

## 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 occasions and have shown few as exemplary images. This has now been clarified in the revised manuscript text (L637 – L639).

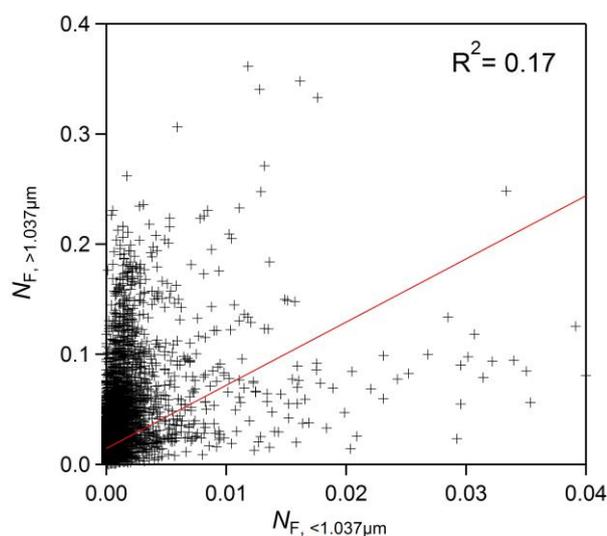
**[RC4]** The measurements have been done with the UV-APS. Regarding the data interpretation, it would be good to acknowledge and point out that the detection efficiency for fluorescent particles of the UV-APS is low especially below approximately 2 microns (e.g. Healy, et al., ACP2014, Saari et al., 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 2 $\mu$ m. 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 (<1 $\mu$ m) VS NF (>1  $\mu$ m) 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.

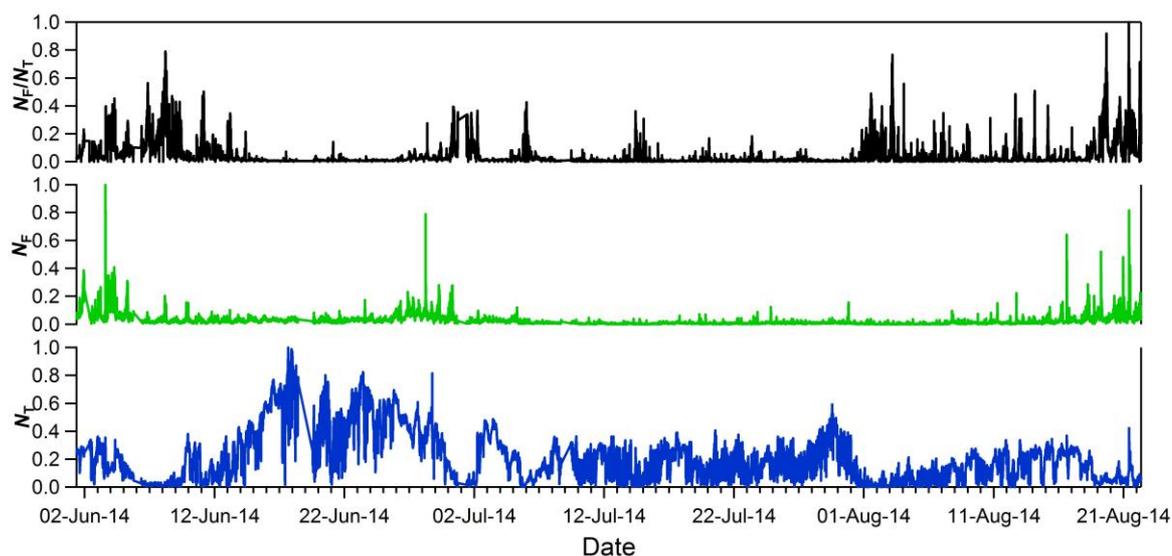
**[AR5]** We understand Reviewers concern. Our intention of carrying out the correlation analysis was to indicate that under certain conditions (mainly clean conditions persisting during monsoon at this site) UV-APS may be used to detect and segregate the particles of biological origin in sub-micron 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 particles 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  $<1 \mu\text{m}$  and  $>1 \mu\text{m}$  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  $<1 \mu\text{m}$  and  $>1 \mu\text{m}$ .

**[RC6]** Line 357-360: The high extreme values of  $N_F/N_T$  actually do result from high variations of  $N_F$ , as evident from fig S2. The presented figures do not support the argued inverse correlation between  $N_T$  and  $N_F/N_T$ . 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 largely 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  $\mu\text{m}$  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 ( $<0.5 \mu\text{m}$ ). However in case of marine aerosols a secondary peak at diameter  $<1 \mu\text{m}$  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.

**References:**

Healy, D. A., Huffman, J. A., O'Connor, D. J., Poehlker, C., Poeschl, U., and Sodeau, J. R.: Ambient measurements of biological aerosol particles near Killarney, Ireland: a comparison between real-time fluorescence and microscopy techniques, *Atmospheric Chemistry and Physics*, 14, 8055-8069, 10.5194/acp-14-8055-2014, 2014.

Saari, S., Reponen, T., and Keskinen, J.: Performance of two fluorescence-based real-time bioaerosol detectors: BioScout vs. UVAPS, *Aerosol Science and Technology*, 48, 371-378, 10.1080/02786826.2013.877579, 2014.

Valsan, A. E., Priyamvada, H., Ravikrishna, R., Després, V. R., Biju, C. V., Sahu, L. K., Kumar, A., Verma, R. S., Philip, L., and Gunthe, S. S.: Morphological characteristics of bioaerosols from contrasting locations in southern tropical India – A case study, *Atmospheric Environment*, 122, 321-331, <http://dx.doi.org/10.1016/j.atmosenv.2015.09.071>, 2015.