

## ***Interactive comment on “Variation in Global Chemical Composition of PM<sub>2.5</sub>: Emerging Results from SPARTAN” by Graydon Snider et al.***

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We thank the referee #3 for these helpful comments. Referee comments are numbered, with our responses, and any changes to the manuscript, subsequently given.

1) 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

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Thank you, we have adjusted the order of paragraphs to reflect your suggestions.

2) A difficulty with the current organization of the paper is that the section on hygroscopicity comes before aerosol composition such that aerosol components (like ASO<sub>4</sub>) 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<sub>2.5</sub> 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.

Thank you, we have arranged sections 3 and 4 as you have suggested, and the corresponding tables and figures. We have also moved lines 158-162 to the beginning of the new Section 5.

Specific comments follow:

3) I recommend instead of “ammonium sulfate”, referring to ASO<sub>4</sub> as “ammoniated sulfate” because the definition doesn’t necessarily assume fully neutralized ammonium sulfate ( $1.375 \cdot \text{SO}_4$ ).

Thank you, we have made the suggested change throughout text.

4) Please point out that PM<sub>2.5</sub> and PM<sub>10</sub> 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.

We have changed ‘maximize’ to ‘characterize’. And have changed a later sentence to (lines 20-23):

“Our methods infer the spatial and temporal variability of PM<sub>2.5</sub> in a cost-effective manner. Gravimetrically-weighed filters represent multi-day averages of fine particulate matter (PM<sub>2.5</sub>), with a collocated nephelometer sampling air continuously.”

5) Line 24: Define AERONET for first time use.

Done, this is now referred to Aerosol Robotic Network

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

We now use your suggestion of using ‘background’.

7) Line 34: What RH?

We have appended “at 35% RH” to the end of sentence

8) Line 41: Define IMPROVE for first time use.

We now refer to IMPROVE as Interagency Monitoring of Protected Visual Environments

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

Our values were higher. Modified sentence to read (lines 42-45)

“Comparison of SPARTAN versus coincident measurements from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network at Mammoth Cave yielded. . .”

10) Line 51: Change “included” to “including”

Done

11) Line 51: What was the standard deviation?

The standard deviation of kappa is 0.04, which has now been included.

12) Line 96: Define CSN

Done. CSN is now defined as Chemical Species Network.

13) Line 97: Define AIM

Done. AIM is now defined as the Aerosol Inorganic Model.

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14) Line 99: Define kappa

We have rephrased lines 102-105 to

“More recently Petters and Kreidenweis (2007, 2008, 2013) have developed -Kohler theory, which assigns individual hygroscopicity parameters to all major components, from insoluble crustal materials to sea-salt.”

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

Rephrased lines 126-129 to

“Section 3 defines categories of aerosol types (crustal and residue material, black carbon, ammonium nitrate, ammoniated sulfate, sea salt, and trace metal oxides) as a function of specific chemical species.”

16) Line 134: Provide years of sampling

Added at line 136-137 “across 13 SPARTAN sites, between 2013 and 2016”

17) Line 147: Define PM10

Rephrased lines 149-150 to

“Air samples first pass through a bug screen and then a greased impactor plate in order to remove particles larger than 10  $\mu\text{m}$  in diameter”.

18) Line 157: Is PM2.5 here gravimetric mass or summed constituents?

Rephrased, and moved to beginning of Section 5 (lines 356-366):

“Gravimetrically-weighed PM2.5 concentrations within the period June 2013 to February 2016 span an order of magnitude, from 9  $\mu\text{g m}^{-3}$  (e.g. Atlanta) to nearly 100  $\mu\text{g m}^{-3}$  (Kanpur)”.

19) Line 166: Add “with other networks”

Changed lines 169-170 to

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“The sites of Atlanta and Mammoth Cave are included for instrument inter-comparison purposes with other networks”.

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

Thank you. We corrected lines 177-178 to read

“These filters are subsequently analyzed for water-soluble ions, trace metals, and surface reflectance to obtain black carbon”.

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

Done.

22) Line 235: Define 1:1 v/v notation

We have amended lines 295-296 to

“A 1:1 volume ratio with water as RH approaches 0% yields  $a = 2$ ”

23) 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).

Thank you, we have now included this reference on line 299.

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

We now define NaCl as ‘Sea Salt’ or ‘SS’ throughout text.

25) 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.

Modified lines 222-223 to

“we generalize that natural CM is approximately  $10 \times [\text{Al} + \text{Fe} + \text{Mg}]$ ”

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

Have rephrased lines 247-248 to

“Residue matter, which is treated as mainly organics, is estimated by subtracting dry inorganic mass (IN) . . .”

27) Line 342: Define NO<sub>x</sub> first use

Modified line 383 to “. . .variation in NH<sub>3</sub> and NO<sub>x</sub> (NO + NO<sub>2</sub>) sources”

28) Line 357: Coarse Zn:Al ratios are discussed throughout the paper but from the composition section, it seems like only PM<sub>2.5</sub> composition was analyzed. Was the coarse mode speciated also measured?

We did measure coarse-mode speciation but focused on fine-mode evaluation for this paper. An upcoming manuscript will contain coarse-mode data.

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

We have now provided correlations between ammonium and nitrate in table 3.

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

Both are now labeled “Prior study”.

31) Line 429: There are several instances when the values in the text are not exactly what are reported in the figures. (PM<sub>2.5</sub> is 69 in text, 70 in figure)

Thank you. We have corrected values to reflect our most recent findings.

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

Fixed, thank you.

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33) Line 472: 17 in text, 18 in figure.

Fixed, thank you.

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

Certain sites did not have comparison figures associated. We aim to order the sites from most to least data available.

35) Line 506: PMc notation has not been used previously.

Have included in Methodology section (lines 150-153):

Aerosols are collected in sequence on a preweighed Nuclepore filter membrane (8  $\mu\text{m}$ , SPI) that removes coarse-mode aerosols with diameters from 2.5 - 10  $\mu\text{m}$  in diameter (PMc), while fine aerosols (PM<sub>2.5</sub>) are then collected on pre-weighed PTFE filters (2  $\mu\text{m}$ , SKC).

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

Discrepancies have been fixed, thank you. Concentrations have also been updated reflecting new data acquired since submissions.

37) Line 554: Does total mass here refer to PM<sub>2.5</sub>?

Yes. Modified line 616-617 to read "...and total mass of PM<sub>2.5</sub> ( $r^2 = 0.76$ , slope = 1.12)."

38) 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.

Added the word 'respectively' to lines 617-619

"Differences between IMPROVE vs. SPARTAN are small for ASO<sub>4</sub> (36% vs. 33%),

ANO3 (2.4% vs. 1.2%), CM (7% vs. 11%), and EBC (3.0% vs. 5.6%), respectively”.

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

Yes. Replaced “total mass” with “total PM2.5 mass”.

40) Line 578: Again PMc notation used here.

PMc is now defined in methodology section, line 151.

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

Modified lines 632-635 to

“SPARTAN component fractions in Atlanta are also consistent with respect to Butler et al. (2003); components CM (12% vs. 10%), ASO4 (23% vs. 28%), ANO3 (3.5% vs 4%) and RM and OM (48% vs 55%) closely match, except for EBC (11% vs. 3%), perhaps reflecting different time periods.”.

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

These discrepancies have been fixed, thank you. Concentrations have also been updated reflecting new data acquired since submissions.

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

Discrepancy has been fixed, thank you.

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

Now makes reference to PM2.5.

45) 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

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composition data?

We tried to clarify at the start of this section that composition-specific  $v$  values are used.

46) Line 678: 0.71 in text, 0.70 in Figure

Fixed, thank you.

Tables and Figures

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

We have changed the order of the tables as suggested.

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

In table 3, PBW is averaged across sites as a percent of total mass. However, it is reported as absolute mass for individual sites.

49) 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,  $0.18[\text{Na}]_{\text{ss}}$  used in the table but  $0.18[\text{Na}]$  used in text (line 287). Define RH, X. Define SSR.

We have now changed NaCl to SS or Sea Salt throughout the text, and added SS subscript. X, SSR, and RH have been defined as well.

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