Responses to Reviewer 1

We appreciate the time and effort of the Reviewer, and we thank the Reviewer for their thorough and constructive comments that help us improve the paper. Specific responses follow.

1. Review comment – "L100: An alternative explanation here is that the particle size varies with the scattering coefficient, leading to changes in the SSA independent of any changes in absorption per particle. The authors should consider alternative explanations, not just one explanation."

Response – Our statement is that the "absorption per particle is on average greater at lower σ_{sp}". In saying that, we neither state nor assume that each individual particle changes to increase the absorption per particle. As discussed later in the paper, we believe that the reductions in SSA associated with lower scattering are indeed related to changes in the size distribution with specific emphasis on how wet deposition impacts this relationship, as suggested by the references to Targino et al. and Andrews et al. We believe this statement is relevant for Arctic haze particles, but we agree with the Reviewer that it may not be generally correct, and we have revised the sentence as follows: "Further, the SSA for particle populations that fall within this constraint have been found to decrease more sharply with decreasing σ_{sp} (e.g. Targino et al., 100 2005; Andrews et al., 2011), making these populations more efficient at warming of the atmosphere".

2. Review comment – "Section 2.2: Not discussed here throughly, or in the cited Sharma et al. (2017) paper, is the potential for positive biases in the CLAP measurements due to non-absorbing particles that are not accurately accounted for by the multiple scattering correction, as discussed by (Cappa et al., 2008;Lack et al., 2008). Some discussion of how such potential biases were considered would be welcome, beyond the brief, albeit incorrect, mention on L323. If the authors believe such potential biases to not have impacted their measurements, convincing discussion to this effect would be helpful."

Response – We agree that the potential for bias in absorption is significant, and that is the reason that we considered two possibilities based on the MAC value. The results adjusted to the more conventional MAC value (approximately 9 m2/g; i.e. $\sigma_{abs}/2$) represent the assumption that absorption is overestimated by the techniques used here. Also, we agree with the Reviewer that we have not thoroughly discussed previous work in this area. To correct this inadequacy, we now reference Cappa et al. (AST, 2008a; AST, 2008b) and Lack et al. (2008) in Sections 2.2 and Cappa et al. (AST, 2008a; AST, 2008b and JGR, 2019), in addition to Lack et al. (2008) in Section 3.2. Further we have significantly modified (added to) the discussion in the fourth paragraph of Section 3.2 to better reflect these and other references. The revised paragraph follows:

"The MAC-vs-BC curves in Fig. 7a and 7b have asymptotes in line with the MAC values determined from Fig. 5 and 6. 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, and dust will not explain the differences in observed and modelled MAC. We consider three possibilities for the higher MAC:

1) 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), and by Yu et al (2019) as discussed above. 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.

2) The higher observed MAC results from the overestimation of absorption by the filter-based measurements, as suggested by a number of observations. Lack et al. (2008) and Cappa et al. (2008a; 2008b) found that σ_{abs} measured with a PSAP ranged from 1.3 to over two times higher than σ_{abs} measured with a photoacoustic technique for organic mass (OM) concentrations above 2.5 $\mu g\ m^{-3}$ and the ratio of OM to the light absorbing component of the carbonaceous components exceeded about 15. To explain the absence of a similar effect on their results by ammonium sulphate particles, they suggested that the liquid nature of non-absorbing OM enhanced multiple scattering across the filter, further increasing absorption by particles on the filter. For the present measurements of rBC and OM (see Fig. 8 and 12), the ratio of OM to rBC (the only significant absorbing carbon) is estimated to be between 15 and 20, consistent with a strong impact of OM on our results. However, Lack et al. (2008) and Cappa et al. (2008a; 2008b) found that for relatively low (OM), the PSAP-based σ_{abs} was only 12% higher than the photoacoustic-based σ_{abs}. At 1 $\mu g\ m^{-3}$ or less (Fig. 12), our flight OM concentrations fall well into the low OM concentration range of Cappa et al. (2008a; 2008b) and Lack et al. (2008). Also, consistent with the lowest level OM in Fig. 12, three years of OM measurements at Alert found OM always less than 0.5 $\mu g\ m^{-3}$ (Leaitch et al., 2018), suggesting that the average impact of this factor on our σ_{abs} may be in the area of 12%. Considering the ambient temperatures (-40°C to -15°C), it is also possible that our non-absorbing OM was in solid forms (e.g. Zobrist et al., 2008), which might render its behavior on the filter more similar to ammonium sulphate. Sinha et al. (2017) studied the effect of volatile material (removed at 300°C) on absorption by particles at two Arctic sites. On average and for particles less than 1 μm, they found that the absorption given by the PSAP was reduced by 22% with the removal of volatile material, which suggests that the present correction based on σ_{scat} from the nephelometer is deficient.

3) The BC is underestimated here using rBC. Sharma et al. (2017) found that filter-based thermo-optical measurements of elemental carbon were an average of 1.9 times higher than rBC measured at Alert. This result will explain the high MAC value of 18.4 m^2g^{-1}, but there are many uncertainties associated with the measurement of BC by the many techniques, and the rBC measurement has been recommended for use (Bond et al., 2013). The size limitation of the rBC measurement was taken into account by Sharma et al. (2017), but not in the POLAR 6 dataset. Schulz et al. (2019) estimate the deficiency in the POLAR 6 rBC mass concentrations due to sizing limitations at 7.5%.

If we assume that our observed σ_{abs} are overestimated by 22% and our rBC are underestimated by 7.5%, the MAC value is reduced from 18.4 m^2g^{-1} to about 13.4 m^2g^{-1}, which is about halfway between our measured value and the more commonly accepted value in the area of 9 m^2g^{-1}. However, because there are a number of potential factors influencing the measurements of both σ_{abs} and BC, we cannot attribute one value with the necessary certainty. For that reason, we employ a range for σ_{abs} by adding to our discussion the POLAR 6 plus Alert (Apr. 1-14) observations (Fig. 5) adjusted to MAC values of 9.2 $m^2\ g^{-1}$ and 7.0 $m^2\ g^{-1}$, respectively, through division of the σ_{ap} by factors of 2 and 2.62. The 9.2 $m^2\ g^{-1}$ is derived from an average of the three MAC values referenced above and the two modelled grids for the Allcore assumption, while the 7.0 $m^2\ g^{-1}$ is an average of the two modelled grids for the Rshell assumption."

3. Review Comment – "SP2: Schulz et al. (2019) give the lower limit as 85 nm (not 75 nm). Also, it is not clear whether the Aquadag or Fullerene soot calibration was ultimately used. The authors mention both, but it seems as if only the Aquadag was used, although Schulz et al. (2018) mention only Fullerene."

Response – We thank the Reviewer for pointing out these discrepancies. The lower limit here is indeed 85 nm and has been corrected accordingly. We apologise for the confusion over the calibration source. By way of explanation, there were two SP2 instruments used on the POLAR 6. One was used for special studies, and the other was used for the general dataset. The general dataset used in the present paper is the same as described by Schulz et al. (2019), and Fullerene soot was the main calibration source as described in that reference. In addition, an intercomparison of the two SP2s and the SP-AMS was done in Bremerhaven during the aircraft integration, and Aquadag, as well as Fullerene, was used in that intercomparison. The present dataset uses only the data used in Schulz et al. (2019). The discussion of the SP2 in Section 2.3 has been modified to read as follows:

"Refractory black carbon (rBC) was measured on the POLAR 6 using a Droplet Measurement Technologies Inc. Single Particle Soot Photometer (SP2). The SP2 detects individual particles using an intra-cavity Nd:YAG laser operating at 1064 nm. Incandescence from components of particles absorbing at 1064 nm (i.e. BC) is detected by a pair of photomultiplier tubes, and the peak amplitude of the thermal radiation is proportional to the mass of refractory material (Moteki and Kondo, 2007; Slowik et al., 2007). The detection range of the SP2 used here is 0.60 fg rBC to 330 fg rBC, or approximately 85-700 nm mass equivalent diameter for a rBC density of 1.8 g cm^{-3} (Bond and Bergstrom, 2006; Bond et al., 2013). Mass calibrations are based on Fullerene soot particles that were size selected using a differential mobility analyser. The mobility diameters were converted to rBC mass concentrations following Gysel et al. (2011). Schulz et al. (2018) describe the dataset used here, and they estimate the uncertainty at ±15%. The rBC measurements conducted at Alert, also made using a SP2, are discussed by Sharma et al. (2017)."

4. Review Comment – "L191: It would be helpful if the averaging time for these detection limits were given. Also, given the various diameters involved, it would be helpful if the authors reported that these were vacuum aerodynamic diameters (if they were). Same for the SP2: these should be indicated as volume-equivalent diameters."

Response – The averaging time of 10 s has been added, and the diameter indicated as VAD. As in the above paragraph, the SP2 diameters are now referred to as mass equivalent diameters. The AMS paragraph now reads: "Non-refractory aerosol mass concentrations were measured aboard POLAR 6 with an Aerodyne High Resolution Time-of-Flight Aerosol Mass Spectrometer (ToF-AMS) (DeCarlo et al., 2006). These measurements are described in detail by Willis et al. (2019). Briefly, the ToF-AMS measured non-refractory aerosol between about 70 – 700 nm vacuum aerodynamic diameter. Detection limits for sulfate and total organic aerosol, based on 10 s time resolution, were 0.01 μg/m3 and 0.08 μg/m3, respectively."

5. Review Comment – "L215: A reference for GADS is needed. I am also generally concerned about the use of GADS (and therefore OPAC) for the calculation of BC absorption. Unless OPAC has been updated (and the ftp server seems to be down so it is difficult to know), the RI for BC in GADS is 1.75 + 0.44i. This imaginary

coefficient is known to be too low and to give too low of absorption, in general. Stier et al. (2007) performed a very nice sensitivity study and found the OPAC values to give generally much lower absorption than when other, more reasonable, RI values are observed. The authors should address this issue. Simply stating that "The imaginary part of the index for BC is lower than some estimates," is insufficient. Additionally, the references given here seem almost arbitrarily chosen. The authors include in their abstract a conclusion that use of a "low imaginary" RI might explain some of the results. But they do not seem to have done anything to address this known issue, instead using a default value with known problems. I think this paper would be much stronger if they directly addressed this issue and, better yet, worked to improve on it."

Response – As shown in a later response, the following reference for GADS has been added: (Kopke et al., 1997). Additional model runs were conducted with a refractive index (RI) of 1.9+0.79i, as recommended by Bond and Bergman (2006). Profiles of the modelled absorption coefficients with this RI are added to Figure 10, shown below. In the text, the following statements (revised line numbers) have been added:

- Lines 237-240 - "Because the imaginary part of the index for BC is lower than some estimates (e.g. 0.65i from Pluchino et al., 1980; Kim et al., 2015), model simulations were also conducted with a refractive index of 1.9+0.79i, as recommended by Bond and Bergstrom (2006). See Liu et al. (2020) for a review of the refractive index of BC."
- Lines – 403-406 - "Modelled profiles of σ_{ap} for April 1-14 and both the Axel and NW Alert grids are shown in Fig. 10a and 10c for the Allcore assumption and in Fig. 10b and 10d for the Rshell assumption; major dust influence is removed, as shown as in Fig. 6a and 6b. The modelled results in Fig. 10a and 10b are for a BC refractive index of 1.75+0.45i. Results for a BC refractive index of 1.9+0.95i are shown in Fig. 10c and 10d."
- Lines – 412-415 - The modelled σ_{ap} for the index with the higher imaginary part are slightly lower, particularly in the 400-600 hPa region. Overall, differences in the modelled σ_{ap} for the two refractive indices are small compared with the estimated range of the observations, and for that reason only the results for the refractive index of 1.75+0.45i are used in the SSA analysis that follows.
- Line 422 – "Only results for the index of refraction of 1.75+0.45i are shown, as discussed above."

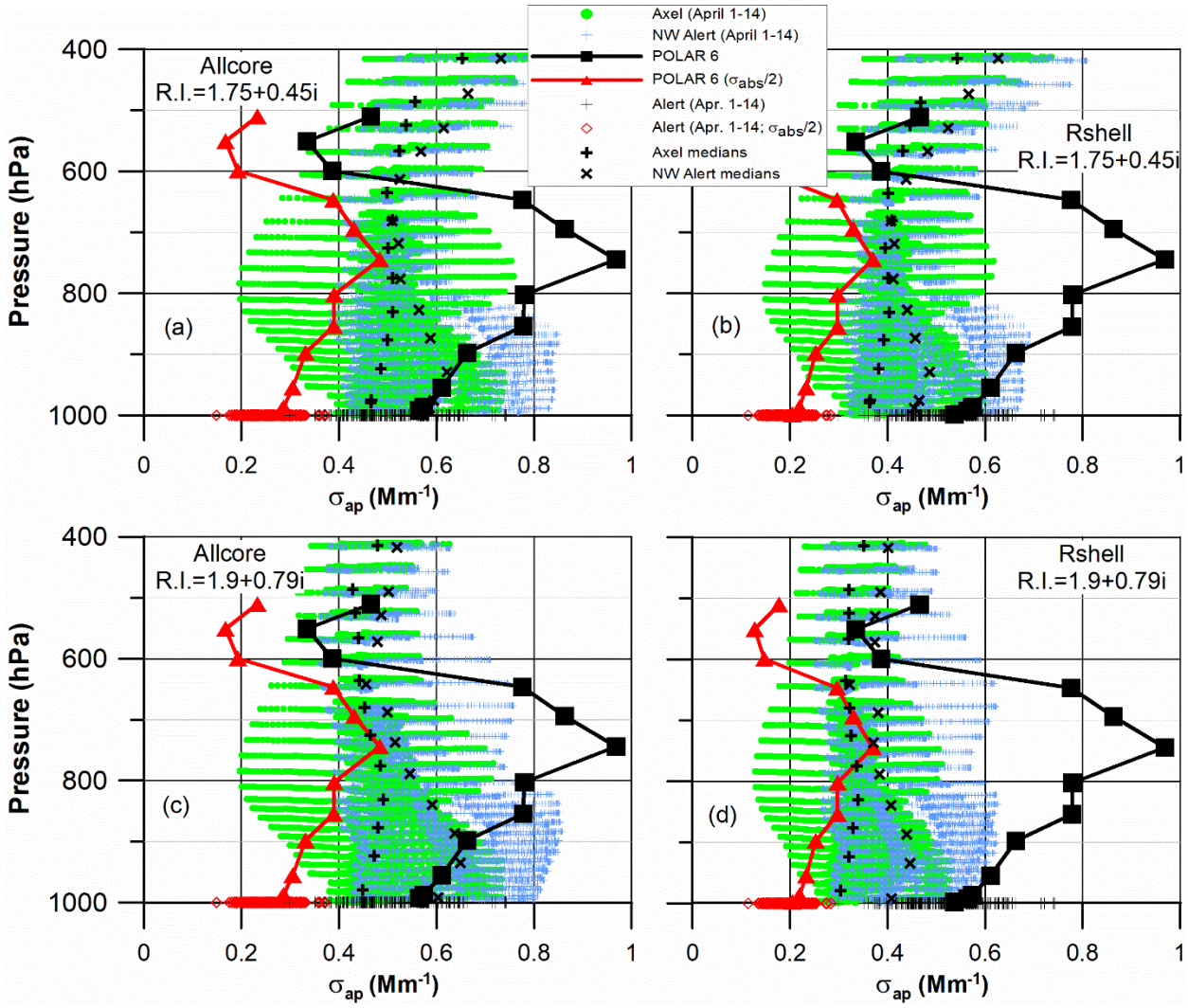

Figure 10. Vertical profile plots of σ_{ap} with atmospheric pressure showing median values based on POLAR 6 observations (black points). Red points in (a) and (c) indicate the assumption of absorption overestimation of the POLAR 6 observations ($\sigma_{ap}/2$). Red points in (b) and (d) indicate the assumption of absorption overestimation of the POLAR 6 observations ($\sigma_{ap}/2.62$). Data from the Alert Observatory for April 1-14 are shown for the absorption overestimation (red squares) and BC underestimation assumptions (black squares). The model results are for April 1-14 with the green dots representing the Axel grid and blue crosses representing the NW Alert grid. (a) gives the model results for the 'Allcore' assumption and the BC index of refraction of 1.75-i0.45; (b) gives the model results for the 'Rshell' assumption and the BC index of refraction of 1.75-i0.45; (c) gives the model results for the 'Allcore' assumption and the BC index of refraction of 1.9-i0.79; (d) gives the model results for the 'Rshell' assumption and the BC index of refraction of 1.9-i0.79.

6. Review Comment – "L216: I think further details are needed in this paper directly regarding the "allcore" and "Rshell" assumptions. For example, it is not clear whether these two conditions are mass conserving. That is, is the total BC mass concentration the same in the two, just with different distributions of BC? Also, the use of different terminology than (Kodros et al., 2018), who the authors cite, makes it less clear what specifically was done. I suggest using consistent terminology.

Response - Figure 2 of Kodros et al. (2018) uses the same terminology as in the present paper, referring to "rshell-constrained" and "allCoreShell". Mass is conserved. We have revised the model discussion to note both:

"refractive indices from the Global Aerosol Dataset (GADS; Kopke et al., 1997). To calculate aerosol optical properties, we assume two of the BC mixing states discussed by Kodros et al. (2018): 1) "Allcore", in which BC is fully mixed with other chemical species in a core-shell morphology within each size section, where BC forms the core of the particle and hydrophilic aerosol species form a concentric shell around the BC core; 2) "Rshell", in which BC is mixed within a particle, again as a core surrounded by hydrophilic species, but the size-dependent fraction of BC–containing particles core sizes and hydrophilic coating thicknesses are constrained by the observed sizes and the modelled BC mass concentration as described by Kodros et al. (2018). These two states are illustrated in Fig. 2 of Kodros et al. (2018), where they are referred to as "rshell-constrained" (Fig. 2b) and "allCoreShell" (Fig. 2d). Particle mass, including BC mass, is conserved. The Allcore state is less realistic because every particle contains a BC core, whereas Arctic observations identify BC in roughly 10-20% of the particles (e.g. Sharma et al., 2017). As a result, the Allcore mixing assumption overestimates absorption (e.g. Alvarado et al., 2016). Rshell, which is based on observations, has a smaller fraction of coating material participating in absorption enhancement, resulting in lower absorption compared with Allcore; Rshell absorption is higher than that for the externally mixed assumption (Kodros et al., 2018). The Mie code of Bohren and Huffman (1983) for two concentric spheres is used to calculate σ_{scat}, σ_{ap} and SSA."

7. Review Comment – "L275: while the scattering threshold applied preserves 98% of the data, the authors indicate on L150 that only about 10% of the absorption measurements were above the DL. I suggest that this distinction is clarified more directly."

Response – We feel that we have been quite clear in the discussions of these points. We would agree that something more is needed here if we were not representing the absorption over the full range of the scattering points, but we do that in Section 3.3. We are not misleading.

8. Review Comment – Fig. 5: I am finding some of the terminology used here unclear. Does, for example, "POLAR 6 data from Alert and Eureka" refer only to the flight data? What does it mean for the flight data to be "from Alert and Eureka?"

Response – We agree that some of the terminology we use is a bit cumbersome, due to the separation of the datasets. In the figures leading up to Fig. 5, we include data from flights at Inuvik. In Section 3.1, we identify the strong dust influence and focus on scattering less than 15 Mm-1, which leads to the removal of most of the Inuvik data, and subsequent focus only on the POLAR 6 (flight) data collected near Alert and Eureka. Because

Fig. 5 bridges these datasets, we clearly identify all POLAR 6 (flight) data with dust removed, POLAR 6 (flight) data near Alert and Eureka, and the Alert Observatory data. Besides demonstrating the MAC values, the point of Fig. 5 is to show the consistency among the datasets.

9. Review Comment – Fig. 6: The y-axes should indicate explicitly which are modeled and which are measured. Response – Added.

10. Review Comment – L291: I suggest it would be very helpful if the authors included MAC values for a "no coating" case such that the fundamental BC-only reference could be better understood and compared with expectations from observations. This is especially important given the use of the, very likely, too low RI values from OPAC. The authors compare their Rshell results to observations for "freshly" emitted BC, but this is in my opinion not a sufficient comparison. The "no coating" case must also be compared. Also, it is to be noted that one of the citations given here (Kahnert, 2010) concludes that "An agreement between observations and theoretical results can only be attained when assuming a fairly high value of the real and imaginary parts of the refractive index." This further suggests the use of the OPAC values is problematic.

Response – We agree that the refractive index is an issue, and we offer some perspective on the refractive index in the revised Figure 10. Our paper points out some of the many factors that lead to differences between modelling of absorption by black carbon components of particles and observations of such absorption. Refractive index is one of those factors, but it does not appear to be capable of explaining the differences alone. We feel that a closer examination of the RI issue is outside the scope of this paper.

11. Review Comment – L295: it is not clear that these positive intercepts are significantly different from zero. Uncertainties are needed.

Response – By using the regression, we are dealing with an average rather than individual points. The revised text reads "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)."

12. Review Comments

- Fig. 7: The smallest [BC] reported here is around 0.02 micrograms/m3. Using the slopes from Fig. 5, this corresponds to an absorption coefficient of around 0.4 1/Mm. Alternatively, the same approximate result (absorption coefficient 0.5 1/Mm) is obtained if the [BC] is multiplied by the MAC values in figure 7. This is lower than the estimated detection limits (L145). It would be good if this issue were reconciled. The authors also note a 60% uncertainty in sigma abs at 1 1/Mm. Presumably, this uncertainty is larger at smaller absorption values. It would be helpful if uncertainties were included in Fig. 7b.
- Fig. 7b: The solid red fit curve does not seem significant to me. Same with the dashed red curve. The authors do not report fit parameters or functional forms. How were these funcational forms determined? Are the fits significant? This links to the statement on L303, where the authors state that dust concentrations increase "slightly" as BC decreases. This conclusion does not seem robust to me.

Response – We apologize for the mistake in the caption for Figure 7b. The smaller values in Fig. 7b correspond to the absorption measurements from Alert, for which the stated detection limit is 0.4 Mm^{-1} (line 152 of ACPD paper). No POLAR 6 values below the stated DL of 0.75 Mm^{-1} (line 145) are included in Fig. 7b. We revised the fits in Figure 7b to be linear, and we include discussion of the fit statistics in the caption and in the text. The revised figure and caption are as follows:

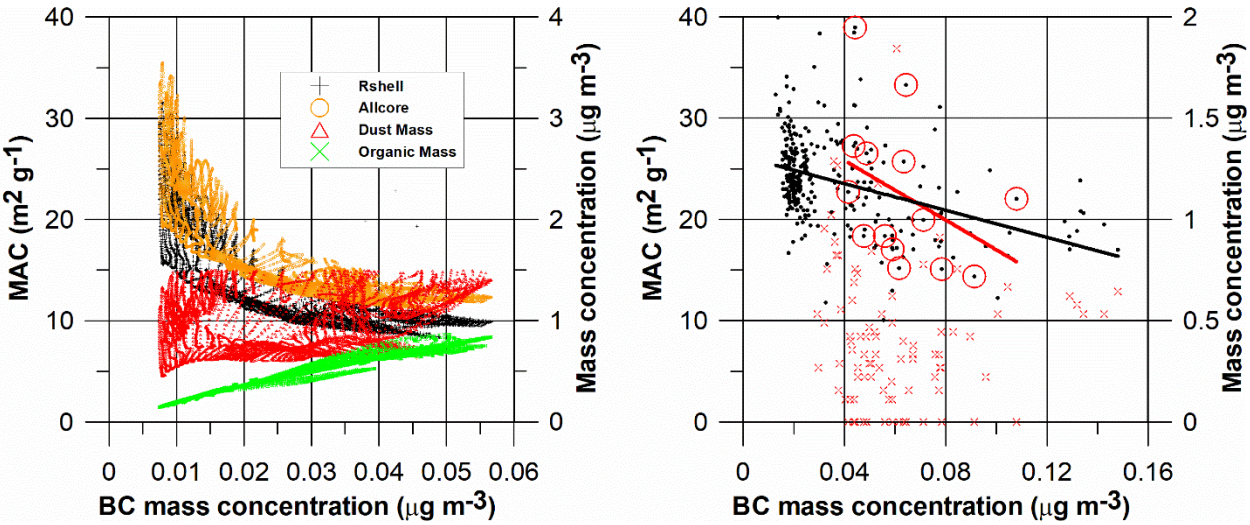

Figure 7. a) Modelled BC Mass Absorption Coefficient (MAC) plotted against modelled BC for the 'Allcore' (black crosses) and the 'Rshell" (orange crosses) 855 assumptions; modelled dust mass concentrations constrained to dust less than 1.5 μg m-3 versus modelled BC mass concentrations (red triangles); 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 crosses); dust mass concentrations (red dots), estimated from particle size distributions onboard the POLAR 6, plotted versus rBC mass concentrations; MAC values associated with zero dust points identified (red circles). The confidence level in the negative slope of the black points is greater than 99%. The confidence in the negative slope of the 14 red-circled points is only 80%.

The revised text is "In terms of the observations, absorbing dust may be present at lower BC concentrations, as shown in Fig. 7b. 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. However, we cannot distinguish whether the composition of these coarse particles is dust or sea salt. In Fig. 7b, the 14 points with coarse particle mass concentrations of zero suggest nothing other than an increase in MAC with decreasing BC (Fig. 7b), implying that dust may not explain the higher MAC at lower BC in this case. Other potential explanations include a greater deficiency in the rBC measurement at the lower BC concentrations, the presence of smaller BC components, as shown in Fig. 8, and/or more complex morphology of BC within the particles.

According to Yu et al. (2019), the latter enhances absorption by factors of 3-16 within Arctic summer particles, when BC is also very low (e.g. Schulz et al., 2018)."

13. Reviewer Comment - L304: The authors state "Also, the higher modelled MAC at lower BC may have a contribution from an increase in the coating enhancement factor as the BC core decreases in size." Did the BC core diameter decrease with decreasing concentration, as implied here? This is not actually shown. This needs to be demonstrated if the authors are to make this claim.

Response – We offer it as a possibility, rather than a "claim". That said, in response to a comment below, we show that the observed mean diameter of rBC decreases with decreasing mass concentration. Also, Sharma et al. (2017) and Kodros et al. (2018) both show that the coating thickness increases with decreasing rBC diameter. It is a possibility that this contributes to the result in Figure 7.

14. Reviewer Comment - L309: The authors state ": : :and the 14 points with coarse particle mass concentrations of zero indicate only a stronger effect on MAC at decreasing BC." This does not seem justified by the data in Fig. 7b. Many of the red-circled points are among the lower values measured. Some are high too, but more are low. I suggest that this conclusion be revised or removed. I also do not think that these data support the authors decision to exclude dust as a potential explanation for the increased MAC values. I suggest that this is a substantial over-interpretation. Further justification is necessary.

Response – As in our response to the above comments from the Reviewer concerning Figure 7, we changed this statement to read "However, we cannot distinguish whether the composition of these coarse particles is dust or sea salt, and the 14 points with coarse particle mass concentrations of zero give no suggestion of any change in MAC increasing with decreasing BC (Fig. 7b). On that basis, dust seems less likely as an explanation for the higher MAC at lower BC in this case,…" However, we have not excluded dust influences: we stated in our conclusions (line 495 of ACPD manuscript) that "The present work suggests the need to consider low concentrations of dust at smaller BC concentrations as well as…"

15. Reviewer Comment - L314: No reference to Yu et al. (2019) is available. Perhaps the authors are referring to this paper: https://www.atmos-chem-phys.net/19/10433/2019/, but it is not clear. Regardless, looking to Schulz et al. (2018) there is no evidence that the BC particles are smaller when the concentration is smaller. It is also not clear what the authors mean by smaller "fragments" of BC. Would these be small aggregates not measured by the SP2 because they are below the detection threshold? More detail is needed.

Response – Our apologies for excluding this reference. Your suggestion is correct, and it has been added to the reference list. We used the term "BC fragments" to represent the BC embedded in the particles, but we have changed 'fragments' to "components" in the revision; see response to your comments on Figure 7. These 'components' may or may not be too small for the SP2. We have added a new figure to the revised manuscript (#8 in the revision) showing that on average the mass-weighted mean diameters of the BC components from these flights are smaller when the BC mass concentrations are lower. The point is that there are many possibilities to explain our observations, which is the substance for our conclusion that more work on BC needs to be more carefully done in the Arctic. The new figure is shown below (with caption), and the revised text in shown in our response to your comments concerning Figure 7.

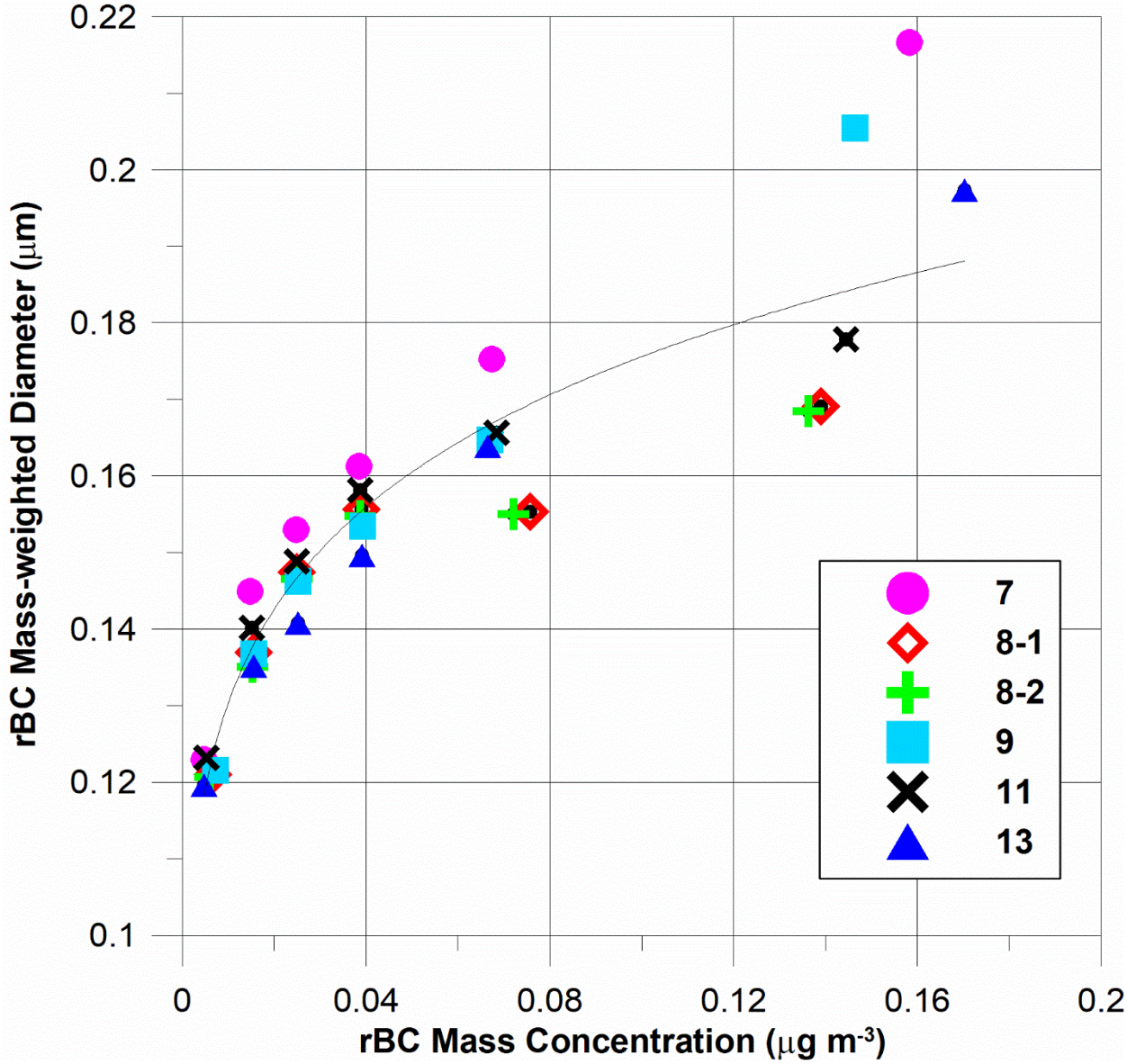

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Figure 8. Mean mass-weighted diameters of rBC measurements versus rBC mass concentrations, assuming spherical rBC components. Points are averages for mass concentration intervals of 0-0.01, 0.01-0.02, 0.02-0.03, 0.03-0.05, 0.05-0.1 and 0.1 to the maximum observed. The power-law fit is through all points. A power law is appropriate for the relationship between diameter and mass of a sphere, which includes the diameter going to zero as the mass goes to zero.

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16. Reviewer Comment - L316: Again, the nature of the curves reported are not stated. What functional form was selected and what was the justification?

Response – We have changed to linear curves in Fig. 7b. We use a power-law fit in Fig.8 and provide an explanation in the caption.

17. Reviewer Comment - L317: The reason for the difference in the MAC when viewed on a point-by-point manner (Fig. 7) and in aggregate with a linear fit (Fig. 5) should not be unclear. It is likely a result of linear fitting over a wide range of values. Linear fits are strongly controlled by values at the extreme. Because most of the high MAC values have the same low [BC], the slope of a linear fit is determined by what happens at higher [BC] relative to these lower values. These are simply two ways of looking at the data. If a histogram of the individual MAC values does not return the same median value (or at least similar) as the result of a linear fit then the appropriateness of a linear fit is in question.

Response – We are unclear what the Reviewer would like us to do based on this comment. In Figure 5 we show fits for three different ranges: higher values (P6); lower values (Alert); all values exclusive of Inuvik (P6 and Alert). Despite the different ranges, the fits are relatively similar, and most certainly similar in the context of our discussion. With Figure 7, we attempt to investigate some of the potential reasons behind our higher MAC, which seems more useful than simply saying this is a consequence of linear fitting.

18. Reviewer Comment - L323: The reference to Lack et al. (2008) indicates a misunderstanding of that paper. The positive bias in the filter-based measurements in that paper was not a result of absorption by organics. Also, it's not clear what the "up to 22% overestimation" refers to. Lack et al. (2008) show that biases of factors of 2 or larger are possible. The discussion here should be revised accordingly. Also, if organics are absorbing, then the measured absorption is not "overestimated." It is what it is and includes contributions from all absorbing particle types. The MAC might be overestimated, but the absorption would not be.

Response – We thank the Reviewer for their comments on this point. As in our above response to "Review comment – "Section 2.2:…", we believe we have corrected this.

19. Reviewer Comment - L329: It is not clear how Bond et al. (2013) support the authors contention here. Bond et al. (2013) do not show that the EC from thermal analysis is a factor of 2 higher than BC from an SP2.

Response – We agree. Again, in our above response to "Review comment – "Section 2.2:…", we have changed the discussion of this point.

20. Reviewer Comment - L331: It is not clear to me what the authors specifically mean when they say "enhancement in absorption by BC due to the morphology of BC as a function of the size distribution." To what does morphology refer? Shape of the BC? Amount of coating?

Response - In our above response to "Review comment – "Section 2.2:…", we have changed the discussion of this point, which includes removal of that statement.

21. Reviewer Comment - L334: it is not clear to me why a higher MAC would lead to higher sigma_abs, here. The authors measured sigma_abs. Are they referring to when the estimated sigma_abs, from the [BC] measurements, are used? If not, then I do not think this is appropriate.

Response – This has been revised, as in the last paragraph of our response to your comment #2 above.

22. Review Comment - L336: I do not find this adjustment to be justified. The authors adjust their measurements to the model results. However, as I've already noted, I think that there are serious issues with the model estimates due to the use of the OPAC refractive indices.

Response – Regardless of any model issues, the lower MAC values (of 7 and 9.2 m^2/g) are consistent with many previous observations (and modelled MAC), as discussed at the beginning of Section 3.2. As such, they offer a reasonable lower limit to the observations, and they offer a comparison with the model unaffected by the optical derivations in the model.

23. Reviewer Comment - Fig. 6c/d: I do not fully follow the reasoning for showing both of these. They are both linear translations of the data in Fig. 5, simply done in reverse.

Response – Plots 6c and 6d were only shown to help clarify our approach. Based on your suggestions, here and below, we have removed them and made changes in terminology (discussed below).

24. Review Comments - L347: It is not clear if the median values referred to here should mean that there is one median point per 50 hPa pressure interval, or, somehow, more than one. Fig. 8 seems to suggest more than one median is obtained, as there is more than one point shown at each pressure interval. Are these the averages calculated for contiguous periods? So there can be more than one, for example, median between 750 and 800 hPa?

Response – There is only one median point per 50 hPa interval. As we state on line 346 of the ACPD manuscript (line 380 of current revision), "we restrict the profiles of σ_{abs} and SSA to median values…" Figure 8 (Fig. 9 in the currently revised manuscript) shows only rBC and modelled BC mass concentrations. There is no need to calculate medians for the rBC because these are all measured points. We calculate medians for σ_{abs} and SSA, because at this point we have used the regression between rBC and σ_{abs} to increase the number of σ_{abs} values, but the individual variation associated with each σ_{abs} and SSA point, based on the regression is not necessarily meaningful. This is discussed on lines 345-346 of the ACPD manuscript.

25. Reviewer Comment - L355: presumably, this is a result of unaccounted for emissions from the regions indicated, not just as a result of emissions from these regions. More broadly, it is not clear what new information is obtained here, given that the authors already indicate that (Schulz et al., 2019) and (Willis et al., 2019) and Xu et al. (2017) have addressed these issues, with the former two using the same dataset.

Response – There is no new information here. We have re-written this paragraph, primarily in response to the comments from Dr. Schnell as shown in our response to him.

26. Reviewer Comment - Fig. 9/10: I find it unclear why the absorption and BC curves would be so very different, given that the authors have worked (Fig. 6c/d) to align these. I don't think it is appropriate to use the rBC*2.62 values and also the higher (20 m2/g) MAC values. The directly measured values should be used if the higher MAC is used. Otherwise, it would seem to me that the adjusted MAC values (7) should be used. I suggest that this discussion and the associated figures require further clarification. It may be that I am simply not understanding the adjustments the authors have done, and how they are being presented, but overall I think this needs to be much clearer as it is a core part of the manuscript. But I would think that this should be: Estimated absorption = [measured BC] * MAC_high, or Estimated absorption = [measured BC] * 2.62 * MAC_adjusted
Also, the nature of the sigma_abs/2 curves is not clear to me, as this is presumably also estimated from the SP2. Overall, I think that much clearer discussion is required.

Response – The absorption and BC curves are not very different, except the absorption (and SSA) are represented by the median values, for the reasons discussed above. The terminology was discussed on lines 335-342. The directly-measured or observed MAC is the higher MAC (18.4 m2/g). However, we agree that this terminology is confusing. As below, we have changed the text as well as the legends and captions in Figures 10 and 11 (previously 9 and 10).

- Lines 333-341 of ACPD manuscript (lines 367-375 of current revision) are revised as follows (also shown in response to a comment above): "If we assume that our observed σ_{abs} are overestimated by 22% and our rBC are underestimated by 7.5%, the MAC value is reduced from 18.4 m^2g^{-1} to about 13.4 m^2g^{-1}, which is about halfway between our measured value and the more commonly accepted value in the area of 9 m^2g^{-1}. However, because there are a number of potential factors influencing the measurements of both σ_{abs} and BC, we cannot attribute one value with the necessary certainty. For that reason, we employ a range for σ_{abs} by adding to our discussion the POLAR 6 plus Alert (Apr. 1-14) observations (Fig. 5) adjusted to MAC values of 9.2 $m^2\ g^{-1}$ and 7.0 $m^2\ g^{-1}$, respectively, through division of the σ_{ap} by factors of 2 and 2.62. The 9.2 $m^2\ g^{-1}$ is derived from an average of the three MAC values referenced above and the two modelled grids for the Allcore assumption, while the 7.0 $m^2\ g^{-1}$ is an average of the two modelled grids for the Rshell assumption."
- Fig. 10 (shown above) and 11 (shown below) and their captions are revised.
- All appropriate text has been revised accordingly.

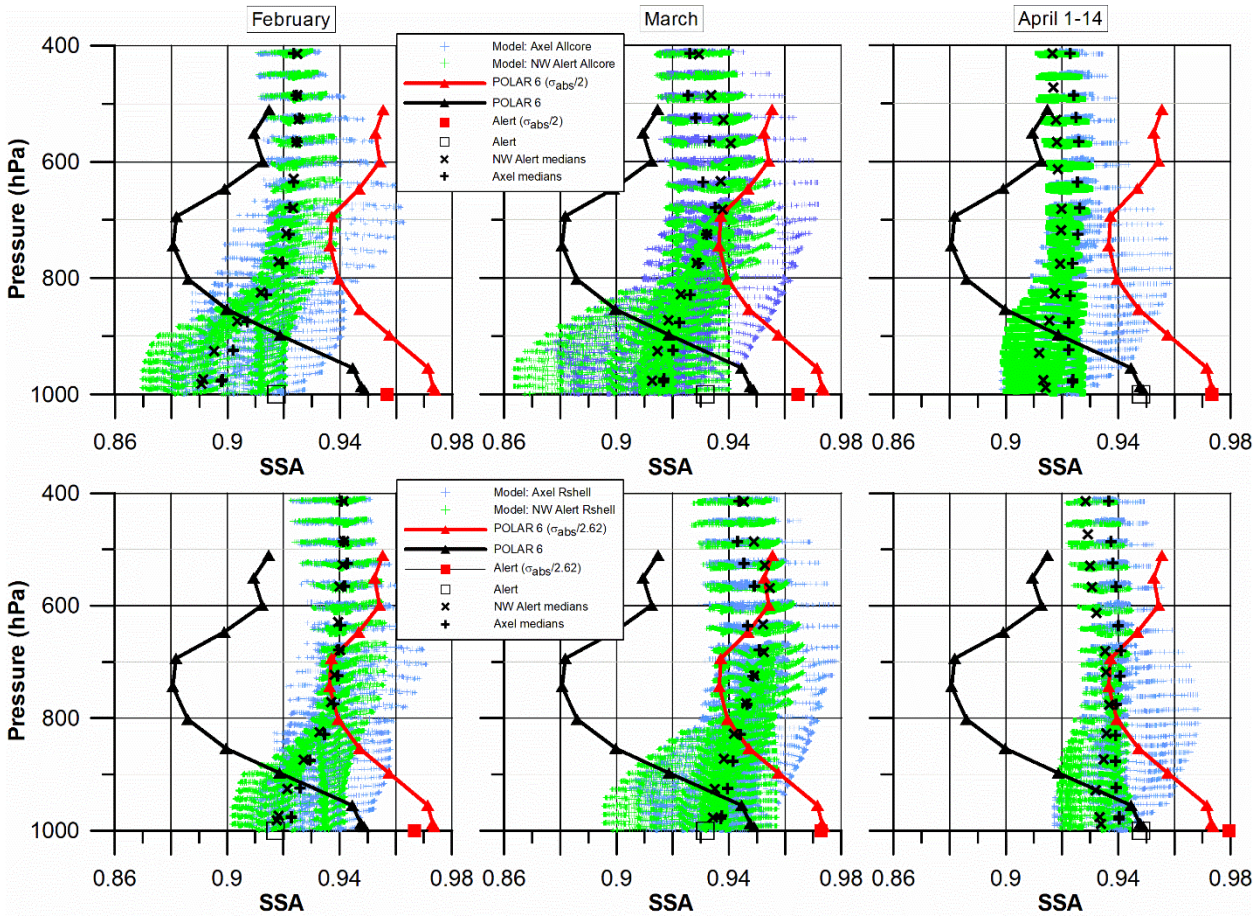

Figure 11. Vertical profile plot of SSA with atmospheric pressure showing modelled results for February (a), March (b) and April 1-14 (c) based on the 'Allcore' assumption and results for February (d), March (e) and April 1-14 (f) based on the 'Rshell' assumption. Median values of SSA from the POLAR 6 observations and the Alert Observatory shown in a, b and c are based on the measured absorption (black points) and the absorption overestimation assumption ($\sigma_{ap}/2$; red points), corresponding to the 'Allcore' assumption. Median values of SSA from the POLAR 6 observations and the Alert Observatory shown in a, b and c are based on the measured absorption (black points) and the absorption overestimation assumption ($\sigma_{ap}/2.62$; red points), corresponding to the 'Rshell' assumption.

27. Reviewer Comment - Fig. 8-10: It would be helpful if averages were also reported for the model results.

Response – As above, they have been added to Figures 10 and 11 (previously, Fig. 9 and 10).

28. Reviewer Comment - L388: It is not clear what is "inconsistent" here. SSA also depends on modeled scattering. Comparison of the absorption is only part of the story. This "inconsisten[cy]" suggests that there is also a discrepancy in the measured and modeled scattering. Indeed, this seems apparent in Fig. 11. I suggest the authors revise the discussion accordingly.

Response – The statement is revised to read "In Fig. 11, the modelled and observation-based SSA have opposite tendencies from the surface to about 600 hPa. In the 700-800 hPa region, there is good agreement between the modelled SSA for the Rshell mixing state and the observation-based SSA for $\sigma_{ap}/2$. However, given that the modelled and measured σ_{sp} are in reasonable agreement in that region (Fig. 12), the SSA agreement seems inconsistent with the underestimation by the model of BC in that pressure region (Fig. 9)."

29. Reviewer Comment - L396: The authors mention here "As above, a relatively low imaginary refractive index...". It is not clear to me where the low RI is fully discussed above.

Response - Based on the additional model runs, we have revised the sentence as follows: "As mentioned above, the ejection of biomass burning particles only into the boundary layer at the source may contribute to the higher modelled SSA in the 600-900 hPa layer. The imaginary part of the refractive index is another model issue, but our simulations with the higher refractive index (real and imaginary parts) did not yield large differences in the σ_{ap}."

30. Reviewer Comment - L413: It would seem as if the authors would be able to directly test the idea of whether the modeled size distributions are smaller than the observations, rather than speculating here. I suggest this would be a good addition.

Response – Thank you. With the revised version, we include a new figure (13) showing a comparison of the model size distributions (Axel grid) with size distributions measured in the Axel grid (flights from April 11 and 13). The text has been modified from "Model underestimation of submicron particle sizes is a possible explanation for lower modelled volume scattering efficiencies. In addition, it is possible that the observed volumes, based on the UHSAS, are underestimated." to "Model underestimation of submicron particle sizes may contribute to the lower modelled volume scattering efficiencies. In Fig. 13, the modelled particle size distributions for the Axel grid, April 1-14 period and averaged over the indicated pressure intervals are compared with measured distributions from the two flights (April 11 and 13) conducted in that grid. The modelled distributions for the 800-900 hPa and 900-1019 hPa intervals are shifted to slightly lower sizes relative to the average of the observations. The modelled distributions for 600-800 hPa are a closer match to the measurements, and for 400-600 hPa, the average of the modelled sizes is a reasonable match to the April 11 measurements, but exceeds the April 13 observations. The overall pattern is generally consistent with the variation of the modelled σ_{sp}, for the Axel grid and April 1-14 period, relative to the observed σ_{sp}, shown in Fig. 12a. In addition, the lower modelled volume scattering efficiencies may result from underestimation of the observed volumes that are based on the UHSAS."

The new figure 13 is shown below.

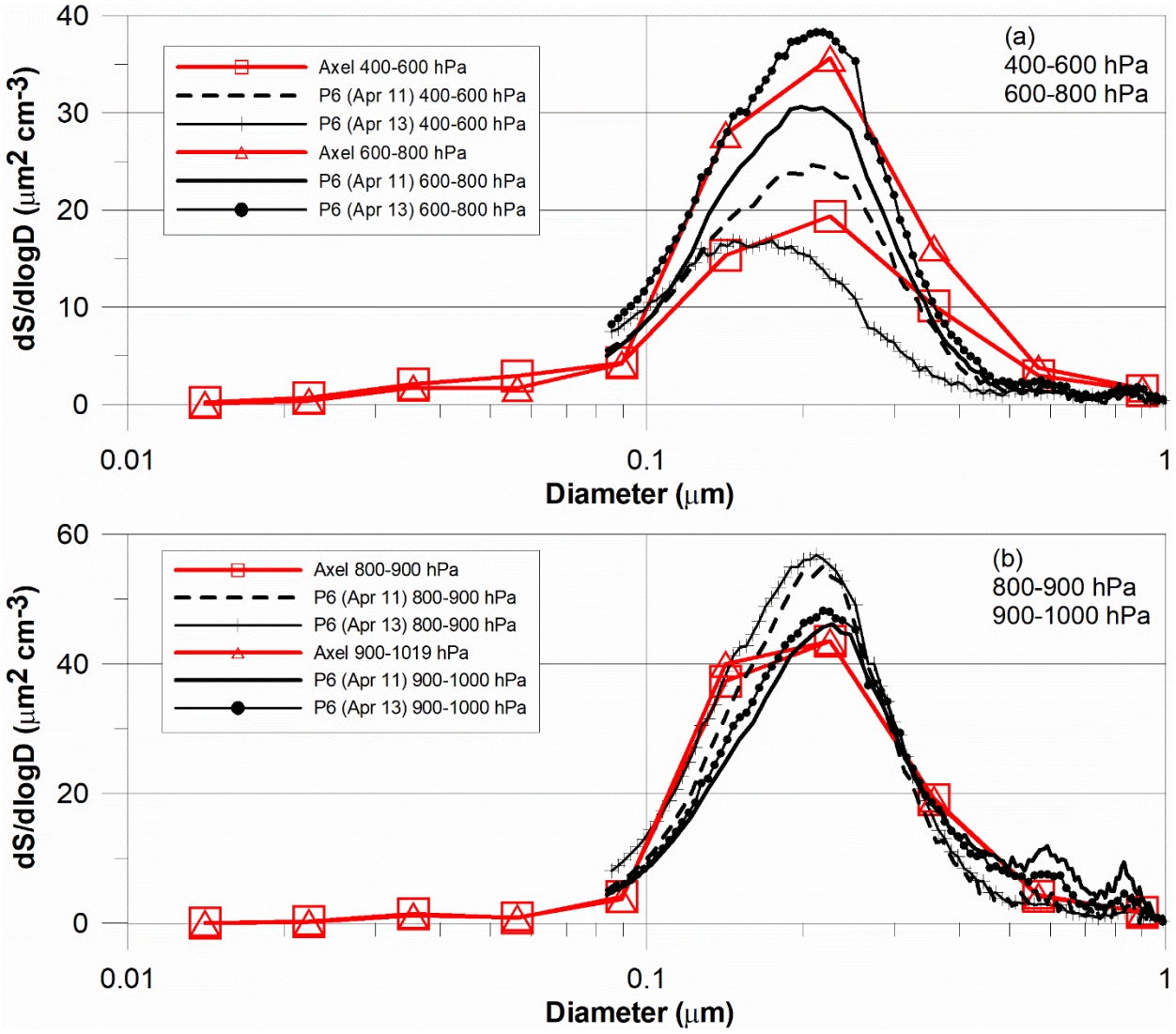

Figure 13. Comparison of modelled size distributions for the Axel grid and period of April 1-14 with the measured distributions from the POLAR 6 (P6) flights on April 11 and 13. All distributions are averaged for the indicated pressure intervals. a) Modelled and measured distributions for pressure intervals of 400-600 hPa and 600-800 hPa. b) Modelled and measured distributions for pressure intervals of 800-900 hPa and 900-1000 hPa.

31. Reviewer Comment - L455: this seems to contradict the authors' decision to exclude dust contributions as an explanation for the slight increase in the observed MAC at low [BC].

Response – This has been re-written as follows: "The results of Targino et al. (2005) and Andrews et al. (2011), suggesting lower SSA associated with lower σ_{sp}, as well as the present results are examples that wet scavenging may enhance the relative absorption by the Arctic aerosol. However, despite our results suggesting dust is not a factor at the lower BC in this case (Fig. 7), the possibility of significant contributions to absorption from small amounts of dust should still be considered."

32. Reviewer Comment - L479: Are the authors here saying that the SP2 underestimated BC substantially? They do not give a clear reason for thinking this might be the case in the discussion above, in my opinion. Also, it is not clear how the authors are concluding that "morphological arrangements of BC components within particles" being "inconsistent with the often-used core-shell concept" helps explain the larger MAC values. The core-shell configuration tends to give an upper-limit for absorption; alternative morphologies give lower enhancements. This would, I think, go counter to the authors' argument.

Response – The statement has been revised as follows: "The higher MAC value may be due to a number of factors, including underestimation of BC, morphological arrangements of BC components within particles that are inconsistent with the often-used core-shell concept, and overestimation of σ_{ap} by our observations. Due to the uncertainties, we evaluated σ_{ap} and SSA assuming an overestimation of absorption in addition to the higher MAC value."

Minor comments:

33. Reviewer Comment - L141: "empirically based" should just be "empirical".

Response – Changed as suggested.

34. Reviewer Comment - I will encourage the authors to avoid use of the red-green color scheme that they seem to favor, as this is difficult to view for color-blind people.

Response – Although we maintain colour in our figures, we have tried to improve the points and lines in the figures more distinguishable by means other than colour.

Responses to Reviewer 2

We are grateful to the Reviewer for their time and for providing constructive comments. Our responses follow the individual comments.

Reviewer comment: The topic of the paper (BC vertical profiles in the Arctic) is importat for climate application. However some issues has to be solved before publication. One of the most important and main lack of the paper is its aim. It just reports data and a comparison with model results but with a poor discussion concerning the origin of the big differences reported. Please first of all details very well the goal and aims of the paper.

Response – We agree that the main objective of the paper may not have been clearly stated. In the last paragraph of the introduction we stated "By constraining the discussion to values of σ_{sp} less than 15 Mm^{-1}, we address the largest component of Arctic haze exclusive of the direct influence from strong plumes. Since most Arctic pollution in April is from long-range transport, the lower σ_{sp} suggests that these particles on average spent longer times in the Arctic atmosphere and thus are more indicative of the "chronic" Arctic haze discussed by Brock et al. (2011). Further, the SSA for particle populations that fall within this constraint have been found to decrease more sharply with decreasing σ_{sp} (e.g. Targino et al., 2005; Andrews et al., 2011), making these populations more efficient at warming of the atmosphere." To clarify our objective, we add, after the above, the following sentence: "Our objective is to further our knowledge of the absorption by BC at these lower σ_{sp} in a region of the Arctic where relatively few airborne measurements have been made."

Reviewer comment: 1-Introduction lines 55-78: most of the reported references (even good) are quite all and the final statment "in part due to the lack of observational data on the distribution of BC with altitude (e.g. Samset et al., 2013)" should be changed considering all the BC vertical profiles reported in the Arctic during the last ten years. They are not reported here. Some examples come from Schwarz et al. (2010), Wofsy et al. (2011), Spackman et al. (2010), Ferrero et al. (2016), Markowicz et al. (2017).

Response – The suggested references has been added. It now reads as follows: "Despite profiles of black carbon and optical properties in recent years (e.g. Brock et al., 2011; McNaughton et al., 2011; Schwarz et al., 2010; Wofsy et al., 2011; Spackman et al., 2010; Ferrero et al., 2016; Markowicz et al., 2017) there remains a shortage of such observational data that limits evaluation of models of Arctic BC and light absorption (e.g. Samset et al., 2013), because the Arctic is subject to transport from many pollution sources at southern latitudes during winter and spring, and variability exists with altitude, with location and from year-to-year." In addition, we have added a reference to Ferrero et al. (2016) in our discussion of the vertical profiles in Section 3.3 as follows: " Also, the lower part of the profile concentration data (<1 km) is similar to the springtime low-level profile BC concentrations from Ny-Ålesund measured by Ferrero et al. (2016)."

Reviewer comment: 2- Introduction lines 88-90: "Airborne measurements of ïA¸s¸ap that are based on transmission of light through a filter, as used here, are constrained by instabilities during changes in pressure (i.e. altitude) and generally higher detection limits (DL) associated with flight conditions". The sentence here is not clear and generate confusion in the reader. Better to remove and details in the method section.

Response – We feel that this statement is reasonably clear. It summarizes an issue with sampling for light absorption based on light transmission through a filter. The statement is appropriate here, because it is fundamental to our objective.

3- Introduction lines 91-100: this part is a methodological part. Please move to the method section.

Response – Again, this short discussion of methods is fundamental to defining the objective, and therefore maintained here.

4- Section 2.1 lines 115-116: "All airborne and model data presented here are referenced to a temperature of 20oC and pressure of 1013.25 hPa". Please remember that are ambient concentrations that determined the final radiative effect. Please add also data in ambient concentrations (at the real T and p) at least in the supplementary.

Response – We agree that the radiative effects are based on ambient concentrations, and we have added the following statement: "As discussed in Section 2.1, the profile data, including σ_{ap} and σ_{ap} (SSA is dimensionless), have been adjusted to a standard temperature and pressure (20oC and 1013.25 hPa) for purposes of comparisons. We note that in-situ values of σ_{ap} and σ_{ap} are appropriate for calculating radiative effects."

5- Lines 193-198: "Model 1.129 measures particles larger than 0.25 ïA¸ m, but only the coarse particle concentrations are used here. As shown by comparisons with a Particle Measuring Systems FSSP-300 probe operated under one wing of the POLAR 6, the coarse particles tend to be sampled less effectively than the submicron particles, but they are still an indicator of the presence of coarse particles, and, more importantly, the coarse particles entering the POLAR 6 sample manifold". There is no reason to avoid the use of submicron data from Grimm OPC. I would suggest to compare the Grimm data with the UHSAS ones on the overlapping measuring region.

Response – The Grimm data were used because those particles were sampled inboard the aircraft, and therefore better represent the inboard aerosol that is the subject of the measurements. Although we have compared the UHSAs and Grimm data, with reasonable results, there is no reason to draw that comparison here.

6- Lines 234-235: The model assumes a refractive index for BC of 1.75-0.45i in the mid visible (Hess et al., 1998). Hess et al. (1998) data are old. Bond an Bengstrom (2006) reported new and accepted values of BC refractive index. There is no reason to use the oldest refractive index. Please, redo the calculations considering the Bond and Bengstrom (2006) data.

Response – In response to comments from Reviewer 1, we have done exactly as you ask. These new results are shown in Figures 10 and 11 of the current revision. Perhaps surprisingly, perhaps not, there is relatively little difference.

7- Section 3.1. Dust episodes in the Arctic are quite important. Please compare your results to other literature papers.

Response – We discuss other results in Section 3.1, and we mention how our dust optical properties with the results of Hallar et al. (2015). We completely agree that dust is important, and we mention that in the conclusions, but the paper is not about dust.

8- Line 289: "Removal of points with modelled dust concentrations greater than 1.5 ïA¸ g m-3 (arbitrary value)". Removing data based on an arbitraty choice can influence results without any scientific criteria. Please details the reason of the 1..5 ïA¸ g m-3 choice.

Response – While the 1.5 value is arbitrary, we demonstrate in Figure 6 that this point of discrimination is at the lower end of the modelled absorption values. Therefore, changing that value would not result in a significant difference.

9- Section 3.2: I see a serious problem here related to the fact that modelled results from which MAC are calculated are based on the hold Hess et al. (1998) refractive index. I suggest to redo the calculations (see my question 6).

Response – As above, we did re-evaluate with the refractive index recommended by Bond and Bergstrom.

10- Figure 6: please also add panels in which only the mass concentrations (either measured and modelled) are plotted one versus the other.

Response – Profile plots of the measured and modelled BC mass concentrations are shown together in Figure 9.

11-Figures 9 and 10: the reason of using half of absorption coeff or doubling it is not clear from the manuscript text. Please details it better.

Response – Agreed. In our response to Reviewer 1, we detail the changes we have made to address this problem.

12- Lines 409-410: "The modelled scattering efficiency (scattering coefficient per unit volume) is significantly lower than the efficiency based on the observations. Near the surface (>900 hPa), the median of ïA¸s¸sp/Volume from the observations is 12.1 ïA¸ m-1" Something appears wrong from a dimensional analysis. Scattering coefficient unit is usually in Mm-1, and volume in m3. How results can be in a lenght at -1 (um-1)? Moreover, the scattering efficiency is a dimensionless parameter (Seinfeld and Pandis, 2006).

Response – Thank you. We have corrected the above sentence “"The modelled scattering efficiency (scattering coefficient divided by volume concentration) is …” The volume is actually a volume concentration ($\mu m^3/cm^3$), as shown in the profile plot of the volume concentration (Figure 12 of current revision), which results in the indicated units.

References:

Bond, T.C., Bergstrom, R.W.: Light absorption by carbonaceous particles:

an investigative review. Aerosol Sci. Technol. 40, 27–67, 2006.

Ferrero, L., Cappelletti, D., Busetto, M., Mazzola, M., Lupi, A., Lanconelli, C., Becagli, S., Traversi, R., Caiazzo, L., Giardi, F., et al.: Vertical profiles of aerosol and black carbon in the Arctic: a seasonal phenomenology along 2 years (2011–2012) of field campaigns. Atmos. Chem. Phys. 16, 12601–12629, 2016.

Schwarz, J. P., Spackman, J. R., Gao, R. S., Watts, L. A., Stier, P., Schulz, M., Davis, S. M., Wofsy, S. C., and Fahey, D. W.: Global-scale black carbon profiles observed in the remote atmosphere and compared to models, Geophys. Res. Lett., 37, L18812, doi:10.1029/2010GL044372, 2010.

Seinfeld, J.H., Pandis, S.N., 2006. Atmos. Chem. Phys. From Air Pollution to Climate Change. Wiley-Interscience edition.

Spackman, J. R., Gao, R. S., Neff, W. D., Schwarz, J. P., Watts, L. A., Fahey, D. W., Holloway, J. S., Ryerson, T. B., Peischl, J., and Brock, C. A.: Aircraft observations of enhancement and depletion of black carbon mass in the springtime Arctic, Atmos. Chem. Phys., 10, 9667–9680, doi:10.5194/acp-10-9667-2010, 2010.

Wofsy, S. C., the HIPPO Science Team and Cooperating Modellers and Satellite Teams: HIAPER Pole-to-Pole Observations (HIPPO): fine grained, global-scale measurements of climatically important atmospheric gases and aerosols, Philos. T. R. Soc., 369, 2073–2086, 2011.

Responses to Reviewer 3

We are grateful to the Reviewer for their time and for their constructive comments.

Reviewer comment: The paper is dealing with the very important topic of the vertical distribution of black carbon and vertical profiles of optical properties. Such measurements are still rare and urgently needed to answer the important questions connected to Arctic warming. The paper is based on a valuable data set which was analyzed in detail and complemented by model results. However, I miss some interpretation and real conclusions. The plots are mainly described by the authors but interpretation is sparse. The structure of individual sections could be a bit clearer, e. g. by introducing subsections. Thus, some more work needs to be done before presenting this valuable dataset to the scientific community. Overall, motivation and conclusions are not clear to me. The paper needs to be checked also for consistency, different symbol or better different indices are used for the same variable if I understood correctly.

Reviewer comment: Various literature is given, missing articles are already mentioned by other reviews, thus I don't want to repeat this now. But I miss a clear motivation for doing this study. What is the open question after all these publications and experiments in the past? This has to be given in the introduction to arouse the interest of the reader.

Response – As in our response to Reviewer 2, we now specifically state "Our objective is to further our knowledge of the absorption by BC at these lower σ_{sp} in a region of the Arctic where relatively few airborne measurements have been made.

Reviewer comment: Obviously, there was no dryer used in the aerosol line? Was there any measurement of rH in the inlet line? Even small changes at low rH may cause changes in particle absorption, in particular in clean environments (Düsing et al., 2019).

Response – The aerosol was sampled at ambient temperatures of -20oC and colder. It was brought into the cabin near the front which was the warmest area of the instrument cabin, and the CLAP was situated beside the nephelometer. The temperatures of the air entering the nephlometer ranged from +14oC to +30oC, and therefore we expect the aerosol was very dry. In addition, we drew comparisons between two in-cabin instruments (both situated farther along the cabin where the air was slightly colder) that indicate the aerosol sampled in the cabin was dry: the in-cabin UHSAS measurements compared well with the Alert aerosol size distributions (which are very dry), as discussed by Wiliis et al. (ACP, 2019); the in-cabin Grimm OPC measurements were compared with the measurements from the outboard (underwing) FSSP-300. The FSSP-300 probe results showed evidence (in some cases) of distributions with particle sizes shifted to larger values compared with the Grimm OPC, and (in other cases) the two agreed well. Also, as stated in the paper, the absorption measurements used in this paper were limited to in-flight pressure variations (as recorded within the nephelometer) of less than 2 hPa over a two-minute period encompassing each one-minute sample. Thus, we have no reason to expect that RH was a large factor in the discrepancy we find. We have added reference to Düsing et al. (2019) as follows: "Recently, Düsing et al. (2019) found that changes in water uptake by filter material used in particle absorption measurements can influence the light absorption measurement. In the present case, because the ambient temperatures were -20oC or colder and the temperatures at the intake of

the nephelometer, which was situated next to the CLAP, ranged from +14oC to +30oC, we expect little influence of relative humidity on our results."

Reviewer comment: Line 140 ff. What was the filter medium used in the absorption photometers? How was the correction done? This is a very sensitive part of the data analysis in the Arctic.

Response - Glass-fiber filters (Pallflex type E70-2075W) were used. This has been added to the paper. They are similar to the PSAP filters except for size. As discussed in Section 2.2, the analysis was done using the algorithm described by Bond et al. (1999) and Ogren (2010). That analysis was done by the first author. The algorithm used for the POLAR 6 data was also applied to the Alert data, and it was found to agree with the Alert data analyzed independently using the NOAA algorithm.

Reviewer comment: Line 170 ff. Why was the volume compared? The surface area is more relevant for optical properties.

Response – The volume comparison was done initially for use in Willis et al. (ACP, 2019) that was examining mass concentrations. However, the comparison was initiated with number distributions that were converted to volume. Since the number distributions also compared well, it follows that the surface area distributions compare similarly well.

Reviewer comment: Line 193 ff. Was there any correction for losses in the sampling line or inlet? It is just mentioned that coarse particles are collected less effectively, but this should be taken into account for the analysis.

Response – No corrections for inlet losses were applied, which is why all results are based on quantities measured inboard. Our assumption is that, because the Grimm instrument was inboard along with the CLAP, Neph, UHSAS, etc., the sampled aerosol was subject to the same inlet constraints. Coarse particles, and hence dust, may have been more abundant in the ambient air, but should not affect our observations-model comparisons, assuming that coarse-particle BC was small. If BC attached to coarse particles was significant, it would only exaggerate the observations-model differences.

Reviewer comment: Please check the symbols: the scattering coefficient is named with the index scat or sp or are these different parameters.

Response – Corrected to sp.

Reviewer comment: Mie model: Do I understand correctly that no measurements of aerosol number size distribution are used? Why?

Response – The model is a global chemical transport model. The particular simulations are described by Kodros et al. (ACP, 2018). As suggested by Reviewer 1, we have added a comparison of measured and modelled size distributions.

Reviewer comment: Figure 1: I see only one star, figure caption says “stars show the center: : :”

Response – The figure has been improved.

Reviewer comment: Figure 3: Figure caption contains < 2 m, while axis and text say > 2 m. I assume the latter one is correct. Are the zero-like number concentrations at the ground stations realistic or could this be also a result of inlet losses?

Response – Thank you. The caption has been corrected to > 2 m. The data points in Figure 3 are all from the POLAR 6 flights. “Alert and Eureka” refer to the flights conducted around those two locations. Similarly, “Inuvik” is for flights conducted out of the Inuvik airport.

Reviewer comment: Line 275: is spt the same as sp ?

Response – Yes, it has been corrected.

Reviewer comment: Line 284 ff. Why is the MAC observed here so different from any other Arctic studies? This should be critically discussed.

Response – The discussion of the differences and possible factors contributing to the differences has been expanded in the current revision; see the responses to Reviewer 1. However, the reasons for the differences are unclear, particularly since the absorption and SSA values from the POLAR 6 agree with the Alert Observatory results.

Reviewer comment: Figure 8 and 9: Model results: why do the model results show these structures? This is not really clear from the plot and text. Please explain!

Response – The work of Xu et al. (2017), using the same model as Kodros et al. (ACP, 2018), found much better agreement with the observations. The main difference between those models was that Kodros et al. injected all biomass burning emissions into the boundary layer. That was mentioned in the original manuscript (lines 359-363) and it is discussed in the current revision on lines 400-402.

Reviewer comment: Most of the figures are just described, I miss some more interpretation. Although the fact that the model does not generally underestimate BC, it is mainly in higher altitudes. This is an important fact and shows that the transport of anthropogenic pollution is by far not well understood and not covered by the models. This should be clearly stated and as a result more measurement for similar regions are needed to close this gap.

Response – The model-observation comparisons are done to provide one modelling perspective on the overall uncertainties in our knowledge of BC and its impact on the Arctic atmosphere. It would be unfair to state here that all models are deficient in some way, even if it is likely. We think that our final conclusion is consistent with your recommendation: “This work typifies the large uncertainty that exists in our knowledge of the contribution from BC to direct warming of the Arctic atmosphere. It suggests a lower level of confidence in assessing direct

absorption by BC, and the need for more detailed efforts if the impact of BC on Arctic climate is to be properly established. Those efforts include improved measurements of BC and absorption, and more vertical profiles of aerosol chemistry, microphysics and optical properties."

Literature:

Düsing, S., B. Wehner, T. Müller, A. Stöcker and A. Wiedensohler (2019). "The effect of rapid relative humidity changes on fast filter-based aerosol-particle light-absorption measurements: Uncertainties and correction schemes." Atmospheric Measurement Techniques 12(11): 5879-5895.

Response to Dr. Russ Schnell – We are grateful to you for providing these references. The inclusion of Hansen and Rosen, 1984; 1985, Hansen and Novakov, 1989 and Hansen et al., 1997 help us to present an improved perspective on Arctic BC. Besides two additions to the introduction, the following has been revised/added as lines 383-397 of the current revised manuscript:

Using version 10.01 of GEOS-Chem (without TOMAS) and its adjoint, Xu et al. (2017) found that BC corresponding to these observations was dominated by sources from eastern and southern Asia. Xi et al. (2017) also found better agreement of modelled BC with these observations; although the same version of GEOS-Chem is used here, all biomass burning emissions were injected only within the boundary layer, potentially accounting for some of the lower modelled BC relative to rBC. The relative increase in rBC in the 600-900 hPa region is consistent with the mean profiles of McNaughton et al. (2011), and the median observed rBC concentration in the 600-900 hPa range is similar to the mode concentration of normally distributed values for "free tropospheric background haze" of 0.06 μg m^{-3} estimated by Brock et al. (2011). The present result is considerably lower than the medians of 0.1-0.5 μg m^{-3} measured in the Arctic in 1983 (Hansen and Rosen, 1984), 1986 (Hansen and Novakov, 1989) and 1992 (Hansen et al., 1997). A decrease in BC at Alert, Nunavut, during the 1990s of more than 50% was associated with a reduction in Eurasian emissions (e.g. Sharma et al., 2019), and the present lower rBC concentrations near the surface may be connected to that reduction. Since east Asian emissions increased during the same time (e.g. van Donkelaar et al., 2008), it is difficult to assess the reason for the present lower concentrations in the 600-900 hPa range. It appears that the present observations represent particles that spent a considerable length of time in the Arctic atmosphere.