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

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We thank the reviewer for their comments and suggestions that have improved the manuscript. Responses to their comments are in bold below.

1) 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.

The title has been changed to ‘The impacts of aerosol loading, composition and water uptake on aerosol extinction variability in the Baltimore-Washington, D.C. region’

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2) 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.

We have included a discussion of the work by Eck et al. (ACP, 2014) who utilized data from DISCOVER-AQ to show large increases in AERONET-measured AOD (on average of 25%) in the vicinity of non-precipitating cumulus clouds in the region. In situ measurements showed increases in aerosol scattering, volume and mass in spirals measured before and after cloud formation. These included a doubling of water-soluble organics and 50% increase in sulfate.

3) 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?

Correct, aerosol loading is used to encapsulate any changes due to variability in aerosol number, size and mass scattering efficiencies.

4) 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?).

Done, the comparison between in situ and remote sensing extinction measurements was based on 668 data points

5) Section 2: In the discussion of the PILS measurements, a few more details are recommended:

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a. (i) is PM1.0, PM2.5, or some other size range being sampled?;

The size range measured by the PILS was largely controlled by the size cut of the inlet (4 micron) and the denuders. The transport efficiency of the PILS integrated onto the P-3B was not fully studied but is believed to give a cut size of somewhere between 1 and 2.5 microns. Measurements by an aerodynamic particle sizer (0.5-20 microns) showed that aerosol in the region was dominated by sub-micron aerosol. A sentence has been added to the text.

b. (ii) were denuders used for the measurements to avoid positive contamination from VOCs and inorganic acids and bases?;

Yes, they were used and a sentence has been added to the text.

c. (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;

I did not mention this previously but LiBr was added to the water supply to measure dilution. A sentence has been added to the text.

d. (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 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.

Done, thanks

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

The UHSAS measured 60nm to 1000 micron and thus not identical to the PILS. However, measurements by an aerodynamic particle sizer (0.5-20 microns) showed that aerosol in the region was dominated by sub-micron aerosol.

7) 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.

Good closure (slope of 0.98 with an R2 of 0.94 for all samples) suggests that any loss mechanisms are equivalent for anions and cations. A sentence relating this has been added.

8) 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.

done

9) 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.

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The molar ratio of ammonium-to-sulfate varied between 1.9 and 2.3 on days with high aerosol loadings suggesting on these days that the sulfate was fully neutralized. On low loading days the ratio is more variable and uncertain. Thus it is not correct to refer to them as ammonium sulfate. Nitrate was a very minor component and excess ammonium and nitrate were not highly correlated except on flight 11 (which had the highest nitrate) when the ratio was 1.15. Because of this uncertainty, wording throughout the text has been shifted to sulfate and nitrate aerosol.

10) Page 23333, Line 21: add units of $\mu\text{g}/\text{m}^3$ to the aerosol concentration.

done

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

done, thanks for catching this error!

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

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