

Author response to reviewer #1

Thank you for your thoughtful review and for the recommendation to publish the manuscript with after minor amendments discussed below. Reviewer comments below are in italics.

Specific referee comments (R) and point-by-point author responses (A):

R1.1. *The first sentence of the abstract is difficult to read past "...in many environments, may thus influence..." and I suggest fragmenting this sentence.*

A1.1. In response to the reviewer's suggestion regarding the first sentence of the abstract, the authors have re-structured the opening sentence of the abstract to read as follows:

“Primary biological aerosol particles (PBAP) can contribute significantly to the coarse particle burden in many environments. PBAP can influence climate and precipitation systems as cloud nuclei while also play a role in the spread of disease to humans, animals, and plants.”

R1.2. *Page 3878 line 15, consider using a comma after "Recently".*

A1.2. The authors would like to thank the reviewers for the recommendation of inserting a comma after the word recently (Page 3878 line 15) and have inserted comma as suggested.

R1.3. *Page 3878 line 17, The acronym IN first appear but is not specified.*

A1.3. *The authors have now specified IN as Ice Nuclei in the text of the manuscript as suggested.*

R1.4. *Please consider adding to the introduction a very brief description illustrating how important the context of the biological aerosol types measured by these instruments are within the bigger biological field, e.g. I would like to be able to have an indication in the text as to whether these instruments capture the full bio-aerosol picture.*

A1.4. A detailed understanding of how the UV-LIF techniques discriminate PBAP is important, but complicated. We introduce some of this complexity within the manuscript by introducing the idea that certain fungal spores (e.g. *Cladosporium* spp.) are not likely detected with high efficiency by either instrument. We also discuss that particle size is an important characteristic for detection, as it is within all optical particle sizing instruments. We appreciate the reviewer's comment, and added a short summary of this information in the introduction to introduce the main aspects of the instruments that contribute to the ability to miss identify particles as non-biological. On Page 3879, Line 20 we inserted the following text:

“In contrast, no real-time technique for PBAP analysis is able to comprehensively detect all classes of biological material. Microorganisms too large or too small for efficient collection by UV-LIF instrument will undercount these particles and some PBAP may fluoresce too weakly to be detected in many circumstances.”

R1.5. *Within the text associated with the description of Figure 2 there are comments about how comparable the Spore concs in (a) are with the measurements FL1-3 and UV-APS (b-e). This is difficult to judge and becomes clearer once the text moves to Figure 6. Maybe this could be pointed out in the text.*

A1.5. To clarify the text as suggested by the reviewer we have added a sentence to P3888, L29:

“Correlation analysis discussed later highlights the agreement further (Fig. 6).”

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Author response to reviewer #2**

Thank you for your thoughtful review and for the recommendation to publish after changes associated with the following comments are made. Reviewer comments below are in italics.

Specific referee comments (R) and point-by-point author (A) responses:

R2.1a. *.....the atmospheric aspect of the presented findings could be emphasized more throughout the manuscript. This mainly concerns the atmospheric observations embedded into section 3.3 “Real-time fluorescence sensors vs. Sporewatch” which should be presented in a section of their own (comparable to section 3.4 “Marine particle influence”). As an example: The authors report on “observed trends that many bioparticle classes correlate strongly with RH and peak at night” (p3891, l18ff). The atmospherically interested reader would expect an explanation (or assumption) here, not only a reference.*

A2.1a. We have added several sentences related to atmospheric observations and have also re-ordered Section 3.3 to make points clearer. From Section 3.3 we have split additional sections “3.4 Diurnal trends and atmospheric implications” and “3.5

Weakly detected *Cladosporium* spores". Examples of added text (underlined text added):

At P3891, L 19: "This is consistent with commonly observed trends that many bioparticle classes correlate strongly with RH and peak at night due to active fungal emission mechanisms that require high humidity to function."

At P3892, L17: "...and the concentration of *Cladosporium* spp. spores, which are among the most common spore types in vegetated areas, shows a relative increase during the middle of the afternoon (peaking approx. 14:00)."

R2.1b. *Also, a lot of technical terms seem to be more familiar to microbiologists or scientists which actively work with bioparticles – but not to the ACP community ("flavine", "hyaline") – and should be explained briefly.*

A2.1b. The authors made an effort to clarify as many additional terms as possible throughout the manuscript. Examples of textual changes are given below:

hyaline: Although the term hyaline was already briefly defined in the manuscript we have expanded this definition to include the following text in the revised manuscript (P3885, L6):

"Further, many fungal spores are hyaline (translucent, glassy appearance when examined by microscope) in nature and are therefore difficult to enumerate via optical microscopy."

flavin: Clarifying text added at P3888, L13: "flavin compounds (naturally occurring pigments, including riboflavin)".

R2.1c. *The atmospheric relevance and origin of the spore species assumed to be detected should be discussed briefly (or noted at the relevant position in the text).*

A2.1c. As suggested the atmospheric relevance of spore species assumed to be detected are now discussed briefly at the relevant position within the revised manuscript, including:

At P3889, L9: "Ascospores are specific to fungi classified as ascomycetes which are thought to be represented in all land ecosystems worldwide. A basidiospore is a reproductive spore produced by Basidiomycete fungi and Ganoderma is a genus of polypore mushrooms which grow on wood"

R2.2. *In section 3.1 the total particle comparison between WIBS-4 and UV-APS is presented, and the authors report on a discrepancy starting at approx. 50 cm⁻³. Is this number expected to be universal, i.e. is there a technical explanation for that? Could it be a coincidence error in the WIBS optic? Particle coincidence is not discussed at all, and as a reader I would expect not only a lower threshold for particle detection, also an upper threshold (in number). Is there any information on that number except from the one obtained from Fig. 1? What means a number of 50 per cc from an atmospheric perspective/typical atmospheric number concentration?*

A2.2. It is unfortunate that the WIBS and UV-APS total particle counts are not exactly equal, but this is very unlikely as two instruments (even of the same type) rarely agree to greater than 5%. The offset above 50 cm⁻³ is likely a result of increasing particle coincidence, as the reviewer points out. A sentence clarifying this technical explanation has been added to the text at P3886, L24:

“The shallower slope to the correlation above 50 cm⁻³ is likely a result of particle coincidence that reduced particle counts within the WIBS at a lower concentration than within the UV-APS as a result of differing physical instrument parameters.”

R2.3. (Minor Point 1) P3879, l9/10: “Emission related excited by . . .” would better read as “Emission related to excitation by . . .”

A23. Thank you for this observation. The typo has been corrected as suggested.

R2.4. (Minor Point 2) P3881/l28: Relates the 0.5 μm to the $D50 = 0.49$ mentioned beforehand, or does this value comes from an independent measure of the optical capabilities of the instrument?

A2.4. The value 0.5 μm is a rounded value of the $D50 = 0.49$ which was determined in a separate study referenced within the text i.e. Healy et al. (2012b).

R2.5. (Minor Point 3) P3884/l1: the acronym “PMT” is used earlier in the text already (e.g. p3881/l18).

A2.5. The acronym “PMT” is now defined at the point it is first used (P3881, L18).

R2.6. (Minor Point 4) P3887/l15 to l25: Is there a way to simplify this statement?

A2.6. The text has been revised to include the following additions (underlined text added):

“There is no doubt that the assumption that detecting fluorescence from these channels implies actively metabolizing cells significantly over-simplifies the perspective of airborne microorganisms (Pöhlker et al., 2012; 2013). The broad nature of fluorescence excitation and emission spectra along with the relative similarity of excitation wavelength between these channels of the two instruments leads to broad consistency between the WIBS FL3 and UV-APS trends.”

R2.7. (Minor Point 5) P3890/l7ff: A short note on how the cited studies “estimated” the concentration of bacteria over vegetated surfaces would be helpful to put the much higher number of measured PBAP into perspective.

A2.7. As suggested by the reviewers, the authors have now inserted a short note within the text as follows:

“The cited studies estimated the concentration of bacteria over vegetated surfaces by considering large number of studies collectively which use different methods of detection e.g. DNA sequencing, when compared to the current work which uses online particle autofluorescence.”

R2.8a. (*Minor Point 6a*) P3890/l9ff: *Here, assumptions are listed why the fluorescence sensors and Sporewatch disagree in number. Lots of “likely . . . but”, “unlikely”, “less likely” makes the text confusing so that the outcome/possible reasons for the disagreement are blurred.*

A2.8. This section was clarified by removing the “likely” terms where possible, as suggested by the reviewer. Additionally, the section text was organized more fluidly by enumerating each of the three hypotheses presented here.

R2.8b. (*Minor Point 6b*) *Also, shouldn’t be the SOA and soot particles much smaller, i.e. below or close to the lower size limit of the WIBS/UV-APS?*

The text as published in the ACPD manuscript mentions that these particles are expected to be $< 2 \mu\text{m}$. We have further clarified the manuscript with the following text (underlined text added):

These particles could be certain types of absorbing brown carbon secondary organic aerosol (Bones et al., 2010; Gabey et al., 2013; Lee et al., 2013) or soot particles (Lewitzka and Niessner, 1995; Panne et al., 2000), as adsorbed coatings of small particles (Huffman et al., 2012) or as discreet particles of size $< 2 \mu\text{m}$.

R2.9. (*Minor Point 7*) P3891/l6: *“This is unlikely...”*. *This sentence does not show any continuity and should be rephrased.*

A2.9. The text has been changed to: “The Sporewatch undercounting is unlikely to ...”

R2.10. (*Minor Point 8*) P3894/l26: *Number of 2nd mode is missing.*

A2.10. This omission was erroneous, and the text has been changed to: “...comprised of 1 and 3 μm modes ...”

R2.11. (*Minor Point 9*) P3894/l10: *“better” or “higher” resolved distribution instead of “more resolved”?*

A2.11. The text has been changed to “more highly resolved”

R2.12. (*Minor Point 10*) P3908/Fig. 1 and P3910/Fig. 3: *“grey” vs “gray” – be consistent with BE vs AE.*

A2.12. All instances of this word have been changed to “gray”.