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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An Evaluation of three methods for measuring black carbon at Alert, Canada" *by* Sangeeta Sharma et al.

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Referee D. Baumgardner on "An Evaluation of three methods for measuring black carbon at Alert, Canada" by Sangeeta Sharma et al. First of all, the authors like to thank Dr. D. Baumgardner for accepting to review this paper with such constructive remarks.

The suggestion of including the aerosol Absorption Angstrom Exponent (AAE) is great as we initially included AAE in the earlier version of the paper but it didn't show any distinction between various combustion source influences at Alert location and was thus removed. The hourly average AAE between March 2011 and December 2013 are shown in Figure below. Values of AAE between 0.5 and 1.5 represent absorption primarily due to fossil fuel BC. A value near 1.0 is considered to be an example of

graphitic carbon particles (Petzold et al., 2009), while AAE values close to 0.5 may reflect different absorption characteristics of elemental carbon (Bahadur et al., 2012). There are brief episodic increases in AAE where values over two are reached, indicating the presence of non-BC absorbing aerosol, but most of the fine mode absorption measurements fall within 0.5-1.5, suggesting that BC is the primary absorbing component with episodic influences of non-BC absorbing components. Mineral dust gives AAE values of three and larger at visible wavelengths (Petzold et al., 2009), which are not evident in the Figure.

Bahadur, E., Praveen, P. S., Xu, Y., and Ramanathan, V.: Solar absorption by elemental and brown carbon determined from spectral observations, P. Natl. A. Sci., 109, 17366–17371, 2012. Petzold, A., Rasp, K., Weinzierl, B., Esselborn, M., Hamburger, T., DÌĹornbrack, A., Kandler, K., Schutz, L., Knippertz, P., Fiebig, M., and Virkkula, A.: Saharan dust absorption and refractive index and from aircraft-based observations during SAMUM 2006, Tellus B, 61B, 118–130, 2009.

Since the AAE is clearly and dominantly influenced by fossil fuel combustion, we are adding this Figure_Supplemental_2a showing 550 nm absorption and Figure_Supplemental_2b showing AAE time-series. The method, uncertainty calculation and discussion of AAE has been added to the Supplement and few sentences to support fossil fuel influence on the aerosol in the paper.

P 16, lines 10-13.... The aerosol Absorption Ångstrom Exponent (AAE) values, as discussed in the supplemental section (see Figure_supplemental_1b), also suggests predominately fossil fuel sources of rBC and little biomass burning influence (AAEavg (April and Oct)=0.75$\pm$0.12). P 17, lines 18-20... The influence of brown carbon may be minimal at Alert as values of the aerosol Absorption Ångstrom Exponent (AAE) are between 0.5 and 1.5 suggesting predominantly fossil fuel influence (see Figure_Supplemental_1b). P 20, lines 6-8, ... "As discussed earlier, the influence of brown carbon due to biomass burning is minimal at Alert during the Arctic haze time for the data collected during the 2011-2013 (AAEavg for April =

0.75±0.12). Thus, that effect of brown carbon will be minimal on the MAC. Also added this to supplemental section: Calculation of Aerosol Angstrom Absorption Exponent: The aerosol Ångström absorption exponent (AAE) was calculated from the PSAP absorption measurements. The AAE is defined as AAE=(lnâĄą((σ_ap (λ_1))/(σ_ap (λ_2)))/(lnâĄą(âĄąãĂŰ((λ_1)/λ_2 ãĂů)) 1 where ïĄň1=467 nm and ïĄň2=660 nm and σap(ïĄň1) is absorption at 467 nm and σap(ïĄň2) is absorption at 660 nm.

Uncertainty in AAE Standard techniques were applied to determine combined uncertainties in the Aerosol Absorption Exponent calculated at two wavelengths; ïĄň1=467 nm and ïĄň2=660 nm. The uncertainty in AAE is determined by Eq. 2 has also been used in Sherman et al. (2015).

ΔAAE(467nm/660nm)=((∂AAE/ãĂŰ∂σãĂů_(ap,467))ˆ2 ΔãĂŰσ_(ap,467)ãĂůˆ2+(∂AAE/ãĂŰ∂σãĂů_(ap,660))ˆ2 ΔãĂŰσ_(ap,660)ãĂůˆ2+2*corr(σ_(ap,467),σ_(ap,660)) ãĂŰ*(∂AAE/ãĂŰ∂σãĂů_(ap,467))*(∂AAE/ãĂŰ∂σãĂů_(ap,660))*Δσ_(ap,467)*Δσ_(ap,660)))ãĂůˆ(1/2) (2) where (∂AAE/ãĂŰ∂σãĂů_(ap,467))=2.26/σ_(ap,467) and (∂AAE/ãĂŰ∂σãĂů_(ap,660))=(-2.26)/σ_(ap,660)

The time series of hourly light absorption measurements from the PSAP at Alert at 550 nm wavelength is shown in Fig_Supplemental_1a. The light absorption has been corrected according to Bond et al. (1999) and also Ogren (2010) for loading and scattering interferences. Episodic increases in absorption during winter/spring reach as high as 4 Mm-1 and overall lower values are measured during the summer and fall. Dust and brown carbon each have strong wavelength dependences, but BC does not. The impact of non-BC light absorbing species will appear as deviations from near unity (1.1±0.3) in the Absorption Ångstrom exponent (AAE) if the non-BC light absorbing species make up more than 40% of the BC (Lack and Langridge, 2013). At Alert, non-BC light absorbing species may include brown carbon and dust. At Alert, absorbing OC (POC, i.e. brown carbon) is more than 40% of the total absorbing carbon for most of the time. The hourly averaged AAE values between March 2011 and December

2013 are shown in Fig._Supplemental_1b. Values of AAE between 0.5 and 1.5 represent absorption primarily due to fossil fuel BC. A value near 1.0 is considered to be an example of graphitic carbon particles (Petzold et al., 2009), values between 1 and 1.5 are due to total carbon, while AAE values close to 0.5 may reflect different absorption characteristics of pure elemental carbon and increase with varying amounts of OC (Bahadur et al., 2012). There are brief episodic increases in AAE where values over two are reached, indicating the presence of non-BC absorbing aerosol, but most of the fine mode absorption measurements fall within 0.5-1.5, suggesting that EBC is the primary absorbing component with episodic influences of non-BC absorbing components. Mineral dust gives AAE values of three and larger at visible wavelengths (Petzold et al., 2009), which are not evident in Fig._Supplemental_1b. Addition of Supplemental_Fig1:

Additional comments: P6, line 7: “There are no other scattering or absorption corrections,. . .” I don’t understand why corrections are not being applied when further on PSAP is corrected. Our main purpose for using the Aethalometer data is in its “unmodified form” to see how well it compares to other more absolute mass techniques for measurement of “black carbon”. There is enhancement in the absorption in Aethalometer due to filter matrix as well as scattering components on the aerosol and it could be as much as by a factor of 3 as recently estimated by Backman et al., 2017 for several Arctic locations including Alert (which includes loading and scattering correction). Magee uses a much higher MAC value than needed for aerosol in the atmosphere. A MAC value of 16.6 m2g-1 at 880 nm has been used in the Aethalometer firmware by the manufacturer to compensate for these artifacts and give best estimate of EBC. We wanted to demonstrate how well is Aethalometer measuring EBC with Magee’s MAC value used at 880 nm. We have long term trends in the EBC measurements derived from Aethalometer at Alert. In the past, we have compared EBC to EC to apply a correction to EBC at Alert and the two techniques agree quite well during the Arctic haze time. Modified by adding these lines on P6, lines 8-11: “There are no other scattering or loading corrections applied to Aethalometer data because a comparison of unmodified EBC mass to best estimate of “BC” mass values are also determined

in this paper. The enhancement in the absorption due to total scattering has been compensated by using a higher αap value used by the Aethalometer firmware. Also added to Section 4.1.1, pg 17 lines 21-25: “In addition, the Aethalometer response depends on filter loading and multiple scattering by the filter medium and sampled aerosol particles. Scattering correction thus becomes important in cases when the aerosol has higher scattering with respect to total extinction (absorption+scattering), i.e. absorption is low. This is not the case at Alert especially during the Arctic haze time. Summertime measurements could fall into this scenario.”

Also Pg 18, lines 1-2: EBC (unmodified) needs to be evaluated due to these reasons in comparison to absolute measurements mass techniques. PSAP absorption coefficients have been corrected by using scattering data measured by a 3-w TSI Nephelometer. The main purpose for applying scattering corrections for PSAP absorption was to derive a MAC values at this location by using best estimate of averaged mass of EC and rBC measurements. The scattering correction was absolutely necessary for this purpose.

Added on P6, lines 30, P7 lines1 &2 Aerosol light scattering, ïĄşsp was measured at Alert by using a TSI nephelometer at three different wavelengths: 450, 550 and 700 nm. The truncation error of the nephelometer, which is due to an angular integration restriction to 7 and 170o (Anderson and Ogren, 1998) was estimated and applied to scattering measurements. Scattering correction was applied to absorption measurements as shown in equation 4.

P7, Line 6: How was PSAP measurements converted from 530 to 550 nm? Added on P7, line6, “by using (ïĄň)-1 relationship to the wavelength. . ..”

Section 2.4: The uncertainty estimates should be added in Table 1. Uncertainties were added to column #1.

Section 4.1.4: Should explain why EBC is not used in the best estimate of BC

We haven’t included EBC in the best estimate of BC as it is light attenuated inferred mass measurement. Our comparison at the end of the paper tells us that these measurements are very close to best estimated absolute mass measurements. Added to P19, lines 17-18, “Considering all arguments, including EC and rBC being more specific direct mass measurements than EBC, which is light attenuation inferred mass indirect measurement,. . ...”

Added to P19 lines 23-25, “EBC mass is not used in the determination of best estimate mass of “BC” as it is an inferred mass derived from optical measurements and need to be evaluated with more direct mass measurements techniques at Alert, presented in the later section.”

Page 21 Supplement figure. Fixed the caption as shown below.

"....(green and red triangles are for data during spring and winter)"

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-339/acp-2017-339-AC2-supplement.pdf

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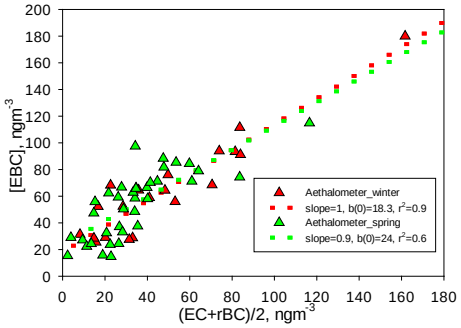

Supplemental_Fig-2: Improved agreements were obtained between the best estimated black carbon mass and masses obtained by optical technique such as Aethalometer (green and red triangles are for data during spring and winter). EBC Aethalometer and rBC data were averaged to EC sampling times.

Fig. 1.