

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.

Anonymous Referee #1

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General comments: In this work, the authors study the concentration and particle size distribution variation of twenty free amino acids (FAAs), DNA concentration and the DNA concentration of specific microorganism (bacteria, *Pseudomonas* and fungi) in size-segregated aerosol samples collected in a boreal forest (Hyytiälä, Finland) between February and October to understand their potential source. For this purpose, two sets of aerosol samples were collected in consecutive days: the first set of aerosol samples were analysed for DNA and microorganisms-DNA and the second set for FAAs. The FAAs were determined by liquid chromatography-tandem mass spectrom-

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etry, the DNA was extracted with a commercial DNA extraction kit and the concentration and purity was measured spectrophotometrically, and specific bacterial and fungal DNA were quantified with quantitative polymerase chain reaction (qPCR). In order to study their atmospheric implication, the authors also assessed the statistical strength of the linear correlations between FAA and microorganisms concentrations with meteorological parameters (e.g., air temperature, soil surface temperature, soil surface water content, wind direction and speed. Permutational multivariate analysis of variance (PERMANOVA) was also performed to evaluate the changes in the concentration of different FAAs according the season and the aerosol filter size. In my opinion this article is relatively well written and it is well within the scope of the Atmospheric Chemistry and Physics journal. The analytical techniques used and the analytes studied (FAAs and DNA concentrations), are not often associated, which allows for a deeper understanding of the concentration of primary biological particles in atmospheric aerosols. Furthermore, from an environmental point of view it is extremely relevant to know in detail the contribution of these biological particles to the atmospheric aerosol pool to identify their potential sources and the biosphere-atmosphere interactions. Moreover, I would also like to highlight the quality of the Supporting Information. The supporting information contains a very exhaustive set of data with details both on materials and reagents as well as validation procedures and results associated with the determination of amino acids. The results are well within the expected for a fit for purpose method and namely the recovery experiments and the assessment of expanded measurement uncertainty supports the concern of the authors for obtaining high standards for the analytical control quality. Finally, I would like to emphasize that, taking into account the correlation study, one cannot assert that the statistics used, provide a strong support to the conclusions drawn, particularly since these linear correlations normally range between 0.5 and 0.7 (positive and negative) and consequently will be "moderate" correlations, the most. Nevertheless, since the authors often use several references to support their claims, the study turns out to be well substantiated. Considering my opinion above, I believe that this study should be accepted in the Atmospheric Chem-

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istry and Physics journal. Nevertheless, below I present some suggestions and notes I would like to see clarified.

Specific comments: In page 2 line 42, the authors start the introduction with the statement "Bioaerosols are emitted directly from the biosphere into the atmosphere (Després et 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. Primary biological aerosol particles in the atmosphere: a review. *Tellus B* 64, 1–58. doi:10.3402/tellusb.v64i0.15598), recommends the use of the term " primary biological aerosol particles (PBAP)" instead of "bioaerosol". Therefore, I suggest the authors to check the definition presented Després et al., 2012 and consider its revision throughout the article.

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.

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

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

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), 5457–5469, 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, cys-

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teine 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?

In line 423, the authors state that "there is relatively strong evidence that cloud-active particles larger than $\sim 1 \mu\text{m}$ are biological in origin". In my opinion, to be a "strong evidence" needs to be better justified.

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

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