

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

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Reply to reviewer #2, anonymous is as follows on “An Evaluation of three methods for measuring black carbon at Alert, Canada” by Sangeeta Sharma et al.

We would like to thank reviewer #2 for accepting to review this manuscript and providing such positive and constructive remarks. One major concern reviewer #2 has is that why was the scattering correction not applied to Aethalometer data. Our main purpose for using the Aethalometer data in this paper is to assess how unmodified EBC compares to other more absolute mass techniques for measurement of “black carbon”. That’s the reason we used unmodified EBC. We also realize importance of making scattering correction to that data but only if absorption measurements are used. There

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is enhancement in the absorption in Aethalometer measurements due to filter matrix as well as scattering components on the aerosol and it could be as much as by a factor of 3.2 (which includes loading and scattering) as recently estimated by Backman et al., 2017 for several Arctic locations including Alert. Magee uses a much higher α_{ap} value than needed for aerosol in the atmosphere. A α_{ap} 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 according to the manufacturer gives the best estimate of EBC. We wanted to demonstrate how well Aethalometer is estimating EBC with Magee's α_{ap} value at 880 nm as 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 made in this paper. And that how well an enhancement in the absorption due to total scattering (loading and scattering by aerosol) has been compensated by using a higher α_{ap} value by the Aethalometer firmware. This is further discussed in section 4.1.1.”

Also section 4.1.1 explains this well on P17 lines 15 to 28: “EBC will overestimate BC if there is absorption from coexisting components and/or coatings of the rBC cores, such as brown carbon (e.g. Kirchstetter et al., 2004; Lack et al., 2013; Lack and Langridge, 2013) and fine particle dust (Weingartner et al., 2003; Müller et al., 2011). However, the influence of brown carbon may be minimal at Alert as values of the Angstrom Absorption Exponent (AAE) are between 0.5 and 1.5 suggesting predominantly fossil fuel influence (see Supplemental section and Figure_Supplemental_1b). 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 (absorp-

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tion+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. Recently, Backman et al. (2017) proposed a reduction of a factor of 3.2 in the light absorption coefficients derived from the Aethalometer due to multiple scattering enhancements associated with particles collected on the filter. These enhancements are considered, at least in part, in the EBC estimate by the τ_{Aap} value used with the Aethalometer, but there remains uncertainty in τ_{Aap} , including the use of a constant value for all conditions. 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 correction for PSAP absorption was to derive 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 for the PSAP absorption.

Added on P6, lines 30, P7 lines1-4: “Aerosol light scattering, τ_{Assp} 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 170° (Anderson and Ogren, 1998) was estimated and applied to scattering measurements. Scattering correction was applied to absorption measurements as shown in equation 4.”

Detailed comments: P1, L3 use the same sig figs. Not sure what the reviewer means for P1 L3, you mean P2, L3 where I mentioned 8 ± 4 etc. Yes fixed to two sig. figs. 8.0 ± 4.0 , 8.0 ± 4.0 , 5.0 ± 2.5 and 9.0 ± 4.5 . You mention P3, line 9 but it is P3, L19, changed to “...light attenuation is converted to BC using a MAC value...” P6, L15 ECC? It is an acronym for Environment and Climate Change Canada, name of our government organization. P9, L17, you mean P9 L25 dATN cannot be >2 if you use the definition of ATN in EQ. (1) – the manufacturer presents ATN in percents. “...estimate that for $\Delta\text{ATN} \geq 2$ a relative uncertainty ($d\sigma_{\text{atn}}/\sigma_{\text{atn}}$) is 2.5% (for noise

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only)." The $\Delta ATN \geq 2$ is in "units" as reported by the instrument which are the ATN measurements multiplied by 100 for numerical convenience. Added to text for clarity, P9, L25, "...2%"

P10, L9 " Equation 7 is rewritten in Eq. 6" Should this be "Equation 4 is rewritten in Eq.7"? P10, L17, changed to Equation 4 is rewritten in Eq. 7.

P1, L20, Eq. 9. I don't understand how you get the numbers 0.02, 1.44 and 0.24. The reviewer means P10, line27 $\Delta \sigma_{(ap,cal)} = \sigma_{(ap,meas)} / (0.02 * \omega_0 / (1 - \omega_0) + 1.44)^2 * ((0.02 * \omega_0 / (1 - \omega_0))^2 + (0.24)^2)^{1/2}$

"The constants in equation 4 were derived by Bond et al. (1999) as $K1 = 0.02 \pm 0.02$ and $K2 = 1.22 \pm 0.20$ ". These constants were determined in Bond et al., 1999 for the PSAP instrument. I have added this statement to the paper on P11, L1-4,

"Where $K1 = 0.02 \pm 0.02$ and $K2 = 1.22 \pm 0.20$ and uncertainties in the calibration constants are $\Delta K1 = 0.02$ (Bond et al., 1999) and $\Delta K2 = 0.24$ (from Bond et al., 1999 with Ogren 2010 adjustment)."

P10, L25 now on P11, L9, modified to, "... Weekly zeroes are performed on the PSAP at Alert flushing particle-free air for the time period of one to two hours through instrument."

P11, L1-17. The uncertainty analysis of the SP2 is very short compared with that if the PSAP. The numbers are simply given. Can you give any more details? Is, for instance the number 19% the uncertainty of the slope? Thank you for pointing this out. P11 L13-31 to P12 L1-4 We are replacing one paragraph "There are a number.... underestimated" with "Uncertainty in the rBC mass derived from SP2 measurements arise from several sources. As described in section 2.2 mass calibration for all three SP2s was carried out using Aquadag as an external standard. Uncertainties in the slopes of the Aquadag calibration curves give rise to uncertainty the rBC mass calculated for each detected particle. This uncertainty is dependent on the individual particle size

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and ranges from around 5% for the largest particles to around 35% for the smallest particles (based on the calibration with the largest uncertainty). When the individual particle masses are combined to give a 1-hour mass concentration, the overall mass uncertainty arising from the calibration curves is on average 12%, 11%, and 16%, for SP2#58, #44, and #17 respectively. Another uncertainty in the rBC mass arises from using Aquadag as a standard. Due to the SP2's enhanced sensitivity to Aquadag (discussed in section 2.2), the calibration curves were scaled by dividing by a factor of 0.70 ± 0.05 . After this correction is applied, the combined 1-hour mass concentration uncertainty (arising from uncertainty in the fits of the calibration curves and from the uncertainty in the Aquadag correction) is 19%, 18%, and 23% for SP2#58, #44, and #17 respectively. Additionally, there is an uncertainty of approximately 12%, 7%, and 15%, for SP2#58, #44, and #17 respectively, which arises from the process of fitting the mass distribution and using this fit to estimate how much rBC mass lies outside the instrument detection range. This results in overall mass uncertainties in the range of 25-38% depending on the instrument used. In some cases calibrations were not carried out over the full detection range of the instrument and had to be extrapolated to higher rBC masses. Uncertainty from this extrapolation is not accounted for; however, the linear correlations between rBC mass and peak height are relatively strong (as shown in Fig. 2) suggesting that this is not a large source of error."

P13, L13-14. "... a factor of 3 higher for all data..." Why is this ratio so different from the slopes _1-2 presented in Table 2? A factor of 3 is arising by using comparisons between EBC/EC and rBC measurements by taking the ratios of all data above detection limit and doing pairwise comparison. Table 2 includes all data in the regression including the below detection limit values as these are regressions. The caption also states that seasonally averaged values include negative values too. In Table 3 if you compare, all data concentration in column 2, where also negative values are included, the ratio EBC/rBC=2.2 and EC/rBC=1.8. These are more comparable to Table 2 regressions. Adding this line to clarify the two in Table 3 caption: "Table 3: Statistical parameters such as mean, median and standard deviation for all data, Mar 2011 to

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Dec 2013 and data with only above detection limit values included. The ratios are only meaningful for data above the detection limit values. Also pairwise statistics available for data set when both pairs in comparison had data. In comparison to Table 2 where all data including negative values are used in the regression analyses, Table 3 values in first column labelled "All data conc." includes negative values too and are more comparable. Also added in Summary and Conclusions, P23 L6, "...on average results of pairwise analyses show both. ..."

P15, L8, L10. What is "rate of thickness increase" and "thickness rate"? Replacing on P15 L23-26 "As rBC cores decreases below 115 nm, thicker coatings are required to produce a measurable scattering signal. In all panels, the apparent coating thickness increases with decreasing rBC core. Below about 120 nm, the apparent rate of thickness increase with decreasing rBC is larger than above 120 nm. Since only thicker coatings can be measured at smaller sizes, some of the increase in thickness rate is attributed to bias in the elastic scattering detection system toward thicker coatings when the rBC cores are less than 115 nm. Overall, there is some increase in coating thickness with smaller rBC core sizes, but the thicknesses represented at rBC core sizes of less than 120 nm are overestimated." With "In all panels, the apparent coating thickness increases with decreasing rBC core. As rBC cores decrease below 115 nm, thicker coatings are required to produce a measurable scattering signal. As a result, when the rBC cores are less than 115 nm, the coating thickness is overestimated due to bias in the elastic scattering detection system toward thicker coatings." P16, L6. What do you mean by "events"? Event is short for particle detection of different types e.g. incandescent and scattering. P16, L12-L17. Also somewhere earlier: could you estimate the mass or volume fraction of BC in the size range where you do have the coating data?

On P17, L1-2 Changed this statement by adding:

"For rBC cores in the 160-180 nm range, the average particle coating thicknesses in April 2012 and in 15 October-November 2013 were estimated to range from 1.25 to 1.4

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(this corresponds to a mass fraction of rBC ranging from 0.51-0.36, assuming a 170 nm rBC core).”

P17, Section 4.1.1 EBC. Here you should give some kind of an estimate of uncertainty due to scattering. See my general comments. Made these changes to section 4.1.1 as per earlier main comments. “EBC will overestimate BC if there is absorption from coexisting components and/or coatings of the rBC cores, such as brown carbon (e.g. Kirchstetter et al., 2004; Lack et al., 2013; Lack and Langridge, 2013) and fine particle dust (Weingartner et al., 2003; Müller et al., 2011). However, the influence of brown carbon may be minimal at Alert as values of the Angstrom Absorption Exponent (AAE) are between 0.5 and 1.5 suggesting predominantly fossil fuel influence (see Supplemental section and Figure_Supplemental_1b). 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. Recently, Backman et al. (2017) proposed a reduction of a factor of 3.2 in the light absorption coefficients derived from the Aethalometer due to multiple scattering enhancements associated with particles collected on the filter. These enhancements are considered, at least in part, in the EBC estimate by the τ_{λ} value used with the Aethalometer, but there remains uncertainty in τ_{λ} , including the use of a constant value for all conditions. EBC (unmodified) needs to be evaluated due to these reasons in comparison to absolute measurements mass techniques.”

P17-18, Section 4.1.3 rBC You fit a single lognormal mode. Fine, I would also. However, do you have any info on whether there might be larger, coated BC particles? In all the work we have done with the SP2, I have not seen any data that suggests a larger mode of coated BC. P21, L7. MAAP measures at 637 nm, see Müller et al. See Massling et al., 2015 paper, they state that their aerosol light absorption coefficient was measured

at wavelength of 670 nm. Perhaps they made a mistake in their paper. Changing it to 637 nm in our paper on Pg 22, L9.

Table 2. In some columns there is lin.reg. Are the associated numbers slopes? Why don't you give the slope and offsets and their uncertainties – regression codes give them. Replaced in Table 2, Lin. Reg. with Lin. Reg. \pm std. err. and added slope and std error in the slope to the regressions. Also added in the text on P13, L25, "...intercepts and standard errors,...". Fig 6. The y axes start from < 0 which implies negative thickness. Can't be true. Made these changes to the caption text: "...Negative coating thickness indicates that the measured scattering for a particle is less than the Mie calculations predict for an rBC particle of that diameter with zero coating. These values arise from the inherent noise in the data, as well as assumptions made about the rBC and coating refractive indices, and particle morphology. "

Added this to the references. Anderson, T.L. and Ogren, J.A.: Determining aerosol radiative properties using TSI 3563 integrating nephelometer, *Aerosol Sci. Tech.*, 29(1), 57-69, 1998.

Please also note the supplement to this comment:

<https://www.atmos-chem-phys-discuss.net/acp-2017-339/acp-2017-339-AC1-supplement.pdf>

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