Understanding atmospheric aerosol particles with improved particle identification and quantification by single particle mass spectrometry

By Shen et al.

General:

This paper characterizes a new approach to scaling single particle mass spec data to mass concentrations then compares the mass to the AMS. This work is of interest to the atmospheric chemistry and aerosol mass spectrometry community. This paper should be considered for publication after the following revisions:

Major Comments:

1. The concept of the ODE needs to be clarified in the work. Several studies [Allen et al., 2000; Dall'Osto et al., 2006; Gross et al., 2000] have identified the need to account for the transmission efficiency of the nozzle and skimmers through a scaling factor and the ability of the wavelength of the LDI laser to be absorbed by the laser and induce the ionization and detection of individual chemical compounds through a RSF. Does the ODE account for both physical and chemical factors? This was not clear in the manuscript. In particular, the authors address the issue of converting dm to dva; however, the issue of scaling factors are never explicitly addressed.

2. More detail of the laboratory studies performed needs to be included in the paper. More detail is given in the figure caption for figure 1 then in the methods. For instance, how were ODEs from laboratory particles translated to ODEs for ambient particles? How were ODEs averaged or “mixed” across particle types like the biomass burning-soil particles which presumably behave like both dust and organic particles?

3. Matrix effects need to be more explicitly addressed in this work. For instance, the ODE for compounds like NaCl and ammonium sulfate, which are poorly detected by SPMS, can be increased by the inclusion of a compound or substance that is well-detected by SPMS. How did the authors account for matrix effects and the fact that ambient particles have a larger variety of chemical compounds that may show higher ODEs than their laboratory counterparts?

4. I suggest that the authors re-write section 3.1 to more explicitly include how the ambient particles were “fit” or “assigned” to different laboratory mimics and to show how this affected the ODEs in a figure. This would be more useful then showing the spectra of the particle classes.

Specific Comments:

Abstract:

1. The goal of this work, if I understand correctly, is to scale the SPMS data to mass without the need of a reference instrument. If I understand this correctly, then this goal should be more clearly stated.

Introduction:

1. I suggest that the introduction be reorganized to explicitly address the transmission biases associated with the nozzle and skimmers and the need for a scaling factor, then biases associated with the ionization and detection of individual chemical compounds and the need for a RSF, then how comparison of SPMS data to a reference instrument has been used to overcome these limitations.
Methods:
1. The authors should discuss any biases associated with scaling to just an SMPS, which cannot size the supermicron particles detected by the SPMS.
2. Page 5, lines 38-39, in addition to Shen et al, please cite [Allen et al., 2000; Dall’Osto et al., 2006; Qin et al., 2006]
3. Section 2.2, too much emphasis is placed on the shape corrections. It would be much more useful to know how scaling factors and RSFs were addressed with the ODEs.
4. There is only one sentence on the laboratory studies used to create the ODEs applied to the field data. This data is critically important and, as stated in the abstract, is a unique aspect of this study. The laboratory studies need their own section in the paper with details of what was studied, how it was studied, how the ODEs were experimentally determined, if RH affected the ODEs, and detail of how Figure 1 was generated.
5. I am surprised the ODEs look so similar for the different particles types in Figure 1. Do the ODEs also account for RSFs?

Results:
1. While interesting, the particle types discussed in section 3.1 don’t seem to be the main focus. I suggest reducing the discussion of the particle types and placing most of this text in the SI. Instead, focus on what the ODEs were for ambient particles compared to the laboratory particles since that is the novel aspect of this work.
2. Figure 2 has a lot of text, I suggest reducing to just a few characteristic ion peaks.
3. More citations are needed in section 3.1 on previous work showing these particle types including, but not limited to [Ault et al., 2010; Gard et al., 1998; Gaston et al., 2011; Gaston et al., 2013; Pratt et al., 2009; Pratt and Prather, 2009; Qin et al., 2012; Silva and Prather, 2000]
4. The “sodium salts” appear to be sea salts. Why not call them sea salts (both fresh and aged) instead?
5. Page 8, line 2, missing this citation [Pratt et al., 2009]
6. Page 8, line 18, cite [Pratt and Prather, 2009] for particle coatings that can mask ion peaks.
7. Page 8, line 36, denote the ion peaks for sodium, zinc, copper, etc
8. Section 3.2 first paragraph seems to imply that the ODEs are scaling factors only meaning they only account for size and not RSFs. Is this accurate?
9. Section 3.2, how were the SPMS particle classes compared to the mass concentrations from the AMS? Several ion peaks for the SPMS were listed for comparison with the AMS, but how were the individual compounds scaled to mass? Were just SPMS ion peaks compared to AMS mass concentrations?
10. Page 11, line 26, what is the “compound donor class”?
References:


