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Comment on amt-2021-284

Anonymous Referee #2

Referee comment on "Relative errors in derived multi-wavelength intensive aerosol optical properties using cavity attenuated phase shift single-scattering albedo monitors, a nephelometer, and tricolour absorption photometer measurements" by Patrick Weber et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2021-284-RC2>, 2021

This paper compares uses from different combinations of instruments to assess uncertainties in spectral optical properties (both measured (scattering/absorption/extinction) and derived (single scattering albedo and scat/abs/ext Angstrom exponents). They find that using the differential method (i.e., ext-scat) for absorption is very unprecise/uncertain at absorption loading levels $<20 \text{ Mm}^{-1}$ (the wavelength of absorption for that constraint is not stated). The uncertainty leads to uncertain absorption Angstrom exponents (AAEs). The authors recommend only using the differential method to obtain AAE when absorption $> 50 \text{ Mm}^{-1}$. This suggests that the differential method is only applicable in the most polluted atmospheric conditions for absorption-related parameters (see for example Figure 5 in Laj et al., (2020) which suggests that even GAW monitoring sites in urban areas rarely reach absorption loading of 20 Mm^{-1} at 550 nm). In contrast, the greater robustness of extinction and scattering observations means the wavelength dependence of these properties (e.g., scattering and extinction Angstrom exponents) are trustworthy at extinction levels $\sim 20 \text{ Mm}^{-1}$ (though again the wavelength for this constraint is not stated)? The authors suggest that either method they use for calculating single scattering albedo (CAPS PM_{ss}a or nephelometer+filter-based absorption instrument) is within the desired uncertainty for SSA.

General comments

While the authors are primarily focused on assessing the differential method for intensive aerosol optical properties in a lab setting, it would greatly expand the relevance of their paper if they could comment more on the implications of their findings for the many long-term monitoring sites around the globe that make aerosol optical property measurements (e.g., ACTRIS network). I'm not 100% sure that's a fair question as, in their study, the authors haven't considered actual atmospheric aerosol in all of its infinite variety (e.g., other absorbing species such as dust and 'brown carbon'; internal mixtures; etc.), but

those limitations could be acknowledged in such a discussion. Filter-based measurements of absorption are often dismissed as lacking (e.g., Lack et al., 2008) <https://www.tandfonline.com/doi/full/10.1080/02786820802389277>), but here it seems they perform quite well.

Please have the manuscript read/corrected by native English speaker or technical writer with eye for detail.

Science comments

The Abstract suggests that EAE and SAE are trustworthy for extinction coefficient values $> 20 \text{ Mm}^{-1}$. That result is not supported by anything presented in the main manuscript - none of the figures show SAE coloured by extinction and extinction coefficient loading is not mentioned in the discussion of SAE. (Figure 11 shows the SAE coloured by absorption not extinction.) Please elaborate. What are the results that lead to this conclusion? Extinction at what wavelength? and for what averaging time?

Line 115 - presumably then the time resolution of the other instruments was also reduced to 10 min? or was only 10min data recorded for the nephelometer while the highest resolution for the other instruments was used and then averaged to 10min when comparisons involved the nephelometer?

Line 165++ Error propagation - I refer you to the supplemental materials in Sherman et al., 2015 (www.atmos-chem-phys.net/15/12487/2015/) where the error calculations for these variables (χ AE and SSA) and for scattering and absorption are described in detail.

Line 211 - the uncertainty of absorption measured by the CLAP depends on the aerosol SSA and to a lesser extent on the averaging time. See equation 7 and figure 9 in Ogren et al. (2017). The Mueller paper does not consider the CLAP (or TAP). Are the PSAP uncertainties/precisions reported in Mueller assumed to be the same?

Related to the two notes above - for what averaging times are the uncertainties in the manuscript calculated? Would your results/conclusions change if longer averaging times were considered? I ask because long-term monitoring sites (e.g., GAW sites) typically use nephelometers and filter-based absorption photometers to obtain extensive and intensive optical properties. The GAW sites typically report hourly averaged data.

Line 206 '*Thus, the SAE drops to 0,76 for 130 nm AQ particles.*' I think the SAE drop is more due to the larger contribution of particles due to the spread in the size distribution for AQ than to the median diameter. Collaud Coen et al. (2007)

(<https://agupubs.onlinelibrary.wiley.com/doi/epdf/10.1029/2006JD007995>) show no spectral dependence for 130 nm particles in their Figure 7. AQ (and soot) size distributions range almost to 1000 nm in figure 2. Why does the SAE not drop as much for soot particles which have a slightly larger median diameter than the AQ?

Line 243 and Line 489 - make clear that the particles are externally mixed in the experiments (if that is the case). I think the AAE would vary if the absorbing particle was internally mixed with purely scattering aerosol.

Editing/wordsmithing comments

I started making editorial comments but quickly decided that too many were needed and that I should focus on the science. This is an important paper: please have a technical editor and native English speaker spend some time on it to make it as clear and accessible as possible.

Line 44 '*trough*' to 'through'

Line 48 '*TAP*' to '*CLAP*' Ogren et al. (2017) was only about the CLAP. The TAP is based on the design of the CLAP but there are some differences related to spot size, flow rate and potentially filter used. Please make this more clear.

Line 50 add Ogren, 2010, Bond et al., 1999. Also, Virkkula et al., 2010 is the corrigendum to Virkkula et al., 2005 which actually describes the full methodology used to develop corrections to the PSAP. Please cite both Virkkula papers.

Line 63-65 It is unclear what references go with what use of Angstrom exponent. please reorganise sentence. Use semi-colons between different clauses and group Foster and Angstrom references within the same parentheses if they are both for wavelength adjustment.

Line 75 '*classify*' to 'classifying'

Line 90 - should Ammonium in Ammonium sulphate be capitalised?

Line 90 '*where*' to 'were'

Line 90-91 Rephrase these two sentences - I think really what is meant is that the ammonium sulphate liquid solution concentrations were not changed so that the dry aerosol size distributions would remain constant.

Line 99 '*Downstream the production the aerosol*' to 'Downstream of production, the aerosol' or 'Downstream the production aerosol'

Line 101 '*connected to using*' to 'connected using'

Line 103 - Strange line over period.

Line 106 - Both '*was used*' and '*we used*' are used in the manuscript. Please be consistent with passive or active voice but not both.

line 106-107 '*small-sized*' suggests there are other sizes of TAPs. Recommend removing 'small-sized' - there are no comments on sizes of other instruments.

Line 107 - give manufacturer of PSAP

Line 120 - give refractive index (indices) of PSLs?

Line 123 - '*validating the same factor*' - what factor?

Line 123 - the neph was directly calibrated with CO₂ (and filtered air), correct? The way the sentence is written it sounds like a calibration 'factor' for CO₂ was derived from the CAPS and applied to the neph. Please clarify.

Line 126-127 - says truncation is not necessary for particles < 200nm but Figure 2 shows significant presence of particles > 200 nm for some species. Make clear that that comment applies to the CAPS. For the neph the Anderson and Ogren truncation will range from 13% (450 nm, SAE=0.76) to 2% (700nm, SAE=3). Onasch et al. (2015) suggests the truncation characteristics of the CAPS PM_{ss} are similar to those of commercial nephelometers, so perhaps CAPS truncation correction is necessary?

Line 126 - '*since highest*' to 'since the highest'

Line 126 - '*amount were*' to 'amount was'

Line 127-128 - '*drifting shift of baseline*' to 'drifting baseline' Also - presumably no data were used during the warm up period? Please clarify.

Line 127 - '*Onasch et al., 2015a*' to 'Onasch et al., 2015'

Line 131 - filter spot does not need to be capitalised, remove comma after 'selected'

Table 1 - should cite both the original Virkkula paper in 2005 and the 2010 Virkkula paper, which is the correction to the original paper.

Line 136 - I would not call the Anderson and Ogren correction a data inversion - the scattering coefficient is multiplied by a correction value C where $C=a+b*SAE$ where a and b are constants and SAE is the uncorrected scattering Angstrom exponent for that particular scattering data point.

Line 136 - nephelometer does not need to be capitalised

line 136 - '*alterned*' to 'altered'

Line 139 - what is PMex? presumably you mean σ_{ep} ?

Line 141 - same comment as above: should cite both the original Virkkula paper in 2005 and the 2010 paper which is the correction to the original paper.

Line 141 - what filter was used with the TAP? The Virkkula 2005;2010 and Bond 1999 and Ogren 2010 corrections were all done with the Pallflex filter. I believe the TAP is shipped with Azumi filters from Brechtel. See discussion in Ogren et al 2017 about differences in correction for the different filters. (Perhaps that is taken care of by the TAP software, but should still be stated what filters were used.)

line 143 - '*Probertites*' to 'Properties'

Equation 3 and line 158 - use the same variable as in equation 3a - i.e., σ_{xp} . You could also put an x in front of AE: $xAE = -\log(\dots)$

Line 178 - '*with minimised size distribution chances*' - unclear what is meant - do you mean 'with minimal size distribution changes'?

Line 187 - '*its spherically shape*' to 'it is spherically shaped'. Split this sentence into two or maybe three sentences. It is hard to follow.

Line 189 - '*approved by DMA and OPC.*' to 'confirmed by DMA and OPC measurements.'

Table 3 - Change table title to 'Overview of the aerosol types used and measured parameters'

Line 205, 206 - Inconsistent notation for numbers - sometimes European notation, but elsewhere use American notation. Should be consistent.

Line 212 - extra parenthesis, also ACP standard for refs is (Anderson and Ogren, 1998; Massoli et al., 2009).

Figure 2 caption - change to: 'Size distributions measured by DMA and CPC ...), Also, use a lighter blue for soot - hard to distinguish from the black.

Line 228 - '*data points averages of at least 100 seconds*' - earlier it's noted that the neph time resolution was 10 min (6000 seconds). Please clarify.

Line 233 - '*for 450nm ... wavelength*' to 'for the 450nm ... wavelengths'

Figure 3 - are there points with $SSA > 0.9$? I recommend using a different colour for those points than the error band colour. Could a consistent SSA gradient be used? - the SSA steps vary between 0.07 and 0.08 in the colour code legend. (Also the colour scale for SSA appears to change amongst the plots - e.g., for figure 3 the highest SSA value is

0.90, while for figure 4&5 it's 0.90 and 0.95. The figure 3 caption suggests that the colour code represents the SSA at 630 nm even if the plot is for 450 nm. Please correct if that is not the case.

Line 262 - '*nor a strong shift for high or low volumetric cross-section values*' do you mean loading?

Line 294 - tables should be numbered in the order in which they are mentioned in text. Here Tables 7-9 are referred to before Tables 5 and 6.

Line 296 - '*delivers reliable SSA*' - the SSA results have not been reported yet in this paper. Should this statement have a citation?

Line 300 - '*1 Hz*' measurement - but the neph is averaged to 10 min which is not 1 Hz.

Line 310 - is the variance for $\sigma_{ap} < 10 \text{ Mm}^{-1}$ not shown? Table 5 only shows ensemble and variance $> 10 \text{ Mm}^{-1}$? for what wavelength is the filtering done?

For both Table 5 (630 nm) & Table 6 (450 nm) is the $\sigma_{ap} > 10 \text{ Mm}^{-1}$ variance for the wavelength that the table is for (i.e., 630 nm for Table 5 and 450 nm for Table 6)? or do both tables assume variance for wavelength $> 630 \text{ nm}$?

Line 316 - '*As a result, this increase also the errors associated with the differential method*' Is this saying that the higher loading is leading to higher uncertainty in absorption?

Line 320 - for what wavelength is the filtering done?

Line 385 - why is the CAPS(scatter)/NEPH(ext) combo used as the SSA reference rather than CAPS(scatter)/CAPS(ext) which would measure in the same volume and not have the flow/averaging issues of the neph? Onasch et al. (2015) lists the advantages that the SSA from the CAPS PM_{SSA} has over calculating SSA from two separate instruments.

Line 385 - 'often used combination' - often used by who?

Figure 7 - as suggested above - use different colour for points than is used for the error band.

Figure 9 and Figure 10 are not referred to in the text.

Figure 10&12 - why are points not coloured by loading?

Line 482-483 '*Here again NEPH shows higher SAE values compared to CAPS by a factor 0.9.*' Please check. Both Figure 12 and Table 12 suggest the CAPS SAE is typically higher than the NEPH SAE - most CAPS points are above the 1:1 line and the CAPS/neph ratios in Table 12 are greater than 1. Also, where does the factor of 0.9 come from?

Figure 14 - '*EAE(TAP)*' to '*AAE(TAP)*'. Are these the same points as are shown in Figure 13?

Line 514 - '*EAE(TAP)*' to '*AAE(TAP)*'.

Line 547-549 - make clear that the constraints '*Low single scattering albedo values (<0.5) and, more importantly, high particle loads of at least 50 Mm⁻¹*' are specific to the differential method and don't appear to be relevant for the filter-based method of obtaining AAE.

Line 551 - the conclusion states that the largest disagreement for absorption coefficient is due in part to filter correction schemes, but this was not shown/discussed in the manuscript.

Line 561 - The suggestion to use the CAPS PM_{ss} as a substitute for the TSI nephelometer doesn't seem logical to me - first - three CAPS would be needed to provide the same spectral coverage at 3 wavelengths that the TSI nephelometer provides in one instrument. Second, the CAPS PM_{ss} doesn't provide backscattering coefficient. The Ecotech Aurora 3000 (or 4000) nephelometer seems to be a more reasonable TSI nephelometer replacement as it provides the same functionality as the TSI instrument. If someone already has a CAPS then yes - it could be used as a stand-in for the TSI nephelometer total scattering for one wavelength, but I would hesitate recommending a CAPS in lieu of a nephelometer - it would depend on the scientific question that was being addressed.