Atmospheric Measurement Techniques Discussions

Atmos. Meas. Tech. Discuss., referee comment RC2
https://doi.org/10.5194/amt-2022-278-RC2, 2022

Comment on amt-2022-278

Anonymous Referee #2

Referee comment on "Performance evaluation of portable dual-spot micro-aethalometers for source identification of black carbon aerosols: application to wildfire smoke and traffic emissions in the Pacific Northwest" by Mrinmoy Chakraborty et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2022-278-RC2, 2022

The authors report on the performance of the MA300 aethelometer relative to the AE33 aethelometer in terms of measurement of effective black carbon concentrations and wavelength dependences. Overall, I think the measurements are of good quality and the topic worthy of consideration. However, I think that the manuscript would be greatly strengthened if it were reframed to focus entirely on the directly measured properties rather than on the derived particle concentrations from source apportionment. I have some fundamental concerns about the interpretation of the source apportionment results and the overall method used to do the source apportionment. I realize that the authors are not introducing the method, but using something from the literature. Nonetheless, I find it detracts from the primary focus of the manuscript, namely the performance of the MA300 relative to the AE33.

Major comments:

Section 2.6: I have a fundamental issue with the methodology applied here. In particular, the assumption that biomass burning particles have a fixed absorption Angstrom exponent. This is known to be false. The AAE for biomass burning varies tremendously dependent on the fuel source and burn conditions. Any apportionment that assumes a fixed value for the AAE for biomass burning is, in my view, inherently flawed. I realize that the authors here are not developing this method and are taking these values and the approach from the literature. Nonetheless, in my view the method is poorly equipped for

true source apportionment since one of the end members is unknown/non-constant. Related, it is not clear why the authors would state on L199 that the AAE for BC is 1 but then in their apportionment use a value of 0.9. It is established that BC has an AAE that is close to 1 but that is not necessarily exactly 1 depending on the particle size or coating state. It would be nice to see a recognition of these issues in the discussion of the apportionment method.

L221: In my view, it is not correct to refer to "eBC_bb" and "eBC_ff" from the source apportionment. The method is not separating different BC sources, but is separating absorption and specifically separating BC (the absorption properties of which are largely independent of the source) and the contribution of brown carbon (BrC). As such one cannot apportion "eBC_bb." One can apportion "eBC" and "eBrC" if some assumed value for the MAC for BrC is determined. Or one can apportion the absorption. But one cannot apportion the absorption into BC from fossil fuel and biomass burning. As currently presented I do not think that this is accurate or correct.

Fig. 2 vs. Fig. 1: The slopes in these two plots should be, largely, directly related. The main reasons that the slopes would change is because of (i) noise differences between wavelengths and (ii) the difference in the assumed MAC values. In particular, the slopes in Fig. 2 should be pretty similar to the slopes in Fig. 1 multiplied by the ratio of the MAC values between the MA300 units and the AE33 units. The authors might specifically acknowledge this relationship. They might consider normalizing the slopes to the value in the UV or IR, as the focus here seems to be on the wavelength-dependence of the slopes and not the absolute values (since, again, the absolute values just come directly from Fig. 1). Related, I personally did not find the discussion in Section 3.2.2 particularly valuable, as it is really just reiterating the results already shown for the eBC concentrations, but here in absorption space. I suggest removing this section or refocusing it on the AAE. Or at least merging it with section 3.2.3, as I see these as largely redundant discussions.

L358: The authors state that "in Section 3.3, we identify that the UV channel absorption measurements, babs,UV , are subject to higher error than measurements at higher wavelengths." This is not robustly true. In particular, it is evident in Fig. 2b that the slope relative to the reference instrument is closer to unity for the UV channel compared to other wavelengths. So I do not understand what the authors mean here when they say that the UV has higher error. Higher error relative to what? What is the metric? Also, in Fig. 4 the UV channel has a slope closer to unity than the IR channel and has a higher R2 value.

L384: The authors need to demonstrate that these statements and conclusions regarding biomass burning contributions during the Reg period are robust to a rigorous assessment of measurement uncertainties and to uncertainties inherent in the assumptions made during the apportionment. What if the BC AAE were 1.2 rather than 1.0, for example? This is within the realm of reasonableness. How would the conclusions here change? This points to a bigger need for a more rigorous assessment of uncertainties in the context of the apportionment that goes beyond noise characterization which dominates the manuscript.

Section 3.4.2: I strongly encourage the authors to reframe this entire discussion to focus on the AAE values and how they compare between the instruments. Everything in the apportionment comes back to the AAE. The apportionment is just a mathematical transformation that then brings in the assumptions regarding AAE values for different particle types. A focus on the inherent measurement, the AAE, rather than subsequently derived properties would, in my view, greatly strengthen the manuscript. To me, I see the apportionment discussion a distraction from the core assessment of instrumental performance.

Other comments:

L40: Rather than making the distinction between "traditional" and new measurement methods, I suggest that the authors simply say that there are three main methods for characterization. The cited paper regarding LII is from 2006, and thus it has been around long enough and used by enough people that it could easily be considered part of the "traditional" canon.

L52: I suggest stating that at 880 nm light absorption is predominantly due to BC rather than it being "only" due to BC, as the former is more formally correct.

L70: The authors might note that, if it is assumed that the scattering correction factor is wavelength independent (which is not a given) that the accuracy of the *C* parameter impacts primarily the determination of the absolute absorption and derived concentrations and not the apportionment.

L87: All AE33's have the same properties and thus "typically" can be removed.

L100: Further details regarding the nature of the "non-linearity arising from flow" would be welcome. As is, it is difficult for a reader to understand what is meant by this.

L154: What is meant by the "(0-120)?" It is unclear.

L156: Convention is generally that sigma means a molecular cross section and that the MAC is simply referred to as MAC. I suggest adopting this convention.

L160: This should refer to Equation 2, not 3.

Eqn. 2: It should be given in the main text. Also, what doe the periods mean? Are they meant to indicate multiplication?

L161: Does the scattering correction factor, C, depend on wavelength? If not, why not, as scattering is a fundamentally wavelength-dependent property?

L170: The authors might note that the MAC values for both instruments are much higher than known (realistic) MAC values for BC, and thus are best considered as effective values rather than actual values. I think this is an underappreciated aspect of these instruments: the absorption measured is not the true absorption because if it were then realistic MAC values would be used. In many ways, the absorption coefficients determined here should be called "effective absorption coefficients" in the same way that the BC is referred to as "effective BC."

Eqn. 6 should technically use a proportional too symbol, not an approximately symbol.

L230: I suggest that the authors retain these high PM event days as they provide an additional test of the comparability of the two instruments, which is the overall aim of this paper. The exclusion of these leaves the reader wondering if the relationship between the two completely broke down during this period, leading to questions regarding the overall performance. Including this period as an additional case study would strengthen things, in my opinion.

L233: I strongly suggest referring to this as "relative accuracy" rather than "accuracy."

This assumes that the AE33 is accurate, yet it has its own uncertainty.

L234: R2 is not a measure of accuracy. It is a measure of goodness of fit, or in this case of the cross-precision of an MA300 and the AE33. I suggest this be revised. The slope is a measure of the accuracy (assuming that the AE33 is, in fact, accurate).

L250: I am unfamiliar with the term "periodical average," and suspect many other readers will be too. I suggest this be defined.

L250: I think that grammatically this should be "during the Reg period" and "during the WF period." This is a general statement for the remainder of the manuscript. It seems that most of the time the author does this.

Table 1: Please report the actual wavelengths, rather than using e.g., "red." Also, the number of sig figs is more than seems appropriate given the standard deviations.

L263: I believe these are average values for the diurnal profile. It would be useful to have this clarified.

L266: In what way are the R2 values "estimated?" Aren't they simply "calculated?" This is

a general statement that also applies to statements like “the estimated coefficients” such as on L269. These are calculated.

L281: As there are only three slopes, I suggest just reporting these three rather than stating a range.

L295: If the authors still have access to the AE33 and MA300’s I encourage them to put a filter on the inlet and measure the standard deviation for particle free air for their specific models.

Fig. 3: We know that the absolute absorption value should vary with wavelength, so I think that it is appropriate to use a distinct x-axis scaling for each wavelength considered. As it stands, by using a constant range it is difficult for the reader to see the data as well at the longer wavelengths.

L333: The authors state that “The slope’s variability in spectral measurements shows similar trends across the MA300 units, indicating the effect of instrumental sensitivity in resolving multiwavelength

babs.” It is not clear to me how instrumental sensitivity comes into play here. Can the authors expand or clarify?

L335: The authors state “In Figure 3, we identify that the unit-to-unit variability ranged 20–23% during Reg and 17–19% during WF period.” It is not clear to me that this is what

the figure shows. The variability would be relative to the average of the three instruments and not relative to the reference instrument. Using the appropriate reference would change the slopes and numbers, but probably not the general conclusions.

L339: What is meant by "large offsets in the light absorption measurements."? I find this to be unclear.

Fig. 4: I find myself somewhat confused regarding these data and the slopes shown. These slopes should, presumably, be related to the slopes shown in Fig. 2. Yet they are not the same. This should be clarified.

L365: The authors here refer to aging affecting wildfire smoke and seemingly exclude aging of fossil fuel-derived particles. Yet, fossil fuel-derived particles also age and can become more hygroscopic over time, so I do not follow this argument. I think that the argument needs to be strengthened or removed.

L368: The authors here state that measurements are not useful when they are too noisy. This is a very general statement appropriate for any measurement and so I don't see the value in including this here.

L370: Again, I do not see where the authors have demonstrated the greater accuracy of the blue measurements relative to the UV. This statement is not consistent with Fig. 2.

Fig. 5 caption and Section 3.4.1: To reiterate an above point, I do not think it is appropriate to state that the method measures a fraction of eBC mass from biomass burning. That is not what is apportioned. The method does not differentiate between BC sources. It looks at the influence of brown carbon on the eBC determination and ultimately apportions the absorption between BC and BrC.

L390: I would go further here to say that not only "may" an assumption of the constant AAE for the biomass component not accurately separate the components to state that it definitely "does not." This links to my earlier point that the AAE values are not fixed for biomass burning and any method that assumes it is a constant is inherently flawed. This goes to the authors finding that the blue-IR apportionment method gives different values than the UV-IR apportionment. The authors used distinct AAE values for each wavelength pair but they have not done any robust assessment of the reasonableness or robustness of these numbers. There is an epistemic uncertainty associated with these values making it impossible to actually know what they "should" be in any given situation. This comes back to my point above that there needs to be a more rigorous assessment of known and unknowable uncertainties.

L405 and Fig. 5: The authors focus in this manuscript is on assessment of the MA300 units. Thus, I find it a surprising choice to show the AE33 apportionment results in the main text (Fig. 5) while putting the MA300 apportionment results in the supplemental. To me, this seems backwards. I also do not fully see the need for the discussion in Section 3.4.1 as this is all from the AE33 and thus not a core focus of this manuscript.