

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.

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We thank the three reviewers for their thoughtful feedback on our manuscript and enclose responses to the various questions and suggestions posed in their reviews. Alongside the specific comments, all of which are gratefully received and will be addressed, we would like to answer some of the queries and suggestions.

Review 1

Given that the WIBS3 has a relatively small "needle" inlet tube with a flow rate of only 0.238 litre per minute, were any tests conducted to determine the potential losses of larger particles in the inlet tube? It would be good to clarify the dimensions of the inlet

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tube. Does the inlet ever partially plug during sampling and need cleaning?

The inlet tube inner bore is actually 1.2 mm (1.65 mm outer diameter), so "needle" is perhaps an inappropriate description on our part. Regarding particle losses, while the sample flow is relatively small (238 ml /min) the total flow bringing the aerosol to the sample tube is much larger (2380 ml /min), so sedimentation is reduced accordingly. The flow velocity through the sample tube is typically 15 m/s so deposition on the walls has never been an issue and to date there has never been a report of blockage in any of the WIBS-3 or WIBS-4 instruments. When cleaning the tube with an alcohol swab has taken place it has revealed no visible contamination. WIBS-3 concentration generally agrees with that reported by a GRIMM 1.108 in the size range 0.8–20 µm and a TSI APS 3321 at $D \geq 2 \mu\text{m}$

We are a little concerned about the statistical significance of the concentrations of fluorescent particles as large as 8–20 micron because of the relatively low WIBS3 sample rate of 0.23 liter per minute. For the linear scales presently used in Figs 3 and 6, some of the size distribution plots for the concentration of particles contain essentially no useful data beyond 4 micron size. The form of the size distributions of both non-fluorescent and fluorescent particles for these larger particles might be more clearly evident if the (left) vertical scales in Figs. 3 and 6 were logarithmic.

The plots will be re-scaled accordingly in the revised manuscript. We feel that the concentrations of PM10 in the Manchester urban setting and the enhanced FBAP supermicron concentrations observed in the Borneo rain forest allow a statistically representative picture to be constructed when coupled with the long integration time and regular diurnal features observed.

Regarding the discussion of the dependence of fluorescence intensity on particle size, it has been demonstrated that for moderately absorbing homogeneous spheres in the 1–10 µm diameter range, fluorescence intensity is approximately proportional to the square of particle diameter (see Hill et al, 2001). [Small weakly absorbing particles

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have intensity approximately proportional to the cube of diameter]. If the fluorescence data in Fig. 7, 8 and 9 were displayed on a log-log scale, the departure from intensity proportional to diameter squared would clearly be evident, and would be a more quantitative measure of a decreasing relative concentration of fluorors (or a transition to weaker fluorors associated with different composition) with increasing particle size.

We will bear this in mind for the next iteration of the manuscript, although we are balancing the need to show this linearity (or non-linearity) with the need to show the structure of the curve at smaller sizes.

The comparable populations of N_{F1} and N_{F2} particles in Manchester is consistent with the previous findings (Pinnick et al, 2004) that spectra of fluorescent particles (excited by 263nm light) in an urban aerosol (Washington DC, USA) have about equal populations that emit in the 300-400 nm region as compared to the 400-600nm region.

This will be noted in the manuscript, and it is also interesting to note that the relationship between the fluorescence intensity between F1 and F2 does not show any linearity in Manchester, whereas the F2 vs F3 intensity curve exhibits a clear linear relationship.

The N_{F1} data for both Borneo and Manchester appearing in Fig 7 is duplicated in Figs 8 and 9; the N_{F3} data is duplicated in Fig. 8, and the N_{F2} data is duplicated in Fig. 9. The authors might consider combining figures 8 and 9 and eliminate the duplications. In figures 8 and 9 the authors might consider plotting $F1/F3$ on a logarithmic scale.

One initial point of clarification (which may simply be a typo in the review): Figures 7-9 do not refer to N_{F1} etc (the number concentration) but the intensity of fluorescence. We will make this distinction clearer in the nomenclature using, for example, I_{F1} to refer to intensity.

Whilst the plots are very similar in appearance, there is reason for the apparent duplication: Figure 7(i), for example, shows the intensity of every particle that is fluorescent in channel F1. No other constraints are placed on which particles are included in this

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plot, and these particles could be fluorescent in both, one, or neither of the other channels. Figure 8(i), however, shows the F1 intensity when we require fluorescence from each particle to be present in F1 and F3, and these may or may not be fluorescent in F2. If this criterion is not introduced in Fig 8, the ratio will not be finite for all particles, skewing the results. The same process occurs in Figure 9 for “F1 and F2” type fluorescence. Finally, $\log_{10}(F1/F3)$ is plotted on a linear scale in Figure 8, which is effectively a logarithmic scale. All of these features will be made clearer in the plots and text. We will look into placing the extra plots into an appendix so as not to deluge the reader with information but still offer completeness.

Although the summaries of WIBS3 data appearing in Figs. 3, 6, 7, 8 and 9 are interesting and informative, the results smear to some extent the information content of the single-particle data measured with the WIBS3 diagnostic tool. The authors might consider, either here or perhaps in a future publication, a principal component analysis approach where for each particle measurement a vector in a multiple-dimensional space is created, where one dimensional could be the elastic scattering signal, and other dimensions could be the various fluorescence channels (perhaps normalized by the elastic scattering). A principal component analysis might be a more powerful tool to better understand and interpret the WIBS3 measurements.

In answering this point it is necessary to mention the fact that this spread of values in the data is present even at a single-day level, and appears to be a combined feature of the variability of the aerosol fluorescence and any noise in the detectors. the manuscript for the entire campaigns. Such plots, of course, do not tell us whether there is structure in the data being summarised. Broadly speaking there is little in this regard, and we will try to clarify this in subsequent iterations of the work.

We agree that PCA is an interesting approach for particle classification, and are considering this approach as a “second generation” analysis once data for a number of environments have been collected. As total fluorescence intensity across broad wave bands is measured by the WIBS-3, there are few dimensions in which to perform this

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analysis compared with Hill et al (2011), who performed PCA on dispersed fluorescence spectra following excitation at similar wavelengths the WIBS3 uses.

The lack of published fluorescence measurements (particularly regarding shorter-wave fluorescence that may relate to Tryptophan) is the reason we feel it is important to discuss the features of fluorescent aerosol at each site in such a broad manner. This is so a base line can be established for future interpretations. To go beyond this and investigate the interdependence of the three fluorescence intensities would require fluorescence to be present in all three channels, which limits the scope to 1.5% of the total number sampled in Manchester. We hope to apply a PCA analysis to the data from the next version of the WIBS which will have enhanced sensitivity for both smaller and larger particles in order to better highlight some of these differences. This is work in progress.

Review 2 This study analyzes measurements of aerosol properties, size and fluorescence, in two locations with very different sources of particles. The primary objective of the study is to evaluate the differences in the particle size distributions and fluorescing intensities in the context of PBA versus non-PBA particles. The goal is to find metrics that can differentiate the two types of fluorescing aerosol particles (here after referred to FAP) so that, in the long run, these types of measurements can better quantify the properties of PBA, their sources, health impacts, etc. Of course, given that we also know quite little about the non-PBA FAP, being able to better characterize their properties would also improve our ability to identify and "fingerprint" these types of particles. Overall, the material is organized and presented concisely with each step leading logically to the next one. From the beginning it is clearly understood that the authors intend to use the F1, F2 and F3 intensities, and some relationships among them, to derive metrics that separate PBA from non-PBA FAP and, in the end, there appears to be an argument for using the F1/F3 ratio as a way to find "Borneo-like" particles in Manchester. To this reviewer, however, to end the paper with this conclusion leaves this reader, and potential user of a WIBS-like instrument, hanging, i.e. if I am now to

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run the WIBS in Mexico City, and find that my F1/F3 ratio distributions look neither like Manchester or Borneo, what have I learned?

I think that the authors have a rich data set that could and should be pushed a bit further than they have done to this point in order to better understand exactly what are the links between the biochemical properties of the FAP and their size, fluorescence intensities as a function of wavelength and other characteristics that better differentiate PBA and non-PBA on a single particle basis, rather than on an ensemble.

The paper touches upon, but never returns to, the problem of actually determining how much fluorescing material there is in a particular particle. In the single particle soot photometer, SP2, we are challenged similarly, i.e. we have an optical diameter measured by light scattering, similar to the WIBS and an incandescence signal, related to the amount of refractory black carbon (rBC) mixed in the particle, similar to fluorescence in the WIBS that is related to the amount of UV active material mixed in a particle. In order to get a handle on the fraction of rBC in a given particle with respect to the rest of the non-refractory material, we derive a mass equivalent diameter (MED) from the rBC mass that we obtain from the incandescence signal. Here, of course is where there is a major difference between the WIBS and SP2. In the SP2 we have what we think is a fairly good relationship between rBC and the intensity of incandescence because of calibrations with rBC reference materials (these, of course, are the focus of a lot of healthy discussions). The point is, however, that we are able to compare the optical diameter with the MED and this gives us a relative indicator of the amount of rBC mixed with non-refractory material. We find some very significant differences from location to location, depending on the source of the rBC and their age, in these mixing indicators.

I recommend an expansion of the analysis of the current data set to take a similar approach when evaluating the Manchester and Borneo measurements. Just as in the SP-2, we look at two different incandescence wavelength to estimate the temperature, the authors are using three different fluorescence wavelengths to separate NADH and

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Tryptophan type of characteristics. The obstacle to following an approach parallel to that which is used in the SP2 is that apparently there is no reference material that relates fluorescence intensity to an intrinsic property of the FAP. This raises my first three questions:

1) Why can't the WIBS be calibrated with known FAP, i.e. PAHs that are known to fluoresce and that can be produced with mono-dispersed distributions?

One shortcoming of our data presentation is that the lack of structure in the raw data, which is the reason for the statistical summaries, is not shown. In the data gathered, the dependence of fluorescence on size is not a series of distinct relationships that can be matched to laboratory test particles, but simply a general upward trend with a significant degree of scatter. There is also the likelihood that fluorescence intensity from a given PAH at a particular wavelength is neither unique to that PAH nor distinct from PBA, given that ambient conditions have been observed to influence the fluorescence intensity from a particle. A clear example of this potential ambiguity is the spectrum from cigarette smoke particles and BG spores shown by Pinnick et al (1998).

Given the lack of structure in the ambient data and this non-specificity, "calibrating" the instrument in this way would not necessarily provide a clear basis for classification and may be premature. This is the reason we have produced this "top-down" analysis of fluorescence properties in each location rather than a bottom-up one from individual laboratory samples (which has been performed using PBA previously by other researchers, e.g. Hill et al 2011). If systematic deviations from what was found at each site are observed, atypical events may then be identifiable.

2) If at the moment the analysis is restricted to the use of arbitrary units (a.u.) how do the intensities measured during the Manchester trials equate to those measured during the Borneo trials? Put another way, what was the transfer standard used to calibrate the F1, F2 and F3 channels so that any differences were really in the FAP themselves and not changes or shifts in the optical or electrical system?

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The performance of the instrument has been evaluated periodically using conventional PSLs, which are slightly fluorescent in the F1 channel, allowing this channel's response to be compared directly between campaigns. This was only performed for the first time some months after the Borneo campaign, but to date it has changed by less than 10% despite extensive instrument use and cleaning of the optical components taking place in between tests. The side-scattering measurement uses the same detection optics as the F2 and F3 channels, and the size response of the instrument is the same between the campaigns, indicating consistency in this detector. The function of F2 and F3 channels can be verified only in a binary sense using fluorescent PSLs, since these saturate both detection channels, but this has been observed consistently before, during and after both campaigns.

The relative xenon pulse intensity is recorded during each excitation and varies by 3-8%. This is not corrected for in the analysis since there is no detectable dependence between fluorescence intensity and xenon power within this regime.

There may be systematic differences in aerosol fluorescence intensity caused by external factors (i.e. different relative humidity, particle age) between sites, but this cannot be accounted for by calibration, and this is one reason that the intensity values recorded are given less attention in the discussion of results than the ratio between channels and the form of the intensity versus D_p curves.

3) Were the background fluorescing values the same in both experiments? In the current analysis, fluorescence is shown plotted in Figs. 7-9 with lines representing median values and quartiles, but these graphs mask what may actually be going on that makes the Borneo data distinct from the Manchester measurements. The spread in fluorescence values, that is quite large, may be linked to other processes that could be related to the source of the difference. We know for example, that there can be photon quenching effects, depending on what the fluorescing material is mixed with. Without knowing the relative fraction of fluorescence material to total non-fluorescing material, this effect cannot be very well assessed. So, my first three suggestions:

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As discussed previously the spread in values is quite large in general in Manchester, and quenching is one factor that may contribute to this. However, we do not attempt to establish how much fluorescing material there is in each particle because this is not possible with the technique and the fluorophore concentration does always not have a monotonic relationship with fluorescence intensity because of self-quenching at high concentrations (Hill et al, 2001).

The effect of particle composition on UV and fluorescence transmission would dictate the optical “species” of particle, and it is this we actively seek to distinguish in each environment, while reserving particle size as another degree of freedom. Considering the relative amount of each fluorophore, which varies somewhat between basic PBA types (bacteria, fungal spores and plant material) and little between biological species has yielded some positive initial results e.g. Sivaprakasam et al (2001), in the past, so it was our intention to see if this revealed any structure in ambient data.

i) Analyze the number concentration as a function of F1, F2 and F3 fluorescing intensity.

Fluorescence histograms actually show quite little variation with time and primarily low values (see below) were obtained in Manchester, with a better-defined mode in Borneo, and these will be added in the revised paper – possibly as an appendix.

ii) Normalize F1, F2 and F3 with the scattering intensity to get [F1], [F2], [F3] then look at the relationships between [F1], [F2], [F3], e.g., scatter plots of [F1] vs [F3] with color encoding using the intensity of [F2].

We are happy to add this type of analysis to the revised manuscript as it would illustrate one further difference between the aerosol at each location: the relationship between [F1] and [F2] in Manchester is much stronger than in Borneo. In fact, the [F2] vs [F3] scatter plot is clearly linear and this will be added to the manuscript.

iii) If fluorescence is expected to be proportional to the cross sectional area of the particle, then derive a surface equivalent diameter (SED) that can be compared with

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the optical diameter. This can be arbitrary as well until some there is a way to better calibrate the fluorescence channels, i.e. derive a scale factor between fluorescence and D_{opt}^2 to get an SED \leq optical diameter. The objective is to find a way to estimate a relative fluorescence fraction that is then used to more clearly compare FAPs on the same basis.

If we understand this suggestion correctly, the same can be achieved using the fluorescence to elastic scattering intensity ratio. Again, this is complicated by the lack of structure between the two channels, but it can be used to set an upper and lower limit analogous to the F1/F3 limits applied for the Borneo data in each size bin in the original analysis. We will discuss the use of this metric in a revised manuscript.

Other comments, questions, suggestions: *Abstract, line 14. Start out defining sizes as radii or diameters. Page 533, Line 22-23. It is not clear what is meant here by the excitation and emission bands being quite well separated. They seem to have overlapping emissions and excitations. Some clarification needed. Page 537, Methods. Forward and side-ward light is measured. Are the two summed? Which is used for sizing? Clarify? Size range? Uncertainty? Sample line losses? Number of size bins? Is the Xenon radiation filtered to remove any light between 310-400, or 400-600nm? Are the activation wavelength really that precise? Lacking essential information here to understand the remainder of the discussion.*

More details will be added regarding these aspects.

Page 543, Line 20. Based on what? Show a microphotograph? Explain what one looks for in a PBA.

Another reviewer has also requested example ESEM images, and some will be added in an appendix to indicate that, for example, iron-rich particles resemble simple spores.

Page 547, lines 19 – To my eye these distributions don't look log-normal. What criteria are being used to make this observation?

C1892

The logarithm of the ratio is plotted on a linear scale, effectively making a log-scale, on which is shown a normal distribution. This will be made clearer.

Page 550, Line 13 – Although in their summary the authors qualify their conclusions that PBA have a minor influence in Manchester, until they further stratify or parse their results and have a more statistically reasonable set of filter analyses, I think that this conclusion should be put on hold.

A more rounded approach will be taken to arguing that the PBA influence in Manchester is likely to be much smaller than that in Borneo. The ESEM data is primarily to indicate that soot and soil dusts (which can contribute false-positives) are found in the coarse mode in Manchester, and this will be backed up with references based on measurements in other UK cities (e.g. Harrison et al 2004), who find similar particle compositions to the EDX analysis performed here. We will also point out that larger PBA (spores and pollen) are small in number in urban regions than rural. The fact that the Manchester fluorescent diurnal cycle is linked to rush hour also suggests dust resuspension may be contributing.

Page 551, lines 15-17: I would argue that the relationship between fluorescence and diameter is not linear, as stated here by the authors. A positive trend, yes, but on a linear-log plot, a straight line is not linear. This raises the question, what would all of these data look like on linear-linear scale plots?

The lines do appear linear (with deviations at certain) on linear-linear scales, but the discussion of this will be reworked in terms of the single-particle data, which does not show a strong relationship but a weak general increase with particle size.

Review 3

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

C1893

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 Pinnick (2004) reference is in fact present in our manuscript, and the Poschl work was rather new when our manuscript was submitted to ACPD in October 2010. These and the other omissions will be addressed in the revised manuscript.

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?

C1894

This manifests as a high concentration of $0.5\mu\text{m}$ particles, which are readily filtered out by applying a minimum size criterion. Since they force the instrument to trigger at a high rate, no real aerosol particles are measured when the phenomenon occurs, and short gaps in the data occur but it should be stressed that these are less than 1% of the time series and only affect the Manchester data.

In our laboratory tests and field comparisons with a GRIMM 1.108 OPC, we find the WIBS3 i) detects few calibration particles smaller than $0.8\mu\text{m}$ and (ii) tends to report the few it does detect as being around $0.8\mu\text{m}$. The size criterion should therefore not skew the results as reported by the instrument. If an entire time bin contains no data it is omitted from the averaging, so it should not introduce bias in this regard either.

As mentioned in Table 2 of the manuscript, the highly fluorescent (saturating) particles account for only a few percent of those fluorescent in each channel, and 0.6% or less of the total recorded aerosol number in Manchester, so we do not anticipate a large bias. We will also make it clearer that “the analysis” in this case refers only to when fluorescence intensity or ratio is discussed (Section 3.2 onwards) otherwise zero/infinite intensities and ratios would appear. These particles are not excluded from the fluorescent size distributions or number concentrations in the earlier sections because this part of the work only deals with the question of fluorescence being present, not its intensity. If they were excluded here, a suppression of the fluorescent number fraction at the largest sizes would result, particularly in F1 in Borneo.

Table 1 lists the “particle categories” that the authors discuss at length in the results section. 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.

This section will be strengthened accordingly.

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

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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.

We will reassess how best to describe the channels concisely. The basis for combining different fluorescence channels will be made clear. Briefly, it is based on the fact that F1 and F3 type fluorescence would be expected of many PBA, and particularly bacteria and fungal spores. The F3 channel was expected to be the limiting factor for bacteria, as the NADH peak (or an NADH-like) emission is seen less often than the Tryptophan peak (Hill et al., 2009). “F1 and F2” is included more for completeness than an expectation of a particular relationship. The ratio F1/F3 was calculated as it allows some distinction to be drawn between particles while removing particle size factors. This type of distinction has been noted, for example, by Sivaprakasam et al. (2004), who monitored total fluorescence after excitation at different wavelengths.

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.

This will be added and the text simplified.

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.

These are intended to demonstrate that no new size features appear when multiple

C1896

channels are combined. They also supplement the discussion of fluorescence intensity ratios.

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?

These observations are characteristic of a UK city (e.g. Harrison et al., 2004, find the coarse mode features a mixture of elemental carbon, organic carbon and inorganic dusts). Sea salt is often found inland so this is also not unexpected. We will add references to the manuscript for better context, as well as the characteristic ESEM images (most likely as an appendix) as they help illustrate how iron oxide particles resemble simple spores before the EDX measurement is performed.

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 material?

Two separate features are seen in the non-fluorescent aerosol cycle in Borneo: a small rise at midday that is anti-correlated with fluorescent particle number at this time, and spikes in mid-afternoon that coincide with the strong increases in fluorescent number.

The former is thought to be material entrained from above the canopy, and the latter is thought to be associated with the PBA release.

The size distribution plots show only 10 size bins, and this makes some of the authors conclusions about the 1.2 μm peak a bit difficult to clearly see. Is it possible to reduce

C1897

the steps between size bins, or is there something inherent about the instrument that limits this?

The size channels are representative of the size distribution reported by the WIBS3 when measuring monodisperse calibration spheres (polystyrene latex and glass) and a comparison with a GRIMM 1.108, which offers similar performance.

Page 546: What would you expect this curve to look like for PBA and non-PBA? What does this mean? A fresh source of homogeneous fluorescent aerosol (PBA or non-PBA) would be expected to feature increasing intensity with d_P^2 , and limited spread. As the Borneo dataset has been concluded in Gabey et al (2010) to be rich in PBA, this is our reference PBA curve.

We interpret the deviations from this in Manchester to arise because of the contribution of different fluorescent particle types and atmospheric processes, each of which will result in more variety in the fluorescence intensity.

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.

The size distributions discussed earlier are an attempt to illustrate this. We will look into combining these pieces of information more effectively.

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.

It is in fact performed and the results are reported briefly. This will be made clearer in the manuscript.

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.

C1898

If a scatter plot between fluorescence intensity in each channel is shown then this becomes clear, and this will be included in the revised manuscript. The phrasing of this conclusion will be revised.

The observation that fluorescence and D_p have a linear relationship is a very interesting conclusion, given the stated reference that for bacteria a D_p² relationship would be expected. What do the authors think this means?

This may indicate that the optical absorbance of non-bacterial aerosol is larger than bacterial aerosol, or that multiple sources are contributing at different sizes.

Responses to some specific comments *What do you mean by "discernibly" in the second line of the introduction?*

This is a semantic problem analogous to defining life, and the sentence will be changed to a list of accepted PBA types.

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.

The use of "consistent" refers to the fact that Tryptophan, for example, is fluorescent regardless of whether it is bound to a protein. It can thus be detected using fluorescence in more microbiological configurations than other amino acids. This will be clarified for Tryptophan and NADH separately as there are some subtle differences in the nature of the "consistency".

I don't understand the reasoning behind the sentence on Page 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.

A larger separation between the excitation and emission bands is useful because exci-

C1899

tation pulse and fluorescence measurement occur simultaneously. The excitation pulse is 50 nm wide in the WIBS3 because of the Xenon lamps used, and the fluorescence would be lost amidst this if the overlap between the absorption and emission peaks was small.

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?

To clarify, the scattering power from a dedicated diode laser is used for sizing rather than light from the flash lamps. The size resolution of the instrument is reflected by the chosen size intervals in the analysis. A summary of the weaknesses of optical particle sizing will also be included to address this point.

What do you mean by "typically" here? (Page 541, Line 8).

This refers to the 10th and 90th percentiles of number concentration for the period specified.

What was the justification for using certain filter periods for sampling? This was arbitrary and simply designed to cover each hour of the day in case systematic variations took place between daylight and nighttime.

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.

The intended meaning is that the concentration found during the filters is larger than average, but these events are not unique in the dataset.

Figure 3: What is the size transmission through the WIBS detector? The total number appears to peak at 1 um, but I would guess this is an instrumental artifact and that the

C1900

"true" peak is likely much lower in the accumulation mode. Is this likely? This should be (very briefly) addressed.

We will discuss the size response of the WIBS3 in a little more detail to indicate that some smaller particles are classed in this size bin and the counting efficiency falls off at $0.8 \mu\text{m}$. The size distributions are plotted to the logarithmic mid-bin, which exacerbates this "enlarging" effect.

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