Response to referee comment on amt-2017-170 by Savage et al.

Anonymous Referee #1
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Note regarding document formatting: black text shows original referee comment, blue text shows author response, and red text shows quoted manuscript text. Changes to manuscript text are shown as *italicized and underlined*. Bracketed comment numbers (e.g. [R1.1]) were added for clarity. All line numbers refer to discussion/review manuscript.

General Comments: The manuscript is very well written and I believe of great relevance to the bioaerosol scientific community. The authors present very interesting and novel work testing a Light induced fluorescence (LIF) instrument (WIBS-4A) whilst attempting to display the data in new ways. Thus I believe the paper should be published upon the correction of some minor technical/specific issues discussed below.

Author response: We thank the referee for his/her positive assessment and summary.

Specific/technical comments:

[R1.1] L196 I believe that this line is misleading, while a value of 0 does indicate a particle is a perfect sphere values just above this do not indicate that they are rod-like as directed by the sentence "Whereas larger AF values greater than 0 and less than 100, indicate rod-like particles" What is the average/median AF value seen for PSL for instance? I doubt they are seen to be 0. Values increasing towards 100 do indicate an increasing rod-like morphology however Indeed placement of the AF values of the PSL sphere in table one would be useful.

[A1.1] As requested, we added median values (± standard deviation) of AF to Table 1 for PSLs.

To clarify the statement we added text in this paragraph at L198 (italicized text added): "A perfectly spherical particle would theoretically exhibit an AF value of 0, whereas larger AF values greater than 0 and less than 100 indicate rod-like particles (Kaye et al., 1991;Gabey et al., 2010;Kaye et al., 2005). *In practice, spherical PSL particles exhibit a median AF value of ~ 5 (Table 1).* It is important to note that *the AF* parameter is not rigorously a shape factor like used in other aerosol calculations (DeCarlo et al., 2004;Zelenyuk et al., 2006) and only very roughly relates a measure of particle sphericity."

[R1.2] L 302 What is a blade of air? Blast perhaps?

[A1.2] We added text at L302 to clarify the description of the experiment. "For each experiment, an agar plate with a mature fungal colony was sealed inside the chamber. *A thin, wide* nozzle was positioned so that *the delivered air stream approximated* a blade of air *that* approach*ed* the top of the spore colony at a shallow angle in order to eject spores into a *roughly* horizontal trajectory."

[R1.3] L 337 What was considered sufficiently fine?

[A1.3] We added clarifying text at L337:
"The setup was modified (method P2) for a small subset of samples whose solid powder was sufficiently fine to produce high number concentrations *of particles (e.g. >200 cm^{-3}) and that contained enough* submicron aerosol *material to* risk coating the internal flow path and damaging optical components of the instrument."

[R1.4] Table 2 Pyrdoxine particle 7 in Biofluorophores has no number in the saturated column

[A1.4] We added missing values for Pyrdoxine in Table 2.

[R1.5] Were there any issues with contamination whilst using a NAD?

[A1.5] There were no contamination issues while running NAD, but the fear of contamination was one reason we employed aerosolization method P2. Between each sample, the instrument ran pumping for about 10 min to prevent contamination. If the baseline of that ambient data collected in those 10 min was higher, other measures were taken to ensure the optical cavity was not coated.

[R1.6] L555 Are intact pollen not counted? Or do they saturate the sizing detector and are thus mis-sized?

[A1.6] Intact pollen that make it into the instrument are counted. Most pollen grains are much larger than the upper size limit of the instrument (~20 µm), however. Thus, species of pollen with large grain sizes exhibit a size mode in the WIBS near this upper size limit. (e.g. Pollen 1, 2, 5, etc.). Any particles larger than this are integrated into the largest sizing bin, which saturates the sizing detector. A clarifying sentence was added:

L557: "… upper size limit of particle collection (~20 µm as operated). *Particles larger than this limit saturate the sizing detector and are binned together into the ~20 µm bin.*"

[R1.7] L560-3 Given that the pollen are disrupted, they now have the intine of the pollen exposed. Thus is it this rather than the fraction of the pollen that is radiated the most important?

[A1.7] The intact pollen and fragmented pollen indeed present different types of material to the excitation pulses and may, therefore, present different emission properties as a result. We believe the following, existing text clarifies this point:

L557: "It is important to note that excitation pulses from the Xe flash lamps are not likely to penetrate the entirety of large pollen particles, and so emission information is likely limited to outer layers of each pollen grain. Excitation pulses can penetrate a relatively larger fraction of the smaller pollen fragments, however, meaning that the differences in observed fluorescence may arise from differences the layers of material interrogated."

[R1.8] L609 should the line say "adds either A and C" rather than "adds either B and C"

[A1.8] This was a typo. The text was modified to correct this error:

L 609: "The "pathway" of change, for Pollen 9, starts as A-type at small particle size and adds B and eventually ABC (A→AB→ABC), whereas Pollen 8 starts primarily with B-type at small particle size and separately adds either ~~B~~ *A* or C en route to ABC (B→AB or BC→ABC)."

[R1.9] L647 tryptophan does not appear to follow A –> BC –> ABC pathway from visual inspection of the associated graph.

[A1.9] This was also a typo. The pathway listed for tryptophan was correct, as follows:

"For example Biofluorophore 1 (riboflavin) follows the pathway B→C→BC while Biofluorophore 11 (tryptophan) follows the pathway A→~~BC~~ *AB*→ABC."

[R1.10] Similarly in the discussion of the pathways for riboflavin the particles appear to have either B or C character to start with before gaining the required character to become BC. The pathway you describe does not suggest this. It suggests that particles pass from B to C to BC

[A1.10] The referee brings up a good point here. The concept of "pathway" here does not make sense to move from B to C to BC. Instead, there is a population of B particles and a separate population of C particles, each of which can separately move to become BC particles as particle size increases. To clarify this, the text has been changed as follows:

L646: "For example Biofluorophore 1 (riboflavin) follows the pathway B *or* C→BC …"