

Interactive comment on “The fluorescence properties of aerosol larger than 0.8 μm in an urban and a PBA-dominated location” by A. M. Gabey et al.

Anonymous Referee #3

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General Comments The manuscript by Gabey et al. entitled “The fluorescent properties of aerosol larger than 0.8 μm in an urban and a PBA-dominated location” describes work utilizing an online fluorescence sensor for detection of biological aerosols in two disparate environments. The experiment is unique and scientifically important, because seemingly few measurements have reported detailed properties of atmospheric PBA. The contrast of a polluted urban environment and a pristine rainforest environment is a very interesting aspect of the work that adds to its quality. The authors previously reported aspects of the same study in Borneo in a previous ACP publication, but this manuscript goes beyond what was reported at that point. The experiment seems to have been performed in an appropriately detailed manner and the manuscript

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is generally organized well and progresses logically. I have no doubt that this study is worthy of publication in ACP and I think it could be very useful to researchers interested in airborne biological particles. However, I think that parts of the manuscript need to be improved with respect to clarity and depth. A number of sections beg the reader to ask questions unanswered by the manuscript, and I felt myself asking “What does this mean?” or “Why do we care specifically about this?” at several points. I suggest clarifying the main messages of the manuscript and highlighting these more specifically, rather than presenting a wide survey of the pieces of information that could be taken from this dataset. Late in the manuscript authors suggest areas of analysis that could be expanded upon, but these would potentially strengthen the arguments within this text without significant additional work. The wording of many paragraphs also contains vague or confusing statements that should be revised. I have highlighted several in specific, but I suggest making sure the authors carefully read through the text to make sure the wording is clear and concise. Several sections could also use additional references, and I have suggested specific examples when available and appropriate.

Specific Comments - Many places in the text need better support through citation. I've seen at least a couple recent papers that report biological aerosols in rainforests, including: Zhang, Environ. Research Letters, 5(2): 024010, 2010. and Poschl, Science, 329(5998): 1513-1516, 2010. Work well beyond what the authors report in Section 1.4 has also been done measuring urban bioaerosol, for example: Pinnick, Atmos. Environ., 38(11): 1657-1672, 2004; Bauer, Atmos. Environ., 42(2): 5542-5549, 2007; Despres, Biogeosci, 4(6), 1127-1141. - The authors mention two interesting situations within the instrument. “Transient rises in the noise floor . . . led to large concentrations of nonexistent” aerosols (P539, line 20). How is this dealt with in the data analysis, and what result might it possibly have on the final data? Also, the first two lines on page 540 mention a situation where particles saturate the fluorescence detector and are excluded from analysis. What kind of bias might this introduce into the data such that they are thrown out? Or what does it change now that they are excluded? - Table 1 lists the “particle categories” that the authors discuss at length in the results section.

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These particle types need to be rigorously and specifically introduced within the text (P539), because the authors assume that these represent different particle types and draw many of their conclusions based on these differences. The authors also mention “tryptophan-like” and “NADH-like” particles. While I understand that they are trying to link the signal from the WIBS detector to the fact that those biological molecules also fluoresce at the same wavelengths, to make the stated assumption that the particles are “like” NADH or tryptophan is somewhat overstated. You can still draw a mental line between the two, but I suggest you not state the relationship as concretely. This is why these signals (F1, F2, etc.) need to be clearly introduced before going further. Within this section I suggest discussing the difference between the meaning behind signal from F1, F2, and F3. And why do you later combine F1 with F3 and F1 with F2? No justification or indication was given for what this scientifically represents. The “tryptophan-like” wording occurs at multiple times, including the conclusions. - The text makes lengthy mention of the diurnal cycles within the Manchester (and Borneo) data. I suggest showing a diurnal plot for the Manchester data, because it would significantly shorten a long section of complicated wording on page 541-542. It could potentially be show in an online supplement if desired. - What do the size distributions of the combination of F1 and F2 (etc.) show? This is an example of a place where I was left wanting to know what the point of the plot was. - Can you show characteristic SEM pictures? This could be very interesting and strengthen the manuscript. - How confident are you that the aerosol in Manchester is derived from anthropogenic sources, dust, sea-salt, based on this microscopy survey? Are there any references that could be cited to confirm this trend, or is this new information? - Add “In winter” after “Manchester” in the last line on page 543. This strikes me as a small, but important omission. - On page 544 the authors state that the non-fluorescing aerosol has a different diurnal pattern than the fluorescing aerosol, but that these are attributed to the same source (lines 10-11). I don’t understand how they draw this conclusion. - Following that, how do you know that the “non-fluorescent material” (last lines of page) is associated with PBA emission? Does it peak at the same point in the diurnal cycle than the fluorescing

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material? - The size distribution plots show only ~10 size bins, and this makes some of the authors conclusions about the 1.2 um peak a bit difficult to clearly see. Is it possible to reduce the steps between size bins, or is there something inherent about the instrument that limits this? - Page 546: What would you expect this curve to look like for PBA and non-PBA? What does this mean? - Pages 547-549 are dense with information, but these sections especially need more justification. There are lots of numbers, but not much explanation of what scientific information one hopes to gain from these plots. I suggest shortening the text and clarifying the main messages that the author wants to convey here. - Regarding the ratio of fluorescence intensity: Would it be helpful to take the concentration of particles in each size bin be into account as a weighting factor? The ratio is one thing, but how much it impacts the signal is another. - Section 4: This section discusses a possible further analysis that could be performed. Why is it not employed now, however? I think this could help flesh out purpose from the fluorescence ratios, but shouldn’t take a great deal of effort. - I’m not sure I necessarily agree with the conclusion that “the F2 and F3 channels detect the same fluorophores.” I agree that this is a possibility, but I don’t think you can say with surety that this is the case. - The observation that fluorescence and Dp have a linear relationship is a very interesting conclusion, given the stated reference that for bacteria a Dp2 relationship would be expected. What do the authors think this means?

Additional Comments - What do you mean by “can, in principle, be distinguished” in the 21st line of the abstract? This, to me, implies more differentiation than is possible with the techniques you report. While the text goes into much explanatory detail on what they mean, but with uncertain impact. For the abstract I think it is important to be clear, since this is potentially a big statement as written. - What do you mean by “discernibly” in the second line of the introduction? - What do you mean by “consistently fluorescent” on Page 533, Line 16? Are they consistent in time, space? This implies to me that the fluorophores occasionally fluoresce, but sometimes don’t. I think you meant that these fluorophores are most commonly considered by researchers to be fluorescent, or something like this. - I don’t understand the reasoning behind the sentence on Page

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533, Line 22-23. What does the fact that the excitation and emission bands are well separated have to do with the ability to determine PBA? The separation of bands simply implies how good your elastic scattering filter needs to be for the instrument to work. - Page 533, Line 23-26: Re-word this sentence a bit (change the word "brings"?). - The first line on Page 534 the authors state that "a large proportion of PBA are detected successfully by UV-LIF . . ." Is this really true? I think this sentence should be a bit more specific, with given references. - Page 534, Line 20-22: Remove pronouns (e.g. this, it). - Citations needed: Page 535, Line 5, 11 - Page 536, Line 12-13: Remove "using two excitation . . . and NADH" (redundant with earlier text) - Page 536, Line 14-16: shorten text by saying: "diurnal cycle of fluorescent particles below the canopy (~xx l-1) but not above (~xx l-1)." - Within the WIBS instrument the amount of scatter from a flash from the lamp allows "particle diameter to be estimated." (Page 537) How much uncertainty is wrapped up within this estimation, and does it change with optical parameters (i.e. composition) of the particle? - Remove word "any" from Page 537, Line 14. - What is "it" on Page 537, Line 17. - Need citation for Page 537, Line 26. - Change to ". . .is the mean baseline (no particle detected) . . ." on page 539, Line 1 - What do you mean by "typically" here? (Page 541, Line 8). - What was the justification for using certain filter periods for sampling? - The wording of lines 19-20 on page 541 is a little confusing. What do you mean? You consider the time periods when filters were used to be sufficiently representative, because the number fractions are only 40% reduced from the total averages? This seems to indicate a difference, rather than a consistency. - Page 543, Line 10: replace with "surveying a variety of" - Please provide a reference for the type of EDX analysis done here (Page 543). - Same page: "prominent signal" = most concentrated elements? - Same page: "classed separately". This was confusing, but after looking at the plot I see what you mean. It could be clarified within the text, however. - Page 545, Line 1: change 'introduction' to 'contribution' - Page 545: "The same cannot be said of the Borneo dataset." But what can be said? - Page 549: Dp typo x2 (not sub-scripted) - Figure 2: labels on top should be amended to be i, ii, iii, iv (they currently repeat) - Figure 3: What is the size

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transmission through the WIBS detector? The total number appears to peak at 1 μm , but I would guess this is an instrumental artifact and that the "true" peak is likely much lower in the accumulation mode. Is this likely? This should be (very briefly) addressed.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 531, 2011.

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