Round four responses to Reviewer

Reviewer comments:

I disagree with their interpretation that "For the Reviewer's argument to be valid, all absorption data in Figure 5 must be noise, however, the Reviewer did not criticize Figure 5." This is not true. For my argument to hold the *variability* around the line is driven by noise. This is not the same as all the absorption data being noise or the overall trend being entirely noise; if this were true Fig. 5 would just be a blob of points. Clearly it is not and clearly there is a relationship between absorption and BC. I have never questioned that. My criticism is that they show the MAC increasing with decreasing [BC], and I believe this is an artifact.

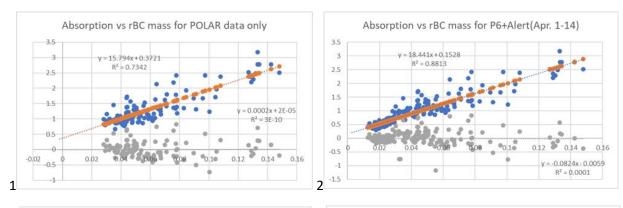
To the extent that Fig. 8 is just a transformation of Fig. 5 (which it is), my comments apply to both. But specifically my issue is with the interpretation that results from their presentation of this data in Fig. 8 and the fact that they have just deleted points below the detection limit, using a detection limit based on only one of the variables. I'll note that if they applied an equivalent threshold to the BC data then the increase in the MAC will likely go away. They mention the positive intercept; I'll note that the thought experiment I did included the positive intercept. The way they treat their values below the detection limit is what is driving the upward trend, I believe, not the positive intercept. Although now that I've looked again, there is an issue with Fig. 5, which is that the line shown for the fit to P6 (dust removed) does not actually have an intercept of 0.35; it appears to have been forced through zero.

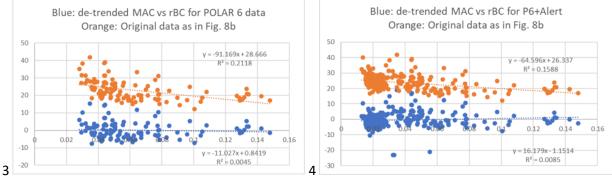
There are a wide range of opinions on how to treat data below the detection limit when performing statistical analysis. However, about the one thing that is never recommended is simply deleting values. While any method to fill in values below the detection limit will introduce some bias, deleting the values can dramatically skew results. This is especially true when one takes a ratio, but then applies a screening based on only one of the variables.

Responses:

- Concerning Figure 5: The intercept of the linear regression for the "P6 (dust removed)" curve was mistakenly labelled as 0.35, when it is actually 0.035. I thank the Reviewer for pointing out the issue with Figure 5. I apologize to both the Reviewer and the Editor for this and too many other little mistakes that were present in the paper.
- 2) The mistake in one of the intercepts in Figure 5 does not change the result shown in Figure 8b, since Figure 8b uses the points representing "P6 (no dust; Alert and Eureka only) and Alert Obs. (Apr. 1-14)". For the data shown in Figure 8b, the intercept from Figure 5 is 0.15, and statistically it is greater than 0.10 at a confidence level of greater than 99%. Above, the Reviewer makes two claims: 1) "For my argument to hold the *variability* around the line is driven by noise"; 2) the way the authors treat the data below detection limit is responsible for the upward trend in the MAC values with decreasing BC mass. We have a relatively high DL for the data from the POLAR 6 measurements, but the DL for the Alert Observatory data is much smaller, due to the more stable environment in which the measurement is conducted. One of the main reasons the Alert data are included is to take advantage of the lower DL to account for smaller absorption (and rBC) values. These values were and remain included in Figure 8b. In an attempt to consider the noise issue described by the Reviewer, I de-trended the absorption data using the linear regressions, in order to see how the de-trended absorption varies with rBC. I did this for the POLAR 6 data only, using its regression of Abs=15.79*rBC+0.372 (not shown in

the manuscript), and I did it for the combined POLAR 6 plus Alert (April 1-14) data, using the regression of Abs=18.44*rBC+0.153 shown in Figure 5. Plots are shown below. The first and second plots are of absorption vs rBC for the POLAR 6 data only and the POLAR 6 plus Alert (Apr. 1-14) data, respectively. The grey points in the first two plots are the de-trended absorption values. The third and fourth plots are the corresponding MAC values, based on the de-trended absorption data. Linear fits are included for all as reference. There is a suggestion of an increase in MAC with decreasing rBC in the POLAR 6 only plot, but it is relatively small compared with the result in Figure 8b, and any such suggestion is eliminated by the addition of the Alert data (plot 2). If the de-trending curve is based on a fit forced through zero, then a more significant increase in MAC with decreasing rBC results, because the fit is pulled down increasingly relative to the data points at lower rBC. However, the best fit does not go through zero. This approach indicates that the increasing tendency in MAC with decreasing rBC shown in Figure 8b is mostly the result of the intercept.





3) In the changes below, I acknowledge that BDL values, mostly at lower rBC mass concentrations, should be carefully considered as well as the significance of the intercept. One of the points of the paper is to examine why the SSA decreases at lower scattering values. Most speculation about this goes to relative precipitation scavenging efficiencies of BC, but the suggestion that the presence of other absorbing components may have a relatively greater impact as BC decreases is another and reasonable factor to consider.

The following changes have been made to the manuscript:

- 1) Correct the intercept of the "P6 (dust removed)" linear fit from 0.35 to 0.035.
- 2) In Figure 8b, change the x-axis to read "rBC" rather than "BC".

- 3) Also, in Figure 8b, remove the curve fit to the POLAR 6 data only, leaving just the fit to the combined POLAR 6 and Alert (Apr. 1-14) data.
- 4) Re-write the paragraph starting at line 317 as follows: "The σ_{ap} -rBC regressions of the observations and the simulations have positive intercepts (the standard error of the intercept for the POLAR 6 plus Alert (Apr. 1-14) fit, based on the measurement error, is 0.034, and statistically the intercept is greater than 0.1 at a confidence level of >99%). The intercept is due to increases in individual MAC values with decreasing concentrations of BC, as shown in Fig. 8a for the model and Fig. 8b for the observations. In these plots, MAC should be constant across all BC if absorption is solely due to BC without enhancements from factors such as lensing or other absorbing components. The relative increases in absorption at lower BC will contribute to reduced SSA values at lower σ_{sp} . The persistence of lower dust concentrations at low BC concentrations in the modelled MAC values (Fig. 8a) likely contributed to the increase in MAC at lower BC: as the BC concentrations decrease up to 100 times below the dust concentrations, absorption by dust with its lower imaginary refractive index (0.0065 in the model) may approach absorption by BC and increase the apparent MAC for BC. Because the modelled organic aerosol (OA) concentrations decrease proportionately with decreasing BC (Fig. 10 and 15) and the imaginary refractive index is lower (0.0065), absorbing OA cannot explain the increase in the simulated MAC at lower BC. For the observations (Fig. 8b), smaller amounts of potentially absorbing dust also may be present at lower BC concentrations. The mass concentrations of coarse particles, estimated from the POLAR 6 size distributions assuming a density of 2 g cm⁻³, are present across all rBC concentrations. We cannot distinguish whether the composition of these coarse particles is dust or sea salt, but we cannot rule out smaller amounts of dust as a factor in the increasing MAC value at lower BC concentrations. Also, factors that might enhance absorption by BC cannot be ruled out: for example, a relative increase in the coating thickness surrounding smaller BC cores at lower BC mass concentrations might contribute to an increased enhancement factor. We add that the absolute absorption values at lower BC are small, and there is substantial uncertainty associated with the large amount of absorption data from the POLAR 6 that lie below the detection limit. Despite the significance of the intercept for the POLAR 6 plus Alert (Apr. 1-14) data (Fig. 5), it is an extrapolation based on the assumption that the absorption data below detection limit follow the same linear fit."