

Interactive comment on “Variation in Global Chemical Composition of PM_{2.5}: Emerging Results from SPARTAN” by Graydon Snider et al.

Anonymous Referee #3

Received and published: 17 March 2016

Review of “Variation in Global Chemical Composition of PM_{2.5}: Emerging Results from SPARTAN” by Snider et al.

This paper summarizes measurements of aerosol composition, estimates of hygroscopicity, and particle bound water at 12 sites across the globe. The results suggest that on average across all sites PM_{2.5} mass constituents were (highest to lowest): residual material (assumed to be organics), ammoniated sulfate, crustal material, equivalent black carbon, particle bound water, ammonium nitrate, sea salt, and trace element oxides. The results represent considerable effort and a significant contribution to understanding particulate matter constituents at urban sites in different environments. The authors have done a commendable job of summarizing and presenting consistent measurements, and for making these data available to the community. The work is scientifically sound and for the most part the methods are well-described. The impor-

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tance of water biases in filter sampling can be significant and a strength of the paper is the estimates of particle bound water and hygroscopicity based on chemical composition and assumptions of molecular speciation and hygroscopic properties. Strengths of the paper include the careful, consistent approach to summarizing data from many different sites, and interpreting the data in the context local and regional sources, including comparisons to other studies when available. The paper could benefit from some reorganization to help with clarity, and careful reading and editing to account for discrepancies between values stated in the text and reported in figures. Some discussion of the values of the average mass scattering efficiencies used for converting light scattering to mass should be included, and whether these were appropriate for varying composition.

Organizational Comments

The introduction is a bit disjointed. As a suggestion, I recommend moving the paragraph that begins on line 102 to line 78 (becomes 2nd paragraph), followed by the paragraph starting on line 93, with the sentence starting as “Furthermore, no global...” on line 80-82 added to the end, so that the RH effects are included in 1 paragraph. The rest of the paragraph (line 83-91) can become the 4th paragraph, followed by the paragraph at line 114, finishing with the paragraph on line 121. The order of the discussion would then be 1) Health (2) Chemical composition (3) Humidity effects (4) Satellites (5) SPARTAN (6) Purpose

A difficulty with the current organization of the paper is that the section on hygroscopicity comes before aerosol composition such that aerosol components (like ASO₄) are being discussed before the reader knows how the authors define them. It would help to follow the development of the method if the description of the assumed aerosol components came first, and the kappa development followed. I recommend switching the order of section 3 (aerosol hygroscopicity) and section (4) PM_{2.5} aerosol composition. Also, define a new section for mass speciation results (4.10 and onward). In accord with this reorganizing, switch the order of Figure 1 and 2, and Tables 1 and 2.

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Specific comments follow:

I recommend instead of “ammonium sulfate”, referring to ASO₄ as “ammoniated sulfate” because the definition doesn’t necessarily assume fully neutralized ammonium sulfate (1.375*SO₄).

Please point out that PM_{2.5} and PM₁₀ are gravimetrically weighed.

Line 19: I’m not sure what “maximize the chemical and physical information” means? It seems like the project is characterizing the chemical and physical attributes of aerosols from filter samples.

Line 24: Define AERONET for first time use.

Line 28: Consider replacing “baseline” with “background” or “rural/remote”.

Line 34: What RH?

Line 41: Define IMPROVE for first time use.

Line 42: From the slope, which network had higher mass?

Line 51: Change “included” to “including”

Line 51: What was the standard deviation?

Line 96: Define CSN

Line 97: Define AIM

Line 99: Define kappa

Line 126: “As a function of chemical speciation” seems redundant.

Line 134: Provide years of sampling

Line 147: Define PM₁₀

Line 157: Is PM_{2.5} here gravimetric mass or summed constituents?

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Line 166: Add “with other networks”

Line 173: This sentence implies that surface reflectance is used to obtain all of the following constituents, not just black carbon.

Line 189: Add “K+” here, assuming that the potassium discussed later is from the IC.

Line 235: Define 1:1 v/v notation

Line 239: Replace the IMPROVE convention with a reference, perhaps Pitchford et al., 2007. (“Revised algorithm for estimating light extinction from IMPROVE particle speciation data”, JAWMA, 57, 1326-1336).

Line 273: Check notation in table 2 and make sure it is the same as in the text for each species.

Line 295: This is a little confusing. I assume based on Table 2 that the authors are saying $0.1 \cdot CM = Al + Fe + Mg$ but it isn’t immediately clear from this sentence.

Line 318: Point out that RM is assumed to be organic matter.

Line 342: Define NO_x first use

Line 357: Coarse Zn:Al ratios are discussed throughout the paper but from the composition section, it seems like only PM_{2.5} composition was analyzed. Was the coarse mode speciated also measured?

Line 395: What was the site average? It would be useful to add a column to table 3 with this information for each site.

Line 404: What is the significance of “Study A” and “Study B”? Why are they referred to in this way?

Line 429: There are several instances when the values in the text are not exactly what are reported in the figures. (PM_{2.5} is 69 in text, 70 in figure)

Line 463: 25% in texts, 24% in figure.

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Line 472: 17 in text, 18 in figure.

Line 483: Recommend discussing the sites in the same order as displayed in the figure.

Line 506: PMc notation has not been used previously.

Line 514: 55% in text 59% in figure. 18% in text, 19 in figure, 7% in text, 7.4% in figure.

Line 554: Does total mass here refer to PM2.5?

Line 556: add “respectively” to these comparisons so the reader knows which is which. The order of the comparison switched for CM (note 11% in text, 10% in figure) and EBC.

Line 570: Does “total aerosol mass” here refer to PM2.5?

Line 578: Again PMc notation used here.

Line 592: And the Butler et al value of 55%.

Line 594: CM 10% in text, 11 in figure, ASO4: 21% in text, 24% in figure; ANO3: 3% in text, 3.6% in figure.

Line 610: Replace BC with EBC. Also, 9% in text, 10% in figure.

Line 629: Does this Zn:Al ratio refer to PM2.5 or PM10?

Line 663: What were the average mass scattering efficiencies applied here, and were they consistent with major mass compositions during the same time periods? There are periods with fairly high biases between the mass estimates. Are the assumptions of ~constant mass and density appropriate during these periods, based on composition data?

Line 678: 0.71 in text, 0.70 in Figure

Tables and Figures

Line 1112: Another reason for switching the order of the hygroscopicity and aerosol

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composition sections would be that the species in Table 1 are not defined until Table 2. Switching the order would help to interpret Table 1.

Line 1112: Are the values of PBW averaged across all sites?

Line 1121: There are some discrepancies with notation of species mass in this table and the text. I recommend using “sea salt” instead of “NaCl” since it is used in the text (line 279). Also, 0.18[Na]ss used in the table but 0.18[Na] used in text (line 287). Define RH, X. Define SSR.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-62, 2016.