

Improved identification of primary biological aerosol particles using single particle mass spectrometry

By Zawadowicz et al.

General:

This paper explores the use of phosphate and organic nitrogen markers and their ratios in order to improve the separation of biological and other phosphate-containing aerosols using single-particle mass spectrometry. Overall, this paper is well-written and should be considered for publication after major revisions are made to the manuscript.

Major Comments:

The major concerns that I have are that the methodology used in this manuscript is not given in enough detail, which makes evaluating this method and the error analysis performed impossible. The paper would greatly benefit from the addition of a table with statistics of how many particles were analyzed, how many positive spectra, negative spectra, and what ion peak thresholds were used. There is also no mention of the methods used to determine misclassifications. This manuscript needs to be rewritten to include these critical details.

My other concern is that the title and introduction imply that bioaerosols will be distinguished from dust and fly ash using this method. However, it is stated in several areas of the results and discussion that what is really distinguished are biological or organic phosphorus and inorganic phosphorus. The abstract and title should be updated to reflect what is actually being measured in this paper.

Finally, given the prevalence of mixed biological/dust particles observed in ambient observations, a more detailed discussion of experiments used to characterize these mixtures is needed.

Specific Comments:

Abstract:

1. The authors should mention the prevalence of mixed biological and dust particles.

Introduction

1. The introduction needs to be substantially revised. In its current form, the introduction first details ice nucleation, which was not explored in this work, then discusses other methods used to identify bioaerosols, then provides a very short introduction to single particle methods of bioaerosol detection. The introduction should be more focused on methods used to distinguish bioaerosols and dust, and focus more heavily on single particle methods.
2. The last paragraph of the introduction should be cut.
3. What is the distinction between goals 1 and 2 listed in the introduction? These two goals seem quite similar to me.
4. The first paragraph of section 4.2 and a condensed version of the first paragraph of section 4.3 both belong in your introduction.

Methods

1. More detail is needed here. A table containing the statistics of how many particles were analyzed, how many positive spectra, negative spectra, and what ion peak thresholds were used.
2. How were misidentifications determined?
3. Was the same laser fluence used for all experiments including the ambient work? This could affect ion peak ratios.
4. Can it be confirmed that your experiments with illite and spores did indeed contain internally mixed particles?
5. Was a sensitivity analysis performed to confirm that your algorithm was indeed optimized for distinguishing particle types?
6. Add lines 19-20 on page 10; lines 13-15 on page 11; lines 19-22 on page 11 here. These are details of your methods.

Results

1. Page 10, lines 9-13. It seems that positive ions can also be used to filter by particle type, as was done using other single-particle methods. The author should comment on this.
2. Page 10, lines 19-20, why were only the organic nitrogen and phosphate peaks used to distinguish these classes of aerosols. From your mass spectra, it seems that the addition of other markers could help improve the separation between different classes of aerosols.
3. Page 10, lines 23-27: do you have an explanation for your observed changes in the phosphate ion ratios for inorganic and biological phosphorus?
4. Page 10, line 29: how are misclassifications identified and quantified?
5. Page 12, lines 7-10: it seems that this method also relies on a Boolean type of classification and not just ion peak ratios in order to distinguish aerosol types similar to the ATOFMS methods. The authors should mention that both methods are helpful for distinguishing particle types with similar ion peaks (e.g., fly ash and soil dust in this case).
6. Page 12, lines 13-15: why is the discussion of your experiments with mixed biological and dust particles not mentioned in this section? Clearly your ambient data shows that these particle mixtures are atmospherically relevant.

Discussion

1. Page 14, lines 23-26: would you be better able to distinguish bioaerosols if you applied a similar filter (e.g., if you looked for spectra containing Ca, Na, organic carbon, organic nitrogen, and P then applied your ion peak ratio determinations?)
2. Section 4.3 belongs in the results section and should be discussed in greater detail since this particle type appeared to be the most atmospherically relevant.

Technical Comments

1. Replace "species" with "compounds". Species denotes something biological.
2. Page 9, line 28, change "contamination" to "contaminant".
3. Page 16, line 2, change "and" to "an"