

Comments to the Referee 1#

We acknowledge the reviewer for his/her notable comments and pensive feedback on our manuscript. Stylistic and typographical errors have been noted and will be corrected in the revised manuscript. The grammar and the English of the manuscript will also be improved in the revised version. Referee comments (given in italic and blue) and our specific responses can be found below.

Specific Comments

P17615, L24: Does your baseline have any diurnal cycle? This is something I have encountered in other datasets. Just briefly state the answer somewhere here.

Since we did not aim to investigate the diurnal change on background fluorescence, we do not have enough background data to answer this question. We operated the instrument in forced trigger mode for only 5 minutes from time to time. However, we observed higher variability on F3 channel compared to F1 and F2 channels. This observation will be added to the revised manuscript.

P17619, L3: "our definition" – I presume you mean the one on p17613, L 22. This was confusing. It appears that you are just defining the acronym "FBAP" rather than a definition of category of WIBS measurements. You also clearly state that it's the definition of Gabey et al. rather than "yours". You do not refer to Table 1 anywhere here.

We used exactly the same definition given by Gabey et al. (2010), namely the combination of F1 and F3 channels for defining FBAP. However, we thank to the reviewer for his/her comment on this point. We think that the explanation given in this paragraph should be rewritten. We therefore have revised the entire paragraph starting at P17619 L2 in the revised manuscript.

I found the particle category "FBAP" as defined in this table to be very confusing. You talk about N_FBAP of dust or ammonium sulphate measurements which are obviously not FBAP. I think it would be much clearer to give this a more generic name, say N_F1,F3 or something, give its full definition at the beginning of Section 3.1, and then, at the end of the laboratory characterization section, say that this is a good definition for detecting FBAP. I don't mind you referring to particles which meet this definition as FBAP in the ambient data set, because I think you are right. However, it is very confusing during the laboratory discussion. I also have a problem with the statement that this definition only includes viable species. I don't think that at this point you have explained why it may not include non-viable species (but then you also haven't defined it very explicitly, so maybe that's why), but presumably it's because the definition relies on both FL1 and FL3. I don't know of any evidence in the literature that the WIBS can successfully distinguish between viable and non-viable PBAP. Either you

have to reference another study that I don't know about, make it clear that you have proven this to be the case, or make it clear that this is speculative. I suspect you need to tone down the language to state something like "The discrepancy may be explained if our definition of $N_{FL1,FL3}$ is insensitive to unviable fungal spores. This may be the case given the dependence of FL3 on NADH, which degrades in non-viable PBAP." Also, do you have any evidence or a reference that you might expect 80% of your fungal spores to be non-viable?

In the case of laboratory tests, we agree with the referee that the particle category "FBAP" is a bit confusing. As suggested, we will use the acronym $N_{F1,F3}$ for the laboratory characterization to prevent any misunderstanding for the reader. Since there is not any published study investigating the capability of WIBS-4 to differentiate viable and unviable aerosol, we can only speculate that, the FBAP definition used in this study should be insensitive to non-viable organisms due to the fact that NADH is the metabolic product of microorganisms and degrades in non-viable PBAP.

Figure 3a clearly shows that we are able to detect almost all fungal spores by using the F1 as indicator. The N_{F1}/N_T value is calculated to about 35% which corresponds to ~90% of the added fungal spores. Initial and final total aerosol number concentrations will be included in Figs. 3a and b to make this statement clearer for the reader. Furthermore, if we look at Fig. 3b, we see that in case of $N_{F1,F3}$ this ratio drops to ~ 20% (50% of added fungal spores) which is likely due to the fact that in comparison to channel F1, channel F3 has a lower sensitivity. It is also possible that some fungal spores may exhibit weaker NADH-like signal and cannot exceed our notably high background threshold. This is still an open question, whether our Xe lamps provide enough energy to penetrate the outer shell of the fungal spores and reach the biofluorophores tryptophan and NADH with 100% efficiency. Therefore, we suggest doing more research on individual microorganisms to answer these open questions in the future.

P17620, L14: You state here that less than 6 particles per litre is a small concentration, but, while it is relatively small compared to the number of dust particles, it is significant compared to ambient concentrations of some PBAP. This is also (to a lesser extent) the case with the soot particles. This needs to be stated and discussed.

If we assume that Saharan dust aerosol exhibits strong tryptophan-like and NADH-like peaks (like the sample used in our study), WIBS-4 may misclassify a significant amount of non-biological aerosol as biological. However, new laboratory studies show that other mineral dust aerosol samples only weakly fluoresce in both detection ranges, F1 and F3, and their fluorescence intensity is considerably weaker compared to the biofluorophores (Pöhlker, 2012, Toprak et al., unpublished study). Thus, the surprisingly high fluorescence measured in the Saharan dust sample may be explained by the fact that this sample was collected from surface and may have been contaminated during collection or transportation.

Technical Comments:

All the technical comments were taken into account and will be corrected in the revised manuscript.

P 17608, I 13: “excitation wavelengths/detection wavebands”

Corrected

L21: “conditions prevail.”

Corrected

L23: “was always higher” – is this really the case? “Always” is a strong word for a seasonal dataset. Would “usually” or “almost always” be more precise?

Here, we will prefer “almost always” since we discuss the general behavior of total aerosol number concentration and underline our statement in Sect. 3 where we discuss the seasonal behavior of the ambient aerosol in detail.

P 17609, L4: “solid particles” – I think this is unnecessarily precise. Is something like a virus solid or is it a liquid? In reality it’s probably a bit of both, so I think just “particles” would be better.

Term “solid” will be removed.

L11: “they affect public health”

Corrected

L12: I can’t find the reference to Pöschl 2005

The reference to Pöschl 2005 will be added to the revised manuscript.

L14: “aerosol –cloud processes” i.e. remove the spaces, otherwise it’s a dash and it means something different.

Spaces will be removed.

L21: “EDX” is not defined

“EDX” will be defined as “Energy-Dispersive X-ray spectroscopy and added to the revised manuscript.

L25: “only to a specific size range” Can you briefly state that. It sounds a bit cryptic as it is.

We agree with the reviewer and revise the last paragraph (P 17609, L15-25) and include any size information for corresponding studies.

P17610, L7: "bacterial concentrations of less than the limit of detection, up to 1.4 ..."

Corrected

P17611, L6: "The release of wet actively discharged"

Corrected

L9: "The online methods, which have been developed..."

Corrected

L10: "from the ambient atmosphere possible"

Corrected

L12: "instrument able to detect"

Corrected

L15: "particles with an aerodynamic"

Corrected

L16: Could you state approximately what time of day the peak was? I presume from this that it was around midday?

P17611, L16 will be revised as follows: ... size distribution had shown a strong diurnal cycle (at around 08:00 a.m., local time) with a dominant peak ...

L24: "In Borneo, they found the minimum and maximum..."

Corrected

P 17612, L8: "studies cover only relatively short periods" – monitoring for a couple of weeks with the WIBS is shorter than your study, but it is longer than most PBAP studies, which use filters.

The most important point we wanted to underline was that these studies do not include any information about the seasonal behavior of the PBAP. We will make this statement also clearer in the revised manuscript and remove the statement "relatively short periods".

P 17613, L4/5: This assertion either needs a reference, or it needs to be qualified by adding "We will show that the combination: ..." or something similar. L23 and elsewhere: The WIBS does not record "shape". I realize that you do not want to use "AF" here as you go on to define it after, but perhaps "sphericity" is a more precise term.

First of all, we agree with the referee and use the “sphericity” term instead of “shape”. A reference will be introduced to L4/5 (Healy et al. 2012) and will be rewritten as follows: “We will show that the combination of measured fluorescence signal ...”

P17614: The AF has been described in a few papers previously. I would be perfectly happy for you reference those papers, and leave most of the first half of this paragraph out, including the equation.

As suggested by the referee, we will shorten this section accordingly.

L24: “manufacturer’s instrument calibration was checked periodically”

Corrected

P17615 L6: “always records a finite”

Corrected

P17616, L7: “aerosol that consists of substances that are able to fluoresce, despite being non-biological in nature, which are a well-known interference to UV-LIF detection.”

Corrected

L14: I think “false triggers” it’s a bad choice of words as this normally refers to the baseline measurements. I presume you are referring forward to your results section in these two sentences here. This is unclear – it sounds like you are talking about the literature. You say “it was shown” but you haven’t shown it yet! I also don’t know what the sentence beginning “It was shown that these ...” is meant to say – it talks about false triggers (false positives) at the same time as saturation. Its also ambiguous what “these” refers to. – FBAP, non-FBAP, both? I don’t know what the first sentence in the paragraph is for. I would suggest rewriting that paragraph something like this (assuming this is what you are trying to say). “We present a laboratory analysis of several representative biological and non-biological particles. We will show that non-biological particles produce few false positive FBAP or saturated measurements, even under high concentrations. On the other hand, the biological particles used never saturate the fluorescence detectors. Hence, we propose a new threshold analysis which excludes particles which saturate a fluorescence channel, as well as those falling below the fluorescence baseline (Eq. 2)” The final two sentences really belong somewhere in the conclusions.

This section will be rewritten as suggested by the referee.

P17617, L2: The csv files do not contain the missed particle counts – that is calculated by the analysis code.

This explanation will be added to the revised manuscript.

P17618, L19: Could you give details of the manufacturers of the spores, and how they were nebulised.

The fungal spores (*penicillium notatum*) were provided from the company “allergon” (Allergon AB, Sweden). They included their mycelium which is the vegetative part of a fungus, consisting of a mass of branching and thread like hyphae. We used small-scale powder disperser (SSPD 3433, TSI) to disperse the fungal spores into the NAUA chamber. This information will be added to the revised manuscript.

L23: state the size range of the CPC

We used the CPC 3022 (TSI), which has a maximum detectable size of around 3 μm . The minimum particle sizes for CPC 3022 can be given as 0.007 μm (with 50% counting efficiency) and 0.015 μm (with 90% counting efficiency). This information will be added to Section 3.1.

P17619, L18: “were performed”

Corrected

L19: Give details of dust source/manufacturer/nebulisation technique.

We added the following sentences to the manuscript: “The Saharan dust (SD2) is a surface sample, which was collected about 50km north of Cairo city, Egypt. Elemental composition of the dust sample and some other details can be found in Möhler et al. 2006. We used SSPD to disperse the Saharan dust into the NAUA chamber.”

L4/5: This makes very little sense and I don't know what it means. Fluorescence itself doesn't contribute to the number of particles, as such, and you haven't given your “definitions of biological particles” yet. Please rewrite it.

L4/5 will be rewritten as follows “Fig. 3a,b is represented side by side to compare the N_{F1} with $N_{F1,F3}$. By comparing panels (a) and (b) of Fig. 3, it can be seen that the combination of F1 and F3 channels (ref. Table 1) results in a much better discrimination of the biological particles.”

P177620, L5: “were found”

Corrected

L21: Again, “false triggers” is a poor choice of words – “false positives” is better, here, and elsewhere. Also, “or it could be”

Corrected

P17622, L18: This is a good discussion of the potential for viable/non-viable detection.

Thanks

P17619, L3 needs to be much more like this.

This sentence will be rewritten in the revised manuscript.

P17622, L25: "admit" implies that you would rather that this result was not the case, which isn't the right tone for dispassionate scientific observers. "we show that there was a" is better.

L25 will be rewritten in the revised manuscript as suggested.

L26/27: It could be ... or it could not be. Please add the alternative explanation that it's just a limitation of the technique.

L26/27 will be changed to: "This could be either a first indication for biological species residing on the surfaces of the dust particles or a limitation of the technique, which may be overcome by altering the fluorescence threshold."

P17623, L1: "to CAST fossil fuel"

Corrected

P17623: Please add information on the length of all these inlets, as well as any information you have about transmission efficiency. Line losses is a very big issue for coarse mode aerosol.

We will add the following sentences to P17623/L24: "The overall length of the sampling line is about 3m. Note that the sampling system is strictly vertical, i.e. there are no horizontal sampling sections which avoids sedimentation losses."

P17624, L2: "global solar radiation", I presume you mean "terrestrial" rather than "global".

Global solar radiation is the scientific term for the sum of the direct and diffuse amount of solar radiation that falls on a horizontal surface.

L4/5: You contradict yourself by saying the instrument sampled continuously, but that it stopped sometimes.

To be honest, due to the instrumental or technical problems, or to check the flow we had to stop sampling not longer than 10-15 minutes. However, because of some technical problems and malfunctions, this period sometimes lasted up to several days until the problem was observed and fixed. Except those days, the instrument sampled continuously. We think that during a one-year sampling this is an acceptable situation.

L20: "in good agreement". Also, here and elsewhere, all the seasons should have a preposition before i.e. "In Spring", "During Summer" etc.

We agree and will change accordingly.

Fig. 9: Can you comment on the spikes at/before about 16:00 during spring?

They are most probably due to some reconstruction events, which took place around the sampling site in spring term. This information will be added to the caption of Fig. 9.

L21: "In addition, the decrease in the ..."

Corrected

Table 2: I would like some information on the distribution of the data in this table. I have previously found that displaying data as mean(median) _{25th} percentile ^{75th} percentile as in 25(24)₂⁵³ is a good way to present a lot of data in a clear way (where [^] denotes a superscript and _{_} denotes a subscript.)

Although, the Fig. 8 represents the 25th and the 75th percentiles of the entire measurement period, Table 2 will be also extended to show the 25th and 75th percentiles for each season.

P17626, L3: Not quite the right use of "analogy". Try "change of FBAP which was similar to that of the spring term."

Corrected

Figure 11: Is the image plot at the bottom all particles, or just FBAP? Please add this to the caption.

The image plot at the bottom shows just FBAP. This will be added to the caption.

P17627, L10: "quite eye-catching" is not formal enough. Try "notable" instead. Also I think at the end it should read, "(indicated by distinct diurnal trends of the terrestrial solar radiation)" assuming I have understood correctly.

L10 will be corrected in suggested way.

L13: "obviously" is a dangerous word in science. I would use "apparently" instead.

Corrected

Figure 17: What is the hatched area indicative of? Add this to the caption.

The hatched area represents the particles smaller than 1 μm . The reader should keep in mind that those particles may refer to non-biological fluorescent aerosol. This information will also be added to the caption.

P17629, L18: "counting statistics were better", like I say, "obviously" is almost never the right word in a scientific paper.

Corrected

P17630, L7: Keep the WIBS-4 abbreviation the same as it was earlier in the paper.

We will use WIBS-4 abbreviation here.

L16/17/18: This sentence doesn't make sense.

We will rewrite that sentence as follows: "Furthermore, it is also possible to distinguish biological aerosol from mineral dust, which is an important component of the atmospheric aerosol and affects atmospheric processes in several ways."

P17631, L10: "For most of the time ..."

Corrected

L16: This assertion needs a reference. I appreciate you discuss this at the beginning of the paper, but it would be useful to the reader to re-reference them here, as this statement is somewhat left hanging.

A reference will be added here. (Elbert et al., 2007)

L24: "how the fluorescence of biological aerosols changes under different ..."

The sentence will be corrected as suggested.

References:

Gabey, A. M.: Laboratory and field characterisation of fluorescent and primary biological aerosol particles, Ph.D., University of Manchester, 2011a. 6390, 6401, 6402

Healy, D. A., O'Connor, D. J., Burke, A. M., and Sodeau, J. R.: A laboratory assessment of the waveband integrated bioaerosol sensor (WIBS-4) using individual samples of pollen and fungal spore material, *Atmos. Environ.*, 60, 534–543, doi:10.1016/j.atmosenv.2012.06.052, 2012.

Pöschl, U.: Atmospheric aerosols: Composition, transformation, climate and health effects, *Angew. Chem. Int. Edit.*, 44, 7520–7540, doi:10.1002/anie.200501122, 2005.