreases below 2μm. Considering this limitation associated with the instrument we report the number concentrations of FBAPs measured using UV-APS as the lower limit proxy for bioaerosols present in the atmosphere. A line (L200) is now added in the manuscript to point out this limitation in the detection efficiency. We have performed the zero tests using HEPA filter and found no particle was detected during zero test.

[RC5] Line 250 to 273: The authors find that the fluorescent and total particle concentrations do not correlate much, independent of whether the particles are coarse or fine. They then argue that the fluorescent concentration is not affected by non-biological particles. They later hypothesize that particles from combustion or similar activities do not get transported to the measurement site. This might well be the case, but what are the high concentrations of fine non-fluorescent particles (figure S2) then? Maybe a scatter plot of NF (<1um) VS NF(>1 um) would be useful. I would expect the submicron biological particles to correlate with the supermicron biological ones somewhat. Maybe use this point for shortening the paper and just state that there seldom are major sources of fluorescent non-biological coarse particles and therefore the numbers reported are relevant. I hypothesize that the lack of correlation for fine particles is at least partly caused by the low fluorescent particle detection efficiency of the UV-APS unit.

[AR5] We understand Reviewers concern. Our intention of carrying out the correlation analysis was to indicate that under certain conditions (mainly clean conditions persisting during monsoon at this site) UV-APS may be used to detect and segregate the particles of biological origin in sub-micron range from non-biological aerosol particles. However, interference from non-biological but fluorescent particles needs to be carefully analyzed. Further, as pointed out by

Reviewer there may be a high number concentration of the particles in sub-micron range, which are non-biological and non-fluorescent (as in our case); these particle do not likely affect the number concentration of fluorescent aerosol particles. In our case we believe, as supported by TAP number size distribution and SEM images, the non-biological and non-fluorescent particles in submicron range were mostly dominated by sea-salt and mineral dust. As suggested by Reviewer we have prepared the scatter plot of N_F for particle size range of <1 µm and >1 µm and is shown below. We infer that the particles in submicron size range, which are of not likely of biological origin are not introducing any interference in the fluorescence.

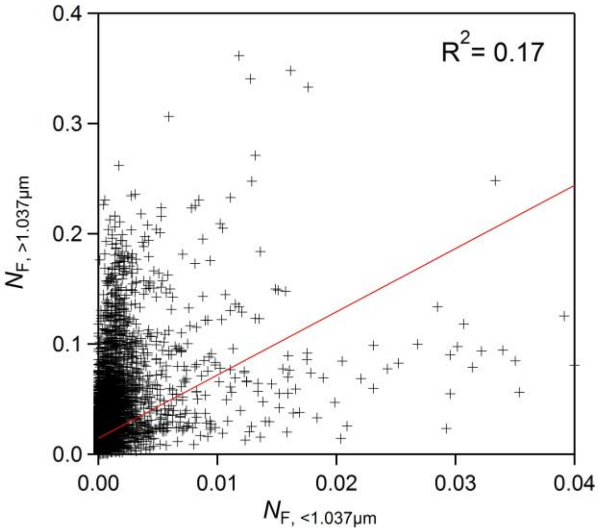

Figure R3.1: *Scatter plot between N_F concentration integrated for particle size range of <1 µm and >1 µm.*

[RC6] Line 357-360: The high extreme values of NF/NT actually do result from high variations of NF, as evident from fig S2. The presented figures do not support the argued inverse correlation between NT and NF/NT. The argument should be backed with a figure or a calculation or removed.

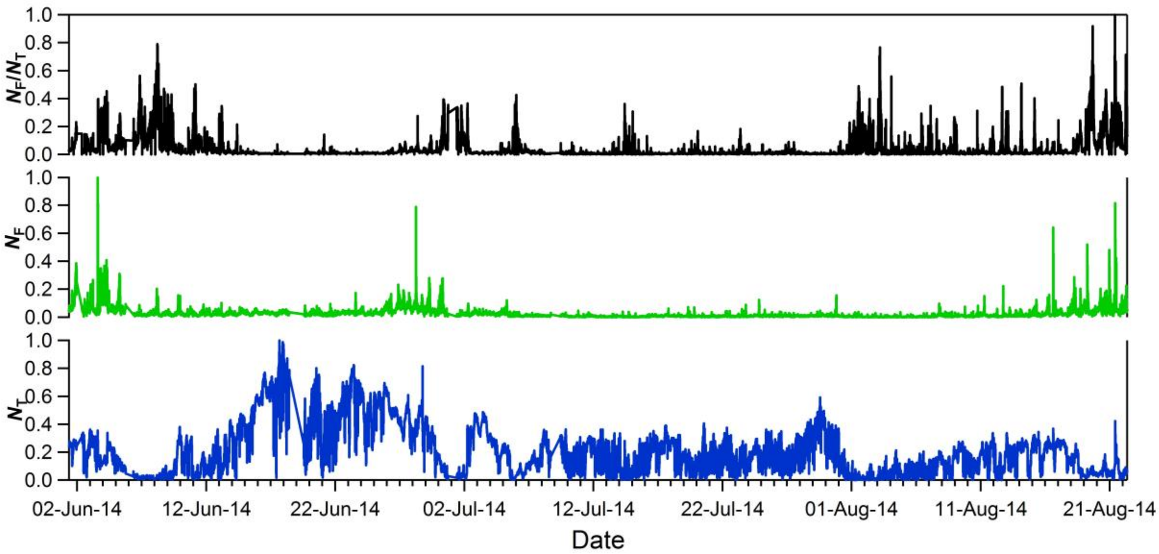

Figure R3.2: *Variability in N_T, N_F and N_F/N_T averaged over the course of entire campaign.*

[AR6] We are unable to understand what Reviewer means to point out. Note that we have mentioned that "The time series of relative contribution of FBAP to TAP number during the campaign overall exhibited the similar temporal variability to N_F. However, pronounced extreme values of N_F / N_T, on the other hand, resulted from strong variability in the concentration of N_T. Please refer to Figure R3.2 above, where it clearly shows the high variability in N_T (lowest panel) is much pronounced and higher during the entire measurement period. However, the corresponding variability in N_F much less pronounced and episodic. Hence, resulting variability in N_F / N_T is to an extent shows similar temporal variability as like N_F but clearly shows the inverse relation with N_T. We have mentioned the equation to derive the variability in the manuscript [L768 - L770]

[RC7] Line 703-729: The NF axis in fig 13 is such that it is very difficult to spot small changes in concentration. However, the slightly higher concentration starts at 17.00, not at 20.00. Apparently this data does not support the nocturnal sporulation argument. The argument on humidity is later supported by the scatter plot, but it might be good to show the diurnal pattern of RH here also.

[AR7] We thank Reviewer for pointing this out, which resulted from wrong choice of scaling. This error has now been corrected in the revised version. We have also removed the sentence mentioning the nocturnal sporulation of spores to avoid confusions. As suggested by Reviewer we have now added the diurnal variations in RH, temperature, and wind speed in the revised figures. We hope that changes will be acceptable to the Reviewer.

Technical

[RC8] Line 75: The first paragraph of introduction is rather long. It would be good to separate the latter part into a new paragraph, starting on line 75 from " It is likely that the surface". This latter part should also reformulated, as it is very difficult to follow the line of thought now:

***[AR8]* Done.**

[RC9] "surface structure, ice nucleating proteins, and other characteristics" – characteristics of what? Of PBAP, bacteria, bacterial spores?

[AR9] We meant to mention that surface structure, ice nucleating proteins, and other characteristics of bioaerosols can influence heterogeneous ice formation. The sentence has been modified accordingly.

[RC10] "Other bioaerosols like pollen" Other than what?
"..Other bioaerosols like pollen and fungal spores are often using air as the transportmedium.". By definition, all atmospheric aerosol particles use air as the transport medium. Maybe: Plants and fungi use the air as a transport medium for their pollenand spores

[AR10] We agree to the Reviewer and have modified the sentence as per his/her suggestion.

[RC11] "Play an important role in public health.." It would be good to convey the idea that the role is negative.

[AR11] As per Reviewers suggestion the sentence has been modified accordingly.

[RC12] Line 123: The authors should either explain the relevance of the present study to Indian agriculture (or vice versa) or remove the sentence.

[AR12] The sentence has been modified to emphasis the importance of present study to Indian agriculture.

[RC13] Lines 192: The description of the drop of the detection efficiency is tautological with the text starting on line222.

[AR13] We thank Reviewer for indicating this. To avoid repeatability we have removed this statement from line L192.

[RC14] Line 195 on: For particles in the size range of 15-20 microns, the aspiration and transport efficiency of the sampling system probably is a more important issue than the calibration of the APS.

[AR14] This has been removed to avoid further confusion.

[RC15] Line 225: It would be good to state that the 1 micron as the fine particle size limit is adhoc.

[AR15] The sentence has been modified accordingly.

[RC16] Line 316-318: This is plausible, but should be written so that it is clear that there is no direct evidence within the present study.

[AR16] The sentence has been modified accordingly.

[RC17] Line 476-478: This is not due to the calculation. The mode mass will peak at higher diameter than number for any atmospheric particles one could find.

[AR17] To avoid any confusion, we have now removed this sentence.

[RC18] Line 484-486: Although the size limit is the same, the authors should warn the readers that the mode largerly absent in NT as submicron is present in MT as supermicron.

[AR18] The reason behind such a shift from a mode in N_T in submicron range to a mode in M_T in supermicron range is clearly explained in revised manuscript (L422 – L423).

[RC19] Lines 496, 659: Note that the downwards slope of the APS detection efficiency might cause a peak to appear at around 0.9 um even when the mode would actually peak at much lower particle diameter.

[AR19] The actual peak in the size distribution of TAP would generally occur at lower diameters (<0.5 µm). However in case of marine aerosols a secondary peak at diameter <1 µm was also reported which was mostly contributed by the sea salt particles. In the present study since the air masses were of marine origin we believe it's important to report this peak.

[RC20] Line 536: Should this be figure 14?

[AR20] This description was with reference to the figures reported in the paper Valsan et al., 2015.

[RC21] Line 591 and elsewhere: The date format should be homogenized between the text and figures

[AR21] The date format has been homogenized.

[RC22] Line 625: "As expected, the NF was highest during the high bio period.."! This should be reformulated as the period was specifically chosen to be high bio.

[AR22] The sentence has been modified accordingly.

[RC23] Line 669 on: The medians in distributions do not make much sense for channels that exhibit a high number of zero values.

[AR23] For the completeness and consistency of the proper representation of figures we Request reviewer to retain us the same format of the figure.

[RC24] Line 770 on: The pollution/concentration rose figures are hard to read and difficult to use in backing up quantitative arguments. The interpretation instruction text in the caption of fig 15 is not helpful and the numbers seem to be wrong. Overall, it would be helpful if someone came up with a better way of displaying the correlation of measured quantities and wind. Why not start with this MS?

[AR24] We thank Reviewer for pointing this out. The caption of Fig. 15 (now Fig. 12) has been corrected accordingly and the scales shown in the wind rose diagrams were also revised for better understanding.

[RC25] Figures 10-12: If the median values are shown, it would be good to continue to show the mean/median legends on the figures.

[AR25] As indicated by the Reviewer, the mean/median legends were added on the appropriate figures.

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Fluorescent Biological Aerosol Particle Measurements at a Tropical High Altitude Site in Southern India during Southwest Monsoon Season

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Abstract

~~Primary Biological Aerosol Particles (PBAPs) like fungal spores, bacteria, pollen, etc. are reported to constitute large fraction of the atmospheric aerosols. They are responsible~~An ultraviolet aerodynamic particle sizer (UV-APS) was continuously operated for the ~~spread of organisms~~ first time during two seasons to sample the contrasting winds during monsoon and ~~diseases throughout~~winter to characterize the ~~biosphere and may impact atmospheric processes and the hydrological cycle by acting as ice nuclei (IN) and giant cloud condensation nuclei (CCN). Despite their importance in the biosphere and climate, continuous measurements of PBAPs in high time and size resolutions are not available for the Indian subcontinent. Here we report the first measurements~~properties of fluorescent biological aerosol particles (FBAPs~~) in India. The measurements were carried out using an ultraviolet aerodynamic particle sizer (UV-APS) in Munnar,~~), at a high altitude ~~tropical~~ site in ~~southern India. The study was conducted for three consecutive months during the Southwest monsoon season (1.June.2014 – 21.August.2014), which is marked by heavy and persistent rainfall and strong Westerly/Southwesterly clean winds.~~

India. Averaged over the entire monsoon campaign (June 1, 2014 – August 21, 2014) the arithmetic mean number and mass concentrations of coarse-mode ~~FBAP (>~~ (>1 µm) FBAP were 0.02 cm^{-3} and 0.24 µg m^{-3}, respectively, which corresponded to ~2 and 6 % of total aerosol loading, respectively. Average FBAP number size distribution exhibited a peak at ~3 µm, which ~~was most likely contributed by~~is attributed to the fungal spores, as supported by scanning electron microscope (SEM) images, and ~~the~~these results are consistent with previous studies made for FBAP. During eleven weeks of measurements the ~~corresponding~~ variability of the total ~~(TAP)~~ coarse mode particle number (TAP) concentration was ~~highly variable in contrast~~high

compared to ~~the variability~~ that observed in FBAP number concentration.~~ Averaged over the entire campaign the TAP number and mass concentrations were 1.8 cm^{-3} and 7.0 µg m^{-3}.~~ The TAP and FBAP number concentrations measured at this site were strongly dependent on changes in wind direction and rainfall. During ~~the period~~periods of ~~continuous and~~westerly/southwesterly winds with heavy persistent ~~rainfalls~~rainfall, the TAP and FBAP concentration exhibited very low ~~concentration levels~~values (1.3 cm^{-3} and 0.005 cm^{-3}, respectively) with no ~~observed~~significant diurnal variations. ~~Averaged over the entire campaign~~Whereas during periods of Northerly winds with scattered rainfall FBAP exhibited ~~a moderately~~relatively high concentration values (0.05 cm^{-3}) with pronounced diurnal ~~variation with highest concentration during early morning hours (~06:00 – 08:00 hrs).~~variations, which were strongly coupled with diurnal variations in meteorological parameters. The campaign averaged FBAP number concentrations were shown to correlate with daily patterns of meteorological parameters and were positively correlated with relative humidity (RH; R^2=0.58), and negatively with temperature (R^2=0.60) and wind speed (R^2=0.60). We did not observe any significant positive correlation with precipitation as reported by previous researchers from selected areas. These measurement results ~~confirms~~confirm the fact that ~~fraction~~the ratio of PBAPs to TAP is strongly dependent on particle size and location and thus may constitute a significant proportion of total aerosol particles.

1 Introduction

Aerosols are generally defined as a colloidal system of solid or liquid particles suspended in a gaseous medium (Fuzzi et al., 1997; Pöschl, 2005) and are ubiquitous in the Earth's atmosphere. The term "Primary Biological Aerosol Particles" (PBAPs; sometimes also referred as bioaerosols or biological aerosols), describes a subset of ~~aerosol particles, i.e. the~~ solid airborne particles originating from biological organisms, including viruses, pollen, microorganisms (bacteria, fungal spores, etc.) and, protozoa or algae, etc., together with fragments of biological materials such as animal dander, plant debris etc. (Artaxo and Hansson, 1995; Coz et al., 2010; Després et al., 2007, 2012; Elbert et al., 2007). Bioaerosols can range in size from a few nanometers to few hundred micrometers in aerodynamic diameter, D_a, (Coz et al., 2010; Després et al., 2012; Jones and Harrison, 2004; Matthias-Maser and Jaenicke, 1994)~~ with viruses being the smallest in size amongst the PBAPs followed by bacterial and fungal spores, while pollen, and plant and animal fragments represent the largest in size. Depending upon size and ecosystem PBAPs can~~. PBAPs have been shown to constitute 14 – 70% of total number of coarse mode particles and around 20 – 24 % of total mass of PM_{10} (particulate matter with size ≤10 μm; Elbert et al., 2007; Després et al., 2012; Pöschl et al., 2010; Huffman et al., 2012). ~~Bioaerosols are present in the ambient atmosphere either as a single particle, or as agglomerates (Valsan et al., 2015) and exhibit a variety of shapes and morphological characteristics.~~ Further, it is likely that the surface structure, ice nucleating proteins, and other characteristics of bioaerosols can influence ~~substantially~~ the heterogeneous ice nuclei formation at ~~various~~relatively high temperature levels (Morris et al., 2004, 2014)~~ and they~~. Bioaerosols can ~~also~~ thus act as giant cloud condensation nuclei (GCCN~~)~~), thus affecting the hydrological cycle (Andreae and Rosenfeld, 2008; Möhler et al., 2007). ~~Other bioaerosols like~~It is also known that pollen ~~or fungal~~ and spores associated with various plants

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and fungi are ~~often using~~dispersed in air ~~as~~resulting in the ~~transport medium for~~ distribution and transfer of genetic material ~~and thus can travel and get transported~~ over large distances (Huffman et al., 2010; Elbert et al., 2007; Hallar et al., 2011; Burrows et al., 2009). A side effect of such ~~a~~ transport and distribution~~, however,~~ is that they ~~are produced and spread in large quantities and~~ can play ~~an important~~a negative role in public health ~~as they can cause allergies.~~. Pathogenic fungi have long been recognized as major threats to animal health and plants including crops severely jeopardizing the food security (Fisher et al., 2012 and references therein).

~~Since the last century numerous studies have been conducted in different parts of the world to understand the abundance and diversity of bioaerosols using various sampling and measurement techniques, however confining to traditional methods.~~ The last decade has experienced a substantial development and application of advanced online and offline techniques for studying the characteristic properties of bioaerosols in ~~the~~both field and laboratory (Fröhlich-Nowoisky, et al., 2009; DeLeon-Rodriguez et al., 2013; Prenni et al., 2009; Huffman et al., 2010, 2012, 2013; Schumacher et al., 2013; Pöhlker et al., 2012, 2013).

Instruments utilizing laser-induced fluorescence (LIF) have been frequently deployed ~~to~~in the field, enabling real-time characterization of the number size distribution of PBAPs ~~in~~with high time and size resolution. However, instruments based on LIF do not provide detailed information directly about PBAPs or the origin of particles, but ~~rather~~ provide broadly categorized information due to a mixture of biological fluorophores, each detected with varying efficiency (Pohlker et al., 2012, 2013). Most FBAP~~–~~ measurements have shown that the dominant size range for PBAPs number size distribution is 1 – 4 μm with concentration varying within the factor of 10 (Gabey et al., 2011, 2013; Healy et al., 2014; Huffman et al., 2010, 2012, 2013; Saari et al., 2015; Schumacher et al., 2013; Toprak and Schnaiter, 2013; Yu et al., 2016). As

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studied and described by Huffman et al., (2010)), based on long-term PBAPfour months of measurements in central Europe, the signal detected by UV-APS (Ultraviolet Aerodynamic Particle Sizer) in ambient settings was defined as Fluorescent Biological Aerosol Particles (FBAP), and the resulting quantification of FBAP was further discussed and it was concluded that FBAP represents an approximate lower limit of actual abundance of PBAPs present in the ambient air sampled by the UV-APS. Thus, for the consistency and simplicity we use the similar terminology as suggested by Huffman et al., (2010).). Hence, the term FBAP is used as a lower limit proxy for primary biological aerosol particles (PBAPs), biological aerosols, biological aerosol particles, bioaerosols and similar terms mentioned in this study.

Despite such instrumental advancements described above, the studies related to the quantification of bioaerosols and their role in climate and human health have been extremely limited in space and time. Particularly,This is particularly true for the Indian subcontinent, which constitute around ~18% of the world's total population, where studies related to the bioaerosols are relatively few and with spotty analysis performed only by traditional techniques (Bhati and Gaur, 1979; Chakraborty et al., 1998; Gangamma, 2014; Srivastava et al., 2012; Sharma and Rai, 2008; Pachauri et al., 2013; Valsan et al., 2015; Ansari et al., 2015; Adhikari et al., 2004). Thus, sources,The abundance, and properties of bioaerosols, which areis strongly dependent on location and season, remains poorly characterized over the Indian subcontinent and need to be addressed systematically.

InvestigatingAdditionally, investigating and quantifying the role of bioaerosols over the Indian continent is not only important because of the scarcity in the literature but also due to its diverse land-use pattern and the unique climatic condition experienced by the in terms of two Monsoon monsoon seasons associated with two distinct synoptic scale wind patterns. Indian agriculture is

~~strongly dependent on the Southwest Monsoon, and is the largest livelihood provider in India and contributes a significant figure to the Gross Domestic Product (GDP). Therefore, it is very important to better understand and quantify the role of bioaerosols in cloud and precipitation formation during Monsoon and convective rainfall. The concentrations~~The concentration of fluorescent aerosol particles in a semi-arid forest in the Western US was shown to increase during and after rainfall ~~in a semi-arid forest in the Western US~~ (Huffman et al., 2013~~), but the same pattern was not observed in a similar study in the Amazon basin (~~). Rainfall-triggered increase in bioaerosol concentration can potentially enhance further precipitation by convective upward movement of bioaerosols into clouds where they can serve either as IN or giant CCN (Shcumacher et al., 2013; Huffman et al~~, 2012).~~., 2013). Thus, the bioaerosols emitted during monsoon ~~season~~ rainfall could potentially play an important role in cloud and precipitation formation ~~as shown by~~ over India (Ansari et al~~. (~~., 2015).

Therefore, it is very important to understand and quantify the role of bioaerosols in cloud and precipitation formation during monsoon and convective rainfall. Additionally, bioaerosols over the Indian sub-continent can ~~have a direct societal impact where huge set of population may~~ directly ~~get affected by~~impact society through the spread of diseases and ~~covertly~~indirectly due to ~~the~~increased risk of loss ~~in~~of agricultural output~~.~~ ~~Thus, studies~~ due to emerging diseases caused by the fungi (Fisher et al., 2012).

Studies involving characterization of bioaerosols using advanced techniques over this region are important to understand and quantify the impact of bioaerosols on regional biodiversity with larger implication towards human and ecosystem health. With this motivation we have deployed ~~an~~a UV-APS for the detection and measurement of number size distribution of PBAPs at a high-altitude site of Munnar in Western Ghats of southern tropical India during Southwest monsoon season for ~3 months. To our knowledge this study presents the first ~~multi-month~~ ambient measurement ~~investigations~~investigation involving UV-APS for multiple months over the Indian subcontinent.

2 Methods

2.1 Site Description

Measurements were performed to sample the air masses (see section 2.2) from a high-altitude site (Munnar; 10.09°N, 77.06°E; 1605 m amsl – above mean sea level – Fig. 1) located in the Western Ghats region of Southern, tropical India, just 90 km away ~~as the crow flies~~ from the Arabian Sea ~~in the Southern part of tropical India.~~. The observational site is located on a hill with a valley towards the South and a small mountain towards the North surrounded by dense vegetation including tea gardens and Eucalyptus trees. Climatologically this region is classified as subtropical highland with dry winters and is listed as the Shola forest-grass ecosystem as defined in the land-use type terminology~~.~~ (Fig. S1). The Western Ghats, one of the eight mountain ranges in India and identified as one of the ~~hottest~~most significant hot spots of biodiversity (Myers et al., 2000) in the world, originates near the border of Maharashtra and Gujarat running ~1600 km towards South, parallel to the Western coast through the states of Gujarat, Maharashtra, Karnataka, Kerala, and Tamilnadu ending at the Southern tip of India near ~~Kaniyakumari.~~Kanyakumari. This mountain range separates the coastal plain from the Deccan plateau making Western coastal plain a narrow land strip with a maximum width of ~ 110 – 120 km, sandwiched between the Western Ghats and the Arabian Sea. During the SW Monsoon season (June – September) the Southwesterly, moisture laden winds are intercepted by the Western Ghats causing persistent and heavy rainfall on the windward side of these mountains.

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This causes the wash out and wet deposition of the pollutants in the coastal strip (Kerala) emitted due to anthropogenic activities, thus bringing clean marine influx with minimum impact of anthropogenic emissions (Satheesh and Srinivasan, 2002). Therefore, during this particular season this observational site can be regarded as relatively pristine, as compared to any other operational high-altitude observatory/site in Indian tropical region (Shika et al., 2016).

2.2 General Meteorology

Southern India nominally experiences two Monsoon seasons, the ~~Southwest~~SW monsoon ~~(SW; June – September)~~ and the Northeast monsoon (NE; November – January), which are strongly associated with the movement of Inter-Tropical Convergence Zone, the ITCZ (Kanawade et al., 2014). The SW monsoon winds ~~are dominant during June to September bringing almost anthropogenically "~~bring relatively clean~~" (not affected by human activities)~~ marine influx over the continent from Arabian Sea when the ITCZ moves Northwards reaching 30°N during July (Naja and Lal, 2002). These air masses originate over the Indian Ocean and travel thousands of kilometers over ~~oceanic water~~the ocean, including the Arabian Sea, before reaching the observational site. The Southward movement of ITCZ reaching up to the equator is associated with the NE monsoon, which is also marked as winter season in India occurring during October to January, when the prevailing winds are predominantly blowing in the NE direction. The measurement site of Munnar receives more than 85% of its annual rainfall during SW monsoon season and experiences scattered rainfall events during NE monsoon ~~season~~. The detailed meteorological parameters measured during the field measurement campaign carried out during SW Monsoon season at Munnar are discussed below.

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2.~~2~~3 Real-time fluorescence measurement

~~The biological~~Biological aerosol particles ~~from a high-altitude relatively pristine site~~at Munnar were measured using ~~an~~ a UV-APS (TSI Inc. Model 3314~~; Serial Number: 71331023~~) as per the standard instructions given in the technical manual. The detailed description about the instrument including operating principles, field operation, data analysis protocol, and critical operational parameters are discussed elsewhere (Kanaani, et al., 2007, 2008; Agranovski et al., 2003, 2004, 2005; Brosseau et al., 2000; Huffman et al., 2010, 2012; Hairston et al., 1997).

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Briefly, the instrument is capable of measuring ~~the~~ aerosol particles in the aerodynamic diameter (D_a) range of 0.~~54 – 19.81~~5 – 20 µm over 52 channels by ~~means of~~ measuring the time-of-flight of particles between two He-Ne red lasers (λ=633 nm). Once the particle size is determined, ~~the same~~ each particle is ~~further~~ excited using ~~a third~~an ultraviolet Nd:YAG laser (λ=355 nm) and ~~emissions are~~ fluorescent emission is measured in the range of 420 – 575 nm. The spectrally unresolved total fluorescence is recorded for each individual particle in to one of ~~the~~ 64 channels ~~with increasing order of fluorescence intensity. Huffman et al., (2010) described that the counting efficiency of the instrument drops below 100% at D_a< 0.7 µm (counting efficiency ~50% at 0.54 µm), hence, the particle number concentration values reported for particle sizes of <0.7 µm are lower limit of the actual concentration of the air sample. During analysis presented in this paper the particles detected in the size range of 15 – 20 µm were included and the reported number concentration values should be considered as the lower limit of the actual values present in the air sample, due to limitations in the size calibration for particles of this size.~~. The UV-APS ~~measurement cycle was initiated with~~ measurements were obtained at 5 minutes interval (~~including the full diameter range scan for 285 seconds and 15 seconds of back-scanning~~ recording a total of 22280 sampling points during entire measurement campaign) ~~where air~~

~~sample was drawn~~ with a volumetric flow rate of 5 ~~L min^{-1} (lpm)~~Lpm at ambient temperature and pressure. All ~~the~~ times reported in this study ~~are local time pertaining to~~ were Indian Standard Time (IST~~; GMT+5:30~~).

~~The UV-APS~~Sampling was ~~placed next to the window inside~~performed at a ~~room in~~building of the College of Engineering, Munnar, Kerala~~ located on a hill. A stain-less steel tubing with ¾" OD (outer diameter) and TSP~~. The sampling inlet was ~~used to construct the inlet unit for air sampling, which was ~9 m and ~~~approximately 2 m above the rooftop of the building and 8 m above the ground ~~and rooftop, respectively. Thus the sampled air masses were expected to have minimal influence caused by the dynamics associated with the building structure.~~level. The sampling inlet was connected to the UV-APS, which was placed next to the window inside a room using 3m of ¾" OD stainless steel tubing. To minimize the particle losses, due to impaction resulting from sharp bends, ~~the~~ electrically conductive silicon rubber tubing (~1.5 m; 12 mm inner diameter) was attached to the ~~stain-less~~stainless steel tube just outside the window (Fig. S1).~~S1) avoiding the sharp bends. Before the sampled air~~ The air sample was passed ~~to the instrument,~~through a diffusion dryer (~1 m length) with silica gel ~~(orange color indicating) was used to dry and maintain~~before entering the UV-APS, thus maintaining the relative humidity of inlet air to <40%.~~ Thus combining all the tubing involved in the air sampling the sample flow~~ The residence time of sampled air in the inlet tube was calculated to be ~ 20 seconds~~. The sample~~, and the flow ~~through all the tubing~~ was ~~expected~~calculated to be laminar ~~during~~ in the entire sampling ~~period and hence~~line. Hence, diffusion losses ~~are~~were expected to be negligible for all the size-ranges of the sampled particles~~.~~ (average penetration efficiency of 99.8% at 290K and 840 hPa; Baron and Willeke, 2005).

For the present study ~~we derived~~the number size distribution of fluorescence biological aerosol particles~~, ~~ (dN_F/dlog D_a~~,~~), for each size bin was derived by summing up the particle number concentration from the fluorescence channel numbers 3 – 64 and similarly the total particle number size distribution~~, ~~ (dN_T/dlog D_a~~,~~), was derived from channel numbers 1 – 64. In the present study we have used 1.0 µm as a cut-off diameter for given dN_F/dlog D_a and dN_T/dlog D_a to calculate the fluorescence biological aerosol number and total aerosol number concentrations, N_F and N_T, respectively. This is mainly due to the fact that particle counting efficiency of the UV-APS drops below unity at 0.7 µm (counting efficiency ~50% at 0.54 µm) and the interferences~~ due to fluorescence~~ from non-biological aerosol particles below 1.0 µm can at times be very high (Huffman et al., ~~2010). Also note that the cutoff at 1 µm moreover represents the border between fine (<1µm) and coarse (>1~~2010). Few other studies have reported a decrease in UV-APS counting efficiency for FBAPs < 2 µm based on comparison of ambient FBAPs with another LIF instrument (WIBS and BioScout) using different fluorescence wavelengths (Healy et al. 2014, Saari et al., 2014). In the present study we define 1 µm as the cutoff diameter to distinguish between the submicron (<1 µm) and the super-micron (>1 µm) modes of the particle number size distribution. The subscripts throughout this manuscript text "F" and "T" refer to fluorescent and total coarse mode particles, respectively. ~~Please refer to~~See Table 1 for~~ the~~ abbreviations, notations, and symbols used in this manuscript. The particle mass size distributions (dM/dlogD_a) for total as well as fluorescent biological aerosol particles were calculated for each size bin by multiplying dN/dlogD_a with volume of an aerodynamically equivalent sphere with the geometric midpoint diameter ($D_{a,g}$) and assuming the unit density (1 g cm^{-3}) and unit shape factor. The integral mass concentrations of coarse fluorescent biological aerosol particles and total coarse particles, M_F and M_T, respectively were calculated by integrating the particle mass distribution for D_a >1µm~~; but~~, thus particle mass reported here should be viewed as first approximation as a result of uncertainty associated with the density and shape of the particles (Huffman et al., ~~2010).~~ 2010). To be consistent with previous UV-APS results no standard temperature and pressure (STP) corrections were applied to the concentrations reported in this study. These number concentrations can be normalized to the volume that the sampled air would occupy under dry standard condition (STP: 273K, 1000 hPa, and 0% RH) by multiplying the concentration values reported here with a factor of 1.29 derived using ideal gas law.

Fluorescence of ~~submicron~~sub-micron particles

It has been reported by previous researchers that UV-APS is known to exhibit fluorescence for some fraction of non-biological aerosol particles including soot, PAHs, and ~~cigarettes~~cigarette smoke, which could be erroneously counted as FBAP (Huffman et al., 2010; Pan et al., 1999a, 1999b). ~~It has also been emphasized that such interference can mostly occur for particles less than 1 µm as the contribution from combustion sources at this size range is expected to be dominant.~~ To investigate the contribution of non-biological aerosol particles that are counted as fluorescence biological aerosol particles, Huffman et al., (2010) ~~performed~~showed the correlation between the integrated number concentrations of fluorescent particles (N_F) and total particles (N_T) for different diameter ranges (only for the fluorescence channels >3). ~~They found that the correlation for the submicron particles was systematically linear, whereas the correlation for supermicron particles was more random, indicating that a large fraction of submicron particles showing fluorescence might have been originated from anthropogenic sources, which may not be the case for the supermicron particles.~~ To ~~investigate~~examine the influence of anthropogenic emissions on ~~submicron~~sub-micron fluorescent particles, we performed ~~the~~a

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similar correlation analysis for the entire campaign~~ and, however, found the different results.~~.

The correlation between integrated number concentrations of fluorescent particles (N_F) and total particles (N_T) for ~~supermicron~~super-micron (D_a>1µm) and submicron (D_a<1 µm) diameter range exhibited a very poor scatter (R^2=0.03 and R^2=0.002 respectively; N=22280; Figs. S2) indicating extremely small percentage of fluorescence ~~was~~ contributed by non-biological aerosol particles in ~~supermicron~~both super-micron and submicron particle ranges. This was in contrast with the observations in Huffman et al (2010).

Since certain component of the mineral dust may exhibit a weak fluorescence (Huffman et al., 2010; Sivaprakasam et al., 2004; Toprak and Schnaiter, 2013), we performed the separate correlation analysis for ~~a focus~~the *dusty* period, which was dominated by the transport of mineral dust from West Asia, North Africa, and Arabian region (discussed below). The correlation between integrated number concentrations of N_F and N_T for D_a>1 µm was moderately linear (R^2=0.26; N=3138; Fig. S3a) compared to ~~submicron~~the sub-micron size range during the *dusty* period (R^2=0.007; N=3138; Fig. ~~S3b). As a result, correlation between N_F and N_T indicates~~S3b), indicating that the fraction of ~~supermicron~~super-micron particles exhibiting fluorescence may have been contributed by mineral dust, but ~~this being~~ not~~ the case~~ for ~~submicron~~sub-micron particles.

From these analyses we infer that the contribution of non-biological aerosol particles exhibiting fluorescence was negligible in both ~~submicron~~sub-micron and ~~supermicron~~super-micron (except during "*dusty* period"; discussed below) size ranges. Thus we hypothesize that due to persistent rainfall the ~~submicron~~sub-micron and ~~supermicron~~super-micron particles ~~resulted~~resulting from combustion ~~and other similar~~related activities, were either efficiently removed or were not transported to the ~~observational~~observation site~~, indicating that substantial fraction of the~~

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~~particles in both the size ranges were of biological origin.~~. Thus this ~~observational~~observation site could be ~~potentially~~ termed as relatively pristine and free from anthropogenic emissions during the monsoon season making this site scientifically interesting for investigating the characteristic properties of bioaerosols on long-term basis using advanced online and offline techniques.

~~Please note, however, that~~However, to ~~have~~maintain the consistency and uniformity in the comparison of N_F, N_T, and other similar parameters reported ~~by the~~in previous studies ~~we derived~~, all the statistics associated with dN_F/dlog D_a and dN_T/dlog D_a with a cutoff diameter of ~1 μm were derived.

2.~~3~~4 Meteorological parameter measurement

~~The meteorological~~Meteorological parameters ~~in parallel with the UV-APS measurements were~~ were recorded during the entire campaign using ~~an ultrasonic~~ weather sensor (Lufft WS600-UMB) installed on ~~a~~the rooftop at the same height and a few meters away from the UV-APS inlet (Fig. 1b). These measurements were made concurrently with the UV-APS measurements.~~S1).~~ The weather station was capable of recording temperature, dew point temperature, relative humidity, precipitation intensity, wind speed, wind direction, and air pressure and was set to record these ~~meteorological~~ parameters ~~with every~~at 5 minutes interval with time synchronized to UV-APS measurement clock. The meteorological data ~~from the weather sensor was stored by using an in-house developed external data logger. The~~ obtained ~~meteorological data~~at this site was compared with data obtained from another ~~ultrasonic~~ weather station (Vaisala WXT520) installed within the close vicinity ~~(Valsala make).~~. The scatter plots between the data (10 ~~min averaged~~minute averages) obtained from ~~our~~both these weather ~~station~~

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~~and the one installed in the close vicinity~~stations exhibited very strong agreement for all the meteorological parameters measured/recorded (average $R^2 \geq 0.95$).

2.~~4~~5 SEM Analysis

The samples for Scanning Electron Microscopy (SEM) analysis were collected on a 25 mm diameter Nucleopore® Polycarbonate filter paper with pore sizes of 5 µm and 0.2 µm using a two stage filtering method as described by Valsan et al~~.,~~. (2015). All samples were collected for ~~approximately a~~approximate duration of 60 min at an average flow rate of 5 ~~lpm~~Lpm and were stored in an air-tight container at 4°C until the SEM analysis~~. The five~~ was carried out. More than 100 individual particles analyzed from samples collected on five occasions during the entire campaign, were ~~analyzed~~ investigated using two different scanning electron microscopes~~. 1.~~ – a) Quanta FEG 200 located at the Sophisticated Analytical Instrument Facility (SAIF) and ~~2.~~b) Hitachi S 4A00 located at the Chemical Engineering Department of Indian Institute of Technology Madras. Before loading the filter paper on to the ~~studs~~sample holders, they were cut into small squares of ~1 cm^2 and sputter coated with gold particles. The biological aerosol particles were identified purely based on their morphological features adopting the method suggested by Matthias-Maser and Jaenicke (1991,1994). Detailed description on sample collection and analysis was discussed elsewhere (Valsan et al., 2015).

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3 Results and discussions

3.1 Campaign overview

Figure 2 shows the temporal evolution and variability of the several ~~parameters characteristic for the~~ meteorological ~~conditions~~parameters, FBAP, and TAP properties observed throughout the measurement campaign during SW monsoon season at ~~a high-altitude site of~~ Munnar.

~~Overall~~Several observations regarding the meteorological conditions during the campaign at Munnar can be ~~summarized as follows:~~made. The predominant wind direction was observed to be Westerly/Southwesterly (Fig. 1), which ~~characterizes~~ is characteristic of the monsoon season and bringing ~~almost anthropogenically~~nearly clean marine influx (laden with dust and sea salt particles; Vinoj and Satheesh, 2003; Vinoj and Satheesh, 2003; Satheesh and Srinivasan, 2002; Vinoj et al., 2014; Prospero, 1979) over the continent marked by presence of persistent rainfall, high relative humidity (RH), higher wind speeds, and lower temperatures. ~~During this period the diurnal variations in temperature and relative humidity were totally absent and temperatures almost approached the dew point temperature. Further, the Westerly/Southwesterly air masses arriving at the observational site were free from any anthropogenic influence and were laden with dust and sea salt particles (Satheesh and Srinivasan, 2002; Vinoj et al., 2014; Prospero, 1979). On few occasions, however, Northerly winds were also observed, which was associated with calm winds~~During this period diurnal variations in temperature and relative humidity were very small and temperatures approached the dew point. On a few occasions, however, Northerly winds were recorded, marked by relatively lower wind speeds, lower RH levels, higher temperatures, and reduced rainfall. During Northerly winds the temperature exhibited ~~relatively~~ more pronounced diurnal variations compared to the relative humidity. The average meteorological parameters (arithmetic mean±standard deviation) recorded during entire measurement period were: (840±1.3) hPa absolute pressure, (17.2±1.4)^{o}C ambient temperature,

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(96.4±5.7) % relative humidity, (2.8±1.3) m s^{-1} local wind speed, (270)° local wind direction (vector mean weighted by wind speed), and (4188) mm of accumulated rainfall.

The total of more than five months of bioaerosol measurements ~~in~~with high time and size resolution were performed at this site ~~comprising~~for two contrasting seasons~~,~~ - monsoon (dominated by Southwesterly winds) and winter (dominated by Northeasterly winds). In this study, we present the results from ~~the~~this field campaign carried out during the SW monsoon season whereas the detailed results from the winter campaign from the same measurement site will be presented in ~~the~~a follow up study. We first discuss the characteristic features of the time series as a broad overview of the observed concentration levels, variability, and trends in N_T and N_F. Figure 2 (f,g,h,i,j) shows the time series of geometric mean diameter (D_g~~),~~, N_F, N_F/N_T, N_T, FBAP and TAP 3-D~~ and the~~ size distribution measured with the UV-APS for the entire campaign.

Throughout the measurement period the hourly averaged D_g time series consistently remained in the range of ~2 – 4 µm with almost no diurnal ~~variations.~~variation. During the second half of the campaign, the D_g, however, exhibited relatively high variability with an average mean diameter of 2.6±0.7 µm. Unlike the N_T and N_F the variability in D_g ~~was observed~~did not seem to be ~~not~~ affected by the meteorological parameters except for wind direction (see section 3.~~6~~4.1) on few occasions. The total coarse particle number concentration, N_T, exhibited high and consistent variability during entire measurement period, however, with no distinct diurnal cycle. ~~-~~Averaged (arithmetic mean±standard deviation) over the entire measurement period was N_T was observed to be 1.8~~±~~ ± 1.5 cm^{-3} with lowest and highest concentrations of 0.01 cm^{-3} and 8.6 cm^{-3}, respectively. ~~The average N_T concentration during the months of June, July, and August was 2.7±1.9 cm^{-3}, 1.5±0.96 cm^{-3}, and 0.96±0.77 cm^{-3}, respectively, with highest and lowest values for~~

~~individual months respectively as follows: June: 8.6 and 0.04 cm^{-3}, July: 5.1 and 0.02 cm^{-3}, and August: 3.6 and 0.01 cm^{-3} (Fig. S4).~~ The monthly averaged N_T concentration (Fig. S4) exhibited ~~the~~a decreasing trend from June to August as the monsoon progressed (Tab. 2). In contrast ~~to the total aerosol particle number concentration~~, N_F, exhibited less pronounced but episodic peaks in the time series during the majority of the measurement period, resulting in ~~modest variability and~~a campaign arithmetic mean ~~value was~~of 0.02 ± 0.02~~±0.02~~ cm^{-3}. The highest N_F concentration of ~0.52 cm^{-3} was observed ~~on 3rd~~in June, prior to the onset of ~~June (and few more occasions)~~monsoon rainfall, whereas the lowest N_F concentration (<0.0002 cm^{-3}) was consistently observed on more than one occasion during the months of July and August. The ~~average~~monthly averaged N_F ~~concentration during June and August was 0.03±0.03 cm^{-3} and 0.015±0.02 cm^{-3}, respectively with lowest N_F concentration of 0.007±0.006 cm^{-3} in July (~~concentrations are listed in Tab. 2~~)~~..

The time series of relative contribution of FBAP to TAP number, N_F/N_T, ~~most of the time during campaign~~ exhibited the similar trend in temporal variability ~~to N_F.~~as displayed by N_F for most during the campaign. The ~~pronounced~~ extreme values of N_F/N_T observed on a few occasions ~~resulted from strong variability in the concentrations of N_T rather than resulting from the variations in the concentrations of N_F, indicating the inverse~~corresponded to low values of N_T implying a negative correlation between N_T and N_F/N_T~~.~~ during these measurements. Huffman et al., (2010) ~~have~~ also reported ~~the~~ a similar ~~inverse~~negative correlation between N_T and N_F/N_T ~~from the measurements carried out~~ at a semi-urban site ~~from~~ in central Europe~~. Temporal evolution of N_F, N_F/N_T, and 3-D number size distribution for individual campaign months is shown in Fig. S5. A~~ indicating that the variability in N_F/N_T was associated with changes in N_T concentrations. The campaign overview (including individual months) of FBAP mass concentrations and 3-D size distribution for each five minutes of UV-APS sample averaged over the entire measurement period and individual months are shown in Figure S6. During the first month of measurement campaign M_F exhibited high concentration with sporadic spikes at irregular intervals with broader size distribution (~2 – 8 µm) towards the end of the month (with highest concentration ~ 6.0 µg m^{-3}). As the measurement campaign progressed, with arrival of persistent and heavy rainfall (whole of July and first half of August) M_F exhibited a gradual decrease with minimum value reaching as low as 6 x 10^{-4} µg m^{-3}. After a period of consistent low mass concentration, during the last week of measurement campaign, M_F exhibited an increase with highest mass concentration of ~ 5.8 µg m^{-3}, which coincided with reduced and scattered rainfallS5.

3.21.1 Particle number and mass concentrations

3.2.1 Statistical distribution of The number **concentrations**

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Statistical distribution of five-minute numberand mass concentration measurements carried out at Munnar over the course of the campaign are shown in Fig. 3 and tabulated in Tab. 2. The box plots show statistical representation of five minute averaged data of the time series. Over the entire measurement period the monthly mean of N_T varied by a factor ~3 from a minimum in August (0.96 cm^{-3}) to a maximum in June (2.7 cm^{-3}). In addition to the highest concentration, the; Fig. 3a). The variability of N_T was also found to be highest in the month of June as can be seen from the size of the 5 – 95th percentile bars, which also reflected in Fig. 3a. The relative the high variability inof N_T for entire measurement period was largely contributed by the variability in N_T observed in the month of June.. During the initial phase of Southwest monsoon season, the predominant Westerly/Southwesterly winds are known to transport the mineral dust, which constitute large fraction of coarse mode (also in larger diameter size of fine mode fraction) TAP

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concentration, over the Indian continental region (Vinoj et al., 2010, 2014; Li and Ramanathan, 2002; Satheesh and Srinivasan, 2002; Vinoj and Satheesh, 2003). As the monsoon progresses the persistent rainfall can cause the washout of these dust particles along the path of monsoonal rain, thus reducing the coarse mode TAP concentration (Pranesha and Kamra, 1997a,b; Radke et al., 1980; Moorthy et al., 1991). The monthly arithmetic mean and median average of N_T did not exhibit significant differences. The monthly mean values of N_F varied by the factor of ~4 with consistently highmoderate variability during all the observational months.entire campaign (Fig. 3b). Similar to N_T, the monthly mean average value and variability in N_F was highest in the month of June, with mean of 0.03±0.03 cm^{-3} and high size ofa 95th percentile (with value of 0.086 cm^{-3}), respectively.. The lowest average concentration in N_F (0.007±0.006 cm^{-3}) was observed in the month of July was associated with relatively lower variability as compared to other months of field measurement campaign. Unlike N_T, the arithmetic mean and median average of N_F for individual months exhibited a significant difference as can be seen from the box plot shown in Fig. 3b. The variability of N_F/N_T showed thea similar temporal pattern asto that ofdisplayed by N_F, except that the campaign average mean N_F concentration was higher than that of the August, whereas the campaign averaged mean N_F/N_T was observed to be lower than the mean calculated for August. As can be seen from Fig. 3c, the mean relative contribution of N_F to N_T was lowest in the month of July (~1%) and highest in the month of June and August (~3%). The median and mean for N_F/N_T, over the course of campaign were ~1 and 2%, respectively. (Fig. 3c). The average values of N_F/N_T over this part of the globe were found to be lower as compared tothan previously investigated sites (Huffman et al., 2010, 2012; Bowers et al., 2009; Schumacher et al., 2013; Matthias-Maser and Jaenicke, 1995; Matthias-Maser et al., 2000; Gabey et al., 2010).

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Diurnal patterns of Though, the *number concentration*

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The average diurnal trends for three individual months and the entire measurement campaign were analyzed. Figure 4 shows the median FBAP values for each hour of the day for three individual months and entire campaign, and Fig. S7 shows the corresponding TAP plots. Overall N_F exhibited a moderately diurnal pattern with consistent early morning (06:00 hr) peak at ~3 µm (Fig. 4a) where in the month of July this early morning peak was absent. A relatively weak peak during late evening (20:00 hr) in FBAP concentration at ~3 µm was consistently observed in the month of July. In the month of June the average diurnal N_F concentration started increasing early in the evening (~18:00 hr), which gradually increased through the night and reaching maximum at ~06:00 hr and started decreasing thereafter as day progressed. The average diurnal N_F pattern in August exhibited more or less qualitatively similar features to that of diurnal pattern observed in June. In general the weak diurnal pattern observed in N_F during the month of July was consistent with weak RH and temperature diurnal patterns, and persistent rainfall observed during July. The early morning peak at ~3 µm on the diurnal scale was also reported from pristine Amazonian rainforest environment (Huffman et al., 2012). Corresponding average size distributions for entire measurement period will be discussed in details in Sec. 3.3. The diurnal variations of N_T (Fig. S7), on the other hand were very distinct from those of N_F. The size resolved dN_T/dlog D_a for each individual months exhibited a consistent and flat concentration profile at <1 µm, except for the month of August where a pronounced afternoon peak (~12:00) at ~1 µm was observed. Reduced rainfall and substantial changes in meteorological parameters including the change in prevailing wind speed and shift in direction during later half of August might have caused the appearance of afternoon peak due to particles resulting from local sources. As like N_F, N_T showed the strong quantitative variability amongst each individual month (Fig.

S7). Previous studies where similar instrument was used have reported that pronounced diurnal variations in N_T are strongly coupled with diurnal variations in meteorological variables especially mixing layer depth (Garland et al., 2009; Raatikainen et al., 2014; Du et al., 2013). The absence of pronounced diurnal variations in N_T at this particular site may be a result of weak dependence of coarse mode TAP concentrations on meteorological parameters combined with persistent rainfall causing the washout of these particles (Radke et al., 1980; Raatikainen et al., 2014; Kanawade et al., 2014; Shika et al., 2016). This also indicates the absence of any strong and localized source of anthropogenic emissions during most of the campaign period. Diurnal patterns of N_F/N_T more or less followed the same pattern as that of N_F during all the measurement months owing to complete absence of diurnal variability in N_T. Averaged over the entire campaign the N_F/N_T was found to be highest during early morning hour at ~06:00 hr (~3.2%) consistent with the time of high N_F concentration (Fig. 4). The distinct diurnal pattern in N_F and N_T supports the fact that the sources of TAP and FBAP were different over this region.

3.2.2 Statistical distribution of mass concentration

Basically UV-APS measures the particle number;numbers, the average mass of size-resolved particles particle mass can be derived as first approximationalso be estimated by assuming the particle density equal to 1 g cm^{-3} (unit density). AccordinglyHuffman et al., 2010, 2012). Based on this, the overview of mass concentrationconcentrations of FBAP over the course of measurement period is (M_F) and TAP (M_T) are presented here. The statistical distribution of five minutes mass concentration derived from number concentration measurements over the course of campaign is shown in Fig. 5 and tabulated in Tab. 2.in Fig. 3. The monthly mean values of M_T exhibited the similar trend and temporal variability as that ofshown by N_T with overall decrease in M_T through the course of measurement months as campaign progressed. The highest monthly

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~~average concentration of M_T (~10.6 µg m^{-3}) was observed in the month of June whereas the lowest M_T of ~4.2 µg m^{-3} was observed in the month of August. Averaged over the entire measurement period the~~as campaign progressed (Fig. 3d). The campaign mean M_T at Munnar was ~7 µg m^{-3}, which was comparable to the values reported from a central European city (M_T ~7.3 µg m^{-3}) and higher than concentration of M_T (~2.5 µg m^{-3}) reported from a pristine Amazonian rainforest region measured during wet season (Huffman et al., 2010; 2012). The monthly mean values of M_F, on the other hand, did not exhibit a similar pattern ~~like~~to that shown by M_T, but followed a temporal pattern ~~like~~similar to that shown by N_F~~.~~ (Fig. 3e). The highest mean mass concentration of M_F (~0.4 µg m^{-3}) observed during June ~~and~~ was ~3 and 2 times lower than the concentrations observed at a central European city (~1.26 µg m^{-3}) and pristine Amazonian rainforest (~0.85 µg m^{-3}), respectively. The higher difference between mean and median values of the box plots indicates the higher temporal variability.~~ The relative difference between mean and median of N_F was found be higher than that of M_F indicating higher temporal variability of N_F during all measurement months. Averaged over the course of entire measurement period this trend was found to be consistent.~~ The median and mean for M_F/M_T over the course of entire measurement period were 6 and 3% respectively, which is relatively low compared to previously reported studies for various other environments (Huffman et al., 2010; 2012; Artaxo and Hansson, 1995; Schumacher et al., 2013; Fig. 3f). On the average, the relative contribution of FBAP to TAP coarse mode particle mass was ~3 times higher (~6%) than its contribution to coarse mode particle number concentration (~2%). This is consistent with the observations that FBAPs show enhanced prevalence among the larger aerosol particles (Huffman et al., 2010).

3.1.2 Diurnal Patterns

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The average diurnal trends for three individual months and the entire measurement campaign were analyzed. ~~*patterns*~~ Figure 4 shows the mean FBAP values for each hour of the day for three individual months in the campaign, and Fig. S6 shows the corresponding TAP plots. Overall N_F exhibited a moderate diurnal pattern with a consistent early morning (06:00 hr.) peak at ~3 µm (Fig. ~~*mass*~~4a) except for the month of July, where this early morning peak was absent. A very weak peak during late evening (20:00 hr.) in FBAP concentration at ~3 µm was observed in the month of July. In the month of June, the average diurnal N_F concentration started increasing early in the evening (~18:00 hr.), which gradually increased through the night reaching maximum at ~06:00 hr. and started decreasing thereafter as the day progressed. A similar diurnal pattern was also observed in August but without high FBAP concentrations in the evening hours. In general, the weak diurnal pattern observed in N_F during the month of July seemed to follow the weak diurnal patterns in RH and temperature, in the presence of persistent rainfall observed during July. The early morning peak at ~3 µm on the diurnal scale was also reported from pristine Amazonian rainforest environment (Huffman et al., 2012). Corresponding average size distributions for entire measurement period will be discussed in details in Sec. 3.3. The diurnal variations of N_T (Fig. S6), on the other hand were very distinct from those of N_F. The size resolved dN_T/dlog D_a for each individual months exhibited a consistent and flat concentration profile at <1 µm. Previous studies where similar instrument was used have reported that pronounced diurnal variations in N_T are strongly coupled with diurnal variations in meteorological variables especially mixing layer depth (Garland et al., 2009; Raatikainen et al., 2014; Du et al., 2013). The absence of pronounced diurnal variations in N_T at this particular site may be a result of weak dependence of coarse mode TAP concentrations on meteorological parameters combined with persistent rainfall causing the washout of these particles (Radke et al.,

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1980; Raatikainen et al., 2014; Kanawade et al., 2014; Shika et al., 2016). This also indicates the absence of any strong and localized source of anthropogenic emissions during most of the campaign period. Diurnal patterns of N_F/N_T more or less followed the same pattern as that of N_F during all the measurement months. The distinct diurnal pattern in N_F and N_T supports the fact that the sources of TAP and FBAP were different over this region.

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The diurnal trends in M_F and M_T for individual months and campaign average were also analyzed and are shown in ~~Fig. 6. The corresponding diurnal trends in M_T are shown in Fig.~~ Figures S7 and S8. The monthly averaged diurnal trends in M_F for individual months and entire campaign exhibited similar a trend similar to that shown by corresponding to N_F. However, the prominent peak in dM_F/dlog D_a was observed at higher diameter (~3 – 4 µm~~), which is due to the fact that dM_F/dlog D_a has been derived from dN_F/dlog D_a assuming unit density. As observed for N_F during the month of June, the consistent morning peak was present in M_F with only difference of prominent second peak in M_F, which starts late in the evening at ~19:00 hr and further extends up to morning hours (~08:00 hr). Thereafter M_F concentration steadily decreased as the day progressed reaching minimum at around mid-day. The early morning peak in M_F concentration was consistently observed in the size range of 3 – 4 µm for the all the measurement months. The characteristic distribution of M_T (Fig. S8), however, exhibited distinct behavior as compared to both M_F and N_T. The concentration peak of <1 µm observed in N_T shifted to the higher diameter range of ~2 – 3 µm as increase in mass is more associated with presence of coarse mode particles. For example in June M_T exhibited similar diurnal feature as that of N_T. The flatter trend observed in average M_T during the month of June disappeared during the month of July and August with appearance of less prominent peak in M_T at around 12:00 hr resulting in relatively pronounced diurnal pattern (Fig. S8).~~). The concentration peak of <1 µm observed in N_T shifted to the higher diameter range of ~3 – 4 μm as increase in mass is more associated with presence of coarse mode particles. The distribution of M_T (Fig. S8), however, exhibited a distinctly different trend compared to both M_F and N_T. The distinct diurnal patterns of M_F and M_T showed very less relative contribution of FBAP to TAP mass as compared to other observational sites (Huffman et al., 2010, 2012; Matthias-Maser and Jaenicke, 1995).

3.1.3 Size distribution of particle number and mass

Figure ~~7~~5 shows the number and mass size distributions for TAPs and FBAPs averaged over the entire measurement period. The TAP number size distribution, $dN_T/dlogD_a$, was generally broad and dominated by a peak at the lower end of the measured size range of number size distribution (D_a≈0.9 μm; Fig. ~~7a~~5a). In $dN_T/dlogD_a$, the concentrations exhibited a significant decrease above diameter ~3 μm with a long tail extending on the right hand side of the distribution. This peak may be comprised of mineral dust and sea salt particles, as also evident from SEM images (please refer to section 3.3) and as also reported by the previous studies investigating aerosol composition over India during monsoon season (Vinoj et al., 2014; Moorthy et al., 1991; Vinoj and Satheesh, 2003; Satheesh and Srinivasan, 2002; Li and Ramanathan, 2002). ~~The corresponding monthly $dN_T/dlogD_a$ are shown in Fig. S9. Overall the individual monthly $dN_T/dlogD_a$~~A similar peak in $dN_T/dlog D_a$ at D_a ≈ 0.9 μm was observed in pristine Amazonian rainforest during wet season and was attributed to mineral dust (Huffman et al., 2012; Fig. 5b).The corresponding monthly plots of $dN_T/dlogD_a$ are shown in Fig. S9 and exhibited the similar qualitative number size distribution pattern as that of campaign averaged TAP number size distribution. Averaged over the entire measurement period, the mass size distribution, $dM_T/dlogD_a$ (Fig. ~~7c~~5c), exhibited a broad peak at ~2.6 μm with an extended tail to the left side of the mass size distribution~~, whereas on the right side a second peak started appearing at D_a≈12~~

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~~μm.~~. The corresponding monthly averaged dM_T/dlogD_a are shown in Fig. ~~S10. As evident from the figure~~ S10 and appeared similar to the campaign average TAP mass size distribution ~~appeared generally similar to each of the individual months.~~. For accurate representation of mass size distribution the unit-normalized mass distribution ~~in~~of D_a plotted in Fig. ~~7~~5 (c and d) is expected to shift to larger particle size with increased area under the curve~~, as D_a is directly proportional to square root of density of the particle under consideration~~ (Huffman et al., 2010; DeCarlo et al., 2004).

The campaign average number size distribution of FBAP (Fig. ~~7b~~5b) exhibited monomodal shape with much narrower peak than the TAP number size distribution, with a dominant mode at D_a≈2.8 μm, which was consistent throughout measurement period. The corresponding monthly mean FBAP number size distributions are shown in Fig. S11. ~~This peak was much prominent and narrow in the month of June with highest FBAP concentration and became less pronounced in July, with the lowest FBAP concentration.~~ As reported by Huffman et al., (2010) multiple and broader peaks in dN_F/dlogD_a are most likely to originate from different sources and biological species. In the present study, however, we did not find multiple peaks in investigated FBAP number size distribution, suggesting that observed FBAPs comprised the particles from similar or same sources. The overall qualitative appearance of the average FBAP number size distribution is similar to that has been reported by previous measurements. For a semi-urban site in Central Europe Huffman et al., (2010) reported an average FBAP peak at 3.2 μm. Gabey et al., (2010) observed a ~~similar~~ peak at ~2.5 μm at a tropical rain forest site in Borneo. From a pristine Amazonian rainforest site during wet season Huffman et al., (2012) reported a ~~similar~~ peak at ~2.3 μm. For another pristine observational site in boreal forest in Finland Schumacher et al., (2013) reported a peak in FBAP number size distribution at ~3 μm. A ~~similar~~ peak at ~3 μm was also observed by Healy et al., (2014) at a rural site in Killarney national park, Ireland. This dominant peak in the range of 2 – 3 μm in FBAP number size distribution is strongly attributed to the fungal spores over the continent as reported by numerous previous researchers **(**Huffman et al., 2010, 2012; Schumacher et al., 2013, Li et al., 2011; Artaxo and Hansson, 1995; Healy et al., 2014; Gabey et al., 2010, 2013; Toprak and Schnaiter, 2013**)**. Recently Valsan et al., (2015) investigated the morphological characteristics of PBAPs from the same site during non-monsoon season and found that fungal spores constituted the major fraction of PBAPs and nominally ranged in the size range of ~3 – 10 μm, which roughly translates into equivalent aerodynamic diameter of 2 – 5 μm~~.~~ (assuming particles to be a prolate spheroid) . The scanning electron microscopy images obtained from the filter samples~~ occasionally~~ collected during this field campaign showed the strong presence of variety of fungal spore in the size range of ~~3~~6 – 10 μm (aerodynamic diameter ~~2~~3 – 5 μm; discussed below; Fig. ~~17~~11). As an overview of the comparison, the FBAP concentration values observed at Munnar are compared to the FBAP concentration ranges obtained using similar online measurements techniques from diverse environmental conditions across the globe, and the details are tabulated in Tab. 3. The campaign averaged FBAP mass size distribution is shown in Fig. ~~7d~~5d, which nominally appeared bimodal with very sharp primary peak at D_a≈3.2 μm and very broad but ~~unappreciable~~small second mode at D_a≈4 μm. ~~The distinct presence of particle mass in the higher diameter range (>10 μm) in FBAP mass size distribution was not prominently noticed in Munnar as compared to previously reported studies (Huffman et al., 2010; 2012). In case of TAP mass size distribution the right side tail started showing positive slope at larger diameter whereas FBAP mass size distribution consistently showed the negative slope at larger diameters. Such a distinct shape of mass size distributions for TAP and FBAP reconfirms the fact that the larger particles observed in the TAP~~

~~mass distribution originated from processes that did not produce particles of the biological origin as likewise reported by Huffman et al., (2010).~~ The corresponding monthly mean FBAP mass size distributions are shown in Fig. S12. ~~The~~ The FBAP mass size distribution for individual ~~month FBAP mass size distribution~~months exhibited the similar qualitative shape to that of average campaign. ~~As mentioned above highest FBAP mass concentration was observed in June, which coincided with a very sharp and narrow primary peak in FBAP mass size distribution, while the lowest FBAP mass concentration during July, on the other hand, coincided with a broad primary peak with lower slope.~~

~~The~~ Figure 6 shows the size-resolved ratio of overall FBAP ~~to~~ /TAP ~~averaged over~~for the course of measurement ~~is shown in Fig. 8~~ and corresponding monthly ratios are shown in Fig. S13. The relative contribution of FBAPs (dN_F) to TAPs (dN_T) in each size bin could be used to derive the relative contribution of biological particles to total aerosol particles at each size. As reported by Huffman et al., (2010) the assumption of unit density of each particle implies that the value of the dN_F/dN_T ratio would invariably is equal to dM_F/dM_T. The integrated N_F/N_T and M_F/M_T, however, would have the distinct values. As can be seen from Fig. ~~8~~6 and ~~S13a~~S13 considerable quantitative and qualitative difference in mean (red) and median (green) curve was consistently observed in all individual months, which likely is the result of poor counting statistics and very high variability in ~~FBAP and~~ TAP number concentrations. Based on the results presented by Huffman et al., (2010) the mean (red) curve, best represents the N_F/N_T ratios at the upper particle sizes~~; hence we will stick our further discussion about N_F/N_T ratios for the present study to the mean curve.~~. The mean N_F/N_T ratio curves for individual months and for entire campaign exhibited two dominant peaks persistently in the particle size range ~3 – 4 µm and ~6 – 8 µm. The first prominent peak in dN_F/dN_T distribution at 3 – 4 µm comprised 15 – 16% while the second peak at 6 – 8 µm represented ~14 – 15% of the FBAP material in TAP over the entire measurement period (Fig. 8). As can be observed from Fig. S13, the second peak in N_F/N_T ratios for July was higher (~12%) than the first peak (~10%) unlike other two observational months. The fact that N_F/N_T ratio is approximately zero for the particle sizes <1.7 µm indicated that FBAP mainly comprised of very small fraction of submicron aerosols at Munnar. The statistics for the individual months showed that the first peak in dN_F/dN_T was more or less consistent at ~22% during June and August except for the July when second peak in N_F/N_T ratios contributed more (~12%) than the first peak (~10%).6).

3.42 Focus periods

As described in Sec. 3.1 based on campaign overview the characteristicsThe characteristic properties of FBAP and specifically TAP number concentration exhibited strong temporal variabilitiesvariability, which could be attributed to changes in prevailing meteorological conditions especially wind direction during monsoon season at Munnar. To explore the potential impact of air mass origin on number and size distribution of FBAP and TAP, we highlightThe following three distinct focus periods during the campaign are highlighted as follows:

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1. A "*dusty*" focus period of "dusty episode" was identified when prevailing wind was predominantly Westerly/Southwesterly and air masses mainly came from the Arabian Sea. These air masses, although almost anthropogenically clean, areThese were laden with sea salt and dust particles during the start of the monsoon, which dominate the coarse mode fraction of atmospheric aerosols (Vinoj et al., 2014; Li and Ramanathan, 2002). These dust particles observed over this region mainly originate) originating from West Asia, North Africa, and Arabian region (Vinoj et al., 2014). During our measurement) and not from local anthropogenic

sources. In this campaign, such a dusty period from 14-06-2014 00:00 hr to 25-06-2014 23:55 hr was observed and is between June 14-25, 2014, which was consistent with the description given above and also based on the SEM images, which showed the presence of mineralthe dust, obtained during dusty collected in this period (see Sec. 3.5 below). This period was marked with an accumulated rainfall of ~1015 mm, average relative humidity of 94.4± ± 6.5%, average temperature of 17.7± ± 1.5°C, and average wind speed 2.8±1.3 m s^{-1} (maximum wind speed of 6.7 m s^{-1}).

2. A "*clean*" focus period of "clean period", was observed during latter half of the monsoon season when wind direction was still predominantly Westerly/Southwesterly and air masses originated over Arabian Sea. During this period, which was chosenobserved from 09-07-July 9 – August 7, 2014 10:25 hr to 07-08-2014 23:55 hr, FBAP and TAP concentrations were extremely low with very weaklow variability. This *clean* period was associated with persistent rainfall (accumulated rainfall of 2650 mm), average relative humidity of 99.5±1.4%, average temperature of 16.4±0.5°C, and average wind speed 3.7±1 m s^{-1} (maximum wind speed of 8.3 m s^{-1}).

3. A "*high bio*" focus period of "high bio" comprised three discrete events of high FBAP concentration observed from 01-06-between June 1-5, 2014 09:10 hr to 05-06-2014 18:20 hr;, June 26-06-2014 00:05 hr to 30-06-, 2014 17:00 hr; and August 18-08-2014 00:00 hr to 22-08-2014 08:30 hr. Interestingly this, 2014. This period is marked with the very distinct metrological parameters compared to the *clean* period: accumulated rainfall 194 mm, average relative humidity 93.4±8.4%, average temperature 18.0±2.4°C, and average wind speed 1.2±0.8 m s^{-1} (with maximum wind speed of 4.6 m s^{-1}). Briefly, during "It is suggested that these high--bio" period stagnant periods are due to high variability in relative humidity and temperature, and the

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movement of air masses ~~came from~~with relatively low wind speed, over densely vegetated region located north of observational site~~, and relative humidity and temperature exhibited high variability~~.

3.~~4~~2.1 Particle number and mass concentrations

The statistical distributions of N_T, N_F, M_T, M_F, and ~~M_F~~corresponding ratios for three different focus periods (*dusty*, *clean*, and *high bio*) are shown in Fig. ~~9~~7 and tabulated in Tab. 4. Each of the focus periods discussed here ~~did~~were not ~~represent~~ of equal duration~~ of the observations.~~. The average total particle number concentration, N_T, showed a decrease of ~70% from *dusty* period to *clean* period (~4.2 cm^{-3} and ~1.3 cm^{-3} respectively), whereas the N_T concentration during *high bio* period was ~1.8 cm^{-3}. The high N_T concentration during the *dusty* period caused the high variability between 5th and 95th percentile in N_T when averaged over entire campaign period (Fig. 3a). The fraction of dust in coarse mode aerosol, which is observed to be very high during pre-monsoon and first few days from the onset of monsoon rainfall, gradually decreased as the monsoon progressed likely as a result of wash out and wet deposition due to persistent rainfall in the path of air masses (Hirst 1953; Madden, 1997; Burge and Roger, 2000). The M_T exhibited similar pattern to that of N_T during three distinct focus periods with average mass concentration of ~16.3 µg m^{-3}, ~5.1µg m^{-3}, and ~7.7 µg m^{-3} for *dusty*, *clean*, and *high bio* periods, respectively~~.~~ (Fig. 7d).

~~As expected, the~~The mean N_F ~~was highest~~ concentration during the *high bio* period (Fig. ~~9b) with an average concentration of~~ 7b) was 0.05±0.04 cm^{-3} ~~and~~with high variability in higher concentration range (0.06 – 0.13 cm^{-3}) as evident from the distance between 75th and 95th percentile. The N_F was found to be relatively stable during the *dusty* period with an average concentration of ~0.02±0.008 cm^{-3}. The ~~average~~mean N_F concentration was found to be an order

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of magnitude lower during the *clean* period (0.005±0.004 cm^{-3}) as compared to *high bio* period, whereas corresponding decrease in N_T from *dusty* to *clean* period (~ by factor of 3) was not of similar magnitude. ~~We put forward~~The following is the hypothesis proposed for such ~~a~~ concentration difference in N_F and N_T during the three distinct periods: During the *clean* period the predominant wind direction was Westerly/Southwesterly and air masses came from Arabian Sea bringing clean marine influx marked by persistent rainfall. As a result, the coarse mode aerosol fraction (N_F and N_T) emitted locally were efficiently removed, however, the sea salt particles present in the air masses, which came from Arabian Sea contributed to TAP number concentration (see section 3.~~5~~3). In addition to the efficient wet removal of FBAP due to persistent rainfall, the high RH level (average 99.5%), ~~which~~ causes the dew formation that further inhibit the spore release in turn ~~reduced~~reducing the FBAP concentration (Schumacher et al., 2013; Jones and Harrison, 2004). The mean values of M_F exhibited trends similar ~~temporal trends and qualitative pattern as~~to those shown by N_F, with highest mass concentration of 0.58 μg m^{-3} during *high bio* period, which reduced by ~86% (0.08 μg m^{-3}) during the *clean* period. As anticipated the relative contribution of FBAP in TAP during *dusty* and *clean* periods was almost negligible with N_F/ N_T ratio of ~1%. Whereas during the *high bio* period the relative FBAP number and mass contribution to corresponding TAP was ~5% and 12% respectively.

3.~~4~~2.2 Size distribution of particle number and mass concentration

Figure ~~10~~8a highlights the dN_F/dlog D_a during three distinct focus periods and corresponding dN_T/dlog D_a are shown in Fig. S14. ~~Overall~~In general dN_F/dlog D_a during each focus period exhibited pattern similar to that of campaign average.

The dN_F/dlog D_a averaged over the ~~dusty~~ *high bio* period ~~was dominated by~~ exhibited a ~~narrow~~ very prominent and sharp peak at ~2.5 – 3.~~1~~ μm. The corresponding dN_F/dlog D_a during *dusty*

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and *clean* period was overall broader also exhibited similar bell shaped distribution with less prominent peaks owing to the relatively lower FBAP concentrations as compared to dusty and high bio periods with gradual increase in FBAP number concentration from diameter range of ~1 – 2.3 µm, with a sharp increase thereafter, whereas downward slope exhibited the consistent pattern. dN_F/dlog D_a during the *high bio* period exhibited relatively narrow peak at ~2.5 µm.. Unlike previously reported studies (Huffman et al., 2010; 2012) the peak in dN_F/dlog D_a (D_a≈3 µm) was not reflected in dN_T/dlog D_a mostly due to relatively less contribution of FBAP in coarse mode TAP number concentration. As can be seen from Fig. S14aS14 the total aerosol particle number size distribution, dN_T/dlog D_a, during dusty periodall the three focus periods exhibited a peak at ~0.9 µm, with a high negative slope on the left side of the distribution curve. This peak may be comprised of mineral dust and sea salt particles, as also evident from SEM images (please refer to section 3.5) and based on the previous studies investigated aerosol composition over India during monsoon season (Vinoj et al., almost similar pattern to that of campaign averaged2014; Moorthy et al., 1991; Vinoj and Satheesh, 2003; Satheesh and Srinivasan, 2002; Li and Ramanathan, 2002). A similar peak in dN_T/dlog D_a at D_a≈0.9 µm was observed in pristine Amazonian rainforest and particles were mostly dominated by mineral dust during high dust period (Huffman et al., 2012, Fig. 5b). During clean period dN_T/dlog D_a resembled the similar shape (with higher concentrations peaking at ~0.9 µm) to that of dusty period, however, with lower concentration. The corresponding dN_T/dlog D_a distribution (Fig. S14c), during high bio period, exhibited multiple peaks and appeared noisy for D_a<1 µm with increasing trend in TAP number concentration for the lower diameter range of the distribution. The downward slope for D_a>1 µm exhibited consistent shape (mean curve) compared to distributions observed during other two focus periods..

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The FBAP mass size distribution (Fig. ~~11~~8b) during *dusty* period was dominated by bimodal peaks with prominent peak at ~3 µm and a relatively less pronounced peak in the range of ~4 – 6 µm showing broader tail on the right side of the distribution curve. The dM_F/dlog D_a, during *clean* period, exhibited similar bimodal peaks with extended shoulder in the diameter range from ~4 to 7 µm. The dM_F/dlog D_a distribution during *high bio* period was ~~distinct~~distinctly different compared to the two other focus periods discussed above with a prominent monomodal peak at ~3 µm. The primary peak observed in dM_F/dlog D_a in the range of ~3 to 4 µm was consistent during individual months and different focus periods. TAP mass size distribution (Fig. S15) exhibited similar qualitative pattern to that of campaign averaged dM_T/dlog D_a with peak between ~2.5 to 3.5 µm with an extended tail on the right side, which gradually increased for D_a>13 µm. The statistics representing 5th, 25th, 75th, and 95th percentile for dN_F/dlog D_a and dM_F/dlog D_a during individual focus periods are shown in Fig. S16 and S17.

The size resolved ratio of FBAP to TAP particles averaged for three distinct focus periods is shown in Fig. ~~12.~~9. As evident from the figure the largest fraction of FBAP particles during *dusty* period occurred between ~ 6 – 9 µm (~20%) with relatively small (~7%) contribution in the ~~size~~diameter range of ~ 3 – 4 µm ~~(~7%). The dN_F/dN_T exhibited the sloping tails on both the sides of the distribution with steep slope on the right side.~~. The fact that N_F/N_T is ~~approximately~~near zero for ~~the~~ particle size ~~range~~ below ~1.5 µm is ~~consistent~~in line with previous observations reported from a semi urban site in central Europe and during wet season of pristine Amazonian rainforest (Huffman et al., 2010; 2012). During the *clean* period the maximum contribution of FBAP to TAP number concentration reduced to ~10.5% in the diameter range of ~ 6 to 9 µm ~~with another prominent~~, but ~~relatively smaller contributing~~ the peak~~,~~ appeared at ~3 – 4 µm and remained almost consistent with relative contribution of ~8%.

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~~During~~ Whereas during *high bio* period the maximum contribution of FBAP to TAP occurred between broader size range of ~ 3 – 8 µm with contribution range of ~28 – 19~~% and relatively broad dN_F/dN_T distribution.~~%. Interestingly during the *high bio* period, the highest contribution of FBAP to TAP number concentration occurred at D_a~~≈~~ ≈ 3.5 µm, as opposed to the other two focus periods when the highest contribution was observed in the larger diameter ranges of ~ 6 – 8 µm. N_F/N_T was consistently found to be ~~equal to~~very low, with values approaching zero for the diameter beyond 13 µm, indicating ~~that~~ FBAP ~~comprised~~constituted an extremely small fraction of total aerosol particles (Huffman et al., 2010; 2012). The two prominent peaks observed during the focus periods were clearly evident in campaign-averaged dN_F/dN_T (Fig. ~~8~~6; peaks at ~3.5 and 6 µm).

3.~~4~~2.3 Diurnal patterns

A prominent early morning peak in N_F during the *high bio* period in the diameter range of 1.5 – 3 µm was observed from 06:00 ~~hr~~ to 08:00 ~~hr,~~hrs., which clearly reflected in campaign averaged diurnal patterns at the same hour of the day. The diurnal variations in N_F during the *dusty* and *clean* periods were not so pronounced (Fig. ~~13~~10) as compared to the variations during the *high bio* period. During the *dusty* period, N_F showed slightly high concentration starting from ~~~20~~17:00 hrs. (lowest panel Fig. 10a) and persistently remained high until early morning without any variations, whereas during the *clean* period, N_F concentration consistently remained flat throughout 24 ~~hrs. The evening peak observed during dusty period, however, was clearly absent during high bio period. A moderately pronounced peak in N_F during evening~~ hours ~~at ~20:00 hr during dusty periods might indicate that releasing mechanism of bioaerosols was efficient as a result of nocturnal sporulation. This can further imply that the morning and late evening peaks in dN_F/dlog D_a at D_a≈3 µm most likely resulted from different type of spores (Hirst, 1953).~~. As

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~~listed~~reported by Huffman et al., (2012) the emission and dispersal of bioaerosols is strongly coupled with environmental variables such as solar radiation, temperature, and relative humidity ~~and each~~. Each of these variables ~~have strong~~exhibited relatively pronounced diurnal ~~cycles.~~variations during the *high bio* period (upper panel Fig. 10c). It has been well documented that relative humidity, in particular, plays an important role in active wet discharge of fungal spores (Adhikari et al., 2006; Burch and Levetin, 2002; Elbert et al., 2007; Jones and Harrison, 2004; Quintero et al., 2010; Zhang et al., 2010) , which constitutes major fraction of atmospheric bioaerosols (Ansari et al., 2015; Bauer et al., 2008; Bowers et al., 2013; Fröhlich-Nowoisky et al., 2009; Sesartic and Dallafior, 2011; Spracklen and Heald, 2014). The meteorological parameters exhibited ~~pronounced~~significant diurnal variations during the *high bio* period, where RH decreased to a level ~~(~~~~~60 – 80~~%),~~%, which is considered to be favorable for release of the fungal spores (Jones and Harrison, 2004; Santarpia et al., 2013). During the *dusty* and *clean* ~~period~~periods, the persistence of high RH values in the range of ~90 – 100%, ~~however,~~ might have inhibited the active wet discharge of fungal spore (Schumacher et al., 2013~~;)~~) thus resulting in the weak diurnal variation in N_F. Unlike N_F, N_T remained nearly flat without any pronounced diurnal variations during three distinct focus periods (Fig. ~~S16~~S18). The corresponding diurnal cycle of FBAP mass concentration and 3D size distributions for three focus periods are shown in Fig. ~~S17.~~S19. M_F exhibited similar diurnal patterns to that of N_F during three distinct focus periods. M_T ~~as like~~ and N_T remained flat during the *dusty* period, ~~however~~but exhibited slightly pronounced diurnal pattern during the *clean* and the *high bio* period between 09:00 hrs. and 16:00 hrs. (Fig. ~~S18~~S20).

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3.~~5~~3 SEM images

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Figure ~~14~~11 shows ~~the exemplary~~representative SEM images of different biological particle types often observed during the SW monsoon season at Munnar. The details about the sampling techniques, instrument used, etc. for obtaining these bioaerosol images are discussed in details by Valsan et al., (2015). Note that these images are ~~not~~ being presented here ~~for any quantitative purpose and to draw any specific scientific conclusions but mainly~~ to showcase the particle types consistently observed throughout the measurement period~~. As seen from the SEM images majority~~ and not for quantitative purposes. The presence of ~~the~~mineral dust and sea salt particles ~~are mostly likely fungal spores. Based on their distinct morphology the spores in Fig. 14a-c~~confirms marine influence of the air mass sampled. Many particles observed by SEM were most likely ~~appeared to be of~~ Basidiospores. The appearance of small protuberances on ~~the surface~~their surfaces suggests that the ~~spore in~~spores (e.g. Fig. ~~14a~~11a and c) most likely belonged to the *Hydnaceae* family (Grand and Vandyke, 1976; Valsan et al., 2015). The Basidiospores shown in Fig. ~~14b~~11b and c were seen in abundance in all the samples collected during the campaign. Some of the spores observed appeared to be coated with salt particles (Fig ~~14e~~11e) and might have been carried from a distant source by the SW monsoon winds. The spores shown in Fig ~~14~~11 (d and f) most likely appeared to be spores of Ascomycota division. The particle shown in Fig. ~~14g~~11g was most likely a mineral dust particle sampled during high dusty episode. Similar particles of varying size during the *dusty* episode were consistently observed during SEM analysis. Fig ~~14h~~11h and ~~i~~11i shows the images of the typical sea salt particles observed during samples collected at Munnar during measurement campaign when wind predominantly came from Westerly/Southwesterly direction travelling over Indian Ocean and Arabian Sea.

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3.64 Meteorological Correlations

The results obtained with UV-APS data analysis during the campaign at Munnar were plottedcorrelated with respect to meteorological parameters to investigate factors responsible for bioaerosol release and their variations in the atmosphere.

3.4.1 Impact of wind direction

The wind rose diagrams scaled by N_F, D_g, and $D_{g,T}$ were also prepared for entire measurement period and three distinct focus periods. These plots are in a way similar to the traditional wind rose diagram (Fig. S19S21) except that, instead of wind speed, they are scaled by characteristic FBAP and TAP parameters, which indicate the frequency of occurrence of respective parameter with respect to wind direction (Sherman et al., 2015). As can be seen from Fig. S19S21, predominant wind direction during entire campaign was Westerly/Southwesterly with frequency of occurrence of about ~90%. The wind speed broadly ranged between 2 – 5 m s^{-1} with no prominent diurnal variations. The overall wind direction and back trajectory analysis (Fig. 1) shows that the sampled air masses may have had their originoriginated over the Indian Ocean thereafterand then turning eastward after crossing the equator and travelling several hundred kilometers over Arabian Sea before reaching the observational site (Fig. 1). The predominant wind pattern during the *dusty* (>95% frequency of occurrence; 2 – 6 m s^{-1}) and *clean* periods (~100 frequency of occurrence; 2 – 6 m s^{-1}) was Westerly/Southwesterly. Whereas during the *high bio* period only ~50% of the time winds came from Westerly/Southwesterly direction and rest comprised the stagnant and calmof relatively slower (0 – 2 m s^{-1}) winds from all other directions with highest contribution of northerly winds (Fig. S19). .S21).

Wind rose diagram scaled by FBAP number concentration is shown in Fig. 15.12. During the entire campaign the predominant wind showed that ~85% of the time FBAP concentration

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occurred in the range of 0 – 0.05 cm^{-3} (Fig. ~~15a~~12a) occasionally exceeding 0.05 cm^{-3} and was contributed by Westerly/Southwesterly winds. The occurrence of relatively low FBAP concentration during entire campaign is ~~consistent~~coincidental with low concentration occurrence during the *dusty* (0 – 0.05 cm^{3}; >90% frequency of occurrence) and the *clean* (<0.01 cm^{3}; ~90% frequency of occurrence) periods. During the *high bio* period the FBAP concentration, >0.05 cm^{3} exhibited ~40% frequency of occurrence of which ~50% was contributed by predominant wind from the North and the Northwest.

Similarly the wind rose diagram scaled by geometric mean diameter (D_g) of $dN_F/d\log D_a$, is shown in Fig. ~~16.~~13. The average size of the FBAP particles associated with Westerly/Southwesterly winds when analyzed for the entire ~~the~~ campaign ranged between 2 – 4 µm of which ~65% of the time D_g was observed to be ≤3 µm. During the three ~~distinct~~ focus periods the frequency of occurrence of FBAP particles in the higher size range (3 – 4 µm) was strongly associated with the Westerly/Southwesterly winds (Figs. ~~16b~~13b – d). The corresponding wind rose diagram scaled by geometric mean diameter of $dN_T/d\log D_a$ ($D_{g,T}$) is shown in Fig. ~~S20.~~S22. During entire measurement campaign the frequency of occurrence of $D_{g,T}$ in the size range of 0.8 – 0.9 µm was ~70% and was mostly associated with Westerly/Southwesterly winds. During the *dusty* period, particles in the size range of 0.8 – 0.9 µm diameter contributed for >95% frequency of occurrence for the entire size range, whereas during *clean* period ~20% occurrence of the particles in the size range other than 0.8 – 0.9 µm were also observed. On the other hand during the *high bio* period total particles in the size range 0.5 – 0.8 µm were observed with ~50% frequency of occurrence ~~constituted by varying wind patterns~~ mostly dominated by northerly winds.

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The FBAP concentration exhibited strong dependence on the wind direction for this observational site. During the *high bio* period the increase in frequency of occurrence of FBAP number concentrations >0.1 cm^{-3} coincided with ~~stagnant~~lower wind speed coming from the North and Northwest (Fig. ~~17a~~14a). During the *high bio* period, as ~~like~~in the case of the *dusty* and *clean* periods, the predominant wind pattern was Westerly/Southwesterly, ~~however~~but, with relatively low frequency of occurrence as compared to the other two periods. To have ~~the~~ a better understanding of the relative contribution of wind direction in high FBAP number concentration during the *high bio* period, ~~we prepared the~~ separate wind rose diagrams for FBAP concentration >0.1 cm^{-3} and <0.1 cm^{-3} as shown in Fig. ~~17.~~14. The FBAP number concentration >0.1 cm^{-3} was associated with ~~calm~~lower wind speed (0 – 1 m s^{-1}; ~80% frequency of occurrence) and predominant Northerly winds (Fig. ~~17a~~14a) as opposed to high wind speed (2 – 5 m s^{-1}) and predominant Westerly/Southwesterly winds for the FBAP number concentration <0.1 cm^{-3} (Fig. ~~17b~~14b). The ~~calm northerly~~Northerly winds with lower wind speed coming over from densely vegetated regions in combination with local FBAP sources during the *high bio* period could be the strong reason for the ~~built~~build up resulting in higher FBAP number concentration during this episode, whereas, Westerly/Southwesterly winds were consistently marked by very low FBAP number concentration mostly owing to higher wind speeds. Further, it might also due to the fact that the air masses arriving at observational site ~~originated~~originating over cleaner marine region, which may be potential but weak source of bioaerosols combined with possible wash out/wet deposition due to persistent rainfall during the transport. ~~Nominally the~~The frequency of occurrence of larger particles (3 – 4 µm) during Westerly/Southwesterly winds was high compared to the Northerly winds, where particles were mostly of smaller size (1 – 3 µm). We hypothesize that during the Northerly wind the bioaerosols were mostly comprised of

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Basidiospores, which is consistent with SEM images obtained during measurement period. Frohlich-Nowoisky et al., (2012) reported that, a region with dominant prevalence of marine air masses ~~have~~has larger proportions of Ascospores and in contrast, the continental air masses exhibit higher proportions of Basidiosppres. However, due to technical difficulties associated with sampling we could not establish the ~~fact that~~identity of the spores observed at this observational site during Westerly/Southwesterly winds~~ were dominated by Ascospores~~ and these details will be addressed in follow up studies. The corresponding wind rose scaled by $D_{g,T}$ obtained from dN_T/dlog D_a is shown in Fig. ~~S21~~S23.

As shown in Tab. 5 the wind speed was observed to ~~be~~correlate negatively ~~affecting the~~with N_F during entire measurement period and is consistent with previously reported studies (Hameed et al., 2012; Almaguer et al., 2013; Lyon et al., 1984; Quintero et al., 2010). The increased N_F concentration levels during ~~calm and stagnant~~lower wind speed might indicate that observed bioaerosols were dominated by the local source rather than transported from longer distances (Sadys et al., 2014; Hara and Zhang, 2012; Bovallius et al., 1978; Maki et al., 2013; Prospero et al., 2005; Creamean et al., 2013) as lower wind speed may actually increase emission of some specific type of spores (Huffman et al., 2012; Jones and Harrison, 2004; Troutt and Levetin, 2001; Kurkela, 1997).

3.~~6.1~~4.2 Correlation with relative humidity and temperature

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Correlation coefficient derived between N_F and relative humidity averaged over the entire campaign is shown in Fig. ~~18~~15 and corresponding R^2 values for three distinct focus periods are shown in Tab. 5. In general an increase in N_F concentration with increasing relative humidity was observed with moderate correlation coefficient (R^2=0.58). Depending upon the type of bioaerosols, geographical location, and local climate, N_F has shown varied dependence on

Formatted: Justified, Space After: 0 pt relative humidity and the precise response of the spore concentration to relative humidity is difficult to characterize. For example, a number of studies have shown that spores of genus like *Cladosporium*, *Alternaria*, and *Epiccocum* are known to exhibit the negative correlation with relative humidity (Oliveira et al., 2010; Herrero et al., 1996; Kurkela, 1997; Oh et al., 1998; Healy et al., 2014); while on the other hand, other studies have also found these spores to be positively correlated with relative humidity (Quintero et al., 2010; Hjelmroos, 1993; Ho et al., 2005). ~~Whereas genus~~Genus like *Ustilago* and some other Basidiospores may ~~as well~~ exhibit strong positive correlation with relative humidity (Sabariego et al., 2000; Quintero et al., 2010; Ho et al., 2005; Calderon et al., 1995). ~~Further,~~ Ascospores concentrations are known to increase during and after rainfall (Burch and Levetin, 2002; Elbert et al., 2007; Hasnain, 1993; Hirst, 1953; Toutt and Levetin, 2001; Lyon et al., 1984; Oh et al., 1998) whereas Basidiospores exhibited a strong resemblance to the diurnal pattern of relative humidity (Li and Kendrick 1994; Hasnain 1993; Tarlo et al., 1979; Trout and Levetin 2001). Almaguer et al., (2013) have reported that in a tropical region, relative humidity has greater influence than temperature on the airborne spore counts and may be a pre-requisite for release of spores (Hollins et al., 2004). Thus, the combination of persistent threshold relative humidity (~60 – 95% as reported by Ho et al., 2005) and rainfall can cause the increase in the spore concentration and the excessive and persistent rain, however, tends to wash the spore out of the atmosphere further reducing their concentration levels (Burge 1986; Horner et al., 1992; Troutt and Levetin, 2001). Based on these arguments combined with observed meteorological conditions we expect that the bioaerosols reported here from Munnar mainly consisted of Basidiospores during the SW monsoon season as also evident from SEM images (discussed above). This is consistent with results reported by Valsan et al., (2015) where they found the dominant presence of dry air spora (*Cladosporium*) during relatively dry and warm weather from the same observational site. In general, N_F and N_F/N_T decreased with increasing wind speed (R^2=0.6 and R^2=0.78, respectively) indicating that wind speed may be one of the strong factors for observed high N_F concentrations at this site. As compared to previously reported correlation between N_F and meteorological parameters (Santarpia et al., 2013), the relations shown for this observational site appeared to be more robust and conclusive. ~~For example since the~~The variability ~~derived~~observed in N_T (N_T - $N_{T,min}$/ $N_{T,max}$- $N_{T,min}$; not shown here) was more consistent and high as compared to variability ~~derived~~observed in N_F (N_F – $N_{F,min}$/ $N_{F,max}$- $N_{F,min}$), which was more episodic and hence one would expect the weak correlation between N_T and meteorological parameters (Tab. 5).

~~On the other hand several~~Several studies have reported that in temperate regions, temperature is probably the most important meteorological parameter affecting the spore concentration (Levetin and Horner, 2002; Adhikari et al., 2006) with highest spore concentration during summer season (Emberlin et al., 1995; Hasnain, 1993; Herrero et al., 1996; Hjelmroos, 1993; Li et al., 2011; Schumacher et al., 2013). When the relation between temperature and spore concentration was investigated on ~~averaged~~the basis of diurnal ~~basis, however~~average, spore concentration have been observed to decrease with the increasing temperature (Burch and Levetin, 2002; Calderon et al., 1995; Sabariego et al., 2000; Schumacher et al., 2013; Trejo et al., 2013). Consistent with this trend, we have found significant negative correlation between N_F and temperature (R^2=0.65) averaged over the entire measurement period at Munnar. The correlation coefficient between N_F and temperature for three distinct focus periods is given in Tab. 5~~. The correlation coefficient between N_F/N_T~~ and ~~meteorological parameters in general yielded higher R^2 values. Note, however, that the interpretation presented here based on the correlation analyses performed between N_F~~are specific to this locality of sampling and ~~meteorological parameters were~~

~~intended~~may not ~~to generalize and extrapolate conclusions~~be extrapolated to ~~various~~ represent behavior in other ecosystems ~~(including~~ in the Indian region~~) and different seasons of the year (including non-monsoon in India) but were~~. These results were, however, presented to ~~take an opportunity to~~ formulate preliminary hypothesis about role of meteorological parameters in governing the ~~variaibilites~~variability of ~~bioaerosls~~bioaerosols specific to this observational site for the monsoon season~~ only~~.

4 Summary and Conclusions

~~During these maiden online measurements of biological aerosol particles we operated a~~A UV-APS was continuously operated during the SW monsoon season (~~1.~~June ~~– 21.~~1 - August~~) of~~ 21, 2014) at ~~a high-altitude site of~~ Munnar in the Western Ghats in Southern tropical India. The number and mass size distributions and corresponding concentrations of biological aerosol were quantified for three distinct focus periods namely *dusty*~~ period~~, *high-bio*~~ period~~, and *clean*~~ period~~, identified based on the prominent wind direction. ~~We have analyzed the three month time series of integrated coarse particle number and mass concentrations, as well as particle number and mass size distributions of both, the total and fluorescence biological aerosol particles.~~ Over the course of the entire measurement period the coarse particle number concentration of FBAPs varied in the range of 0.2 × 10^{-3} cm^{-3} to 0.63 cm^{-3} with an arithmetic mean value of 0.02 cm^{-3} (±0.02 cm^{-3}). This average concentration accounted for 0.04 – 53% (mean value 2.1%±4.05%) of the total coarse particle number concentration. The coarse particle mass concentrations of FBAPs varied in the range of 0.5 × 10^{-3} – 4.93 µg m^{-3} with an arithmetic mean (±standard deviation) value of 0.24 (±0.28) µg m^{-3}.

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~~The average FBAP concentration during the entire measurement period was found to be highest in June (0.03 cm^{-3}) and lowest in July (0.007 cm^{-3}).~~ The FBAP concentrations observed at Munnar during SW monsoon season ~~are~~were within the range but slightly on the lower side of the ~~bioaerosol~~ concentrations reported by previous researchers using various online and offline techniques~~. Numerous other studies~~ from ~~different part of the world have reported detailed description about observed biological aerosol particle number concentrations using offline and online techniques from various~~varying environments (Despres et al., 2007; Huffman et al., 2010, 2012; Adhikari et al., 2004; Bovallius et al., 1978; Bowers, et al., 2009, 2013; Lee et al., 2010; Matthias-Maser and Jaenicke, 1995; Matthias-Maser et al., 2000; Shaffer and Lighthart, 1997; Tong and Lighthart, 1999; Wang et al., 2007; Li et al., 2011; Hameed et al., 2009; Bauer et al., 2008; Schumacher et al., 2013; Gabey et al., 2010, 2011, 2013; Saari et al., 2015; Toprak and Schnaiter, 2013; Healy et al., 2014). For brevity, here we compare the number concentrations observed at Munnar only with number concentrations from varying environments ~~reported by previous researchers~~carried out using online measurements. Huffman et al., (2010) have reported coarse mode average FBAP number concentration from four months of measurement to be 0.03 cm^{-3}, which constituted ~4% of total coarse mode particles from a semi-urban site of Mainz in Central Europe. The median FBAP concentration during the wet season of pristine tropical Amazonian rainforest region was found be 0.07 cm^{-3}, which constituted ~24% of total coarse mode particle number concentration (Huffman et al., 2012). By analyzing the full one-year observations from Boreal forest in Hyytiala and pine forest in Colorado, Schumacher et al., (2013) reported highest FBAP concentration in summer of 0.046 cm^{-3} (constituting ~13% of total coarse mode particles) and 0.03 cm^{-3} (constituting ~8.8% of total coarse mode particles), respectively. Healy et al., (2014) reported the average FBAP concentration of ~0.01 ~~cm^{3}~~cm^{-3}

using the UV-APS measurements carried out with in the Killarney national park, Kerry situated in Southwest of Ireland. Gabey et al., (2013) by performing the measurements at a high altitude ~~cite~~site in central France reported averaged FBAP concentration of 0.012 cm^{-3} and 0.095 cm^{-3} using two-wavelength (280 nm and 370 nm respectively) single-particle UV-induced fluorescence spectrometer. Gabey et al., (2010) from tropical rainforest in Borneo, Malaysia reported that mean FBAP number fraction in the size range of 0.8 – 20 µm was ~55% and ~28% below and above the forest canopy, respectively. It is important to note, however, that the measurement results compared here were obtained from different instrumentation operating with different wavelength. ~~Nevertheless, the FBAP number concentrations observed under various environmental conditions are largely comparable to the FBAP number concentration observed at Munnar during SW monsoon season. Note that the relative contribution of FBAP number concentration to total coarse mode particles may show a strong spatial variability.~~

The average observed dN_F/dlog D_a exhibited a peak at ~3 µm, which was consistent even during distinct focus periods with slight quantitative variation in the FBAP number concentration. Such a consistency in the peak of dN_F/dlog D_a during entire measurement period ~~is an indication of the fact~~indicates that sources and type of bioaerosols did not exhibit considerable variability and diversity at Munnar during SW monsoon season. The peak observed in dN_F/dlog D_a in this study is consistent with range of the peaks published by previous researchers. At a semi-urban site in Central Europe the peak in dN_F/dlog D_a was observed at ~3 µm (Huffman et al., 2010). In pristine tropical rainforest region of Amazonia a peak in dN_F/dlog D_a was found at ~2.5 µm (Huffman et al., 2012). Whereas the peak in dN_F/dlog D_a at a boreal forest in Finland exhibited a strong seasonal dependence with different modes at ~1.5 µm, ~3 µm, and ~5 µm indicating differences in the bioaerosol sources (Schumacher et al., 2013). In the pine forest of Colorado the distinct peaks were observed at ~1.5 µm and ~5 µm (Schumacher et al., 2013). The mode at ~3 µm reported for Colorado is likely due to the fungal spore whose release mechanism is strongly governed by the combination of relative humidity and temperature (Huffman et al., 2010 and references therein).

On the diurnal scale a pronounced diurnal cycle with ~3 µm peak with a maximum concentration at ~06:00 hr. was observed when averaged over entire measurement period. This general pattern is consistent with previous studies reporting the early morning peak in FBAP concentration for various environmental conditions (Healy et al., 2014; Huffman et al., 2012; Schumacher et al., 2013; Toprak and Schnaiter, 2013). The early morning peak~~, which in the present case appears to be strongly governed by the diurnal variations in relative humidity, is most likely to be~~ was contributed by Basidiospores as their release in the atmosphere is strongly coupled with relative humidity (Adhikari et al., 2006; Burch and Levetin, 2002; Hasnain, 1993; Healy et al., 2014; Ho et al., 2005; Huffman et al., 2012). This is also consistent with the SEM images shown and discussed above.

The meteorological parameters were observed to correlate significantly with FBAP concentration at Munnar. ~~When investigated on a daily averaged basis (24 hr), however, no significant correlation between N_F and meteorological parameters except moderate negative correlation with precipitation was observed. During the entire measurement campaign, except on few occasions no significant variations in temperature and relative humidity was observed. This in combination with persistent rainfall resulting in the wash out/wet deposition of biological aerosol particles might have caused such a weak correlation for a daily averaged (24 hr) analysis. On a diurnal scale, however, a significant correlation between N_F and meteorological parameters was observed.~~ We observed that N_F followed the similar diurnal trend to that of relative humidity and was anti-correlated with temperature. As reported by previous studies from selected locations (Huffman et al., 2013; Schumacher et al., 2013; Prenni et al., 2013; Hirst 1953) we did not observe any sharp increase in N_F concentration immediately after or during rainfall. We hypothesize that the spore built-up and release of certain species can happen only at certain threshold relative humidity (Jones and Harrison, 2004). ~~Under~~Our results indicate that under the dry environmental conditions where relative humidity levels rarely attain such threshold required for fungal spore release can cause the strong built up of fungal spores inside fungal bodies. Under these conditions precipitation can cause the relative humidity levels to increase up to threshold required for fungal spore release in combination with mechanical splashing due to raindrops, and can cause the sudden and sharp increase in spore concentrations. On the contrary, like in present case, the ~~incessant persistence of~~persistent high humidity conditions can cause the continuous release of the spore without an opportunity for built-up of fungal spores in fungal body to be released during rainfall. It is also reported that persistent high levels of relative humidity can inhibit the sporulation (Schumacher et al., 2013) further considerably reducing the spore release. More detailed measurements are required from the regions where relative humidity persistently remains low (<60%) for extended amount of time and experiences sudden rainfall. The correlation between N_F and wind speed was found to be strongly negative. Since majority of the spore release was dominated by the local sources, the ~~stong~~strong winds coming over from West/Southwest direction, which were relatively clean, might have caused the dilution of air mass thus reducing the spore concentration.

Overall, the long-term measurements reported in this manuscript showed the quantitative and qualitative agreement with previously reported studies. The emissions and abundance of biological aerosol particles in Western Ghats air during monsoon season appeared to be closely

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linked to the ~~variaiblities~~variability in the meteorological parameters. ~~As reported by Huffman et al., (2012) and corroborated by the observations reported in this study, UV-APS is successfully able to detect the aerosol particles of biological origin, however, may pose certain limitations in scientific interpretation from the obtained data.~~ The scatter plot analysis carried out between N_F and N_T for submicron and ~~supermicron~~super-micron particles indicated that submicron particles at this observational site were also dominated by aerosol particles of biological origin, thus indicating the lowest possible interference from particles of anthropogenic origin known to exhibit the fluorescence at the prescribed wavelength used in UV-APS. Hence, given observational site can be termed as relatively pristine while under the influence of SW monsoon season. This emphasizes the need to perform similar measurements under different land-use type during same season over Indian region. The contrasting characteristics of this observational site associated with pollution and interference of non-biological aerosol particles in fluorescence will be discussed in follow up studies. We propose ~~and intend to take forward these~~more studies by means of performing simultaneous online measurements of biological aerosol particle ~~number concentrations in high time and size resolution~~ under contrasting environments during distinct meteorological seasons over Indian region. ~~This future work~~These measurements could be supplemented with advanced offline measurement techniques including SEM analysis, DNA analysis, and fluorescence microscopy of the samples collected in parallel with the measurements. We believe that such a comprehensive approach over Indian region would be helpful in understanding the possible tight coupling between aerosol and hydrological cycle especially during monsoon. This could also help to better understand the implication of biological aerosols on crops and human health where agricultural industry has the major share in GDP to cater the need of 18% of the world's total population.

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Table 1: List of frequently used acronyms and symbols with units.

Symbol	Quantity, Unit
D_a	Aerodynamic diameter, µm
D_g	Geometric midpoint diameter of fluorescent particles
$D_{g,T}$	Geometric midpoint diameter of total particles
DNA	Deoxyribonucleic acid
FBAP	Fluorescent biological aerosol particle
He-Ne	Helium-Neon
ITCZ	Inter Tropical Convergence Zone
Lpm	Liters per minute
M_F	Integrated mass concentration of fluorescent particles, µg m^{-3}
M_T	Integrated mass concentration of total particles, µg m^{-3}
Nd:YAG	Neodymium-doped yttrium Aluminum garnet
NE	Northeast
N_F	Integrated number concentration of fluorescent particles, cm^{-3}
N_T	Integrated number concentration of total particles, cm^{-3}
PAH	Polycyclic aromatic hydrocarbon
PBAPs	Primary Biological Aerosol Particles
RH	Relative Humidity
SEM	Scanning Electron Microscopy
SW	Southwest
TAP	Total Aerosol Particle
TSP	Total Suspended Particle
UV-APS	Ultraviolet Aerodynamic Particle Sizer
λ	Wavelength, nm

Number		June	July	August	Campaign
N_T (cm^{-3})	Mean	2.66	1.54	0.96	1.77
	Median	2.45	1.48	0.73	1.44
N_F (cm^{-3})	Mean	0.03	0.007	0.015	0.017
	Median	0.02	0.006	0.007	0.01
N_F/N_T (%)	Mean	0.03	0.01	0.03	0.02
	Median	0.01		0.01	0.01
Mass		**June**	**July**	**August**	**Campaign**
M_T (µg m^{-3})	Mean	10.61	6.15	4.15	7.17
	Median	9.58	5.55	2.8	5.57
M_F (µg m^{-3})	Mean	0.42	0.11	0.18	0.24
	Median	0.33	0.09	0.1	0.15
M_F/M_T (%)	Mean	0.09	0.03	0.08	0.06
	Median	0.04	0.02	0.03	0.03

Table 2: Integrated number concentrations and mass concentrations of coarse TAP and FBAP (~1–20 µm): arithmetic mean and median for each month and for the entire measurement campaign

Sl No:	Location	Land Use	~~Meaurement~~Measurement Period	Season	Instrument	FBAP Number Concentration	Total Number Concentration	Number Ratio (%)	Reference
1	Mainz, Central Europe	Semi-urban	Aug-Dec, 2006		UVAPS	3 x 10^{-2} cm^{-3}	1.05 cm^{-3}	4	Huffman et al., 2010
2	Central Amazonia rainforest	Tropical rainforest	Feb-Mar, 2008		UVAPS	7.3 x 10^{-2} cm^{-3}	0.33 cm^{-3}	24	Huffman et al., 2012
3	Manchester, UK	Urban	December, 2009		WIBS-3	2.9 x 10^{-4} cm^{-3} (FL1)	1.38 x 10^{-2} cm^{-3}	2.1	Gabey et al., 2011
						5.2 x 10^{-4} cm^{-3} (FL2)		3.7	
						1.1 x 10^{-5} cm^{-3} (FL3)		7.8	
4	Central France	Rural	22 Jun-3 July, 2010		WIBS-3	1.2 x 10^{-2} cm^{-3} (280 nm)			Gabey et al., 2013
						9.5 x 10^{-2} cm^{-3} (370 nm)			
5	Helinski, Finland	Urban	Feb, 2012 (Winter)	Winter	BioScout	1 x 10^{-2} cm^{-3}		23	Saari et al., 2015
			June-Aug, 2012 (Summer)	Summer		2.8 x 10^{-2} cm^{-3}		6	
				Summer	UVAPS	1.3 x 10^{-2} cm^{-3}		8	
6	~~Colarado~~Colorado, USA	Pine forest	June-July,2011	Dry period	WIBS-3			5.8	Crawford et al., 2014
				Wet Period	WIBS-4			15.2	
7	Finland	Rural forest	August,2009 - April,2011	Spring	UVAPS	1.5 x 10^{-2} cm^{-3}	0.43 cm^{-3}	4.4	Schumacher et al., 2013
				Summer		4.6 x 10^{-2} cm^{-3}	0.45 cm^{-3}	13	
				Fall		2.7 x 10^{-2} cm^{-3}	0.41 cm^{-3}	9.8	
				Winter		0.4 x 10^{-2} cm^{-3}	0.47 cm^{-3}	1.1	

	Colorado , USA	Rural, semi-arid	2011-2012	Spring	UVAPS	1.5×10^{-2} cm^{-3}	0.73 cm^{-3}	2.5	
				Summer		3×10^{-2} cm^{-3}	0.44 cm^{-3}	8.8	
				Fall		1.7×10^{-2} cm^{-3}	0.28 cm^{-3}	5.7	
				Winter		0.53×10^{-2} cm^{-3}	0.2 cm^{-3}	3	
8	Karlsruhe, Germany	Semi-rural	April 2010 - April 2011		WIBS - 4	3.1×10^{-2} cm^{-3}	0.583 cm^{-3}	7.34	Toprak and Schnaiter., 2013
9	Nanjing, China	Sub-urban	Oct-Nov, 2013	Autumn	WIBS-4		13.1 cm^{-3}		Yu et al., 2016
						0.6 cm^{-3} (FL1)		4.6	
						3.4 cm^{-3} (FL2)		25.3	
						2.1 cm^{-3} (FL3)		15.6	

Table 3: Comparison with other online measurements carried out under various environmental conditions across the globe.

Number		Dusty	Clean	~~HighBio~~High Bio
N_T (cm^{-3})	Mean	4.2	1.27	1.78
	Median	4.36	1.15	1.4
N_F(cm^{-3})	Mean	0.02	0.005	0.05
	Median	0.019	0.004	0.038
N_F/N_T	Mean	0.01	0.01	0.05
	Median			0.03
Mass		Dusty	Clean	~~HighBio~~High Bio
M_T (µg m^{-3})	Mean	16.34	5.12	7.7
	Median	16.84	4.28	5.85
M_F (µg m^{-3})	Mean	0.36	0.08	0.58
	Median	0.33	0.05	0.47
M_F/M_T	Mean	0.02	0.03	0.12
	Median	0.02	0.01	0.08

Table 4: Integrated number concentrations and mass concentrations of coarse TAP and FBAP (~1–20 µm): arithmetic mean and median for each focus period (Dusty, Clean and ~~HighBio~~High Bio).

	Campaign			Dusty			Clean			High Bio		
	N_T	N_F	N_F/N_T	N_T	N_F	N_F/N_T	N_T	N_F	N_F/N_T	N_T	N_F	N_F/N_T
RH	-0.64	0.58	0.85	-0.25		0.18	-0.66	-0.01	0.13	-0.64	0.5	0.68
Temperature	0.45	-0.65	-0.82	0.34	-0.04	-0.25	0.78	0.02	-0.2	0.43	-0.68	-0.83
Wind Speed	0.4	-0.6	-0.78	0.09	-0.18	-0.31	-0.18	-0.27	0	0.3	-0.61	-0.74

Table 5: R^2 values for correlation between meteorological parameters (RH, Temperature and Wind Speed) and N_T, N_F and N_F/N_T during the entire campaign and each focus periods.

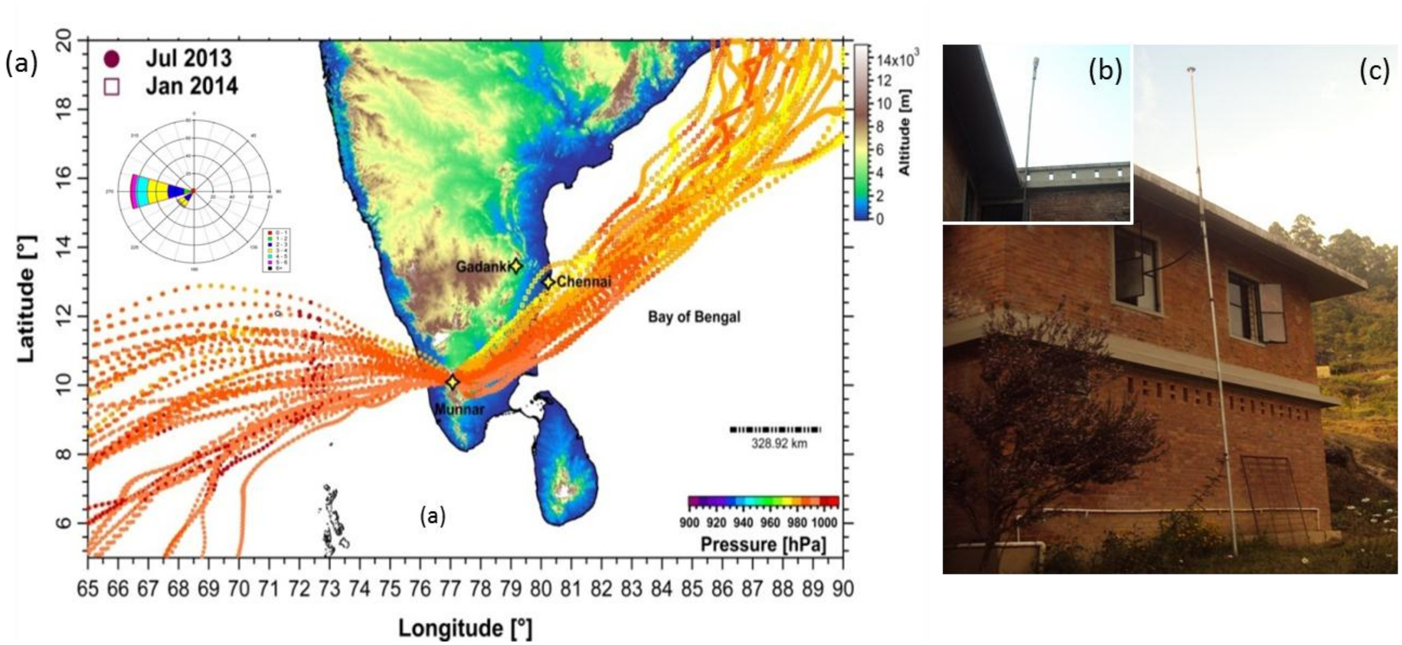
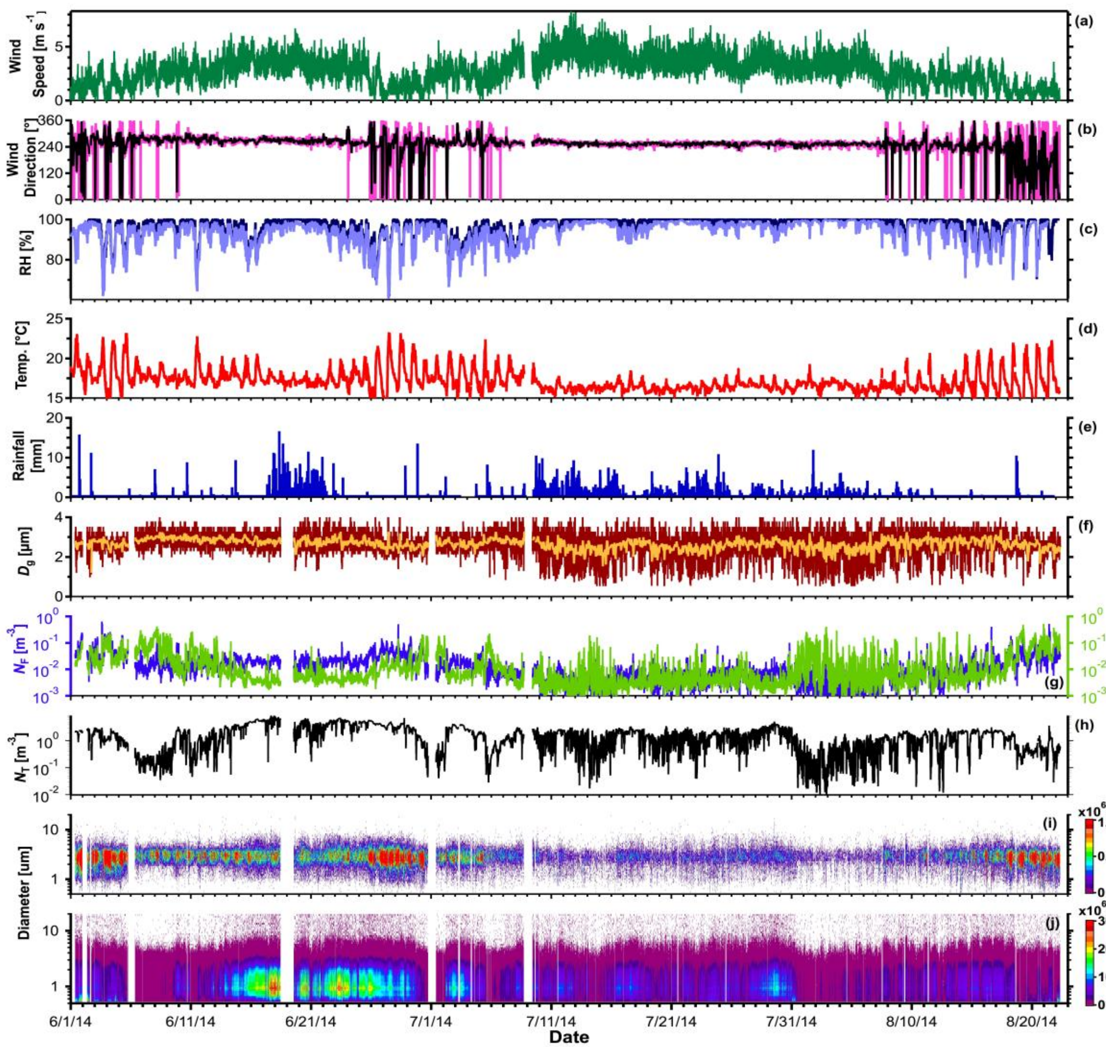


Figure 1: Location of measurement site Munnar (10.09°N, 77.06°E; 1605 m amsl – above mean sea level) located in the Western ~~Ghat~~Ghats mountain range in Southern tropical India with 10 days back trajectories (HYSPLIT, NOAA-ARL GDAS1 model; start height 50 m above ground level; starting time 23:30 local time) illustrating the distinct and contrasting wind patterns during two contrasting seasons; Southwest monsoon season (representative month Jul) and ~~Northeast monsoon~~Winter season (representative month Jan) when field measurement ~~campaign was~~campaigns were carried out. It is evident that predominant wind pattern during Southwest monsoon season was Westerly/Southwesterly bringing the clean marine influx~~. Also shown in inset is~~ as also evident from the wind rose diagram ~~prepared using the data obtained~~shown in inset(a). The meteorological parameters were recorded using the ~~ultrasonic~~ weather station ~~(a).~~installed close to the inlet system (b). The inlet system prepared for sampling the air using Ultraviolet Aerodynamic Particle Sizer (UV-APS) for bioaerosol number size distribution measurement~~. Inset shows the arrangement made for installing the ultrasonic weather station (b).~~ (c). The map shown is color-coded by topography (meters~~).~~


Wind Speed [m s-1]
(a)
Wind Direction [°]
(b)
RH [%]
(c)
Temp. [°C]
(d)
Rainfall [mm]
(e)
Dg [µm]
(f)
NF [m-3]
NF/NT
(g)
NT [m-3]
(h)
Diameter [um]
(i)
x10^6 m-3
(j)
6/1/14
6/11/14
6/21/14
7/1/14
7/11/14
7/21/14
7/31/14
8/10/14
8/20/14
Date

) and trajectories are color-coded by atmospheric pressure level (hPa)

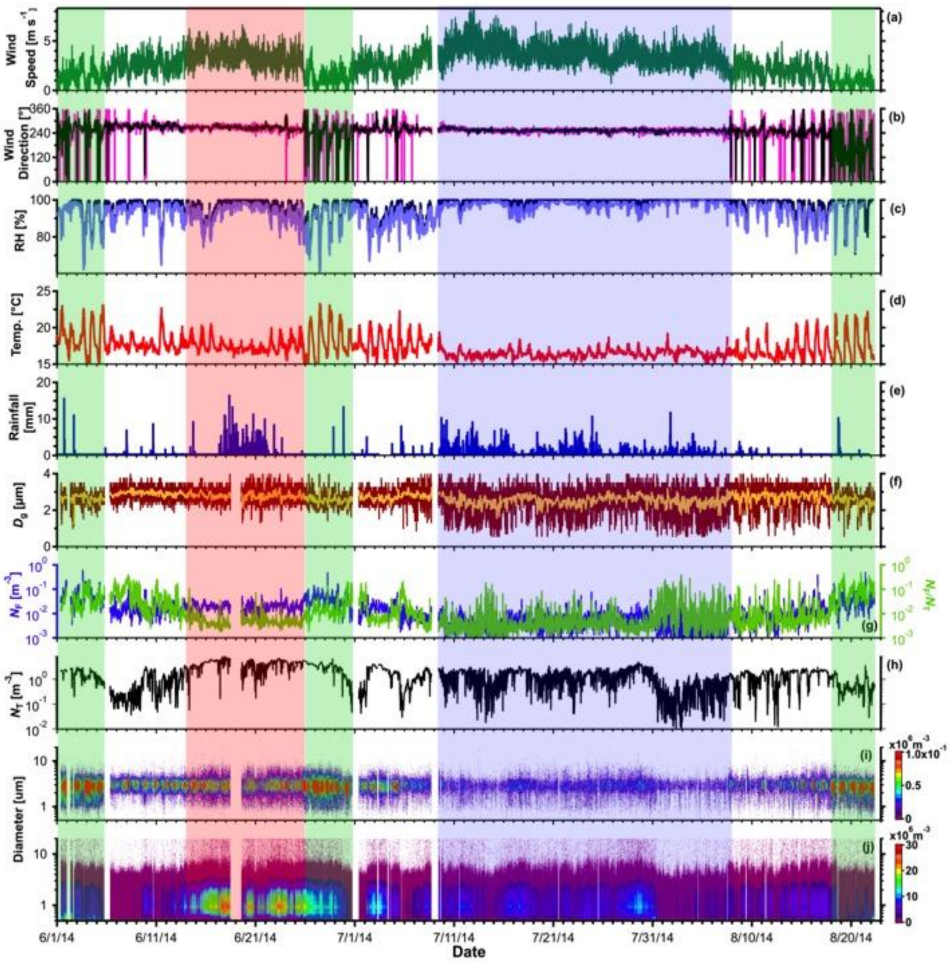


Figure 2: Time series of measured meteorological parameters, parameters derived from FBAP and total particle number size distribution measurements using UV-APS: (a) wind speed, (b) wind direction: five minutes average (magenta) and one hour average (black), (c) relative humidity, (d) temperature, (e) rainfall, (f) geometric mean diameter (D_g) five minutes average (dark red) and one hour average (yellow), (g) FBAP number concentration (N_F; blue) and relative contribution of FBAP to TAP (N_F/N_T; green), (h) TAP number concentration (N_T), (i) a contour plot of FBAP number size distribution ($dN/d\log D_F$), and (j) a contour plot of TAP number size distribution ($dN/d\log D_T$). The shadowed block represents the different focus periods (red for dusty; green for high bio; blue for clean: please refer to text for more details).

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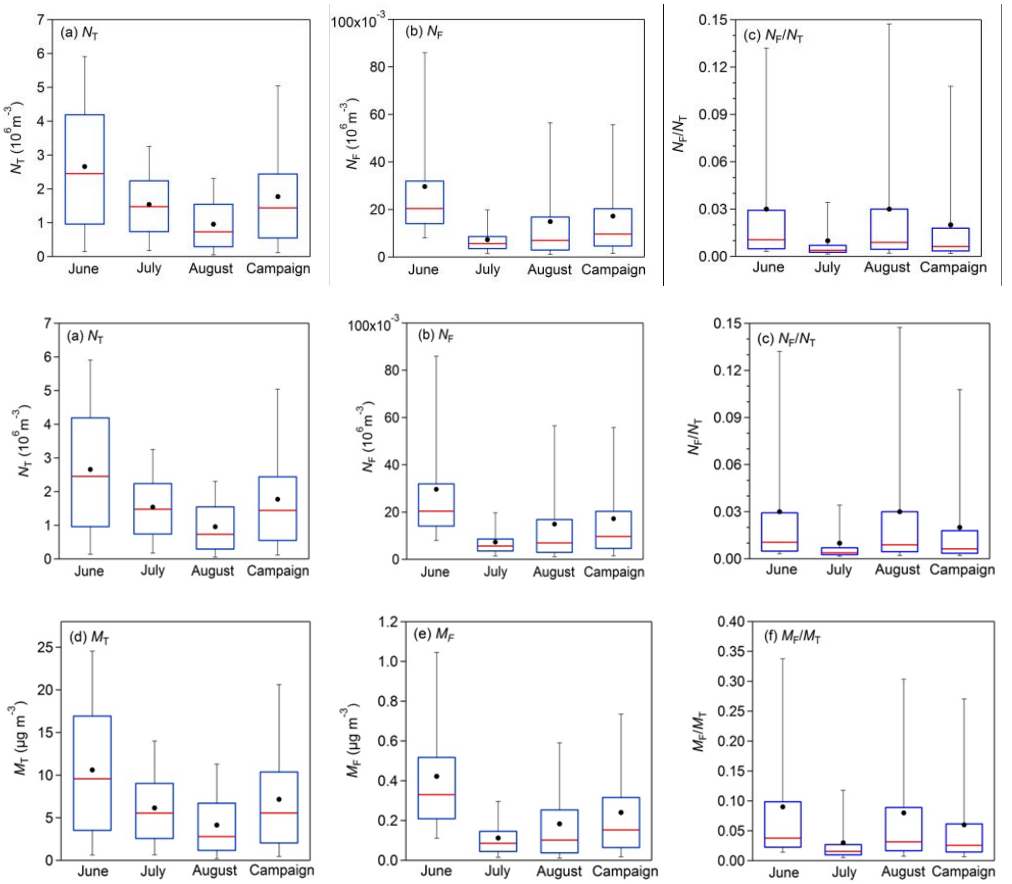

Figure 3: Statistical distribution of integrated (~1 – 20 µm) FBAP and TAP number and ~~contribution of N_F to N_T~~ mass and their ratios measured during each month (Jun – Aug) of SW monsoon season and averaged over the entire measurement campaign carried out at Munnar as box whisker plots: (a) TAP number concentration (N_T), (b) FBAP number concentration (N_F), ~~and~~ (c) contribution of FBAP number concentration to TAP number concentration (N_F/N_T~~).~~), (d) TAP mass concentration (M_T), (e) FBAP mass concentration (M_F) and (f) contribution of FBAP to TAP mass concentration (M_F/M_T).

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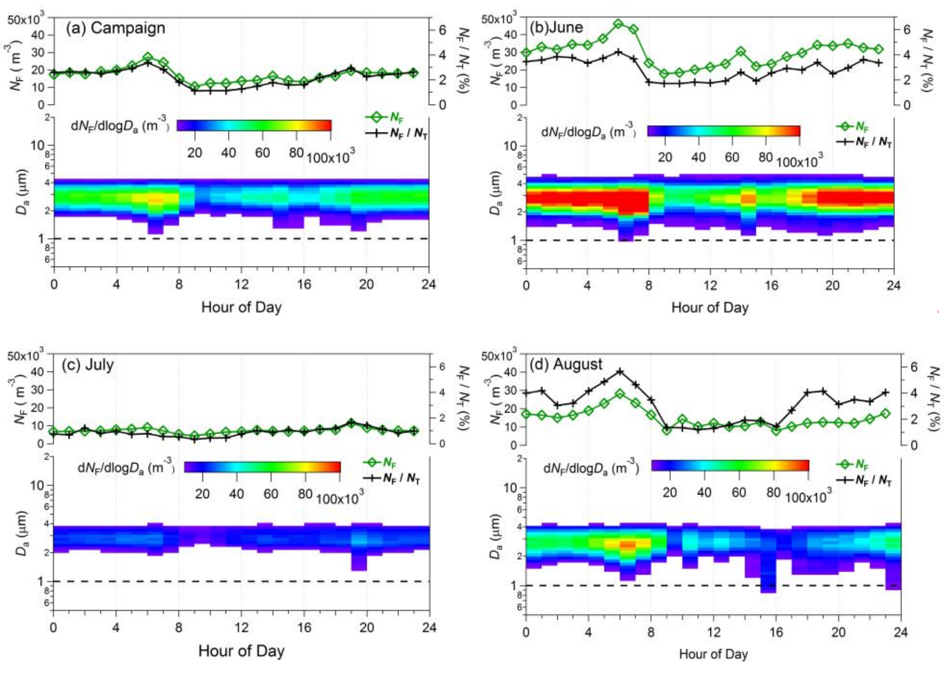
(a) Campaign
(b)June
(c) July
(d) August
N_F (m^{-3})
N_F / N_T (%)
dN_F/dlogD_a (m^{-3})
D_a (μm)
Hour of Day
N_F
N_F / N_T

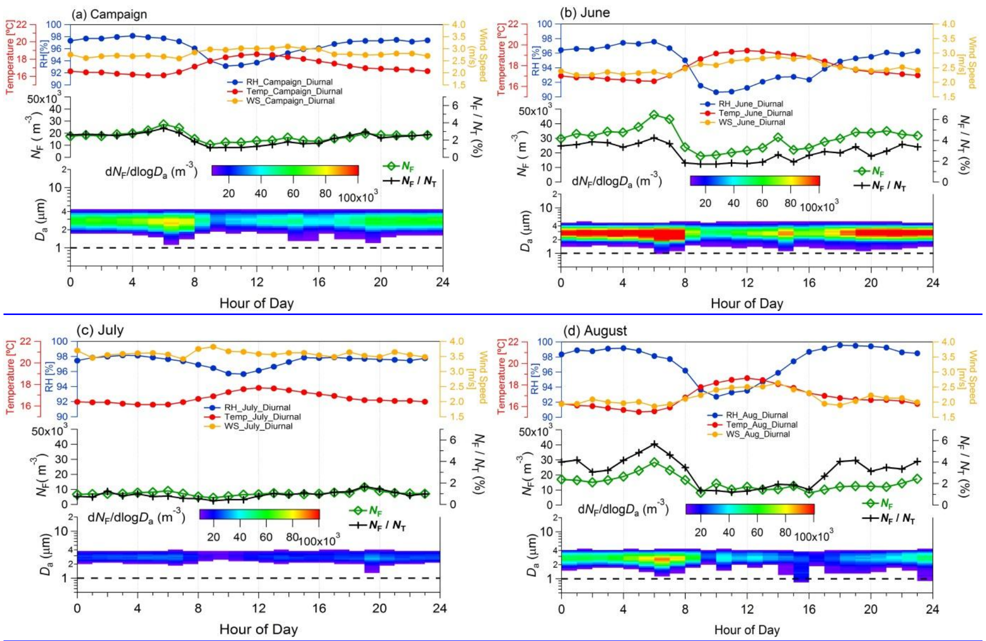

Figure 4: Diurnal cycles of observed meteorological parameters, FBAP number concentrations (N_F) and size distributions averaged over individual month of measurement and entire campaign (hourly ~~median~~mean values plotted against the local time of the day). Upper portion of each panel shows the observed meteorological parameters: relative humidity (%; blue), temperature (°C; red), and wind speed (m s^{-1}; orange on right axis). Middle panel shows integrated FBAP number concentration (~1 – 20 µm; N_F) on the left axis (green color) and FBAP fraction of TAP number (N_F/N_T) on the right axis (black color). Lower portion of each panel FBAP number size distribution (3-D plot) plotted against hour of the day on x-axis, aerodynamic diameter on y-axis and color is scaled for $dN_F/d\log D_a$ indicates the concentration. Dashed black lines in lower portion of the each panel at 1.0 µm shows the particle size cut-off diameter below which fluorescent particles were not considered as FBAP due to potential interference with non-biological aerosol particles. (a) averaged over entire campaign,(b) Jun, (c) Jul, and (d) Aug. Please refer to supplementary Figs. for corresponding TAP plots.

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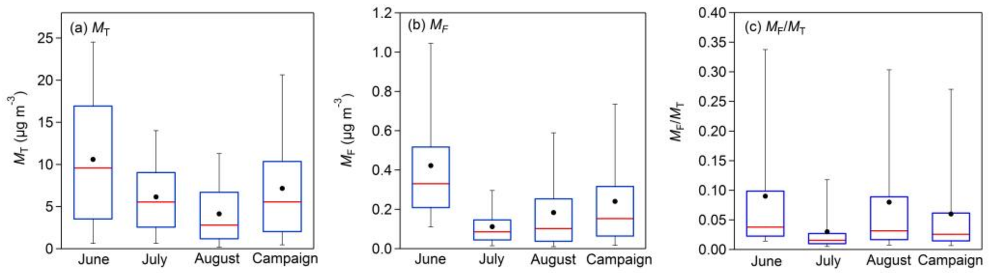


Figure 5: Same as Fig. 3 but for integrated (~1 – 20 µm) FBAP (M_F) and TAP (M_T) mass concentrations derived from number measurements by assuming unit density and shape factor.

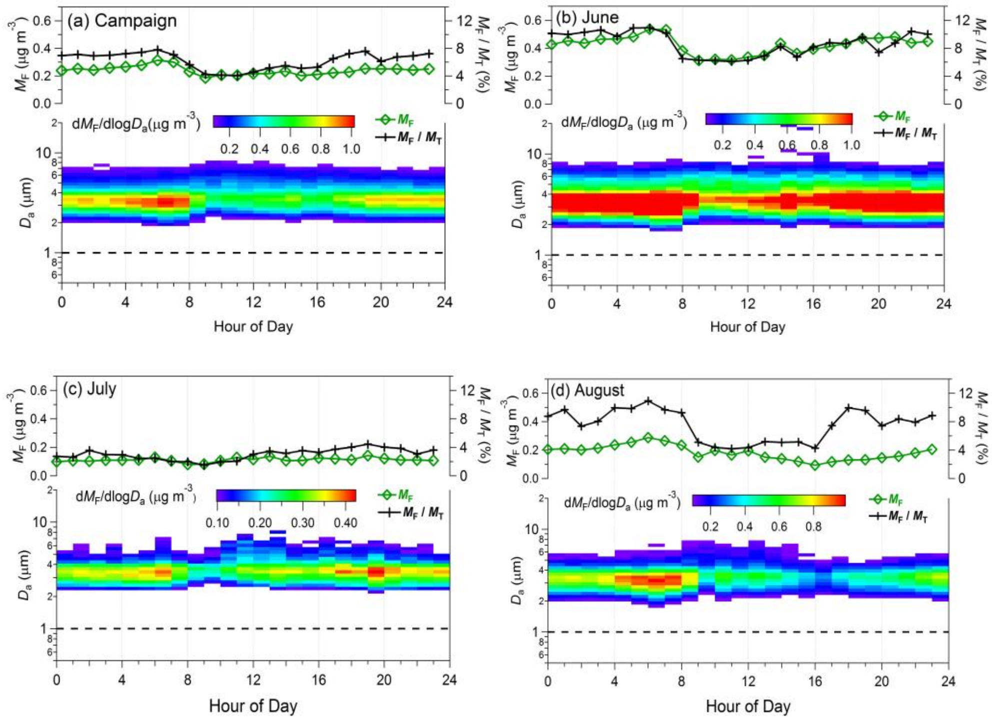


Figure 6: Same as Fig. 4 but representing the FBAP (M_F) mass concentrations.

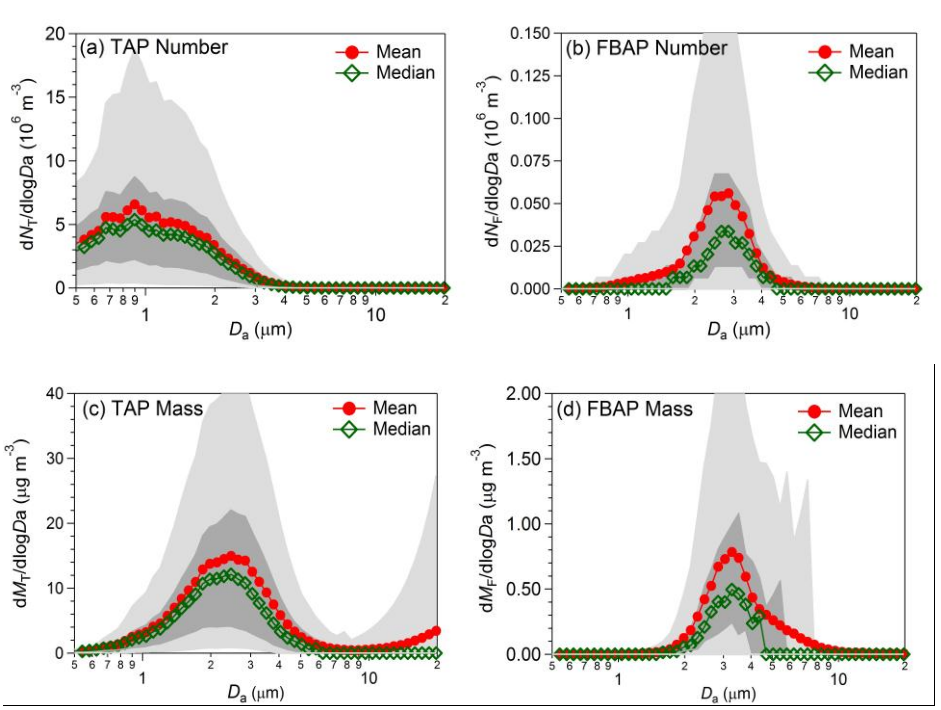


Figure ~~7~~5: Particle number size and unit-normalized number size and mass size distributions averaged over the entire measurement campaign carried out at Munnar. Lower and upper parts of dark and light shaded area represents the 5th, 25th, 75th, and 95th percentile respectively. (a) TAP number (dN_T/dlog D_a), (b) FBAP number (dN_F/dlog D_a), (c) total mass (dM_T/dlog D_a), and (d) FBAP mass (dM_F/dlog D_a).

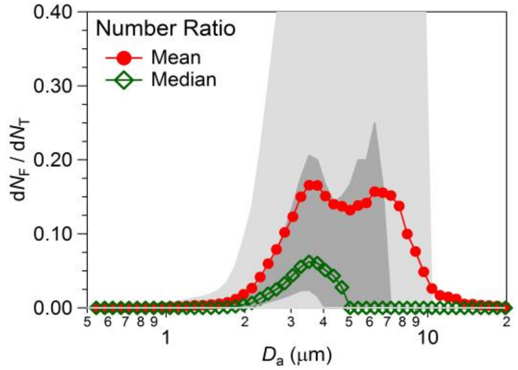


Figure ~~8~~6: Size distribution of FBAP to TAP ratio averaged over the entire measurement period carried out at Munnar (dN_F/dlog D_a= dM_F/dlog D_a). Lower and upper parts of dark and light shaded area represents the 5th, 25th, 75th, and 95th percentile respectively.

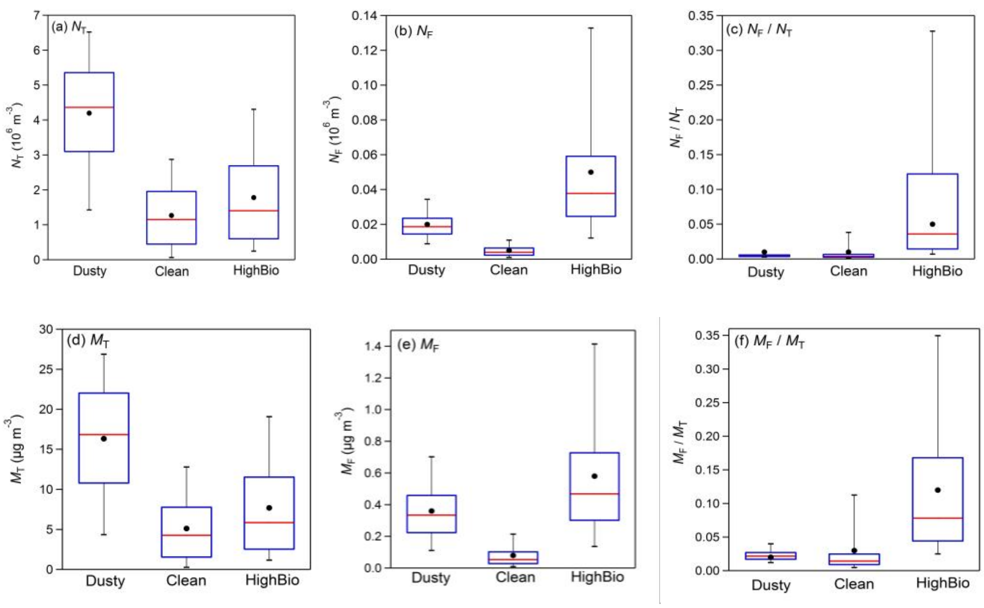
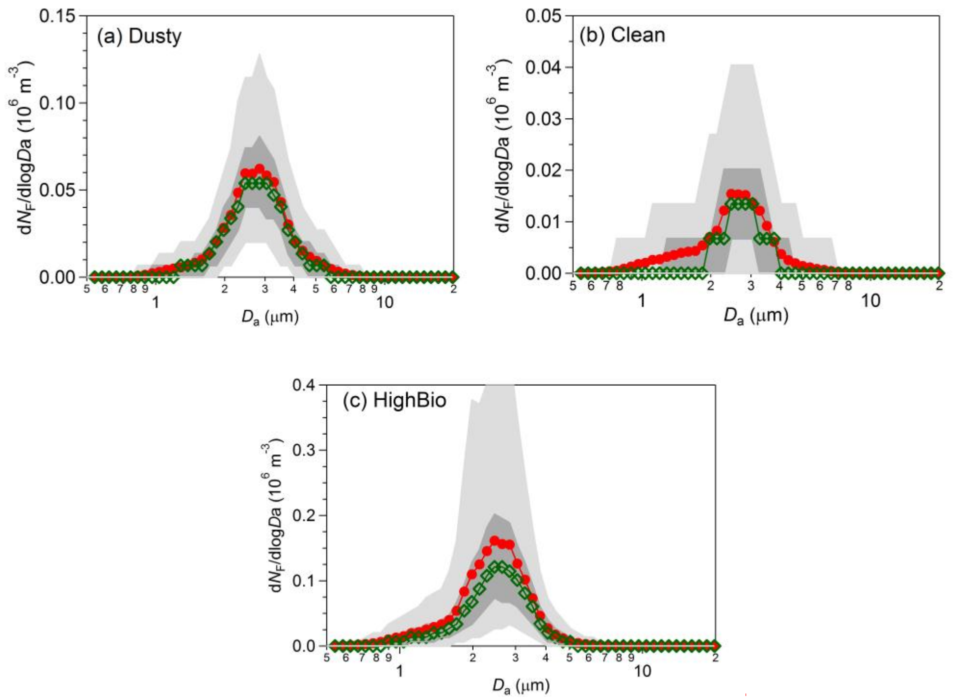


Figure ~~9~~7: Statistical distribution of integrated (~1 – 20 μm) FBAP and TAP number and mass contribution of N_F to N_T, and M_F to M_T averaged over each distinct focus periods (dusty, clean, and high bio; please refer to the text for definitions related to each focus period) measurements carried out at Munnar as box whisker plots: (a) TAP number concentration (N_T), (b) FBAP number concentration (N_F), (c) contribution of FBAP number concentration to TAP number concentration (N_F/N_T), (d) TAP mass concentration (M_T), (e) FBAP mass concentration (M_F), and contribution of FBAP mass concentration to TAP mass concentration (M_T/M_F).



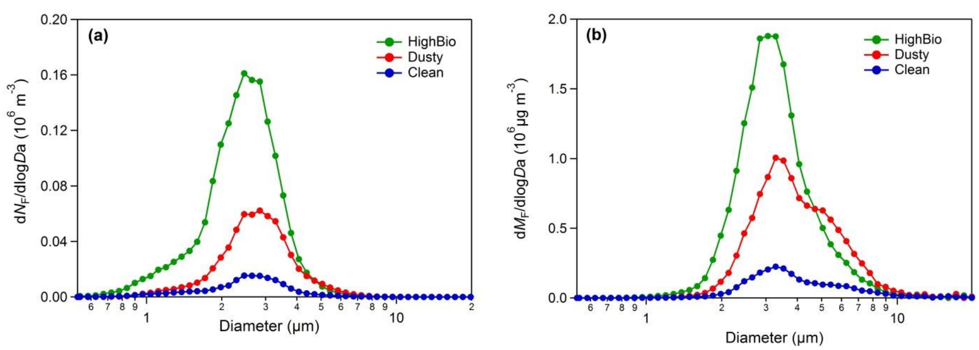


Figure ~~10~~8: FBAP number ~~size distributions~~ (dN_F/dlog D_a) and mass (dM_F/dlog D_a) size distribution averaged over each distinct focus periods during the measurement campaign carried out at Munnar. ~~Lower and upper parts of dark~~(a) number size distribution, and ~~light shaded area represents the 5th, 25th, 75th, and 95th percentile respectively. (a) dusty period ,~~ (b) ~~clean period, and (c) high bio period~~mass size distribution.

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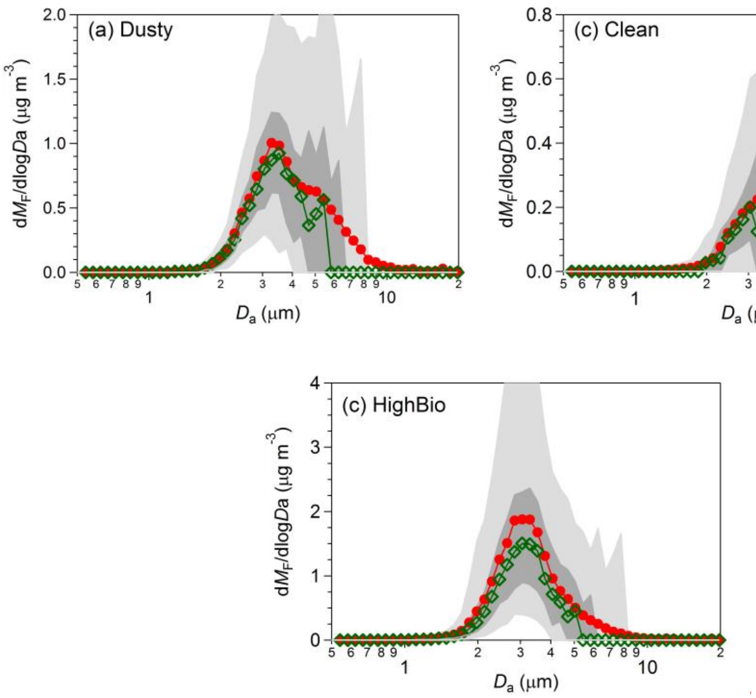

Figure 11: Same as Fig. 10 but representing FBAP mass size distribution (dM_F/dlog D_a).

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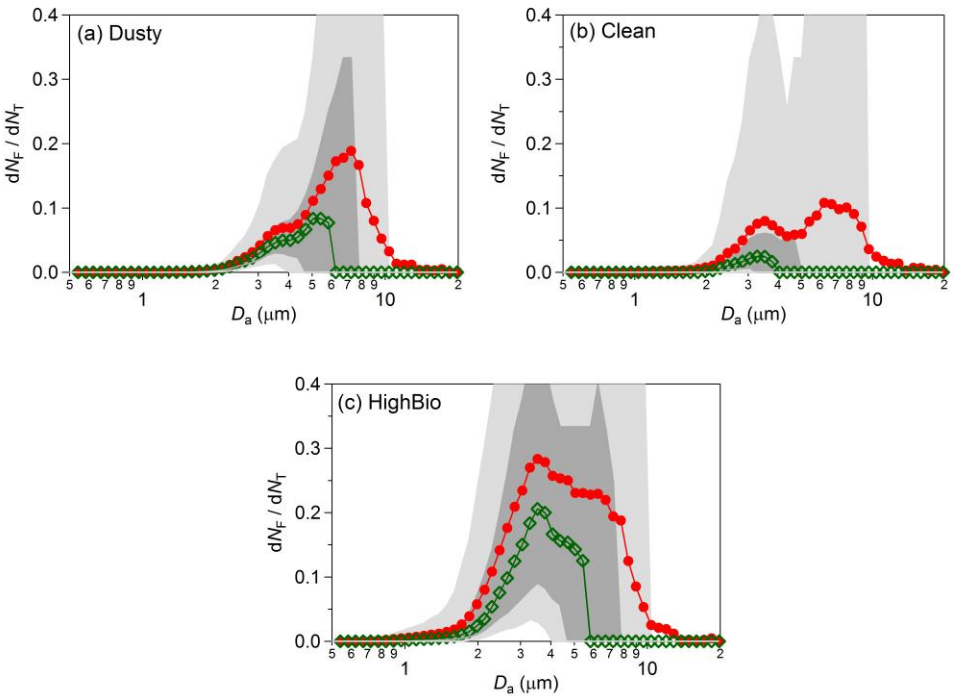

Figure ~~12~~9: Size distribution of FBAP to TAP ratio averaged over the each distinct focus periods during the measurements carried out at Munnar (dN_F/dlog D_a= dM_F/dlog D_a). Lower and upper parts of dark and light shaded area represents the 5th, 25th, 75th, and 95th percentile respectively: (a) dusty, (b) clean, and (c) high bio.

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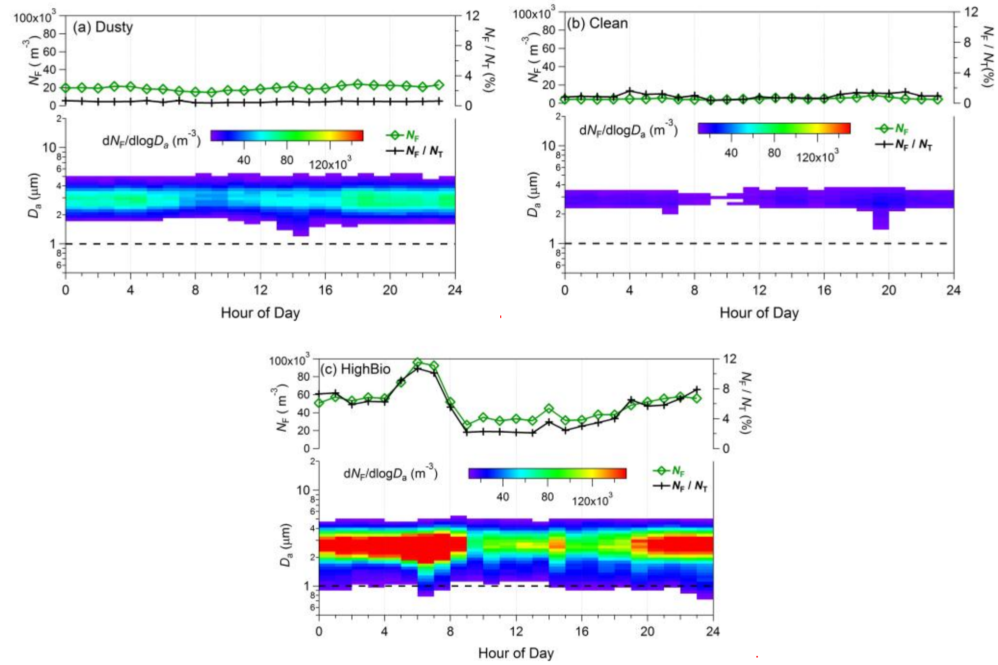
(a) Dusty
(b) Clean
(c) HighBio
N_F (m^{-3})
N_F / N_T (%)
dN_F/dlogD_a (m^{-3})
D_a (µm)
Hour of Day

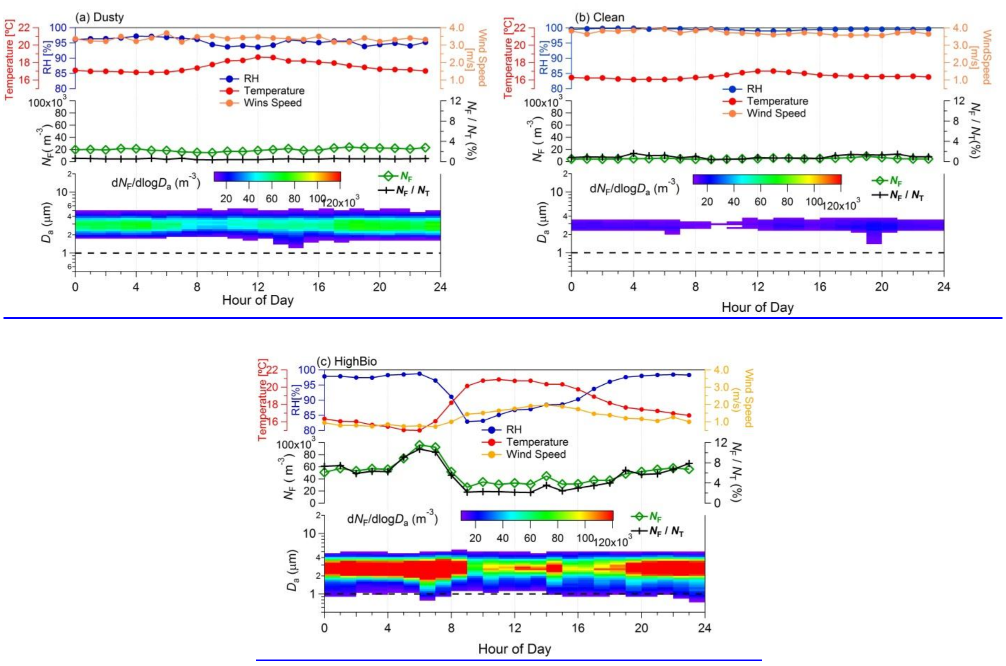
(a) Dusty
(b) Clean
(c) HighBio
Temperature [°C]
RH [%]
Wind Speed [m/s]
RH
Temperature
Wins Speed
Wind Speed
N_F (m^{-3})
N_F / N_T (%)
dN_F/dlogD_a (m^{-3})
D_a (µm)
Hour of Day

Figure 1310: Diurnal cycles of observed meteorological parameters, FBAP number concentrations (N_F) and size distributions averaged over each distinct focus period identified during measurements carried out at Munnar (hourly medianmean values plotted against the local time of the day). Upper portion of each panel shows the observed meteorological parameters: relative humidity (%; blue), temperature (°C; red), and wind speed (m s^{-1}; orange on right axis). Middle panel shows integrated FBAP number concentration (~1 – 20 μm; N_F) on the left axis (green color) and FBAP fraction of TAP number (N_F/N_T) on the right axis (black color). Lower portion of each panel FBAP number size distribution (3-D plot) plotted against hour of the day on x-axis, aerodynamic diameter on y-axis and color is scaled for DN_F/dlog D_a indicates the concentration. Dashed black lines in lower portion of the each panel at 1.0 μm shows the particle size cut-off diameter below which fluorescent particles were not considered as FBAP due to potential interference with non-biological aerosol particles. (a) dusty (b) clean, and (c) high bio. Please refer to supplementary Figs. for corresponding TAP plots.

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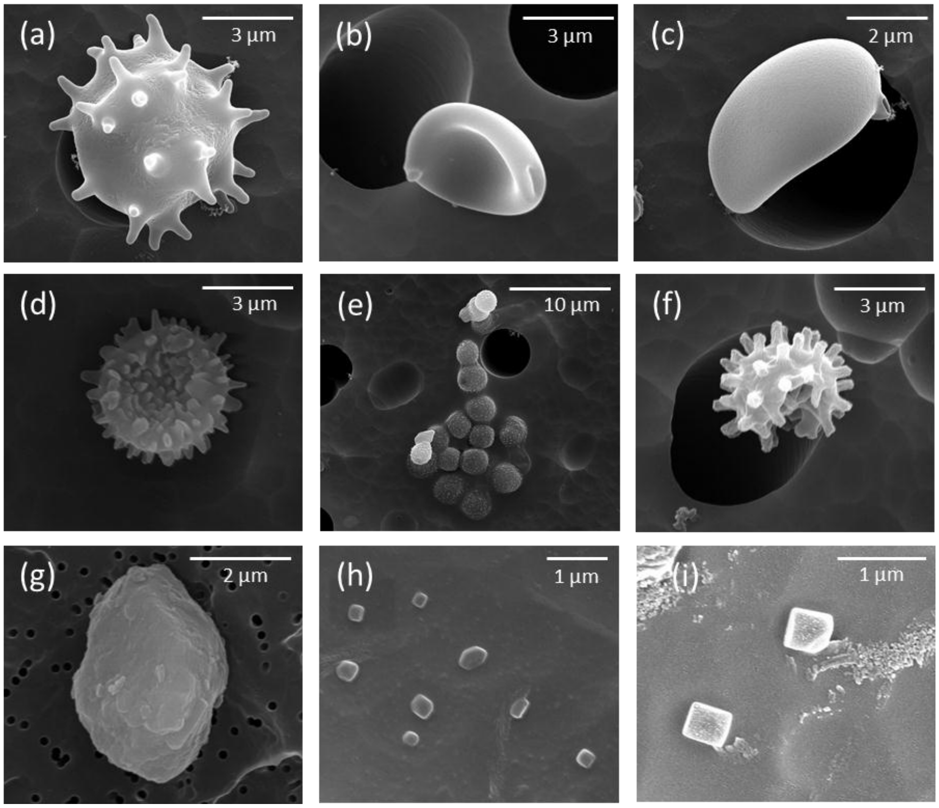

Figure ~~14~~11: Scanning electron microscope images of the exemplary aerosol particles (FBAP and TAP) observed during the campaign at Munnar. The scale bar is shown at the top right corner of each image.

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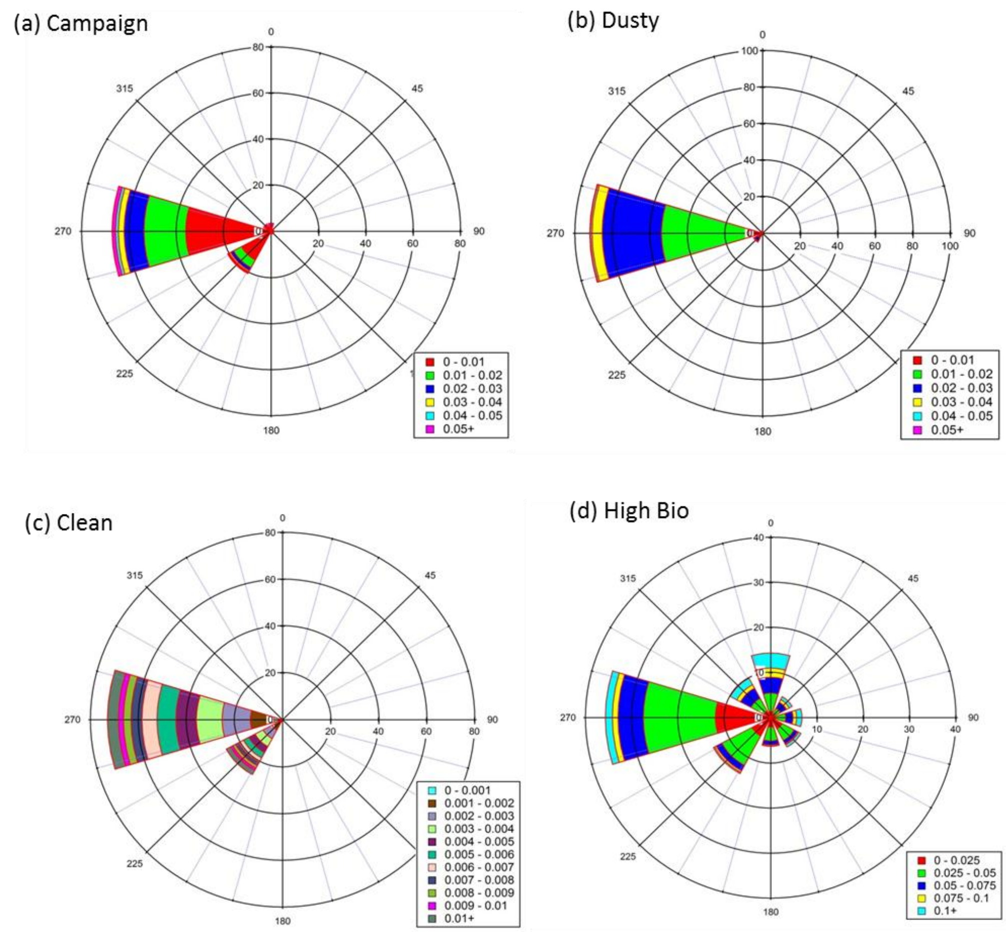
(a) Campaign
0
45
90
180
225
270
315
20
40
60
80
0 - 0.01
0.01 - 0.02
0.02 - 0.03
0.03 - 0.04
0.04 - 0.05
0.05+
(b) Dusty
0
45
90
180
225
270
315
20
40
60
80
100
0 - 0.01
0.01 - 0.02
0.02 - 0.03
0.03 - 0.04
0.04 - 0.05
0.05+
(c) Clean
0
45
90
180
225
270
315
20
40
60
80
0 - 0.001
0.001 - 0.002
0.002 - 0.003
0.003 - 0.004
0.004 - 0.005
0.005 - 0.006
0.006 - 0.007
0.007 - 0.008
0.008 - 0.009
0.009 - 0.01
0.01+
(d) High Bio
0
45
90
180
225
270
315
10
20
30
40
0 - 0.025
0.025 - 0.05
0.05 - 0.075
0.075 - 0.1
0.1+

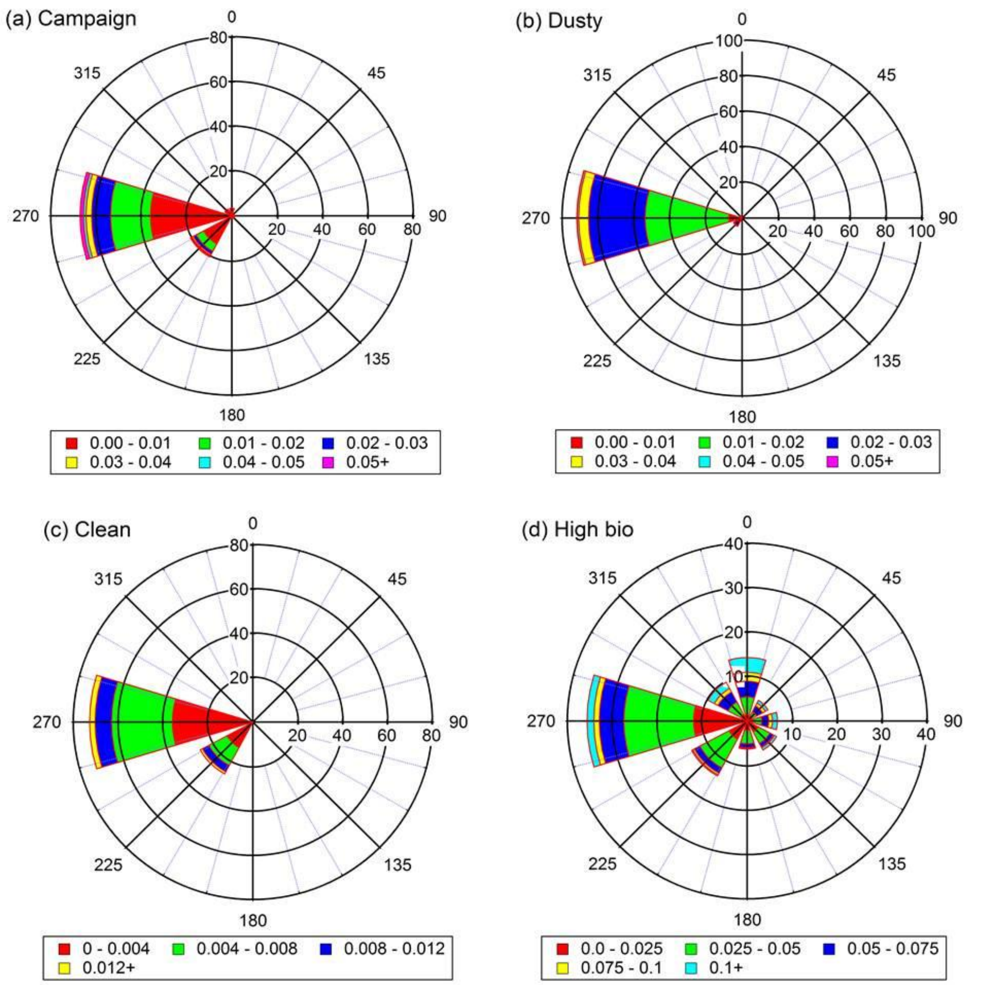

Figure ~~15~~12: Wind rose diagram scaled over FBAP number concentration (N_F). These diagrams in a way are similar to the traditional wind rose diagram except representing the N_F in this case instead of wind speed. These diagram can be nominally interpreted as followed: For example (a) shows that ~52% of frequency of occurrence of N_F concentration in the range of 0 – 0.~~001~~01 cm^{-3} was associated with Westerly/Southwesterly winds and on the contrary (d) indicates that out ~18% of frequency of occurrence of high concentration (N_F>0.1 cm^{-3}) ~16% was associated with Northerly/Northwesterly winds. (a) entire campaign, (b) dusty period, (c) clean period, and (d) high bio period. Note that non-uniform scale of each panel has unit of cm^{-3}.

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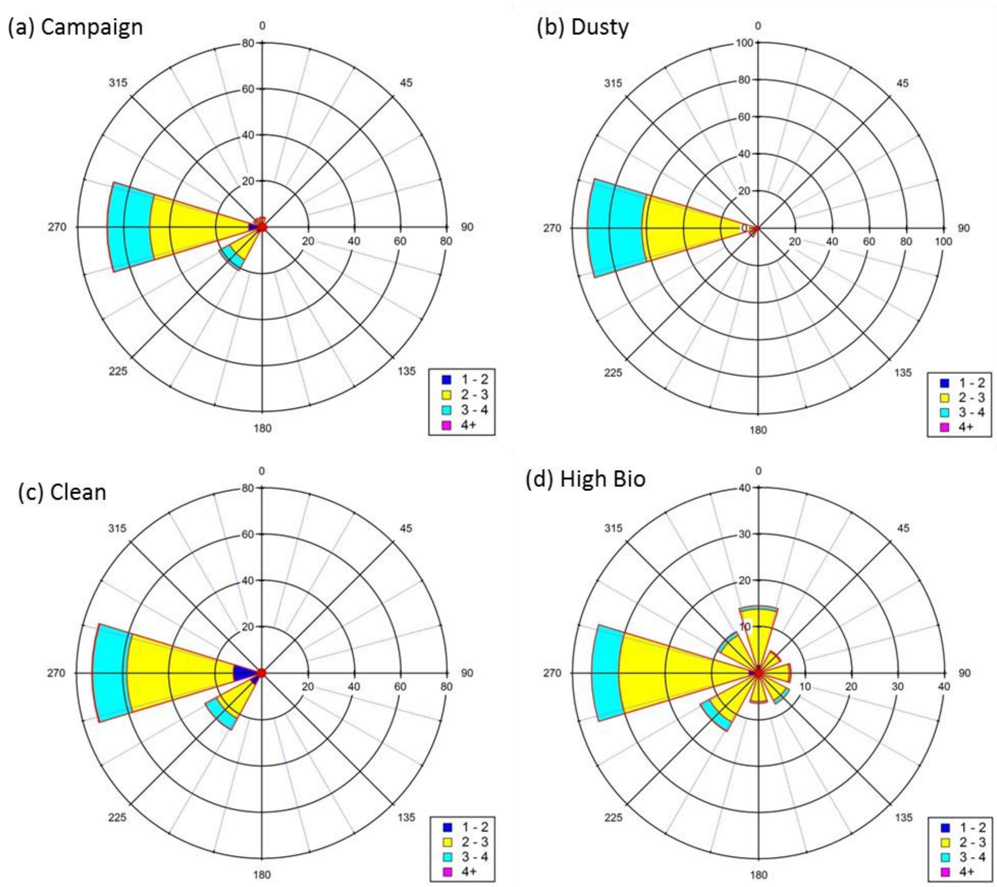


(a) Campaign
(b) Dusty
(c) Clean
(d) High Bio
0
45
90
135
180
225
270
315
1 - 2
2 - 3
3 - 4
4+

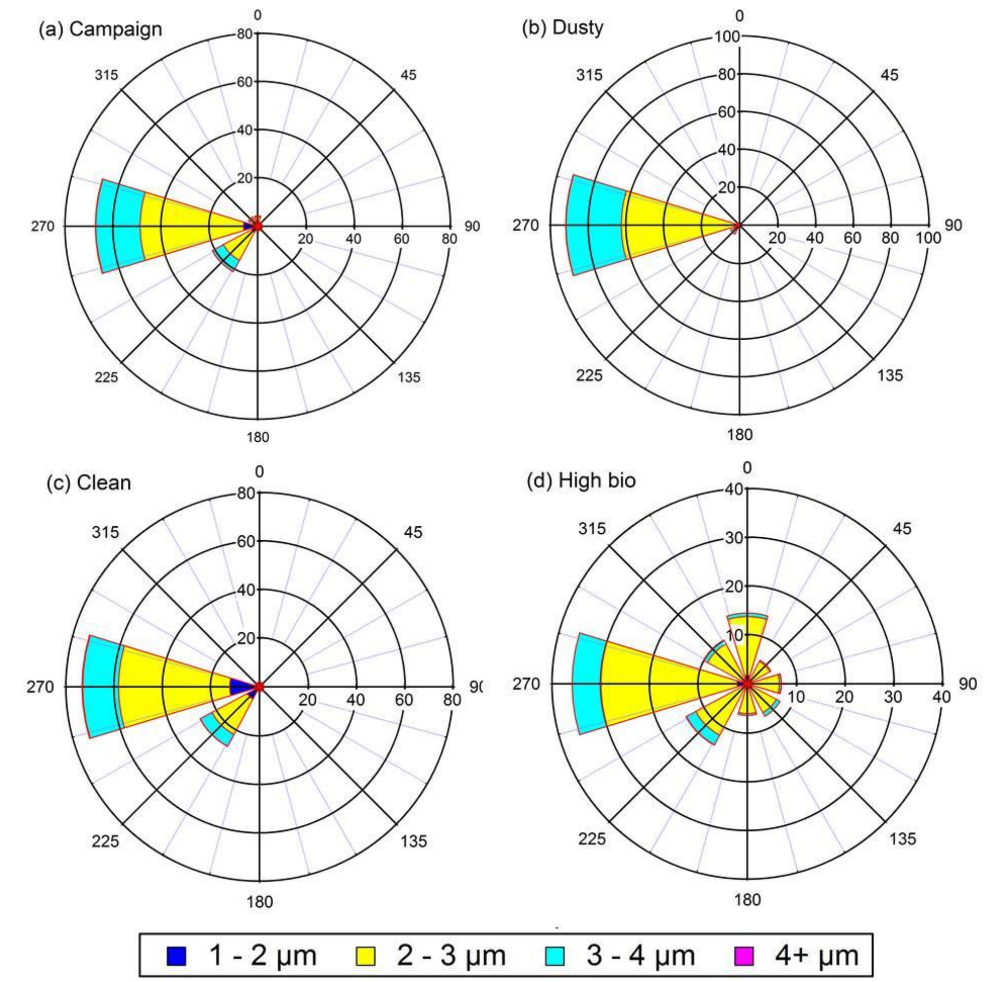
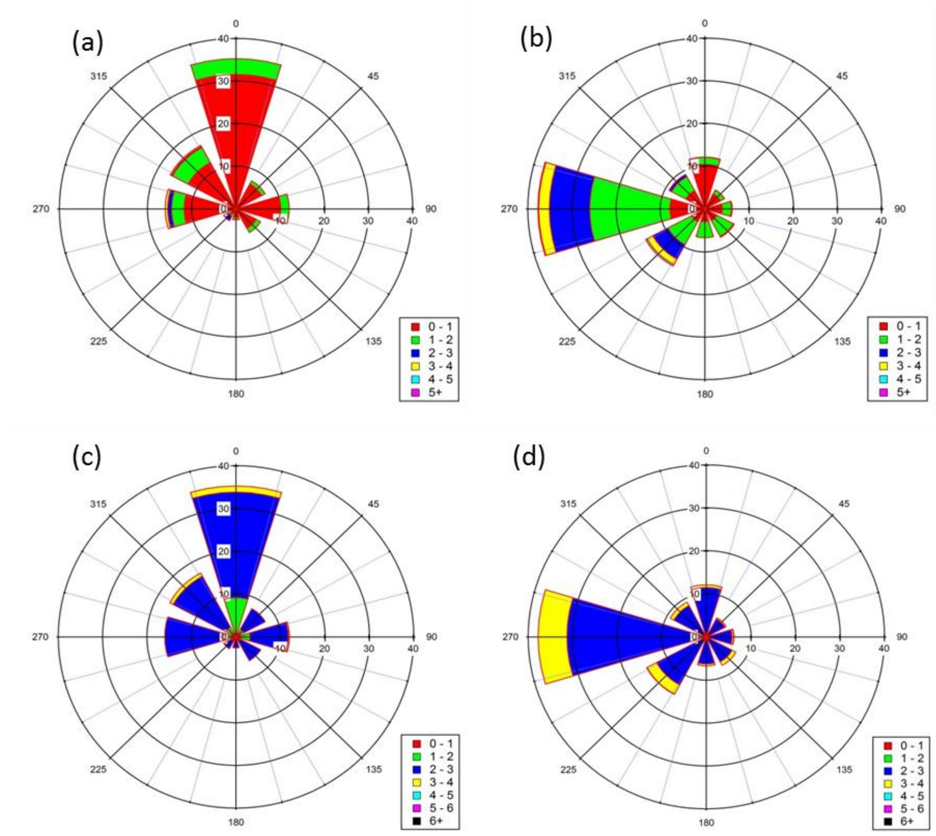


Figure ~~16~~13: Same as Fig. ~~18~~13 but scaled by geometric mean diameter (D_g) of dN_F/dlog D_a. (a) entire campaign, (b) dusty period, (c) clean period, and (d) high bio period.


(a)
(b)
(c)
(d)
0
45
90
135
180
225
270
315
10
20
30
40
0 - 1
1 - 2
2 - 3
3 - 4
4 - 5
5+
5 - 6
6+

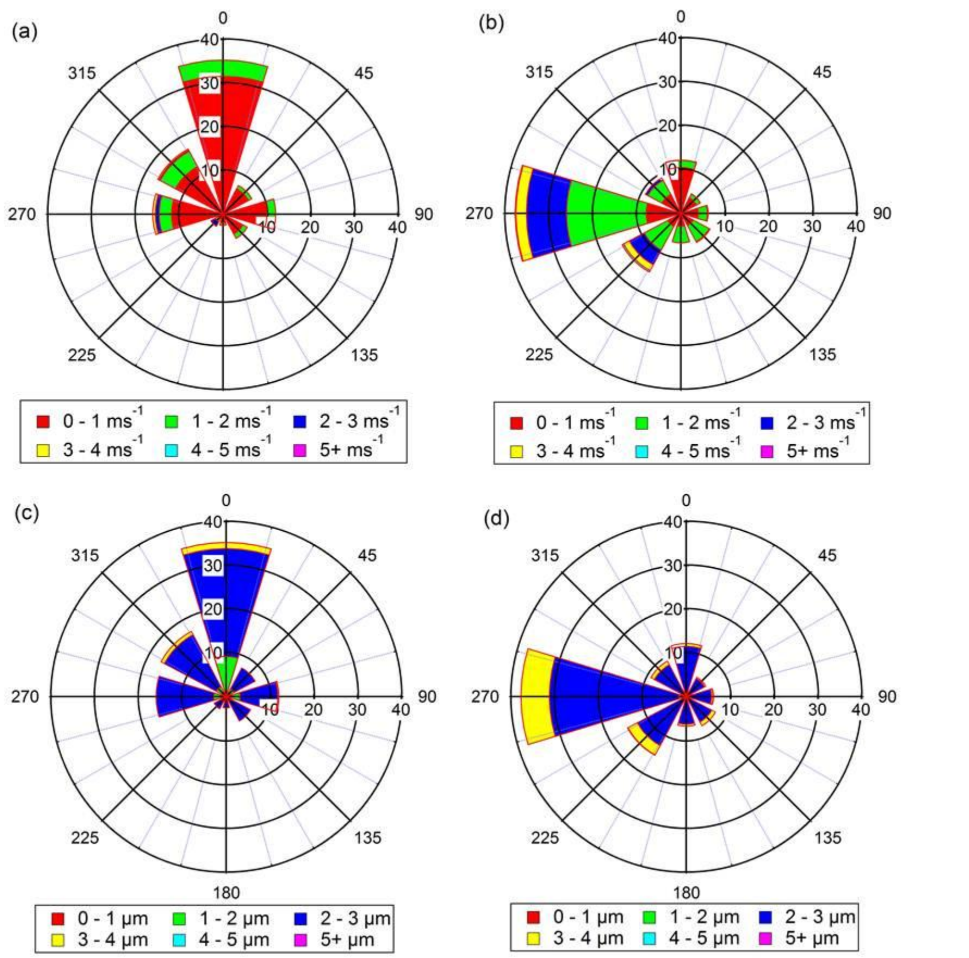


Figure ~~17~~14: Wind rose diagram scaled by wind speed and geometric mean diameter (D_g) of dN_F/dlog D_a. The figures have been separated for FBAP number concentration (N_F) range, N_F >0.1 cm^{-3} and N_F< 0.1 cm^{-3} observed during high bio period. For example: when, N_F>0.1 cm^{-3} ~60% of the time wind was observed to be in the range of 0 – 1 m s^{-1} (a) and ~94% of the time the geometric mean diameter (D_g) of dN_F/dlog D_a was in the range of 2 – 3 μm (c). On the other hand for N_F<0.1 cm^{-3} ~60% of the time wind was greater than 1 m s^{-1} (b), and ~80% of the time geometric mean diameter (D_g) of dN_F/dlog D_a was in the range of 2 – 3 μm (d).

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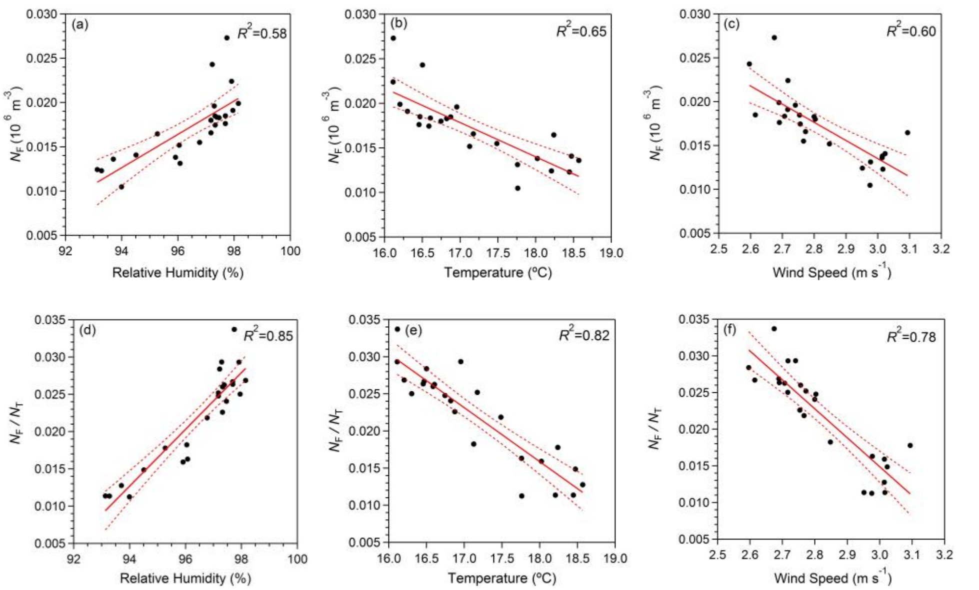

Figure ~~18~~15: Correlation between aerosol particle number concentrations (N_F, N_T, and N_F/N_T) and meteorological parameters (relative humidity, temperature, and wind speed). Red line indicates the best fit to the scattered points and dashed black line indicates the 95% confidence level obtained for the best fit.