

Interactive comment on “Aerosol composition and variability in the Baltimore–Washington, DC region” by A. J. Beyersdorf et al.

Anonymous Referee #1

Received and published: 31 August 2015

The authors present a nice study summarizing airborne measurements of aerosol properties during DISCOVER-AQ flights around the Baltimore-Washington DC area. They take advantage of the sampling design in this field project which was to repeat identical flight paths multiple times to gather statistics to make meaningful claims about spatial scales of variability and relationships between aerosol loading, composition, optical properties, and meteorology. In addition to reporting useful data for this populated region, the broader conclusion reached is of importance for efforts to relate AOD to PM_{2.5}, and this conclusion is that extinction is mainly driven by total aerosol loadings and other factors play secondary roles such as humidity-driven hydration effects. This does not preclude day-to-day changes where hygroscopicity can become more important than other days, but with knowledge of this parameter on a given day, it is

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feasible to use the data for AOD-PM_{2.5} conversions for a very broad area in the region amounting to ~1400 km².

The topic of this paper is important and relevant to this journal. The paper is written well and the methods, analyses, and interpretations drawn from the data are solid. I thought that the title was fine, but it could even be revised slightly if the authors choose to in order to advertise the overall impact of their work even more since they go beyond just reporting composition data in the paper. I have only minor suggestions below that the authors should address prior to final publication:

Major Comments: While this may be out of the scope of this study and up to the authors if they want to address it, this reviewer is curious to know if anything can be said about the impact of aqueous processing (i.e., cloud droplets, deliquesced aerosol) in influencing aerosol composition in the study region.

Specific Comments: Page 23321, Line 8: Just to confirm, aerosol size distribution is not being mentioned here because ‘aerosol loading’ incorporates the impact of varying size distributions. Is this what the idea is here and throughout the paper?

Page 23323, Line 3: The r^2 value would mean more if authors report at least the sample number or some kind of measure of the statistical significance of the correlation (i.e., what % confidence?).

Section 2: In the discussion of the PILS measurements, a few more details are recommended: (i) is PM_{1.0}, PM_{2.5}, or some other size range being sampled?; (ii) were denuders used for the measurements to avoid positive contamination from VOCs and inorganic acids and bases?; (iii) the list of species in Line 7-8 on Page 23324 does not mention lithium and thus it is uncertain as to how the dilution factor was estimated for the two PILS instruments; (iv) no mention is made about the impact of volatilization in the PILS when discussing the mass closure statistics around Line 19 of Page 23324. The readers should refer to this possibility and reference the detailed results in previous work that showed that on average, slightly more than 10% of the ammonium is lost

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in the PILS with a tip temperature of approximately 100 C:

Sorooshian, A., F. J. Brechtel, Y. L. Ma, R. J. Weber, A. Corless, R. C. Flagan, and J. H. Seinfeld (2006). Modeling and characterization of a particle-into-liquid sampler (PILS), *Aerosol Sci. Tech.*, 40, 396-409.

Page 23324, Lines 15-24: are the sizes sampled by the PILS and UHSAS the same?

Page 23324, Lines 15-24: Another interesting piece of analysis that could shed light on the 18% of mass is a simple charge balance of the PILS species measured. It would be useful to see just how well the closure is between anion and cation species charges.

Page 23325, Line 29: It would be interesting for the authors to refer to Table 6 of the Sorooshian et al. paper noted above since the true ratio may have even been higher due to the impact of volatilization on reducing this ratio in the PILS instrument.

Page 23326, Lines 11-14 and other areas: when the authors refer to 'ammonium sulfate' and 'ammonium nitrate', it is assumed that they have confirmed that the molar ratios of their measurements agree with the expected 2.0 and 1.0 ratios for these species for each individual flight that they refer to in such sentences. Since thermodynamically ammonium has a preference to neutralize sulfate first, the excess ammonium needs to then be compared to nitrate mass to confirm that a 1.0 ratio exists. More discussion about this issue is warranted since this reviewer finds it too simple just to refer to 'ammonium nitrate' and 'ammonium sulfate' without some more discussion of flight-by-flight statistics of these molar ratios.

Page 23333, Line 21: add units of ug/m³ to the aerosol concentration.

Figure 4: Some of the species charges are incorrect. Authors should check these, especially for calcium and magnesium.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 23317, 2015.