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Froyd and coauthors describe an approach to quantification of dust and specific aerosol species (organics, sulfate) using single particle mass spectrometer (SPMS) measurement data combined with concurrent scattering-based particle counting measurement data. The latter are used to provide accurate particle number-size distributions in bins as a starting point for the analysis. The approach involves first using mass spectral information to infer density and shape factor for each measured single particle. These properties are then used to convert the aerodynamic diameter of each particle to its respective volume equivalent diameter. Once all of the single particles are binned into volume equivalent diameter bins, the fractional contributions of particle ‘classes’ in each bin are calculated. As the total particle number in each bin is already known as a function of time from the supporting particle counting measurements, here the SPMS data are only used to produce the breakdown of particle compositions within each size bin. Single particle and ensemble sulfate and organic mass concentrations can then be calculated. Total dust mass concentrations are also estimated. Although applied to aircraft datasets in this case, the approach could be extended to ground-based field studies for other SPMS instruments. The uncertainties associated with the method, including particle counting measurement uncertainty, SPMS counting statistics, the use of binned diameters, and assumptions around particle shape factor and density are carefully considered and laid out in detail. This manuscript represents a roadmap for future SPMS users that aim to use their single particle data in a more quantitative way. Although it is a little long, the content here is certainly useful for future applications of the method. I have only minor comments below.

Author reply: We thank the referee for the detailed review of this work and for their suggestions below.

Apart from the uncertainties listed in the Appendix, one issue is particles that are not efficiently ionized (or ionized at all) by the SPMS, because these classes will be absent in the analysis. Differences in ionization efficiencies for different particle classes, or absent classes, will affect the relative counts and fractional contributions of each class in each bin if it is assumed that all particle classes are detected with equal efficiency. Are there known particle mixing state impacts on relative ionization efficiencies for the PALMS instrument? If so these should be discussed and an estimation of the impact of this phenomenon on the quantification uncertainty would be useful.

Author reply:
The reviewer raises a good point regarding potential chemical biases, and we agree it is worth highlighting further. In the submitted manuscript, this issue was only briefly addressed on p.3 line 10:
  “PALMS and other SPMS instruments with sufficient laser power observe a chemical fingerprint for every type of aerosol particle in the atmosphere, minimizing chemical bias.”

If a certain particle type did not produce detectable ions when hit by the ionization laser, a systematic bias would occur where the SPMS instrument would underestimate the concentration of that particle type. PALMS does not suffer from this systematic bias: The atmospheric particles with the lowest ionization efficiency are sulfuric acid/water particles, and in PALMS they yield lower ion current but are still well above background. Since PALMS was originally designed as a stratospheric sampling instrument, early efforts in the 1990’s established thresholds for laser power and wavelength that could reliably ionize sulfuric acid, as well as all other atmospheric particle types. In conjunction with Reviewer 1’s similar concerns we have now have added a paragraph addressing potential chemical biases to the Methods section:

  “An important consideration when measuring fractional and absolute abundance from single-
particle analysis is the estimation of potential sampling biases due to particle composition or physical properties. In one example, if a certain particle type does not produce detectable ions when hit by the ionization laser, a systematic bias would occur that would under-estimate the concentration of that particle type. Additionally, particle mass spectrometers can exhibit a low bias for non-spherical particles due to diverging particle trajectories (Huffman et al., 2005; Vaden et al., 2011). Many aspects of the PALMS instrument design reduce these potential biases (Murphy, 2007). The very close proximity of detection and ionization beams (center separation ~100 μm) enables PALMS to obtain positive ion mass spectra for >90% of particle triggers. In addition, the particle transit distance between the vacuum entrance and the ionization region is shorter than most other SPMS systems. This high targeting efficiency and compact detection region in PALMS minimizes chemical biases from particles of different shape or density that could have diverse trajectories. The PALMS ionization laser produces a mass spectrum for all known particle types in the atmosphere. Even pure sulfuric acid/water particles, which are notoriously difficult to ionize due to their low absorption properties, generate sufficient ions in PALMS for particle classification. For the data presented here, particles were exposed to >2500 MW cm-2 of laser fluence at 193 nm, which is above the ion formation threshold for relatively pure sulfuric acid (Thomson et al., 1997). For organics and sulfate/organic mixtures, ion production is self-limiting due to space-charge repulsion, and total ion current in PALMS is relatively insensitive to laser power. Lastly, particles in the accumulation mode are nearly or completely ablated by the excimer laser (Murphy, 2007). Consequently, PALMS mass spectra represent the composition of the entire particle so that classification routines and relative measurements of chemical sub-components are not biased to physical inhomogeneities such as coatings, phase separation, or surface-enhanced species. It should be noted that although coarse mode sea salt mineral dust particles do not fully ablate and therefore may give an incomplete picture of the particle composition, the mass spectral signatures are still unique to those particle types, and they are classified accurately. We therefore assume that detection biases due to particle chemical or physical properties yield systematic errors that are small compared to other sources of error (see Appendix A).”

Page 2, line 25: Nitrate, ammonium and potassium have also been previously quantified for particle classes and for single particles using similar approaches for ground based measurements (Healy et al. 2013, 2014). Those applications also share the similarity with this work of taking concurrent particle counting measurements as the representative total number-size distribution rather than using size-dependent detection efficiency curves to work up from SPMS counts. It should be noted that that dataset was less challenging for quantification applications however, because only the submicron distribution was considered and crustal/sea salt contributions were minimal in that case. The assumption of equal detection efficiency for all mixing states was also taken in that work, but the spread in relative sensitivities observed for quantified species for each hour of the measurement period indicated that matrix effects associated with mixing state do impact quantification accuracy, at least for ATOFMS measurements.

Author reply:
Thank you for pointing out the Healy et al., 2013 study. We have added that reference to the metals and sulfate/organic studies in the sentence on p.2 line 25, and also added the following text:
“...and non-refractory material such as ammonium and nitrate (Healy et al., 2013),...”

Page 4, line 8: also 50% for 3.2 μm?
Author reply: Yes, and we have reworded this sentence for clarity.
Page 4, line 33: Are the negative spectra used in any way?
Author reply: The negative spectra are not used in the current work to classify particles. However, negative spectra classes can certainly be defined for quantification by PALMS and other SPMS instruments as necessary. In previous studies (referenced in the manuscript) PALMS has used negative spectra to quantify particle components such as organosulfate and sulfate acidity.

Page 15, line 15: Nitrate can be quantified using the approaches laid out here.
Author reply: Agreed, and we have modified this sentence to specifically mention possible nitrate quantification using negative ion spectra.
Page 16, line 8: “with decreasing altitude”
Author reply: Sentence was clarified as “…concentrations decrease steadily with increasing altitude…”

Page 17, line 19: Fig. 10 not included
Author reply: Sentence was corrected to reference the appropriate figures: “…Figures 3, 4, 5, and A1…”

References