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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Fluorescent biological aerosol particle concentrations and size distributions measured with an ultraviolet aerodynamic particle sizer (UV-APS) in Central Europe" *by* J. A. Huffman et al.

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Reviewer 1 Comments:
RC 1.0
Huffman et al. utilize an ultraviolet aerodynamic particle sizer (UVAPS) to measure the size and concentration of fluorescent particles for a long duration at a site in Germany. This is a well-written and very timely paper. The topic of biological particles, which are a sub-set of fluorescent particles, is of interest in many parts of the atmospheric science community. There currently exist insufficient data, especially over an extended

time period, and this manuscript helps fill this gap. I have a few points that I hope can be discussed and which the authors should consider in a revised manuscript.
[Author Response, AR 1.0] The authors thank the reviewer for his/her positive and constructive comments stating that this "very timely paper" will be "of interest in many parts of the atmospheric science community." The comments in this review have been helpful in helping make the revised version much stronger.

[RC 1.1] The paper indicates that the UVAPS data provides a "lower limit for the actual abundance of PBAPs" (Primary Biological Aerosol Particles), e.g. Abstract and Methods. The authors give a solid description which includes the fact that some non-PBAPs can also fluoresce. Indeed, they attempt to remove some of these non-PBAPs to arrive at final concentrations and timelines. However, this discussion invalidates the claim that the UVAPS can be considered a lower limit for PBAPs. The authors cannot claim to know the fluorescence properties of all atmospheric aerosol; therefore there is no way to definitively state that everything they measure is a PBAP (that is to say, they are removing certain groups of particles they know fluoresce but are not PBAPs; there is no way to say if they removed all groups). Indeed, without some other verification method such a statement is unfounded. Therefore, I suggest the "lower limit" statements be removed; simply state that this is a "best estimate".
[AR 1.1.a] We agree that the potential interferences addressed in our discussion paper and in the referee's comment limit the reliability of the measurement techniques. Indeed, we had not intended to claim that every fluorescent particle detected by the UVAPS would definitively be a PBAP, and we will try to clarify this in the revised version as specified below. As detailed in the manuscript, however, all evidence available from our study and related earlier studies suggests that substantial interferences with non-fluorescent particles that exhibit fluorescence are likely for submicron particles, but not for coarse particles. Following up on the referee's comment we intend to clarify that FBAP provide an estimate for viable bioparticles and can be regarded as an *"approximate"* lower limit for PBAP. It would be misleading to designate the FBAP

concentration as a best estimate of the PBAP concentration, because a wide range of PBAP are not viable (e.g. dead cells, plant debris, etc.) and exhibit little or no auto-fluorescence (see references). To clarify these and related aspects, we intend to revise the manuscript as follows. *Italics used for all inserted text, and all page (P) and line (L) numbers refer to ACP Discussions manuscript.*

P 17706, L 8-9: We originally discuss FBAPs with the following abstract text: "which can be regarded as viable bioaerosol particles representing a lower limit for the actual abundance of PBAPs." This sentence will be changed to "which *provide an estimate of viable bioaerosol particles and* can be regarded as an *approximate* lower limit for the actual abundance of PBAPs."

P 17709, L 12-21: "Further investigation will be required to achieve full understanding of the response of the UV-APS to different types of biogenic aerosol particles and to quantify potential interferences with non-biogenic particles and particle components (e.g. soot and polycyclic aromatic compounds, PAHs) that *may* also display fluorescence. As discussed below, such interferences may indeed influence the measurement results obtained for fine aerosol particles (< 1 μm). *In the coarse fraction, certain components of mineral dust (e.g. kaolin) may exhibit weak fluorescence, but at significantly lower levels than most types of biological particles (Sivaprakasam et al., 2004). In experiments with test aerosols in an outdoor environment Ho et al.(1999; 2002)* found that fluorescence signals from *supermicron* aerosols were dominated by viable PBAPs and found no indications that *non-biological* materials contributed to aerosol fluorescence at the wavelengths used. *Indeed,* all available information suggests that coarse fluorescent particles (> 1 μm) measured by the UV-APS can be regarded as "fluorescent biological aerosol particles" (FBAP), *which provide an estimate of viable bioaerosol particles and whose* abundance represents *an approximate* lower limit for the actual abundance of primary biological aerosol

particles."

P 17715, L 17: "*Comparison of UV-APS measurements with filters analyzed with SEM-EDX from Amazonia show similar trends in PBAP size distribution as well as in number and mass concentration, further suggesting that, at least under certain operating conditions, the UV-APS FBAP measurement has no significant bias with respect to PBAP (Pöschl, Sinha, Martin, et al., 2010)*".

Further, and as we will discuss in the revised manuscript, we understand that this technique is not without uncertainty, but that the detection of FBAP by laser-induced fluorescence techniques enables significant advances in the scientific investigation of PBAP (See insert for P 17729, L 13). We feel this "*exploratory*" study (wording added P 17706, L 27 P 17727, L 21) is worthwhile for publication in ACP, because, as the reviewer states, it "helps fill [the] gap" in what is currently "insufficient data."

At the end of the revised manuscript we intend to add the following statement (insert at P 17729 L 24): "*Additional future work will also be to compare UV-APS measurements with other real-time FBAP detectors, culturability and DNA-based techniques, as well as fluorescence and electron microscopy analysis of co-collected samples. We propose and intend to pursue such studies to advance the development of FBAP measurement techniques and the knowledge of the abundance and properties of PBAP in the atmosphere.*"

[RC 1.2]The term "diel" is used extensively throughout the text and figures. This is a rather uncommon term (not incorrect, especially in biological situations, but then not normally used in this manner in atmospheric). I would suggest "daily" be used instead. ***[AR 1.2]*** We understand that the term 'diel' is not used extensively within the atmospheric community. As the reviewer has pointed out, however, the term is also not

used incorrectly in this context. We considered the use of the term 'diurnal' instead, but this more clearly reflects changes in day-versus-night, or on a once-per-day cycle, and the term would therefore be incorrect in this context. For these reasons we propose to retain use of the term 'diel' within the revised version. The suggestion to clarify usage of this term is very useful, however. As a result, we propose to change several instances of the word 'diel' to 'daily' and the first times (P 17706 L 21 P 17717, L 6) 'diel' is used to define it as "diel *(24 hours)*."

[RC 1.3]The results and discussion and the figures could be shortened substantially. Specifically, the description of number and mass concentrations is several pages each. The specific figures are then called out with more description. I suggest cutting this substantially. In particular, figures could be combined in multiple panels for number and mass (and size and mass distributions) instead of separating these. Please attempt to eliminate redundancy in the description.
[AR 1.3] The comment that the results and discussion "could be shortened substantially" was particularly constructive. The discussion of particle number and mass concentrations (Sections 3.1), in particular will be shortened significantly in the revised version (with most removed text moved to supplementary online material, SOM). After the addition of some minor explanatory comments, the total word count will be reduced by 32-percent from 2185 words to 1479 (509 and 197 words reduced in sections 3.1.1 and 3.1.2, respectively). Figures 1 and 4 will be moved to the SOM, in an effort to "eliminate redundancy." We feel that these changes will substantially improve the readability of the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 17705, 2009.