

Interactive comment on “Vertical profiles of light absorption and scattering associated with black-carbon particle fractions in the springtime Arctic above 79° N” by W. Richard Leitch et al.

Anonymous Referee #1

Received and published: 16 September 2019

The authors report on black carbon concentration and light absorption and scattering measurements from the Arctic. They compare their observations to model results, and interpret their observations directly. Overall, I have some concerns regarding this work, in particular (i) weaknesses of some of the core assumptions made, (ii) some lack of clarity regarding what was actually done, and (iii) sufficient distinction from previous work. I believe that substantial revision will be necessary before this work can be published.

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

Printer-friendly version

Discussion paper



per particle. The authors should consider alternative explanations, not just one explanation.

Section 2.2: Not discussed here thoroughly, 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.

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.

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.

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

[Printer-friendly version](#)[Discussion paper](#)

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.

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.

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.

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?”

Fig. 6: The y-axes should indicate explicitly which are modeled and which are measured.

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.”

[Printer-friendly version](#)[Discussion paper](#)

This further suggests the use of the OPAC values is problematic.

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

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 \sim 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 σ_{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 functional 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.

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.

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

[Printer-friendly version](#)[Discussion paper](#)

this is a substantial over-interpretation. Further justification is necessary.

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.

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

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.

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.

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

[Printer-friendly version](#)[Discussion paper](#)

BC from an SP2.

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?

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

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.

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.

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?

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.

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 $r_{\text{BC}} \cdot 2.62$ 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 $\sigma_{\text{abs}}/2$ curves is not clear to me, as this is presumably also estimated from the SP2. Overall, I think that much clearer discussion is required.

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

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.

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.

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.

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].

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

[Printer-friendly version](#)[Discussion paper](#)

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.

Minor comments:

L141: “empirically based” should just be “empirical”.

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.

References

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, *Journal of Geophysical Research: Atmospheres*, 118, 1-173, <https://doi.org/10.1002/jgrd.50171>, 2013. Cappa, C. D., Lack, D. A., Burkholder, J. B., and Ravishankara, A. R.: Bias in filter-based aerosol light absorption measurements due to organic aerosol loading: Evidence from laboratory measurements, *Aerosol Science and Technology*, 42, 1022-1032, <https://doi.org/10.1080/02786820802389285>, 2008. Kahnert, M.: On the Discrepancy between Modeled and Measured Mass Absorption Cross Sections of Light Absorbing Carbon Aerosols, *Aerosol Science and Technology*, 44, 453-460, <https://doi.org/10.1080/02786821003733834>, 2010. Kodros, J. K., Hanna, S. J., Bertram, A. K., Leaitch, W. R., Schulz, H., Herber, A. B., Zanatta, M., Burkart, J., Willis, M. D., Abbatt, J. P. D., and Pierce, J. R.: Size-resolved mixing state of black carbon in the Canadian high Arctic and implications for simulated direct radiative effect, *Atmos. Chem. Phys.*, 18, 11345-11361, <https://doi.org/10.5194/acp-18-11345-2018>, 2018. Lack, D. A., Cappa, C. D., Covert, D. S., Baynard, T., Massoli,

Printer-friendly version

Discussion paper



P., Sierau, B., Bates, T. S., Quinn, P. K., Lovejoy, E. R., and Ravishankara, A. R.: Bias in filter-based aerosol light absorption measurements due to organic aerosol loading: Evidence from ambient measurements, *Aerosol Science and Technology*, 42, 1033-1041, <https://doi.org/Doi.10.1080/02786820802389277>, 2008. Schulz, H., Zanatta, M., Bozem, H., Leaitch, W. R., Herber, A. B., Burkart, J., Willis, M. D., Kunkel, D., Hoor, P. M., Abbatt, J. P. D., and Gerdes, R.: High Arctic aircraft measurements characterising black carbon vertical variability in spring and summer, *Atmos. Chem. Phys.*, 19, 2361-2384, <https://doi.org/10.5194/acp-19-2361-2019>, 2019. Stier, P., Seinfeld, J. H., Kinne, S., and Boucher, O.: Aerosol absorption and radiative forcing, *Atmos. Chem. Phys.*, 7, 5237-5261, <https://doi.org/10.5194/acp-7-5237-2007>, 2007. Willis, M. D., Bozem, H., Kunkel, D., Lee, A. K. Y., Schulz, H., Burkart, J., Aliabadi, A. A., Herber, A. B., Leaitch, W. R., and Abbatt, J. P. D.: Aircraft-based measurements of High Arctic springtime aerosol show evidence for vertically varying sources, transport and composition, *Atmos. Chem. Phys.*, 19, 57-76, <https://doi.org/10.5194/acp-19-57-2019>, 2019.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2019-785>, 2019.

[Printer-friendly version](#)[Discussion paper](#)