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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of free amino acids, bacteria and fungi in size-segregated atmospheric aerosols in boreal forest: seasonal patterns, abundances and size distributions" *by* Aku Helin et al.

Aku Helin et al.

evgeny.parshintsev@helsinki.fi

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We appreciate a lot the work done by the reviewers and wish to express our gratitude to their critical comments. The manuscript definitely benefits from the suggested corrections. Below, we give the detailed answers to the reviewer's suggestions, while text is corrected with track changes in the revised manuscript.

Referee 1

(1) Bioaerosols are emitted directly from the biosphere into the atmosphere (Després et

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al., 2012;Fröhlich-Nowoisky et al., 2016)". Although this statement is not conceptually wrong, one of the cited articles, i.e. Després et al., 2012 (Després, V.R., Alex Huffman, J., Burrows, S.M., Hoose, C., Safatov, A.S., Buryak, G., Fröhlich-Nowoisky, J., Elbert, W., Andreae, M.O., Pöschl, U., Jaenicke, R., 2012. Pri- mary biological aerosol particles in the atmosphere: a review. Tellus B 64, 1–. doi:10.3402/tellusb.v64i0.15598), recommends the use of the term " primary biological aerosol particles (PBAP)" instead of ". Therefore, I suggest the authors to check the definition presented Després et al., 2012 and consider its revision throughout the article.

(2) Both terms are used in the literature, but we agree with the Referee, that PBAP suits better here.

(3) Terms "Bioaerosols" was corrected to "Primary biological aerosol particles" and abbreviated as PBAP throughout the article.

(1) Throughout the manuscript, the authors mention several times that the sampling period was one year. However, in section 2.2, the sampling period was defined from February to October. Although cover all seasons, there are 3 months missing and the sampling period is in fact only 9 months. This information should be corrected in the manuscript.

(2) We agree.

(3) Corrected throughout the article.

(1) Line 203. The bacterial cells and fungal spores have the exact same concentration levels, or there is some mistake in this sentence?

(2) We reported a wide range of typical concentration levels for both fungal spores and bacterial cells based on the references cited in the text. Therefore, the values presented seem to be identical or in the same order of magnitude, and there is no mistake in the reported concentration ranges. However, we acknowledge that in reality, the concentration levels are seldom similar as mentioned in Line 202. The concentration

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levels vary depending on the geographical location, meteorological factors etc. Nevertheless, in our opinion, adding additional comments on varying microorganisms' levels is not relevant at this point.

(3) No changes were made to the manuscript.

(1) Line 339. The presence of particles enriched with FAAs from the sea bubble-bursting phenomena?

(2) In principle, it might be possible that there is some enrichment due to bubble-bursting phenomena, although this is not likely the case here. This was not the only sampling period during which the air-masses were passing the Baltic Sea region (e.g. during non-frozen sea periods, Line 391). When taking into account the presented evidence that supports the contribution of pollen, e.g. pragmatically considering that the filters were visibly yellow, it is not convenient to speculate that the vast peak in concentration levels would be caused by bubble-bursting phenomena.

(3) No changes were made or comments added to the manuscript.

(1) Line 342. Barbaro et al., (Barbaro, E., Zangrando, R., Vecchiato, M., Piazza, R., Cairns, W. R. L., Capodaglio, G., Barbante, C. and Gambaro, A.: Free amino acids in Antarctic aerosol: potential markers for the evolution and fate of marine aerosol, Atmos. Chem. Phys., 15(10), 5457–, doi:10.5194/acp-15-5457-2015, 2015) introduces an argument contradictory to that presented in this manuscript. According to Barbaro et al., (2015), the enrichment of aerosol samples in hydrophobic FAAs (e.g., methionine, cysteine and tryptophan) supports the assumption that long-range transport processes, as the different chemical and photochemical events that occur during long-range transport were faster for hydrophilic than for hydrophobic amino acids. Can authors comment on this contradiction?

(2) As far as we could interpret, there is no such contradiction present between our results and the ones presented by Barbaro et al. In their "hydropathy" index classifi-

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cation, only Met is included in hydrophobic amino acids, whereas Trp and Cys are not accounted for. Nowhere in their article is a statement that the presence of Met, Cys and Trp would be indicative of long-range transported aerosols. Oppositely, in another article by the same authors (Scalabrin et al., ACP 2012, doi: 10.5194/acp-12-10453-2012), they emphasize that Met is not typically observed in long-range transported aerosols. We considered that there is enough references in the current version of the manuscript to support our observations.

(3) No changes were made or comments added to the manuscript.

(1) In line 423, the authors state that is relatively strong evidence that cloud-active particles larger than 1 μm are biological in origin". In my opinion, to be a "strong evidence" needs to be better justified.

(2) We agree that evidence might not be strong.

(3) "Strong" changed to "some".

(1) In conclusion, after reading the Supporting Information, I believe that I understood the estimations made by the authors to reach the percentage of PM that should be of biological origin. However, since the two conversion factors used are subject to high uncertainties, in my opinion, these estimation uncertainties should be emphasized in the text of the manuscript, to avoid misleading the reader.

(2) We agree with the referee. However, detailed explanation on estimation uncertainty does not suit the "conclusion" part. Thus, we emphasized the SI and added "high uncertainty" to the text.

(3) The following sentences were modified: "Elevated PBAP abundances occur during the pollen season (Manninen et al., 2014), and based on our estimation even up to $\sim$77% of total PM may be of biological origin (see SI for details). Even though our estimation is highly uncertain, the magnitude of biological cloud-active particles during this period may be atmospherically relevant (Diehl et al., 2002;Diehl et al., 2001;Pummer

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et al., 2012;Pummer et al., 2015)."

(1) In Supporting Information, section "Validation experiments and quality control "the authors state that of the amino acid calibration curves were forced through origin". Could you please justify this choice?

(2) The relevance of forcing through the zero was not dramatic on the results due to intercepts being close to zero anyway. However, in practice it was noticed that forcing through zero gave more realistic LOQ value, i.e. higher values than compared to not forcing through zero.

(3) No changes were made to the manuscript

Referee 2

(1) As already pointed out by referee #1, the use of the term primary biological aerosol particles (PBAPs) defined in detail in one of the references (Despres et al., 2012) may be considered instead of bioaerosols. At the end of page S21 in the supplementary information it is already in use without prior definition.

(2) We agree.

(3) Corrected throughout the article.

(1) line97word9:weighted → weighed

(2) We agree.

(3) Corrected.

(1) line121end:primers pairs → primer pairs

(2) We agree.

(3) Corrected.

(1) line586:Ozler → Özler

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(2) We agree.

(3) Corrected.

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