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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurements of coarse mode and primary biological aerosol transmission through a tropical forest canopy using a dual-channel fluorescence aerosol spectrometer" *by* A. M. Gabey et al.

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We thank all of the reviewers for their considered comments and interest in the manuscript and welcome the opportunity to expand and clarify on certain points. This reply attempts to address in bulk the concerns and comments raised by the reviewers and outlines the additions and changes to the manuscript. We also provide more information that will be added to the manuscript. Stylistic and typographical errors have been noted. The purpose of the manuscript is to communicate observations of the diurnal variation of coarse mode aerosol and their fluorescence behaviour below

and above a tropical rain forest. Many of the general comments regarding site descriptions and species distributions are being dealt with in the special issue overview paper and associated papers and so have only been discussed briefly. In some cases very detailed technical discussions are beyond the scope of this observation based manuscript, however we have where possible provided additional descriptions, data and references that address these points as far is possible within the limitations of the technique deployed. These will be added to the revised manuscript accordingly.

Responses to specific questions

(H) : J.A. Huffman, (A) : Anonymous Reviewer H1. How does the WIBS3 differ from the WIBS2 and how is particle sizing performed?

The WIBS3 differs from the WIBS2 discussed in Kaye et al., 2005 because it has an additional photomultiplier tube (PMT) split into 4 quadrants. This PMT is dedicated to sensing elastically scattered light in the forward direction. The forward and side-scattered intensity are then compared to a 2-dimensional lookup table which was generated using a Mie theory model and calibrated to the WIBS3 using polystyrene latex (PSL) microspheres. The 4 quadrants of the PMT also allow the Asymmetry Factor to be derived for each particle. This will be discussed later in the response.

H2. Is there any chance that some PBAP go undetected or that false positives contribute to results?; The use of "PBAP" may be too bold.

These are inherent weaknesses of the fluorescence approach to identifying PBAP. While false positives and negatives cannot be ruled out, we agree that framing the results using the FBAP nomenclature proposed by Huffman et al. (2009 ACPD) is beneficial in conveying the weaknesses of the approach to the reader. The likelihood of false positives contributing to our results is discussed more fully in our response to comments by Pinnick et al.

H3. What is the choice of fluorescence baseline and how does this affect uncertainties?

The fluorescent threshold described in the manuscript denotes the minimum fluores-

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cent intensity that the WIBS3 can report reliably. A non-zero fluorescent intensity is always recorded because the optical filters exhibit a small amount of fluorescence. The WIBS3 can measure this by running fluorescence measurements when no particles are being sampled using particle filters fitted to the inlet of the instrument. These filters preclude particles $D_A > 0.2$ μm. The intensity of this baseline is normally distributed and we are able to establish the mode and standard deviation of it. The threshold chosen (applied after the sampling is performed) is 2.5σ above the baseline mean, with less intense fluorescent events placed in the lowest fluorescence bin, much like the lowest fluorescence intensity bin in the UV-APS as discussed in the paper by Huffman et al (ACPD, 2009). In a continuous Gaussian probability distribution, only approximately 1% of non-fluorescent particles would exceed this threshold and this is the basis of our over-counting uncertainty estimate. The uncertainty related to PBAP failing to fluoresce brightly enough is likely to be larger than 1% in each channel, but this is unquantified.

H5. How is the Asymmetry Factor (AF) measurement performed, what values are expected for different particle morphologies and what can be concluded from the AF data collected?; A15. What artefacts might affect the AF results?

As mentioned earlier, the forward-scattering sensor is split into 4 quadrants, each measuring the intensity of light at 4 angular offsets. The fractional standard deviation of the 4 intensities is multiplied by an instrument-defined constant and this is a measure of the scattering asymmetry factor, AF, which can be related to the shape of the particle. Single-particle scattering patterns from differently shaped particles are discussed in detail in Aptowicz, 2006;Kaye et al., 2007 but to generalise: a spherical particle will exhibit a more symmetrical intensity distribution than a morphologically complex particle, so quantifying the symmetry of this pattern allows some distinction between them. The arbitrary AF scale is 1 – 100, where 1 denotes a perfectly spherical particle and 100 is a fibre. Laboratory tests with the WIBS3 using spores with different shapes confirm its ability to discriminate between different classes. We therefore feel the AF measurement is a useful way of comparing the relative shape of different classes of particles in

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an aerosol population.

The AF distributions reported in the paper seem to reinforce the notion that a transition between two particle populations occurs at 2 – 3 μm. One question also concerned references to laboratory tests using PSL to investigate the role of noise. While these were limited experiments, they showed that moving between 1, 2.1 and 3.0μm PSL reduces the modal AF, presumably because of a weak signal-to-noise ratio in the quadrant PMT that records the angularly resolved elastic scattering. Figure 1 shows that recorded AF modes are at 4, 3 and 2 respectively; illustrating that the variation in AF modes of the fluorescent and non-fluorescent particles is not an artefact of a noisy signal but a property of these particles (note that a correction factor was applied to account for a change in the way AF is calculated in the time between the laboratory test and the Borneo campaign).

H6. If the instrument was on the ground in both scenarios, how is it above the canopy in one location?
The above-canopy site was in a clearing at the top of a ridge and the WIBS3 was situated on a small tower. While the clearing was at one end surrounded by trees, the WIBS3 was placed at the opposite end, several metres above the highest foliage.

H7. Is the morphological parameter equivalent to the standard deviation [of the 4 elastic scattering measurements]?
The AF measurement is the fractional standard deviation of the forward-scattered intensity distribution measured at 4 angular offsets, and multiplied by a calibration factor.

H8. How comparable are the measurements from each site, given they were performed at different locations and times?
While 1km apart, the two measurement sites both lay within the same harvesting coupe in 1988 and were each repopulated in the early 1990s in the same way (described by Hewitt et al., 2010; this issue). This leads us to believe that the canopy tree species at each site are comparable. Species survey measurements conducted at the in-canopy

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site support this view and again will be reported elsewhere in this special issue. Net radiometer measurements at the sites show that the typical solar intensity reaching the WIBS3 measurement level below canopy peaks at 50Wm^{-2} at midday in the understorey compared with 1000Wm^{-2} at midday above the canopy.

The largest uncertainty in FBAP transmission calculation arises due to the elapsed time between the end of understorey sampling and the start of above-canopy sampling using the WIBS. This was taken into account by using the continuous data from several other particle counters, deployed continuously at each site, and comparing with WIBS data. In the understorey, data from the GRIMM aerosol spectrometers, which covered almost all of July, showed that the diurnal coarse aerosol cycle recorded by the WIBS3 continued but peak concentrations gradually fell so that the total coarse number had fallen to 0.35 of the value in the averaged understorey dataset outside the hours of 1000 – 1400. This information will be added to the revised manuscript and the discussion will be expanded to include these uncertainties and how they influence the derived transmission factors. Despite this, the general cycle in aerosol number observed below the canopy remains consistent and this is still represented in the smoothed dataset. Furthermore the above-canopy cycle does not appear to be affected in the same way.

We will add additional analysis and discussion of the uncertainty using continuous coarse aerosol number concentration measurements from several other aerosol spectrometers that were deployed at each site. The GRIMM optical particle counter was deployed continuously at the same measurement height as the WIBS3 in the understorey. An aerodynamic particle sizer (TSI APS) was also deployed at the above canopy site continuously. Analysis of this data set shows that when the WIBS3 was above canopy, the understorey nocturnal number concentration for particles in the size range 0.5 – 20 μm falls to approximately 1/3 of the value recorded when the WIBS3 was in the understorey over that period. The resulting gradient between the two sites could therefore be as little as zero at night. This analysis will be included in the revised manuscript since it adds a considerable uncertainty to the derived canopy transfer efficiencies.

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H9. How does RH contribute to PBAP release?
We suspect that the highly consistent diurnal cycle in PBAP number is the result of fungal spores being released from so-called "active dispersal mechanisms" (e.g. Meredith, 1963;Elbert et al., 2007;Ingold, 1939). These mechanisms generally involve the uptake of water from the atmosphere and localised changes in relative humidity, either upward or downward depending on species, trigger the release of spores. We have shown there is a strong diurnal variation in relative humidity which generally rises past 80% at 1500 h each day, approximately the time of the first spikes in number concentration. Similar behaviour is reported in a tropical understorey by Gilbert and Reynolds, 2005.

H10. How well does WIBS3 data compare with that from co-located instruments?
When connected to the same inlet stack as a TSI Aerodynamic Particle Sizer at the above-canopy measurement location the APS reported a secondary mode, suggesting a local source, sized 2 - 2.5μm (which came to be regarded as the PBAP mode) and the WIBS reported its "PBAP" mode at 2 - 3μm. The total number agreement was 70% above 2μm for each instrument, but the basis of the size calculation is completely different: the APS uses the time of flight between two laser beams whereas the WIBS3 uses the intensity of scattered light. As such we feel they cannot be directly compared, although the APS data has been used informally to gain an understanding of what was occurring above the canopy in terms of coarse number concentration. In the understorey the WIBS3 was co-located with a GRIMM 1.108 aerosol spectrometer, and linear regression of all June-July data shows the GRIMM:WIBS mean counting ratio is 0.91 in the GRIMM size interval $0.5 \leq D_P \leq 20$ μm, and the GRIMM reports a secondary size mode at 2 - 3 μm.

H11. What is the meaning of "canopy transfer" in this paper and have similar measurements been performed?
The large horizontal separation of the sites was a logistical factor: Canopy flux measurements were not the primary purpose of the WIBS3 deployment, but instead an estimate of the degree to which the properties of the atmosphere and forest canopy (as a

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source and sink) and how mixing between the two may affect the particle concentration in each location. This will be discussed in greater detail in the paper by Whitehead et al.2010 (this issue) where the connectivity between the two sites will be analysed. A more careful definition of "canopy transmission" will be adopted in the manuscript to make this clear. One benefit of this arrangement is the variety of supplementary measurements that were also carried out in the OP3 project, which were mostly carried out at the above-canopy site and which will be discussed in several associated papers in this special issue.

A1. Is the above-canopy site more affected by wind than the below-canopy site and can wind data be provided?

Understorey wind speed and direction were recorded using a 3D sonic anemometer co located with the WIBS3 inlet. The decision not to show this was taken for brevity because the wind speed is strongly suppressed in the understorey (typical wind speed is 0.1 - 0.2 ms^{-1}) and no causal relationship was found between it and PBAP number. Turbulence data will be discussed in greater detail in further papers, e.g. Whitehead et al. (2010, this issue) as part of the special issue, but we will include a summary figure (such as Figure 2) in the revised manuscript showing wind speed below and at the top of the canopy to allow the aerosol variations to be placed in context with meteorological variables.

Figure 2 shows that wind speed at the canopy top is greater but exhibits a similar diurnal cycle to that in the understorey, peaking at 0.5-1.0 ms^{-1} during daylight. The friction velocity, u*, (not pictured) also rises in the daytime and is evidence of some mixing with the air mass above the canopy. Doppler LIDAR measurements from the clearing some 300-400 m from the in canopy site will be reported elsewhere but this and comparison with the meteorological variables recorded at the GAW Tower site reveals significant decoupling between the sites at night. This decoupling is also evident in the diurnal sensible heat fluxes recorded both below and above the canopy. Again these observations will be reported in the paper by Whitehead et al. (2010 this issue),

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however we will include this information in the revised manuscript. The diurnal variation at 2m however does reflect that at 32m, although there are possible small orographic influences from local hills several km distant observed at the above canopy site. For periods where micrometeorological and WIBS3 data are available, 32m wind speeds were found to correlate positively, albeit weakly, with the non-PBAP number variation (+0.19) and weakly anti-correlated with the PBAP number variation (-0.195). These observations, together with the implications for tropical boundary layer variability and its influences on aerosol mixing, will also be discussed in Whitehead et al (in preparation) along with measurements of eddy covariance particle fluxes in the same special issue.

Changes of above-canopy concentrations might perhaps be alternatively interpreted if wind data were additionally taken into account. Plotting the mean coarse and PBAP number concentration as a function of wind direction (Figure 3, right) and the wind direction histogram (Figure 3, left) show that the wind was almost always from the 130 - 270° directions (North-West and West-South quadrants) and that within this space there is little change in PBAP number loading (red).

Total concentration (in blue), on the other hand, appears to be enhanced when the wind is North-Westerly. Alongside the lag in non-PBAP recovery following rainfall described in the paper, suggest the non-PBAP is transported but the PBAP is locally produced, as one would expect given a large area of forest canopy. There appears to be little correlation between above-canopy PBAP number and wind speed, at least within the small range of wind speeds experienced over the duration of the experiments.

A2. Why were the measurements not performed at different heights at the same location?

The 1 km horizontal separation of the sites was a logistical factor: Canopy flux measurements were not the primary purpose of the WIBS3 deployment, but instead an estimate of the degree to which the properties of the atmosphere and forest canopy (as a source and sink) and how mixing between the two may affect the particle con-

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centration in each location.

A3. How dense and uniform was the vegetation at each location and how much shade and clearings were present at each site; A4. What other minor or major parameters may influence the particle fluxes?

Species survey measurements conducted at the in-canopy site support this view and again will be reported elsewhere in this special issue. Net radiometer measurements at the sites show that the typical solar intensity reaching the WIBS3 measurement level below canopy peaks at 50Wm^{-2} at midday in the understorey compared with 1000Wm 2 at midday above the canopy. The above-canopy measurement site was situated in a clearing at the top of the ridge, and a large clearing was situated approximately east of the below-canopy site over 300 metres distant. Analysis of particle concentrations and turbulence measurements below the canopy as a function of wind direction over the project duration will be included (see Figure 3) to help address this question. Any further discussion of channelling and whether it occurs at this location it is left as a qualitative aspect of the discussion. A more detailed discussion of the flow connectivity between the two sites can be found in the associated paper by Whitehead et al. (this issue) who use fine and coarse aerosol concentration time series from both sites to investigate this matter using auto-correlation lag time analysis to investigate this question. The results of that study do not change the conclusions we present here on coarse aerosol variation.

A5. What is the difference between the WIBS2 and WIBS3?
Please see response to H1.

A6. Can a more detailed description of the instrument set-up be provided (i.e. calibration, inlet properties and flow rate)?
The instrument total flow rate is 2.38L min^{-1}, of which 10% is used for the sample flow. The remaining 90% is filtered and used as a sheath flow to constrain the sample

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flow in the sensing volume. The GRIMM 1.108 aerosol spectrometer has a total flow rate of 1.2 L min^{-1}, all of which is used for the sample flow. The instruments were connected to co-located inlets of similar configuration: 2 metre length with a bend of diameter 15 cm, which is calculated to allow particles up to 10μm to be transmitted without significant losses from particle impaction.

PSL and glass microspheres were used (1μm PSL, 2.1μm, 3μm Green fluorescent PSL, and 2μm glass) to check the alignment of optical components and sizing accuracy. A summary of this information will appear in the revised paper, but the calibration and test particles used were not chosen based on tropical fungi. A number of actual PBAP have been sampled in the laboratory, and this is discussed in our response to Pinnick et al's comments later in this document. Calibration and instrument set-up information will be added to the revised manuscript.

A7. How is humidity likely to affect the instrument's performance?
Large changes in relative humidity can affect the kinetics of certain spore species, as described by Reponen et al., 1996, who report that a change from 30-100% RH will increase the aerodynamic diameter from 1.8μm to 2.3μm in the most extreme case of Cladosporium cladosporioides (fungal spores). Westphal et al., 2003, report size changes of 0.05 –0.1 μm in bacterial spores measuring 3μm in diameter over an RH range of 30% - 80%. The RH range in our study is typically only 70-90% in the understorey with most particles observed in the interval 80-90% RH. Typical RH range is 50-80% above the canopy and we have not observed any intermediate size modes between the two that appear to represent fluorescent and non-fluorescent material. It would be difficult to detect humidity induced changes in coarse mode particle size using the instrumentation described size based as a result. Based on an analysis of fluorescence intensity data from the WIBS3 in the rainforest understorey, the fluorescent intensity of those particles that fluoresce appears to vary only very weakly with RH. As a result we believe it unlikely that RH is not responsible for the apparent appearance of larger, fluorescent particles. No change in detector performance was noted with the

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change in humidity, and we are not aware of any mechanism by which RH is likely to affect the number of supermicron particles reaching the detector in the instrument. Inlet inertial losses were calculated and are predicted to dominate only at particle diameters greater than 10μm.

A8. How would clusters of fluorescent particles influence overall fluorescence?
One would expect this fluorescence to be detected, at least in the NADH channel, if the agglomerated particle is large enough for detection generally. Clusters of bacteria have been measured with the UV-APS in the past (e.g. Agranovski and Ristovski, 2005) although a mechanical stimulus such as a shattering rain drop or high wind would be needed to introduce these into the air.

A9. Can aerodynamic diameter be considered equivalent to optical diameter or can they be inter-converted?
Theoretically the two should not be regarded as comparable, since aerodynamic diameter concerns the amount of time it takes a particle to move a set distance and optical diameter is based on scattering intensity, however both the APS and WIBS3 report modes at similar sizes in their respective number size distributions, albeit with the WIBS3 reporting a less defined distribution. We conclude that at D_A or $D_P \geq 2\mu$m the two can be regarded as similar in practical terms. A mathematical conversion between optical and aerodynamic diameter would have to assume a particular density and refractive index for the aerosol sampled. Both types of diameter measurement are also influenced by effects arising from particle morphology, but not necessarily in the same way. Any conversion would either require much more comprehensive particle information or be subject to a degree of error and, given that the instruments broadly seem to agree on particle size, would in our opinion add little to the manuscript.

A10. Why were other meteorological parameters not recorded, particularly wind?
A suite of meteorological and micrometeorological parameters were in fact recorded (general wind properties are described in the response to question A1) but the decision not to include them in the manuscript was taken for brevity, since we could not identify

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any apparent correlation between them and apparent PBAP number.

A11. The diurnally averaged understory data will be much smoother than that above the canopy. How could this affect the derived result?
We have investigated this point by validating the WIBS aerosol concentration data with several other particle counters that were deployed continuously at the above canopy site, including both Mie scattering optical particle counters and aerodynamic particle sizing (APS, TSI Model 3022) instruments. The biggest difference between the above-canopy time series and the average diurnal dataset is the lack of transient spikes in "PBAP" number each afternoon, which only appear in the understorey WIBS3 data. An inspection of the corresponding above-canopy APS time series of aerosol concentration for sizes $D_A \geq 2\mu$m reveals no such spikes over the duration of the campaign, so the derived transmission efficiency is suppressed at these times, suggesting that the canopy transmission (which is the combined effect of the atmosphere, distance and canopy between the two sites) is very limited. There is also little evidence of a long-term trend in the above-canopy APS data so we believe both WIBS3 datasets to be representative of the general situation at each location for most of the campaign when the decline in understorey concentration is taken into account.

A12. Does canopy transmission efficiency have a size-dependence?
Based on WIBS3 data in each location there does appear to be such size dependence as shown in the graph in Figure 4, however this data is not corrected for the reduction in number in the understorey. That the smaller material above the canopy outnumbers its understorey counterpart suggests that it originates outside the canopy whereas the larger material, as expected, is emitted below the canopy. The relative increase in $D_P > 10\mu$m particles should also be regarded with caution since the uncertainty is large because of low counts throughout the campaign, as shown by the shaded area.

A13. Why do total coarse number concentrations show the same diurnal pattern [as fluorescent particles] and why does PBAP number drop before RH?
The understorey coarse aerosol concentration shows the same diurnal pattern since

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the total concentrations are dominated by PBAP, particularly at night. If the diurnal variation of non-fluorescent particles is examined it reveals a much weaker and different diurnal variation suggesting the dominant diurnal variation is dominated by sources of fluorescent particles, i.e. biological. The nocturnal coarse number concentrations below the forest canopy are commonly observed when relative humidity increases above a certain threshold (see discussion in next section), although this does not always occur, suggesting a more complex relationship between continued sporulation and regeneration. However, in general, higher RH does seem to be associated with active spore release mechanisms which become more effective in these periods (discussed earlier and in Elbert et al. 2007). During the daytime when sporulation mechanisms are weakest, typically between 1000 – 1300 h, on some days we do observe a small divergence between total coarse aerosol number concentration and PBAP number concentration and is likely due to weak turbulent transport of non-PBAP into the canopy. The relative concentrations however are much smaller than nocturnal PBAP concentrations.

The reduction in PBAP number but sustained high RH after 0400 would be consistent with spore release: Once the conditions for active spore release are met, the action of the release mechanism is usually quite short-lived and violent. (the mechanical actions of several fungus species are explored in Meredith, 1963). We believe that 80% RH appears to represent a threshold for spore release by most of the species present at this location. This is also consistent with the single plant measurements conducted (described above). The time series of spore release from the single lichen plant sampled at close range illustrates the semi-sporadic nature of the emission process. An attempt to parameterize this process is ongoing and will be the subject of a future paper.

A14. Can the range of AF values be specified and compared with the obtained values?

The AF measure is calibrated by the instrument manufacturer such that AF=100 represents a fibre and AF=0 represents a perfectly spherical object centred in the sensing

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volume. In general we observe a low AF for PSL microspheres and a higher AF for PBAP, and a general indication of the range of particle morphology is shown in the ESEM images in Figure7, but since this does not quantitatively represent the particles sampled by the WIBS3 it is difficult to map the AF measurements onto a specific particle shape. Instead we opt to discuss the relative sphericity and range of morphologies when the AF distribution contains multiple modes.

Responses to comments by Pinnick et al

On the classification of all fluorescent material as PBAP

To clarify our definition of "biological", we do not rule out non-homogeneous particles. We include spores, bacteria, humic matter, viruses, detritus and pollen. In the existing literature both "primary biogenic" and "primary biological" are used interchangeably when describing such material. We would reserve "biogenic" for aerosols that come about because of biological processes such as sulphates from plankton and secondary organic aerosol from biogenic VOC emissions. Papers dealing with hydrocarbon emissions from ecosystems on land and sulphate emissions from plankton (e.g. Charlson et al., 1987;Andreae and Crutzen, 1997) refer to the resulting aerosol as biogenic, albeit not primary biogenic. As such, the assertion by Pinnick et al's comments that larger particles are more likely to fluoresce because they are more likely to contain fragments of biological material does not necessarily affect our interpretation of the larger aerosol as PBAP, under our definition. Quoted PBAP number or mass in existing literature seems to concurrently describe visually identifiable biological material, culturable material and fluorescent material (albeit much less frequently). We welcome the chance to provide additional clarity in this area and feel that adopting the Huffman et al classification of fluorescent biological aerosol particles (FBAP) is beneficial. It is not immediately clear to the reader that the fluorescence technique tends to report larger PBAP numbers than culturable approaches (e.g. Agranovski et al., 2003) and the ability to convey this, along with the pitfalls of the technique, in the notation may be helpful to the reader.

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Where the current manuscript is concerned, we feel that it would be helpful to change our criterion for PBAP from 'either fluorescence channel exceeding its threshold' to 'both channels exceeding their thresholds' and using the former to denote the upper limit of fluorescent aerosol number and the latter as the principal PBAP number, with a short discussion of possible reasons for the disagreement in number between the channels. The diurnal cycle observed is changed little by this, although as expected the overall reported "PBAP" number falls by 10-20%.

The likelihood that PBAP can represent more than 50% of the coarse aerosol number

Assuming for a moment that the fluorescent particles in this study are indeed all fungal spores the diurnal variation and number concentrations in the understorey of tropical rainforest in Queensland, Australia, are comparable to those obtained by Gilbert and Reynolds, 2005, who observe confirmed spore concentrations $1000L^{-1}$ at night and a steep reduction in the morning to $100L^{-1}$, consistent with the data presented in the manuscript and representing that fungal spore emissions alone can dominate the nocturnal coarse number concentration in a tropical understorey. Gilbert and Reynolds also demonstrate, as have several others (e.g. Grinn-Gofroń and Mika, 2008;Troutt, 2001), that temperature and relative humidity appear to be important factors in predicting spore concentration.

The use of the each of the WIBS3's measurement channels to discern "PBAP"

While no formal combination of the elastic and inelastic scattering measurements was used to separate PBAP, the paper was arranged such that the selection of particles based on fluorescent intensity was a starting point for classification, and the size and Asymmetry factor (AF) measurements independently show contrasting properties of the fluorescent and non-fluorescent material. Furthermore, as we demonstrate, the diurnal variation of fluorescent and non fluorescent particles is very different. At its peak, around 70% of the WIBS3-reported PBAP number lies in the 2 5μm size range, ruling out most pollens and plant fragments since these are too large. Individual bacteria

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are at most around 1μm in size and viruses travel on larger particles. The remaining possibilities are clusters of bacteria, spores and the non-pure PBAP described by Pinnick et al in their review of the manuscript. These cannot be ruled out and will be discussed in the revised manuscript, although we believe that spores originating in the canopy dominate the PBAP number concentration below the canopy because of the lack of strong mechanical drivers such as wind or rain correlated with the coarse number concentration increase each day.

As part of an effort to identify individual sources in the understorey a single plant experiment was conducted in the field using lichen. This was monitored for 18 hours at the end of the measurement period. The time series of total and PBAP (both channels registering fluorescence) number is plotted in Figure 5.

In terms of the agreement between the number of fluorescent particles according to each channel: $N_{NADH} = 0.94 N_{TRYP}$ and fluorescent particles can be seen to dominate the total coarse particle concentration. The size mode during this experiment shifts to between 4-5 μm in this instance, although AF increases only slightly to a mode of 18, indicating the particles released from the Lichen are similar to those measured in ambient air. The number concentration is subject to fluctuations that are more intense and frequent than the overall PBAP number in the understorey since the measurements were made much closer to the source (within 2 cm of the plant) but the same overall diurnal response is still observed: a large initial peak in late afternoon and a decrease in overall number before sunrise (0700). The overall number concentrations measured during these obvious sporulation events are significantly larger than those observed in general in the understorey. It was determined by survey that no individual plants lay within 1 metre of the inlet on the mast where the WIBS was sited. This difference will be due to the fact that there will be significant loss by sedimentation of these coarse mode particles within very short distances from individual sources.

Despite this the similarity in diurnal concentration variation, size and fluorescence characteristics of the particles observed both from the single plant experiment and ambient

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measurements are consistent with the view that the majority of these particles are from biological sources. Figure 6 shows the diurnally averaged total (black line +/- 1 standard deviation in grey), non-fluorescent (dashed line) and PBAP (green line; requiring fluorescence in both channels) in the understorey (upper panel) and above canopy (lower panel).

The non-fluorescent number variation bears little resemblance to the PBAP number in both locations and the fact that above-canopy PBAP number shows a weak correlation with the below canopy PBAP, presumably due to the distance and from the main sources, and the canopy itself strengthens this view.

Validation of WIBS3 data using electron microscopy

In an attempt to verify the WIBS3 readings, Nuclepore filter samples (co-located with the WIBS3) were collected at random intervals throughout the campaigns and analysed using an Environmental Scanning Electron Microscope (ESEM).Although not quantitative in terms of atmospheric number concentrations nor having the temporal resolution of the WIBS data, the majority of particles imaged were seen to be clearly biological and moreover their sizes conform closely to the mode in FBAP measured by the WIBS. Some examples of these biological particles are included in Figure 7 to support this view.

Nearly all of the images obtained in each location showed what appears to be spores or pollen grains, the majority of which are as indicated above between 2-4μm in diameter in their dehydrated state. A semi-random selection from each site is shown, but this is not intended to indicate the relative abundance of particle types. None of the filters in this non-exhaustive analysis showed particles resembling black carbon. Of the subset of particles found on the filters the vast majority resembled PBAP. Interestingly a range of morphologies is observed and this partly informs our statement in the manuscript that the broader AF distribution for larger particles represents a bigger range of morphologies.

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Why thresholds are apparently not based on lab measurements of fungal spores?

As a result of the biodiversity of the rainforest we believe it would be difficult to defend setting fixed "PBAP" criteria in fluorescence or elastic scattering channels when it is unclear what species, or even how many species, are being sampled. A detailed discussion of the fluorescent intensity sampled from each particle is, we believe, beyond the scope of the current manuscript but we are happy to provide additional information about the response of the WIBS3 in-situ and based on extensive tests in the laboratory. Dried Lycopodium, Johnson and Bermuda grass spores (sized 6-10μm) and fresh spores from a household fungus have been sampled in the laboratory using the WIBS3 and exhibit varying fluorescent properties. The mean distributions of fluorescent intensity divided by forward elastic scattering intensity are plotted in Figure 8 for particles crossing the threshold of the Tryptophan and NADH channels in the upper and lower panels respectively.

All biological species sampled crossed the NADH fluorescence threshold frequently and far more often than the Tryptophan threshold, whose recorded fluorescent intensity varied more by species. The most extreme example of this is Lycopodium (yellow line), where fewer than 10% of spores registered any Tryptophan fluorescence whereas more than 90% did so for NADH, although this was by far the most aged (> 10 years) sample used. In contrast the fresh household mould spores (grey line) consistently exhibited fluorescence in both channels. As discussed previously this is also true of the ambient understorey aerosol (green line) and somewhat true of the above-canopy (blue line) aerosol, though not of an urban location (brown line) where combustion aerosols would likely play a bigger role.

Comparing the understorey and above canopy NADH fluorescence histograms, some structure is present above canopy at low intensity though the majority of the curve follows the form of that in the understorey. We do not intend to try and discern different biological species from WIBS3 data, but the fluorescence histograms from Lichen-

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dominated particles (orange plots) have more in common with the ambient tropical aerosol and mould spores than they do with the other spores or ambient urban air. It is certainly conceivable that the recorded instance of lichen contributes to the understorey aerosol although the intensity and regularity of its emissions compared with the understory ambient N_{PBA} time series suggest it is not primarily responsible for what is observed.

In the entire June-July understorey dataset the correlation coefficient between the number of particles fluorescent in the Tryptophan and NADH channels is 0.98. (A linear regression shows N_{NADH} = 0.9665N_{TRYP}). Above the canopy the number agreement is reduced (N_{TRYP} = 0.66N_{NADH}) and the correlation coefficient is 0.90, indicating some similarity to that in the understorey, though less is fluorescent in the Tryptophan channel. This might be expected due to ageing of the aerosol above the canopy. Measurements that we have conducted with the WIBS3 in urban and rural environments in the UK, where one would anticipate more combustion aerosol interference, particularly in the smaller aerosol size mode, generally show a lower coarse number concentration and much smaller fluorescent number fractions (< 10%) and N_{TRYP} is typically 0.25N_{NADH}. This is consistent with the observations reported by Huffman et al from a semi-urban site using the UV-APS (ACPD 2009). We plan to produce another paper including a detailed discussion of how the fluorescence signatures relate to one another in the OP3 dataset. These measurements suggest that most fresh, genuine PBAP fluoresce in both channels but aged PBAP are not detected as frequently in the Tryptophan channel, giving rise to an artificially lower reported concentration if we require dual-channel fluorescence.

The likelihood of understorey fluorescent aerosol originating remotely.
Alongside the existing measurements, 4 GRIMM 1.108 standard aerosol spectrometers were installed in profile at 8m intervals from the understorey to the canopy top. When the highest PBAP numbers are reported by the WIBS3 (1400 – 1800) the profile system reports that the highest supermicron number concentration occurs in the lower

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understorey. Aerosol profile measurements and micrometeorology in and above the canopy are discussed by Whitehead et al in this special issue (paper in preparation). The supermicron number concentration reported by the above-canopy APS is consistently lower than 1000 L^{-1} while the WIBS3 was in the understorey. Finally, the understorey diurnal cycle does not appear to depend on wind direction (recorded above the canopy) or the origins of air masses reaching the measurement sites (using modelled back-trajectories), each of which vary significantly throughout the campaign. As a result, we do not believe the source of fluorescent material to be outside the canopy. The above-canopy WIBS3 measurements are slightly periodic, rising in number at night, but each dataset exhibits the same AF and size modes and a similar size-resolved diurnal cycle is detected in each location (Figure 9), we believe the two locations to share a common PBAP source: the canopy. The plots in Figure 9will be converted to dN/dlogD_P and used to supplement the diurnal number concentration plots in the revised manuscript.

Summary of response

Some additional detail of the measurement locations, how the data was interpreted and the likely errors introduced will also be added to the methodology, as requested, however it should be noted that these are described in greater detail in the associated special issue papers to which our manuscript contributes and in the attendant technical citations. The revised discussion and conclusion will include further discussion on the uncertainty arising from the non-simultaneous WIBS3 measurements above and below the canopy and elaborate on the arguments for excluding certain types of particles in this tropical forest environment which is located in a mosaic of large islands subject to marine influences and is also remote from major pollution sources. It therefore differs significantly from studies reported from other locations, e.g. Amazonian forests.

Summary of planned revisions

• References to the WIBS3 will be supplemented with Pan et al., 2007, Foot et al., 2008 and Kaye et al., 2007. Complete definition of the AF and optical size parameters

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as measured by the instrument will be added.
• We have refined the analysis by requiring PBAP candidate particles to exhibit fluorescence in both UV channels rather than just any one channel and explaining briefly the relationship between the channels and the influence this has on the data sets.
• An overview of current UV-fluorescence aerosol measurements
• Additional introductory material involving estimates of PBAP loadings, supporting measurements conducted into UV-fluorescence measurements (as suggested in comments from Huffman and Pinnick et al) and potential sources of interference.
• Supplementary figures: wind data and size-resolved diurnal number concentration and fluorescent number fraction and re-rendering current graphs in the correct size. A separate graph of the diurnal non-fluorescent particle concentration will be added to emphasise the different mechanisms controlling these different classes of particles.
• Additional discussion about the ambient aerosol in each location when the WIBS3 was not present there and how concentrations were validated using data from other aerosol instruments.
• Reformulated discussion about what canopy effects can and cannot be attributed to the data presented and improved comparisons with previous data (e.g. Gilbert and Reynolds, 2005)
• Brief discussion of the role of wind speed, micrometeorology and boundary layer dynamics with reference to the paper by Whitehead et al. (2010, to be submitted in this special issue).
• More detailed discussion about the derived "transmission efficiency" and its implication.

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Figure 1

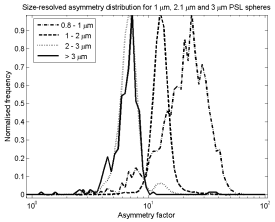

Fig. 1.

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Interactive Comment

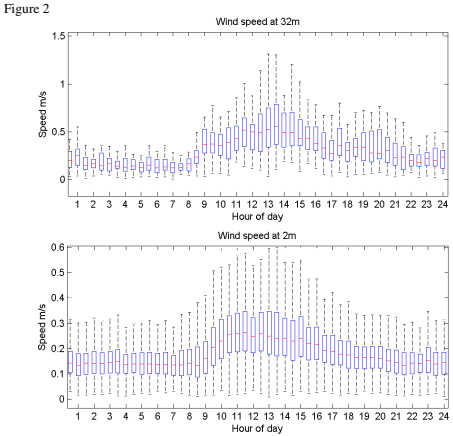

Fig. 2.

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Figure 3

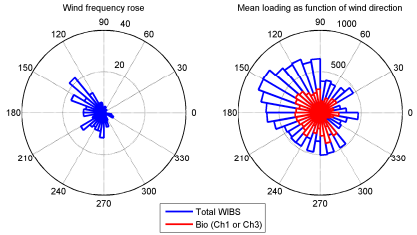

Fig. 3.

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Figure 4

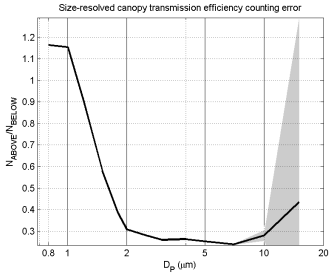

Fig. 4.

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Interactive Comment

Figure 5

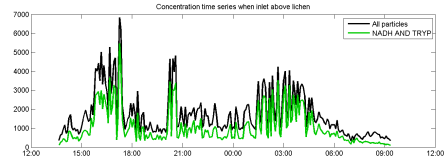

Fig. 5.

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Figure 6

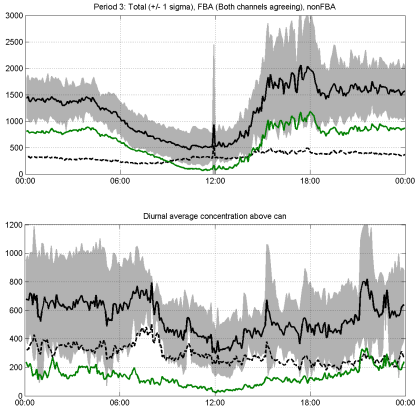

Fig. 6.

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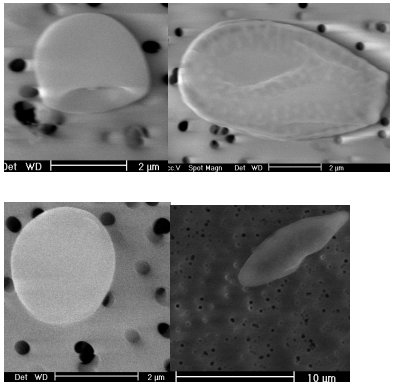
Discussion Paper

Figure 7
Below canopy



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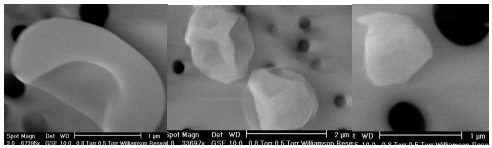

Fig. 7.

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Figure 8

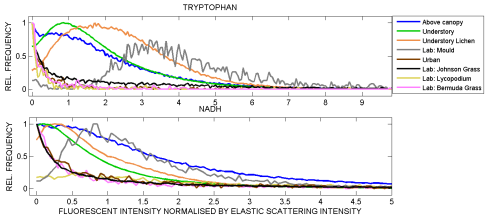

Fig. 8.

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Figure 9

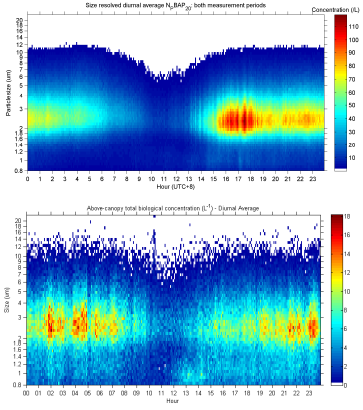

Fig. 9.

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