Response to referee comment on acp-2016-743 by Gosselin et al.

Anonymous Referee #3
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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*. 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 comparing data from modern Light/Laser induced fluorescence (LIF) instruments with molecular tracers such as arabitol and mannitol. The paper also attempts to display the data in new ways scaling particle number to mass concentrations. The paper is very well cited and builds well on previous work. 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 Comments:
Comment 1: L62-63 "For example, asthma and allergies have shown notable increases during thunderstorms due to elevated bioaerosol concentrations" This is indeed true however allergic rates have been climbing in recent years and I feel this should be incorporated. I suggest using the reference. Linneberg, A., 2011. The increase in allergy and extended challenges. Allergy, 66(s95), pp.1-3.

The Linneberg reference was added to L62.

Comment 2: L139 should H2O have a sub-scripted 2

This was corrected in the revised manuscript.

Comment 3: Were the differences in sampling lines of the WIBS and UV-APS calculated? Reynolds number for instance?

We did not calculate the Reynolds number or quantify possible difference in the two sampling lines. The lines from the inlets were somewhat different in length (~4.5 m for the UV-APS and <1 m for the WIBS), but both were arranged to minimize bends and were oriented vertically. Thus, the differences in particle number concentration from the inlets and lines is likely minimal.

Comment 4: Were all particles assumed to be spherical for the density calculations or was the WIBS ability to determine shape utilized?

All particles were assumed to be spherical for particle mass calculations. Particle morphology could impact particle mass calculations, however, the asymmetry factor (AF) provided by the WIBS has not been characterized sufficiently to understand the relationship of this parameter to particle morphology. As a result, we did not utilize AF. To clarify ambiguity, the text was revised at L160-162 as follows:
"Total particle number size distributions (irrespective of fluorescence properties) obtained from the UV-APS *and WIBS* were converted to mass distributions ~~using~~ *assuming spherical particles of* unit particle mass density as a first approximation ~~for all direct comparisons with tracer mass and~~, unless otherwise stated."

Comment 5: Do you believe that cluster 1 is solely a fungal spore cluster, given its size range overlaps with that of some bacteria?

> The organization of clusters from the raw data is a function of the mathematical algorithms utilized and is relatively robust. The assignment of names or sources to the derived clusters is much more uncertain. While Crawford et al. (2015) assigned Cluster 1 to be fungal spores, this should be taken loosely. It is very possible that some fraction of non-fungal particles have been conflated with this cluster. Without direct comparative evidence there is no way to confidently know the source or category of each particle. For example, even the cluster assignment of even polystyrene latex particles of known type was reported as only 98% in a previous publication (Crawford et al., 2015). To clarify this point we have added the following text to the end of L203. "*It should be noted that assignment of names and approximate origin (e.g. fungal spores or bacteria) to clusters is approximate and does not imply particle homogeneity. Each cluster likely contains a small percentage of contaminating particles. For more details see Robinson et al. (2013) and Crawford et al. (2015).*"

Comment 6: Why was a rainfall accumulation threshold of greater than 0.201 chosen?

> A threshold of 0.201 represents a normalized and unitless value that takes into account both disdrometer and tipping bucket measurements. This value was chosen arbitrarily based on the following reasoning. Rain events that presented <0.201 often did not coincide with other indicators of rain such as increased fluorescent particle concentration and RH. When the threshold value was increased to 0.201 we observed more continuity in the measurements that are indicative of rain events.

Comment 7: What did the correlations look like before the manual reclassification of some of the rain/dry periods? How much did this effect it?

> Regarding the correlations, manual reclassification by wetness category increased the R^2 values in all cases. For example, prior to reclassification the mass correlation of arabitol with WIBS cluster 1 during rainy periods was 0.77 after reclassification the R^2 value was 0.82. This trend of increased R^2 was observed with other correlations for both rainy and dry periods.

Comment 8: L 430-431. The Hill 2009 reference does talk about increased wetness effecting the fluorescent properties in comparison to dry samples however in this study wet samples were particles suspended in solution rather than particles at higher relative humidity's. **(a)** I believe that this line should be rewritten. **(b)** Do you believe the particles sampled during wet periods to be in droplets or to have increased moisture content? **(c)** Could a moistened PBAP have increased fluorescence due to fluorescent compounds being extracted/leached to its surface?

> These are interesting questions that were somewhat beyond the scope of the ambient study performed here and thus we did not fully investigate them.
> **(a)** Taking this comment into account we revised this sentence (L 430-431) to be more accurate: "This *could* impact the fluorescence properties of the fungal spore particles *that have different amounts of adsorbed or associated water* (Hill et al., 2009; 2013; 2015)."
> **(b)** As far as the moisture content of individual spores, we have no direct evidence either way. It is possible that some of the spores were fully contained within water droplets, either as a by-product of the high RH and deliquescence or because spores were actively ejected by fungus and thus encased in a small droplet. Upon interrogation within the UV-LIF instruments, however, the spores were almost surely not activated within a droplet, because of the size ranges observed. If they were encased within a droplet the average size would have likely been too large for the UV-

LIF instruments to sample efficiently and we would not have observed the dominant 2-6 µm modes.

(c) We are aware of no studies that directly link increased fluorescence with the leaching of fluorescent compounds from the interior to the surface of a particle. However, (Hill et al., 2013; 2015) showed that the water content associated with bacterial aerosols significantly affected their fluorescence properties, which led to the brief statement quoted above.

Comment 9: Was there much difference in fluorescent intensity for FAP on Dry and Wet periods?

We did not perform this analysis as a part of this study. But, intrigued by the referee's question we calculated average fluorescence intensity from two samples (one Rainy, one Dry) as examples. Hi Vol sample 8 was a dry sample with intensities as follows: FL 1, 872 ± 718; FL 2, 654 ± 277; FL 3, 497 ± 347. Hi Vol sample 16 was a rainy sample with intensities as follows: FL 1, 1687 ± 613; FL 2, 740 ± 333; FL 3, 707 ± 493. In this example, FL1 intensity increased by a factor of 2, FL2 intensity only nominally increased, and FL3 intensity increased by ~40%.

Comment 10:

- L555 Should "Figures 6 c-f" read "Figures 6 d-f"?
  - Corrected
- L560 Should "Figure 6 c, d" read "Figure 6 d, e"?
  - Corrected
- L567 Should "Figure 6 e, f" read "Figure 6 g, h"?
  - Corrected

Comment 11: For the total particulate matter mass concentrations why did you not use the high volume sampler samples to determine the total mass? Instead of the UV-APS measurements.

- Filter mass was not measured before and after sampling and so it was not possible to estimate total particle mass using these filters. As a result, we estimated particle mass using the integrated mass from a particle sizing instrument.

Comment 12: You mention Cladosporium are generally present/released at dry periods was there any evidence that this occurred during this campaign?

The observation that *Cladosporium* spores are present in highest concentration during dry periods has been reported many times and is generally well accepted (De Groot, 1968; Oliveira et al., 2009). For example, it was shown for a study in rural Ireland that both WIBS and UV-APS instruments poorly detected *Cladosporium* particles (Healy et al., 2014). Unfortunately we have no direct observations of this from the campaign. We collected particle by impaction (Sporewatch drum sampler), but it malfunctioned and we have no direct microscopy samples to show relative spore concentrations. The DNA analysis shows relative diversity, but does not provide quantitative evidence that can support the suggestion that *Cladosporium* was present primarily during dry periods.

References

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De Groot, R.: Diurnal cycles of air-borne spores produced by forest fungi, Phytopathology, 58, 1223-1229, 1968.

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