

Referee comments for “SPARTAN: a global network to evaluate and enhance satellite-based estimates of ground-level particulate matter for global health applications”

We thank the reviewer for their helpful comments. We have revised the manuscript according to their suggestions with point-by-point responses below.

Answer to Reviewer #2 comments

SPARTAN: a global network to evaluate and enhance satellite-based estimates of ground-level particulate matter for global health applications Snider et al. Atmos. Meas. Tech. Discuss., 7, 7569–7611, 2014

This manuscript describes plans for a new global network which is intended to help apply satellite measurements to the effort of estimating PM_{2.5} concentrations. The manuscript briefly discusses site criteria and sampling strategies and then focuses on the instruments that will be deployed as sites come on-line. Finally, they present a few results from the ground-based instruments from a prototype site.

Reply: Thank you for the many helpful suggestions in this review.

1. General and science comments: Maybe I missed it but it would be useful to provide a list of potential satellites this network will be used to enhance. Figure 1 mentions MODIS and MISR in the caption, but I think that is the only place where a specific satellite is named. Tied to that, in Figure 3 and a couple other places in the text there are variables with the subscript ‘sat’. As far as I can tell this subscript refers to a measurement made with a ground-based instrument at the time of a satellite overpass. It might be helpful to distinguish satellite measurements from ground-based measurements at the same time as the satellite by using different subscripts (for example, ‘sat’ and ‘sat*’ or something like that.

Reply: In the introduction we now describe the applicability of SPARTAN. Page 7583, Line 26-28:

“We define AOD_{sat} as the ground-measured AERONET AOD averaged from 10:00 to 14:00 and interpolated to 550 nm via the Ångström exponents” We thereafter refer to ground-based measurements at satellite overpass time with the subscript sat. We do not use satellite observations in this manuscript.

2. GAW and NOAA aerosol network observatories are highly instrumented sites in a wide range of locations and are often co-located with an AERONET sunphotometer. These sites typically make measurements of aerosol light scattering at low RH. It would make sense to attempt to collocate some of the SPARTAN deployments at these sites in order to evaluate your ‘dry scattering’ estimates. These sites have personnel, power and often additional measurements which might enhance evaluation of SPARTAN results. NOAA network sites making dry scattering measurements that are collocated with aeronet sites include: mauna loa, cape san juan, trinidad head, mount waliguan, egbert, appalachian state, mount lulin, gosan, bondville, cart_site and barrow.

Reply: Helpful suggestion. We will explore collocation with these sites. It is also worth noting that presently GAW does not have PM_{2.5} available on a global scale, therefore this size range is limited mostly to sites in Europe and North America (e.g. GAW report No. 178, section 4.6.2). Although Europe and NA are intentionally not focal points of the SPARTAN project, we have done some comparisons of PM_{2.5} with collocated BAM, TEOM instruments in Beijing and Halifax, as well as TEOM and a nephelometer run by IMPROVE at Mammoth Cave.

3. P7575 line 21 - ‘...calibrate nephelometer...’ calibrate seems like a strong word here. Evaluation or constrain is probably better. It’s a calibration if you know the answer, i.e., exactly how the filter based measurements can be transformed into scattering. but if you are going from a bulk measurement of aerosol chemistry from filters at weighing RH to scattering at RH close to ambient RH then you are making assumptions about particles size, refractive index and hygroscopicity.

Reply: We agree ‘constrain’ is a better choice of wording and have changed this line in section 3.1.

4. P7578-7579 – discussion of filter samples. What about organics? what about organic material? Zhang et al., GRL VOL. 34, L13801, 2007 suggest that organics make up a significant fraction (25-75% of

aerosol mass).

Reply: We have preliminary estimates of organics inferred from total and reconstructed fine mass. These results are being prepared for future publication.

5. P7579 line 13 – mention of nephelometer angular range - how do you correct for the angular truncation? if the sampled air includes super micron particles the truncation correction can be very large (on order of 20-30%). (at least that's how big it is for TSI nephelometer with similar angular range - Anderson and Ogren, 1998.) Getting the truncation right is important if you are relating ambient scattering from the neph to ambient scattering from a satellite or somehow derived from AOD both of which wouldn't have truncation issues.

Reply: AirPhoton presently does not correct for 0-7° nor 171-180° truncation. Since we are interested in the sub 2.5 micron size range, in particular in the application of equation 2, suppression of supermicron forward scatter will not significantly affect the final results. An upcoming algorithm in development at AirPhoton will compensate for lost backscatter.

Page 7579, Line 12-13: “The forward and backscattering measurements are made independently. Correction for angular truncation is in development”.

6. P7582 - discussion of nu related to term one - it would be interesting to see whether the first term is always the most strongly related to 'nu' and whether the relationship is that strong at other sites. i would recommend finding aeronet sites that are already collocated with dry nephelometer measurements and seeing how the relationship does. one site that would be easy to do this at is Bondville, IL. there is an aeronet sunphotometer, a TSI nephelometer (operated by NOAA) and improve chemistry samples for PM2.5. That nephelometer operates at low RH so you would need to reverse eqn 1, e.g., $bsp_{wet}=bsp_{dry}*f(RH)$

Reply: Thank you for the suggestion. We have done preliminary analysis the variance of terms 1, 2, and 3 with respect to η in section 4.1 and 4.2. We would like to consider this as future work as we continue to evaluate the major influences of equation 4.

7. P7593 line26 - Do you have plans for checking the smoke stain reflectance values against results from instruments that measure light absorption/BC? having more black carbon-type measurements is something that would be very useful to the climate community, but they would need to be tied to something more familiar (PSAP, MAAP, aethalometer...)

Reply: We have found a strong correlation with the BC measurements made by the COSMOS instrument. Presently we are comparing SSR values with OC/EC results from quartz filters in Harvard Impactors sampled for 21 daily values per location (section A2.5). We will soon also be comparing our SSR results to collocated aethalometers in Kanpur and Buenos Aires.

Page 7594, line 15-18: “...quartz filters are analyzed for elemental carbon via an OC/EC analyser (Sunset Laboratory). The EC mass fraction is used to correct PTFE reflectance via the Smoke Stain Reflectometer instrument”.

Page 7578, line 18-20: “Additional collocated absorption measurements, such as with COSMOS in Beijing (Kondo et al., 2009), are being used for further interpretation.”

Text editing/clarification type comments

1. P7571 – lines 23-24 this statement is vague/unclear w/o having read the paper.

Reply: We have changed line 23-25 in abstract to be clearer to a first-time reader.

“Our initial measurements indicate that the AOD-to-ground-level-PM_{2.5} ratio is driven temporally and spatially by the vertical profile in aerosol scattering”

2. P7571 – line 25 – what is the aerosol scattering vertical profile interacting with? this is unclear.

Reply: We modified this abstract line to “Spatially this ratio is also strongly influenced by the mass scattering efficiency.”

3. P7573 line 20 – ‘inform’ is a weird word choice...how about affect or influence or determine?

Reply: We agree and have changed to “influence”

4. P7573 line 28 – aernet being used to validate satellite data - a reference or two here would be good.

Reply: We have also included Remer et al (2005) on line 22 to establish a firmer connection between AERONET and MODIS.

5. P7574 line 7 – suggest changing ‘...midday aerosol measurements...’ to ‘...midday aerosol optical measurements...’

Reply: We have modified line 29, page 7573 to “SPARTAN collects both midday aerosol optical measurements (needed to compare with satellite overpass times) and the 24-h PM_{2.5} averages relevant for health studies”

6. P7577 line 2 – ‘...to prevent their lower capture efficiency the...’ rephrase: ‘...to enhance their capture efficiency...’

Reply: Line 22-24, page 7576 in section 3.2 has been rephrased to “The manufacturer (SPI) coated capillary pore surfaces with a thin layer of vacuum grease to enhance their capture efficiency”

7. P7577 line 18-20 - it’s unclear to me why this sampling strategy increases retention time - you still are collecting some samples during the warm part of the day. it only seems like it would increase retention time for the 6:20-9:00 sample.

Reply: In section 3.2.1, the rationale is that nighttime aerosols would contain a larger concentration of ANO₃ whereas daytime hours would have less condensed-phase ANO₃. Thus in sampling a warm daytime period first, there would be minimal semivolatile losses since there would be minimal deposited semivolatiles.

Page 7577, Line 13-21: “We choose to start sampling runs for each filter in the morning (9:00) when temperatures are lower, to increase retention of temperature-dependent semi-volatile inorganic and organic material that was collected overnight. We tested the behavior of semi-volatile material (ammonium nitrate) in the cartridge to diurnal heating cycles. Based on our experiments with ammonium nitrate, a moderate loss rate can be expected from the PTFE filters while warm air actively flows over the filters (*c.f.* Appendix A1.2), however loss rates are minimal during periods when there is no active sampling. Thus we design the sampling protocol to actively sample for only one diurnal cycle and to avoid daytime sampling after nighttime PM has been collected”

8. P7577 line 20-25 i would move the staggered sampling timing up to where you discuss avoiding day of week biases and before the discussion of volatilization. Presumably part of the staggered sampling time impetus is to avoid time of day biases.

Reply: Thank you for the suggestion. The order of explanation in section 3.2.1, lines 3-21 is now 9-day sampling, time-of-day sampling, and explanation of semivolatile losses.

9. P7577 line 27 ‘...we reduce the sampling time between 15% and 100%...’ does a 100% reduction mean no sampling?

Reply: Line 23-24 has been rephrased to “...we sample between 15% and 100% of each 2 hour 40 minute period to prevent filter saturation...”

10. P7577 line28++ ‘During the staggered air samplings the collocated nephelometer measures particle light scattering continuously.’ i would rephrase this sentence: ‘Unlike the filter measurements, the collocated nephelometer measures continuously.’

Reply: Thank you for the suggestion. We have chosen your phrasing on line 25, page 7577.

11. P7579 line 20 - ‘The inlet is a 10 cm length of copper tubing ending with a plastic bug screen.’ have you calculated losses in the nephelometer inlet?

Reply: We have not directly calculated losses, but found less than 2% loss of particles in a dichotomous

sampler with a longer path length at faster inlet air speed.

Page 7579, line 16-17 “Inlet wall losses for particles below 2.5 μm are expected to be less than 2% (Liu et al., 2011).”

12. P7580 line 12 - using one number for hygroscopic growth seems fraught with peril!

Also, do you apply constraints or eliminate data for hours when there are large changes in RH?

Reply: We agree. This is simply for initial analysis. Future work will refine the approach together with knowledge of $\text{PM}_{2.5}$ composition. Nevertheless there is evidence many multicomponent sub-saturated ambient aerosols show growth curves such that $\kappa \sim 0.1$ to 0.2 . We automatically remove any hour-long periods for which $\text{RH} > 80\%$. See text in section 3.3 for slight clarification.

13. P7580 line17-18 ‘Hourly nephelometer scatter as measured by the nephelometer, is approximately proportional to $\text{PM}_{2.5}$ mass (Chow et al., 2006),’ does the Chow paper refer to dry or ambient scattering?

Reply: Chow measured ‘dry’ RH via a ‘smart heater consisting of a tube wrapped with heating tape that only applies heat when the RH at the outlet exceeds 72%.’ Since we ignore RH data for $\text{RH} > 80\%$, the humidity range Chow refers to is very close to our own.

14. P7581 line1 ‘mean of dry aerosol scatter’ change to ‘mean of dry aerosol scattering’

Reply: Thank you. We have changed “scatter” to “scattering” in section 3.4.

15. P7581 line 4 ‘prediction accuracy was $1 \mu\text{g m}^{-3} + 17\%$ ’ 17% of what?

Reply: The 17% is the mean relative difference in measurement between our $\text{PM}_{2.5}$ estimate and direct $\text{PM}_{2.5}$ measurements. The revised sentence explicitly includes the fine mass concentration in the equation.

Page 7581 line 5-7: “The resultant prediction accuracy was $1 \mu\text{g m}^{-3} + 17\% \times [\text{PM}_{2.5}]$ at three North American sites and for Beijing (*c.f.* Appendix A1.5). Uncertainties from chemical extractions are listed in section 3.2.2. ”

16. P7581 ongoing evaluation - as mentioned previously - should also compare at sites with dry nephelometer measurements and if possible absorption measurements. Again - gaw and noaa network sites seem like a good partner in this endeavor..

Reply: As we are interested in $\text{PM}_{2.5}$ in areas which GAW does not yet include $\text{PM}_{2.5}$ measurements. We have not found any NOAA stations with dry aerosol $\text{PM}_{2.5}$. As more collocated $\text{PM}_{2.5}/\text{AOD}$ are found, we will consider these as part of future work analyzing equation 4.

17. P7582 line1 - why interpolate to 550 to match satellite AOD measurements? Should explain

Reply: The updated section 4.1 page 7583, line 1-3 now reads

All nephelometer scatter and AERONET AOD measurements are interpolated to 550 nm via the nephelometer Ångström exponents to match the wavelengths typically reported for satellite AOD

18. P7582 line 4 ‘...daily-varying concentrations of nu in...’ i am uncomfortable with calling ‘nu’ a concentration even though the units work out. i recommend changing the word ‘concentration’ to ‘value’

Reply: We agree, and have made the change in section 4.1.

19. P7582 eqn 4 - is the parameter you are calling bsp,sat really the bsp from the nephelometer at the time of the satellite overpass? if that is not the case, then the statement that the second term only requires the nephelometer is incorrect.

Reply: “Sat” means sampling constrained between 10 am and 2 pm. See updated section 4.1.

From Page 7583, Line 26: “We define AOD_{sat} as the ground-measured AERONET AOD averaged from 10:00 to 14:00 and interpolated to 550 nm via the Ångström exponents”

20. P7582 line 15 *'The second term describes: : :'* *this term doesn't describe diurnal variation it just helps account for it.*

Reply: We agree and modified line 20-21 in section 4.1 to use the word 'account' rather than 'describes'.

21. P7582 line 17 – *adjusting to 550 nm - what is the point for doing this? AERONET doesn't measure at 550 nm and neither does your nephelometer, so presumably it's the satellite that is at 550 nm. should state this.*

Reply: Good point. We have updated section 4.1, page 7571 lines 26-28: "We define AOD_{sat} as the ground-measured AERONET AOD averaged from 10:00 to 14:00 and interpolated via the Ångström exponents to the wavelength (550 nm) typically reported for satellite retrievals"

22. P7582 line 19 – *'nonlinear regime' what do you mean by non-linear regime? is this referring to RH>80% or scattering>1300 Mm-1 or both?*

Reply: Thank you. We modified page 7592, lines 4-9, to

"Measurements with $RH > 80\%$ were excluded ... In Beijing the prototype AirPhoton nephelometer signal saturated during extreme low-humidity pollution events ($PM_{2.5} > 400 \mu g/m^3$) such that $b_{sp} > 1300 Mm^{-1}$, and these data were omitted from averages. Light scattering performance returned to normal after these events".

23. P7582 line 20 *'The third term is the inverse of the mass scattering efficiency and related...'* *I would suggest adding in the word loosely (or very loosely?) when talking about how the third term is related to aerosol composition.*

Reply: The sentence has now been modified to

Page 7582, line 22: The third term (T3) is the inverse of the mass scattering efficiency, which is a function of aerosol size and composition.

24. P7582 line 23 *'We find that nu is most: : :'* *change to 'We find that, in Beijing, nu is most...'*

Reply: Section 4.1 has been modified to reflect the new use of statistics and have deleted this sentence.

25. P7583 line1-2 – *' : : weakly related to diurnal variation in atmospheric scattering (second term Eq. (4), : :'* *Anderson et al., 2003 used lag-autocorrelation to investigate mesoscale aerosol variability. they found R values (not R2) of around 0.3 for 24 h lag for several sites so it's not surprising there's not much relationship between bsp,1h and bsp,24h, especially once you add in a diurnal cycle in RH. Anderson et al 'Mesoscale Variations of Tropospheric Aerosols,' J. Atmos. Sci., 60, 119-136, 2003.*

Reply: Thank you. We now include this reference in section 4.2 and A1.1.

26. P7583 line27++ *'nu appears to be closely related to the ratio of ground-level atmospheric scattering to column AOD...'* *you would expect this if as you cited earlier Chow et al., 2006 found scattering proportional to and pm2.5*

Reply: We did expect that $PM_{2.5}$ is proportional to scatter; this was another means by which to confirm this hypothesis.

27. P7584 line27 – *Angstrom exponent should be capitalized and have the appropriate diacritical marks.*

Reply: Thank you. We have updated the typography throughout the manuscript.

28. P7587 – *appendix A - again, check out tad anderson 2003 paper about mesoscale variability. in addition to looking at changes in aerosol properties with time he also looked at changes in aerosol properties as a function of length scales.*

Reply: Thank you again for brining this reference to our attention. We've included its mention line 17 in appendix A1.1

29. P7588 line3-4 - *comparison with yang - presumably your measurements were made at the same time (seasonally) as yang's measurements... or there is no seasonal variability in the two components (which*

seems unlikely).

Reply: We have not sampled long enough to elucidate seasonal trends, however Yang's data was the result of a multi-year average. We observe only that our nitrate-sulfate ratios are reasonable values, e.g. their agreement within a factor of 2.

30. P7591 line7 – clarify that DustTrak, Aurora, and Dylos instruments are also nephelometers. were these instruments operated at ambient conditions?

Reply: We have now made it clear in section A2.2 these instruments were operated at ambient conditions.

31. P7591 line13 – ‘: :nephelometer signal saturated during extreme pollution events (PM_{2.5} >400 $\mu\text{g}/\text{m}^3$).’ – provide the scattering value corresponding to this pm2.5 value.

Reply: Page 7592, line 8, we added "...pollution events (PM_{2.5} > 400 $\mu\text{g}/\text{m}^3$) such that $b_{\text{sp}} > 1300 \text{ Mm}^{-1}$ "

32..P7600,P7601 – these tables are hard to read – could you make every other line italic? Or add spacing between sites.

Reply: We have increased the font size in tables 1 and 2 and bolded the font to aid legibility.

33. P7606 Figure 3 – the plot is labeled as ‘AODsat’ you should make it clear here and in text that AODsat relates to the AERONET retrieved AOD at the time of the satellite overpass, rather than the AOD from an (unnamed) satellite.

Reply: We have now emphasized that AODsat is *ground*-measured AOD during satellite overpass hours in section 4.1.

From Page 7583, Line 22: "We define AOD_{sat} as the ground-measured AERONET AOD averaged from 10:00 to 14:00 and interpolated to 550 nm via the Ångström exponents"

34. P7610 Figure A4 - need to do better job cropping figure.

Reply: There was an issue with saving the figure during typesetting we will aim to fix.