Reviewer Comment: I appreciate the effort the authors have put in to their revisions, including the additional figures and calculations. However, I believe a number of concerns remain regarding the authors interpretation and some of their new results that must be resolved prior to publication. My comments follow.

Author Response: We are grateful to the Reviewer for their persistence and very helpful comments.

Reviewer Comment: I appreciate that the authors have added results using a more realistic refractive index for BC. However, I still have strong concerns about how they present their results in the context of the GADS RI's that are used. The discussion of MAC's in Section 3.2 uses only the GADS RI values. The maximum MAC for bare BC when an RI of 1.75 + 0.45i is used 5.1 m2/g, assuming a material density of 1.8. This is substantially lower than the 7.5 m2/g that the authors refer to from Bond et al. for "freshly emitted BC." Thus, when the authors state their "Allcore" results for Axel are "are more consistent with the MAC value estimated for "freshly emitted" BC this is, in my opinion, an apples and oranges comparison. A calculated MAC of 7 m2/g using the GADS RI values implies an enhancement of at least 7/5.1 = 1.37 and for the NW Alert values the modeled enhancement must be at least 1.86. Applying these enhancements, which are much less sensitive to the BC RI used, to the "freshly emitted BC" implies a much larger MAC for the model if more appropriate RI values had been used. I believe that such issues need to be factored in to a much greater extent when interpreting the increase in the calculated MAC with decreasing BC mass concentration (around L315). Given the BC size distributions used, it is possible to calculate a maximum bound on the "lensing" enhancement (assuming a nonabsorbing coating). This can then allow for bounds to be placed on the dust contribution, or on the contribution from OA. The discussion here is very speculative, but it should be made more precise. The authors can disentangle the contributions from lensing, absorption by the organics (since these are slightly absorbing in the model) and absorption by dust. To a reasonable extent, their dust concentrations are relatively constant with BC mass, around 0.8 ug/m3 or so. This, given the RI reported, corresponds to an MAC of around 0.2 m2/g for dust. Given 0.8 ug/m3 and this MAC, the calculated absorption by dust is around 0.16 m2/g. It is then trivial to show that the calculated MAC should increase with decreasing [BC] simply due to dust contributions. In fact, I suspect that the modeled MAC co-varies with the absolute dust contribution in the model (or even better, the dust/BC ratio). All this is to say that there seems no reason for the authors to be so speculative here, or to invoke unproven increases in lensing with decreasing [BC]. As the authors have a model, they can attribute absorption specifically to dust or BC, and I suggest they do so for this discussion. I'll also note that the authors statement on L524 that dust is not a factor at low BC does not seem to hold up, in light of the model behavior. Dust is most certainly a contributing factor.

Author response: We found an error in the model absorption data for the index of refraction of 1.75+0.45i. It had been twice adjusted to STP, which is the reason why the absorption values for that RI were higher than the Bond RI at higher altitudes. Subsequently, we decided simply to use only the modelled absorption and scattering based on the Bond RI, and have removed reference to the 1.75+0.45i, except on line 251 where we mention the difference between the present simulations and those used on Kodros et al. (2018).

We re-plotted Figure 6 based on the Bond RI, but showing only one case (NW Alert, Rshell). We modified the discussion in Section 3.2 (Lines 301-308 of this second revision) to discuss the revised MAC values from the model cases based on constraining the dust content to less than 0.2 mg m^{-3}, as follows: "Modelled σ_{ap} are plotted against modelled BC for the NW Alert grid and Rshell mixing assumption in Fig. 6. These modelled values include a significant dust presence in many cases. We estimate the model MAC values for BC by constraining the dust concentrations to less than 0.2 $\mu g\ m^{-3}$, as shown by the black

points in Fig. 6. For the case in Fig. 6, the MAC value for BC is 9.3 $m^2 g^{-1}$. The modelled MAC value for BC is estimated at 13.1 $m^2 g^{-1}$ for the NW Alert grid and Allcore mixing assumption. For the Axel grid, the estimated MAC value for BC is 8.8 $m^2 g^{-1}$ for the Rshell assumption and 14.0 $m^2 g^{-1}$ for the Allcore assumption. Due to a smaller influence of the coating material on absorption enhancement relative to the Allcore case, the lower Rshell results in a MAC value more consistent with those mentioned above." We have eliminated any discussion of a MAC value associated with freshly-emitted BC.

We do not intend to suggest in this paper that dust is unimportant. To that end, and following your suggestion to "disentangle" the contributions, we added a single figure (revised Figure 7) showing the contributions to absorption from BC and non-BC absorbing components of the particles, split between less than 700 nm and greater than 700 nm. We discuss this in a short paragraph covering lines 309-315 of this second revision. Revised Figure 7 and the accompanying paragraph are as follows:

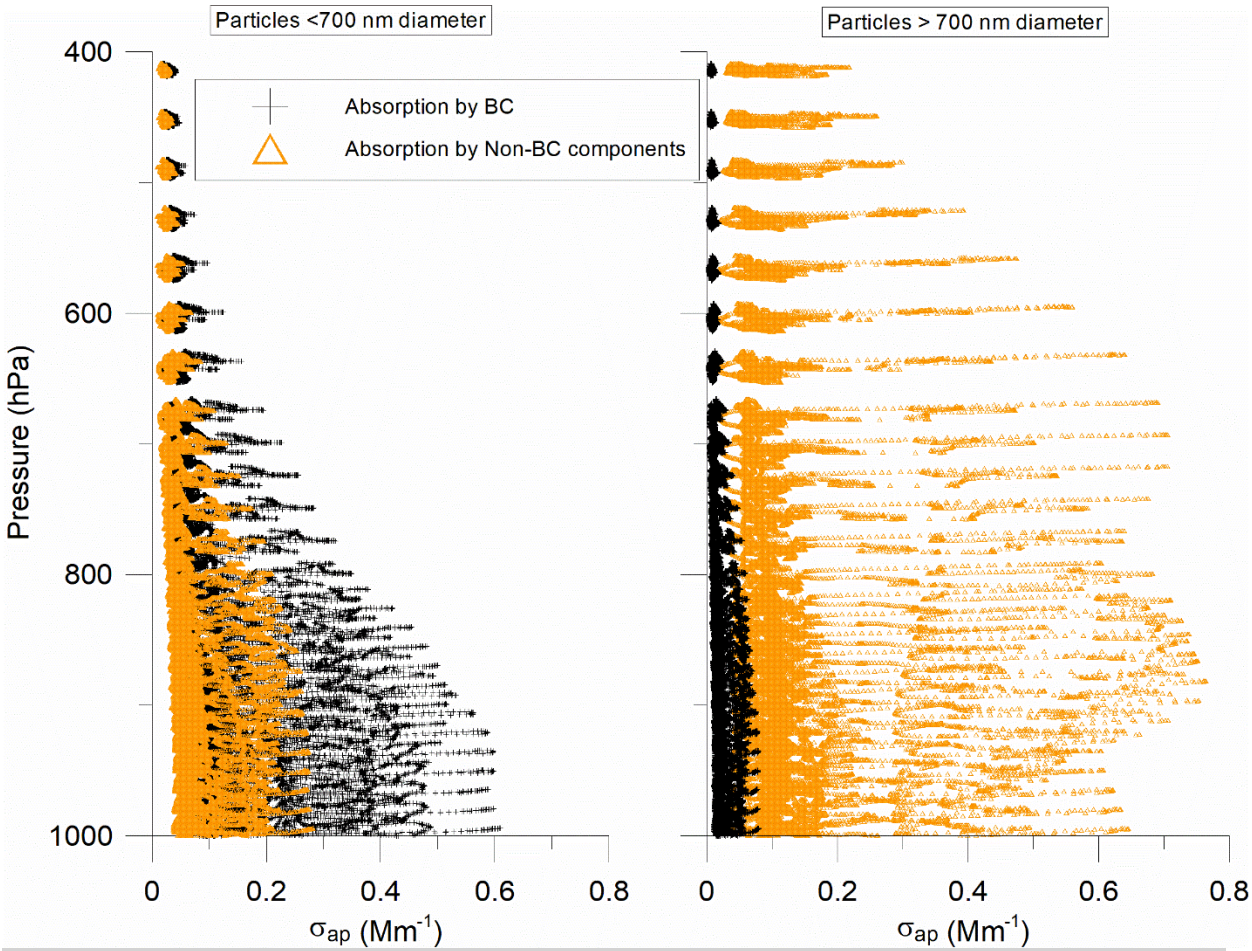

Figure 7. Modelled profile data of the contributions to absorption from BC (black crosses) and non-BC absorbing components (orange triangles) for a) particles less than 700 nm diameter and b) particles >700 nm diameter. The simulations are for the NW Alert grid box and based on an external mixture.

New paragraph (lines 309-315): "We acknowledge that the model indicates a significant contribution from dust and possibly other non-BC components to atmospheric absorption in the Arctic. Average modelled profiles of the contributions to absorption from BC and the non-BC absorbing components of

particles, assuming an external mixture, are shown in Fig. 7 for the NW Alert grid box, split between particles less than 700 nm diameter and particles greater than 700 nm diameter. The non-absorbing components include brown carbon and dust. For particles smaller than 700 nm, the simulations suggest BC is on average the stronger absorbing component of the particles, with contributions from non-BC components approaching those from BC below 600 hPA. For particles larger than 700 nm, absorption by non-BC components, primarily dust, dominates in this model. As shown in Fig. 3, there were relatively few coarse particles were sampled from the POLAR 6 in the regions around Alert and Eureka, at any altitude. This is a discrepancy between the observations and simulations."

Further, with the Bond RI, we remove the $\sigma_{abs}/2.62$ results, and we now compare more simply with σ_{abs} and $\sigma_{abs}/2$.

Reviewer Comment: Associated, I do not agree that the circled data points in Fig. 7 show "nothing other than an increase in MAC with decreasing BC" (L329). As the authors have no error bars on these numbers it is not possible to know whether this is simply noise (as I suspect it is) or some real trend. Also, I suggest that the authors distinguish between the POLAR 6 and Alert measureements in Fig. 7b, as they do in other figures. It seems evident, based on comparison with Fig. 5, that the cluster of points at low BC are all from Alert while the points at higher BC (including the "zero dust" points) are all from POLAR 6, and from Fig. 5 it seems that the precision (single point uncertainty) is much lower for the POLAR 6 data.

Author Response: We have now identified the Alert and POLAR 6 data separately in this figure (now Fig. 8; previously Fig. 7). The slope of the curve is not driven by the presence of the Alert points relative to the POLAR 6 points: the slope of the POLAR 6 data is very slightly more negative than the slope of the curve that includes the Alert points, and statistically the confidence level that the slope of the 108 POLAR 6 data points is negative is better than 99.9%. Based on the reviewer's comments concerning the significance of dust, we have removed the 'zero dust' point curve, and we have revised the discussion as follows:

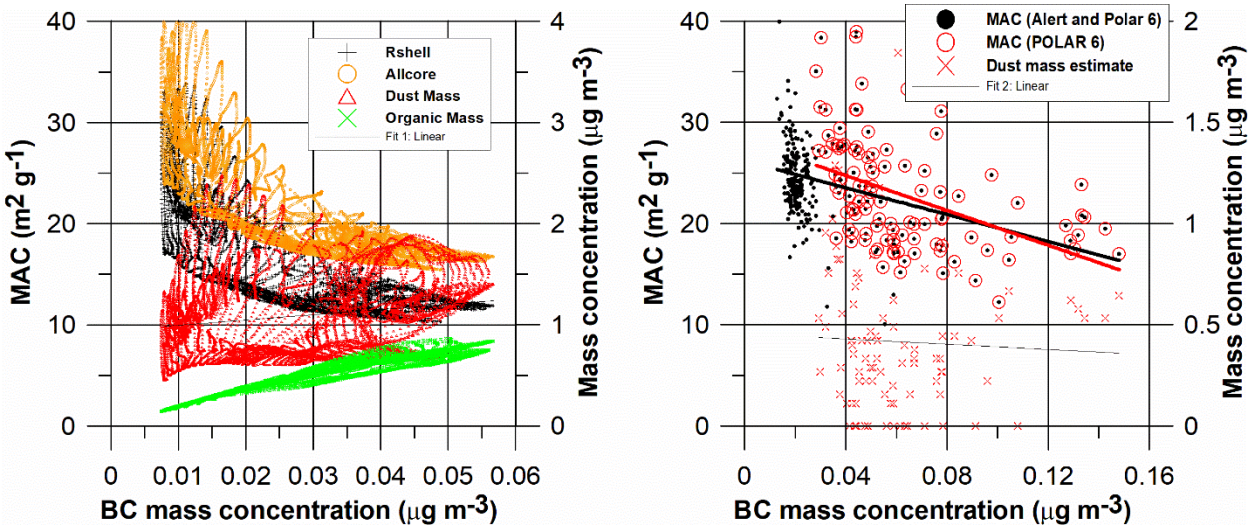

Figure 8. a) Modelled BC Mass Absorption Coefficient (MAC) plotted against modelled BC for the 'Allcore' (orange circles) and the 'Rshell" (black crosses) assumptions; modelled dust mass concentrations constrained to dust less than 1.5 μg m^{-3} versus modelled BC mass concentrations (red triangles); 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). The confidence level in the negative slope of the black points is greater than 99%.

"The σ_{ap}-rBC regressions of the observations and the simulations have positive intercepts (regression-based confidence levels exceed 99.9%; the standard error of the intercepts, based on the measurement error, is 0.034). The intercepts are the result of increases in individual MAC values with decreasing concentrations of BC, as shown in Fig. 8a for the model and 8b for the observations. In these plots, MAC should be constant across all BC if absorption is solely due to BC without enhancements, such as lensing. The persistence of 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. 8a) and the imaginary refractive index is lower (0.0065), absorbing OA cannot explain the increased MAC at lower BC. A relative increase in the coating thickness surrounding smaller BC cores at lower BC mass concentrations could also contribute to an increased enhancement factor. The observations suggest the presence of smaller amounts of potentially absorbing dust may be present at lower BC concentrations (Fig. 8b). 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, in accordance with the model, the observations suggest that dust may be a factor in the increasing MAC value at lower BC concentrations.

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 m^2 g^{-1}) and as in Fig. 6 for the model (8.8 m^2 g^{-1} to 14.0 m^2 g^{-1}). Reasons for the higher observation-based MAC, based on the regression in Fig. 5, are unclear. There is no indication from the blue versus green slopes (Fig. 2) to suggest brown carbon is a significant factor for the Alert and Eureka data. Based on Figures 5-8, we estimate that dust may have contributed to the absorption in the amount of 0.15-0.3 Mm^{-2}. This estimate is significant, but not nearly sufficient to explain the differences in observed and modelled MAC. We consider three additional possibilities for the higher MAC:"

Reviewer Comment: Fig. 10: I appreciate the authors have added calculations using a more realistic RI value for BC. However, I find the new results somewhat confusing. Below is a plot of the calculated MAC versus size for BC assuming either the GADS RI or Bond et al. RI. The Bond et al. values are larger for nearly every size. This remains true if one accounts for coatings. However, in Fig. 10 there are a number of altitudes where the GADS results yield greater absorption than the Bond results. This is not true for all altitudes, however. It is important that these differences in model configurations/results are explained more thoroughly than is done currently. Why should the absorption be lower for the GADS RI values? Did the authors, perhaps, also make some different assumption regarding the OA RI value?

Author Response: As mentioned above, we mistakenly corrected the GADS RI absorption values twice for pressure and temperature. As also mentioned above, we have dispensed with the GADS RI, and now show only the results for the Bond RI.

Reviewer Comment: L333: The authors now cite Yu et al. (2019) as showing that absorption is increased by 3-16 times in the Arctic. This is a misunderstanding of the Yu et al. paper, which makes various unconstrained assumptions regarding the optical properties of the coating material. Only when they assume the coatings are notably absorbing do they get the largest enhancements. This important aspect is not evident in what the authors have written here. The Yu et al. paper is ultimately just a Mie theory-based calculation exercise using as a constraint observed coating-to-core ratios. Associated, I do not think the citation of Yu et al. (2019) on L341 is justified. Yu et al did not actually observe enhancements, and the authors here seem to misunderstand what Yu et al. showed.

Author Response: As above, we have removed this discussion of Yu et al. on the previous L333. As the Reviewer indicates, this Yu reference is inappropriate. We were mistaken in its use. In the section previously on L341, now L344, we reference Wu et al. (2016) that shows absorption due to the coating of soot particle aggregates can be about 60% higher than absorption by uncoated soot particles. The section has been adjusted as follows:
"The higher MAC is reasonable. This is suggested by the recent observations in smoke plumes and in background particles over the continental U.S.A. (Mason et al., 2018) as well as by Wu et al (2016) whose modelling of soot aggregates shows absorption by heavily-coated soot can be about 60% higher than uncoated soot. Another factor suggesting the higher MAC is reasonable is the close agreement between the present σ_{ap} and SSA for Alert with the April summary of Schmeisser et al. (2018) for Alert: the σ_{ap} are 0.50 Mm^{-1} and 0.45 Mm^{-1} for the present Alert analysis and for Schmeisser et al. (2018), respectively; the SSA are 0.95 and 0.95, respectively."

Reviewer Comment: L373: The deficiency in rBC mass concentrations would be a function of the particle size distribution, with a larger bias likely when the mass mode diameter is smaller and closer to the 85 nm threshold. The 7.5% cited here assumes a constant bias, but the authors should consider that the bias varies dependent on the mean particle size.

Author Response: Thank you, we have revised lines 372-374 to read: "Schulz et al. (2019) estimate the deficiency in the POLAR 6 rBC mass concentrations due to sizing limitations at 7.5%, but they did not consider potential bias with respect to particle size. Accounting for size limitations of the rBC measurement at Alert increased the BC estimate by 40-50% during the spring measurement period (Sharma et al., 2017); although the resulting rBC was still a factor of 1.9 lower than the EC measurement."

Reviewer Comment: Response 30 and Figs. 12 and 13: The authors have added a comparison of size distributions, as suggested, as support for their contention that differences in the scattering efficiency between model and measurement at the lower layers might result from model/measurement size differences. I appreciate this addition, however I am not convinced by the argument here. The authors' state that "Model underestimation of submicron particle sizes may contribute to the lower modelled volume scattering efficiencies" (L473) but then a few sentences later that "The modelled distributions…are shifted to slightly lower sizes relative to the average of the observations." These points seem to me to conflict.

Author Response: We think these statements are consistent. The first says that the lower modelled scattering efficiencies may be due to the model simulating particles somewhat smaller than the observations. The second statement indicates that the modelled sizes are slightly lower.

Reviewer Comment: The authors now mention that the sampled air stream warms significantly (by up to 50 deg C). Are the authors concerned that this might impart some bias on the measurements of the more volatile particle components?

Author Comment: It is always a concern. In this case, we feel that the rapidity at which the particles entered the cabin and were sampled more likely maintained the integrity of the organic components, particularly if the OA components were in a glassy state in the ambient. In Leaitch et al. (ACP, 2018), we sampled for the organic aerosol on filters both inside and outside the Alert Observatory, each averaged over equivalent weekly periods. The filters that were sampled outside at the much colder ambient temperature were overall found to have less OA than those sampled inside. Unfortunately, because the inside and outside filters were analyzed by different techniques, we cannot conclude anything about the volatilization issue from that study.

Reviewer Comment: The caption for Fig. 6 is incomplete, not referencing panel b.

Author Comment: The caption has been revised.

Reviewer Comment: Fig. 7: What do the authors mean by "confidence level" for the slopes. Is this from a t-test? I am familiar with confidence intervals, but this is not the same as a confidence level.

Author Comment: Confidence "level" is appropriate here. Based on a linear regression, we are referring to the probability of the slope being negative is at least 99%.