

Again, we thank the Reviewer for their constructive comments.

1) Reviewer Comment: The authors have still not included uncertainties in Fig. 8 on their individual MAC values. The absorption values associated with many of these data points are extremely close to the stated (estimated) detection limits (L159) of 0.75 1/Mm for 1 min averages. For example, [BC] = 0.04 ug/m³ and a BC MAC value of 25 m²/g (both within the range of measurements) the absorption would have been only 0.8 1/Mm. I have made this point previously, but the measurements lack error bars. Without error bars there can be no rigorous assessment of the relationship between the MAC and [BC]. The authors note that they have to throw out 90% of their data (L163) because it is below the detection limit. But I question whether the points here are even really above the detection limit. Or, more specifically, that their observations are not simply driven by noise. Further, because the authors filter for the detection limit this will introduce a negative slope because the detection limit for BC is lower than it is for absorption. A thought experiment. Given a normally distributed noise profile but a parameter that cannot physically be < 0 (such as is likely the case here), if noise dominates the observed variability for two parameters then the ratio between the larger of the two and the smaller of the two will increase as the smaller of the two parameters decreases. This can be shown using fictitious example data (see first figure below). Assume absorption = 1.4 1/Mm with a Gaussian noise profile with a FWHM = 0.4 and BC = 0.06 ug/m³ with a Gaussian noise profile with a FWHM = 0.02. Assume all variability is determined by noise. The ratio absorption/[BC] then has the following form (black points). This looks notably like the observations. If I then cut off all absorption measurements < 0.75 1/Mm the lower MAC values are cut off (red points). One can alternatively calculate a curve that represents the absorption-based threshold for determination of a given MAC value for a given [BC]. This curve is has the relationship MAC = 0.75/[BC], given the detection limit reported. I've overlain this on the authors figure (second figure below). This strongly suggests, at least to me, that the MAC vs. BC relationship is an artifact. I remain unconvinced that the variability in the authors MAC observations is not simply driven by noise and the fact that they are working close to their detection limit.

Response: The primary issue appears to be with Figure 8b. The Reviewer has asked for error bars, which we do not include, as that would lead to a visibly messy plot. Experimental error estimates are fully included in the methods, and do not need to be included in a plot. The experimental error that the Reviewer asks for is random, and it does not change any bias(es) in the data that results from unmeasured quantities or unconsidered processes. In addition, we present counter-arguments to the Reviewer's alternative suggestion for the interpretation of the curve in Figure 8b, which assumes our data are all noise close to the detection limit.

- a) The Reviewer's argument is based on the assumption that the absorption data are close to the detection limits and effectively "noise". This argument is partly based on the fact that we have eliminated approximately 90% of the absorption data. However, the largest reduction in the absorption data was due to the application of the pressure constraint, which was first applied, and not the detection limit. The pressure constraint is completely independent of the detection limit (DL). To clarify this, we have modified the sentence on lines 163-164 as follows: "... 220 absorption data points remain after scrutiny for pressure variations, followed by removal of remaining points below DL." Contrary to the Reviewer's comment that "many of these data points are extremely close to the stated (estimated) detection limits (L159) of 0.75 1/Mm for 1 min averages", 75% of the absorption data lie at or above 1.0 Mm⁻¹ and 30% lie at or above twice the DL (1.5 Mm⁻¹).
- b) Figure 5 clearly demonstrates a significant connection between absorption and BC that contradicts the Reviewer's suggestion that the data are simply "driven by noise". In that figure,

the absorption data are strongly associated with the rBC mass concentrations (the R^2 in Figure 5 is 0.88; if we only include the POLAR 6 data, the R^2 is 0.73). In other words, the absorption data are driven by process. Indeed, it is rare to see correlations this strong with data measured in remote conditions.

- c) The Reviewer states that “This curve is has the relationship $MAC = 0.75/[BC]$, given the detection limit reported.” In our original ACPD manuscript, we explained that the MAC values in Figure 8b should average to be constant across all BC if absorption is solely due to BC without enhancements, such as lensing. We further explained that the curves in Figure 5 have significant intercepts of 0.15 Mm^{-1} for the “POLAR 6 and Alert Obs.” points and 0.37 Mm^{-1} for only the POLAR 6 points (red circles in Figure 8b). As we stated in the manuscript, our reason for introducing Figure 8 was the small but significant intercept of the Absorption vs rBC points. Because of the intercept, a curve is derived that has MAC increasing with decreasing BC, as in Figure 8b. If there was no intercept, the average of the curve in Figure 8b would have no slope. With Figure 8 (both a and b), we attempt to address the potential impact on our average MAC as well as a possible origin of the intercept. It is reasonable that one possibility is the additional presence of a less efficient but more constant absorber (than BC) that has a smaller impact on the absorption at higher BC and a higher relative impact on MAC at lower BC, and that is shown by the model results in Figure 8a. In other words, the reduced SSA at lower scattering efficiencies could also be due to relatively small amounts of dust or other absorbing components beside BC that are difficult to measure. We only attempt to add another possibility to the objective assessment of the lower SSA at lower scattering efficiencies.
- d) The Reviewer states, “Further, because the authors filter for the detection limit this will introduce a negative slope because the detection limit for BC is lower than it is for absorption. A thought experiment.” How do we objectively assess that a detection limit is lower (or higher or similar) to another when the two quantities have completely different units? Even if we assume that the measurement of absorption is more often BDL (and we do not agree that it is) the data points are selected first based on the availability of absorption data. In other words, we do not artificially apply lower BC values, as the Reviewer seems to imply.
- e) The Reviewer questions our detection limits. To that point, we conducted several in-flight zeroes, for a number of reasons, one of which was to be able to estimate the DL for absorption. We note that our DL for absorption is significantly higher than those from others who report such DLs (e.g. McNaughton et al., 2011), and there are results in the literature for which absorption DLs are simply not reported (e.g. Brock et al., 2011).
- f) Overall, we agree with the reviewer that attention has to be paid to validate data with small signals. This was well aware to us, even prior to the campaigns, and it is one reason we conducted all the in-flight zeroes. For all the reasons presented above, we believe in the integrity of these data.

2) Reviewer Comment: L328: Fig. 9 does not show anything about coating thickness, as stated here. The figure shows rBC diameter vs. rBC concentration.

Response: Reference to Figure 9 has been removed.

3) Reviewer Comment: L335: Are the authors referring to Fig. 8a or Fig. 8b? It is not clear. If the former, the MAC does not “approach” the values from Fig. 5 at higher [BC]. If the latter, don’t the MAC values from Fig. 8b derive from Fig. 5, thus necessitating a relationship between the two? Regardless, it is not

clear to me what the authors' mean when they state "as in Fig. 6 for the model." Fig. 6 does not show MAC vs. BC.

Response: Figure 8 is correct, as we refer to both Figures 8a and 8b. The sentence has been modified to read: "At higher BC concentrations, the MAC-vs-BC curves in Fig. 8 approach the MAC values determined from Fig. 5 for the observations ($18.4 \text{ m}^2 \text{ g}^{-1}$) and from Fig. 6 for the model ($8.8 \text{ m}^2 \text{ g}^{-1}$ to $14.0 \text{ m}^2 \text{ g}^{-1}$).", where the blue highlights refer to the changes.

4) Reviewer Comment: L339: The authors now state that dust may have contributed 0.15-0.3 1/Mm absorption (there's a typo that gives the units as Mm^{-2}). At $[\text{BC}] \sim 0.04 \text{ ug}/\text{m}^3$ (from Fig. 8b) and with the low-dust "Allcore" and "Rshell" MAC values, the BC absorption should be 0.37 1/Mm or 0.52 1/Mm. The potential dust absorption is significant in this context and can explain much of the differences in modeled and measured MAC and the apparent increase in the MAC with decreasing BC. If dust absorption = 0.15 1/Mm the MAC attributed to BC would be too large by 30-40%. If dust absorption = 0.3 1/Mm, the MAC attributed to BC would be too large by 57-81%. I do not find that the authors have made convincing arguments that allow them to rule out dust as a potential bias as they do on L341.

Response: The typo has been corrected, thank you. The paragraph starting on Line 335, which contains the statements the Reviewer discusses in this comment, starts out by stating "At higher BC concentrations...". From Figure 5, the absorption values for "P6 (no dust; Alert and Eureka only) and Alert Obs." reach to about 3 Mm^{-1} . If we subtract 0.15 Mm^{-1} or 0.3 Mm^{-1} from all absorption values in Figure 5, we maintain the slope (18.4), and simply push the values through the origin, as expected. The MAC values do not decrease by as much as the Reviewer suggests. We are not attempting to underrepresent the potential impacts of dust. We removed absorption values more strongly influenced by dust by eliminating the Inuvik flights from the discussion.

5) Reviewer Comment: Fig. 11: There's an error in the legend, with the labels for the black and red absorption swapped. There is also a difference between the red curves in panel a and b, yet these should be identical.

Response: Thank you. These mistakes have been corrected.

6) Reviewer Comment: Fig. 8: The caption says that the dust concentrations are shown for when $[\text{Dust}] < 1.5 \text{ ug}/\text{m}^3$. However, there are points on the graph for which $[\text{Dust}] > 1.5 \text{ ug}/\text{m}^3$. And in Fig. 8b the difference between the black and red lines is not indicated.

Response: Thank you, these corrections have been made, as shown below.

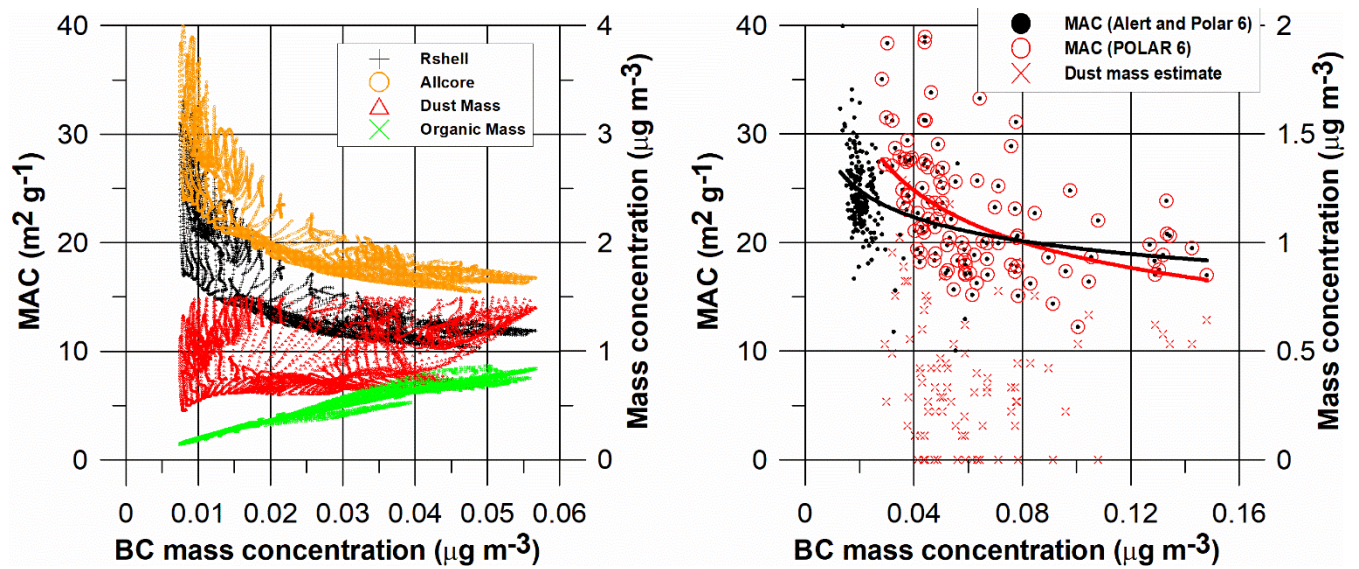


Figure 8. a) Modelled BC Mass Absorption Coefficient (MAC) plotted against modelled BC for the 'Allcore' (orange circles) and the 'Rshell' (black crosses) assumptions; modelled dust mass concentrations constrained to dust less than $1.5 \mu\text{g m}^{-3}$ versus modelled BC mass concentrations (red triangles); modelled organic aerosol (OA) mass concentrations versus BC mass concentrations (green crosses); all modelled values are for April 1-14, 2015. b) MAC values from POLAR 6 flights and Alert Observatory (April 1-14) plotted versus measured refractory black carbon (rBC) mass concentrations (black dots); dust mass concentrations (red crosses), estimated from particle size distributions onboard the POLAR 6, plotted versus rBC mass concentrations; MAC values associated with POLAR 6 measurements only identified (red circles). In b), the black line is a power-law fit to the black points, and the red line is a power-law fit to the red circles. Power-law fits were chosen since, based on the linear fit between absorption and rBC (Fig. 5), the expectation is that MAC will vary inversely with the BC mass concentration. The confidence level in the negative slopes is greater than 99%.