Responses to the Reviewers' Comments

Title: Estimates of mass absorption cross sections of black carbon for filter-based absorption photometers in the Arctic
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The co-authors and I very much appreciate the constructive comments on this manuscript by the reviewers. The comments have been very thorough and useful in improving the manuscript. We have taken them fully into account in revising it. Major revisions made to the manuscript are described first, followed by responses to the comments raised by each reviewer.

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Major revisions

1. Addition of the COSMOS-MAAP comparison at Ny-Ålesund

In the revised manuscript, we have added analysis of COSMOS-MAAP comparison at Ny-Ålesund during 2017–2020 in Sect. 3.2.3 to investigate the possibility of converting b_{abs} (MAAP) to M_{BC} at this site. Similar to the other comparisons at Arctic sites, the time series data of b_{abs} (MAAP) were highly correlated with those for M_{BC} (COSMOS), and the MAC_{cor} (MAAP) values at Ny-Ålesund were estimated to be 10.6 and 10.9 m^2 g^{-1} for the 1-h and 24-h data, respectively. These MAC_{cor} values are about 60 % higher than the manufacturer's default setting value (= 6.6 m^2 g^{-1}). Year-to-year variation of MAC_{cor} (MAAP) has been also examined and yearly MAC_{cor} (MAAP) was stable to within 10%. These results are shown in Fig. 7a and 9, Table 3, 8, and 11, and Fig. S6 in the Supplement.

Responses to Anonymous Referee #1

>> This manuscript addresses observations of black carbon aerosol in the Arctic. Knowing spatio-temporal variation of black carbon (BC) concentration and resulting light absorption is of general atmospheric and climatic interest as motivated by the authors. They present multi-annual and multi-site data sets of parallel measurements with two or more methods. This is a great effort and of large value as starting point for many studies. The methods and data treatment are sound and the manuscript is well written and it provides ample and important information towards consistent interpretation of observations made with different instrument types commonly deployed to quantify black carbon mass concentrations, and also provides approximate values for the conversion factor between light absorption coefficient and black carbon mass concentration, i.e. the MAC value, as far as achievable with filter-based methods. Therefore matching the scope of this journal.

This manuscript was initially submitted to the journal "Atmospheric Chemistry and Physics" (https://acp.copernicus.org/preprints/acp-2020-1190/) where it underwent a first round of reviews, which resulted in transfer to the current journal (Atmos. Meas. Techn.). I already provided a review at that stage (https://acp.copernicus.org/preprints/acp-2020-1190/acp-2020-1190-RC2.pdf). The authors already implemented these comments in an appropriate manner. Therefore, I only have few additional minor and technical comments listed below. Other than that, I can only stress that this valuable manuscript warrants publication.

Answer 1-1: Thank you for the positive comments on our manuscript.

>> Minor and technical comments:
Equation 1: The value(s) applied for f_{fil} should be reported. And/or the ratio "f_{fil}/MAC(COSMOS,SP2)", which is the final and only factor applied to measured b_{0} for inferring M_{BC}.

Answer 1-2: In the revised manuscript, we have added an equation (Equation 2) in Sect. 2.2.2.1 to explain how f_{fil} values are derived:

$f_{\mathrm{fil}}\,(Tr) = \frac{1}{[1.0796\,Tr + 0.71]\,B}$ with $Tr \geq 0.7$, where *Tr* is the filter transmission and *B* (= 1.397) is a scaling factor.

>> Caption of Figure 2: I suggest to add something along the line: "D_{m} is the mass equivalent diameter of bare BC or FeOx".

Answer 1-3: We have added the statement "D_m is the mass equivalent diameter of bare BC or FeO_x." in the caption of Fig. 2, as the reviewer suggested.

>> Section 2.2.3.3: It seems that actual wavelength of the MAAP instrument at Fukue was measured and found to be 639 nm, which is a very slight difference from 637 nm reported in the literature. It might be worthwhile to mention that 639 nm is an actual measured value.

Answer 1-4: In the revised manuscript, we have clarified that the peak wavelength of the light source of the MAAP at Fukue was measured to be 639 nm (Kanaya et al., 2013), which is very slightly different from the previously reported value (637 nm; Müller et al., 2011) in Sect. 2.2.3.3.

>> Figure 3c: choosing logarithmic instead of linear axis scaling might provide better visualization of performance in the lower concentration range, where the majority of data points appear. In the current variant it is not perfectly clear whether the value of the regression slope, which is driven by the high concentration data points due to fixing the axis intercept at the origin, only matches data well at high concentrations or at low concentrations too.
Choosing logarithmic instead of linear axis scaling might be preferable for several figures for the reasons laid down in the previous graph.

Answer 1-5: Following the reviewer's suggestion, we have examined the degree of agreement between M_{BC} (COSMOS) and M_{BC} (SP2) at Alert on a logarithmic scale (Fig. S1a in the Supplement). When M_{BC} (SP2) is relatively low ($M_{BC} < 10$ ng m^{-3}), which corresponds to the monthly-averaged M_{BC} ranges in summer at Arctic sites (Sinha et al., 2017), M_{BC} (COSMOS) tended to be higher than M_{BC} (SP2) by about 1–2 ng m^{-3}. This small absolute difference is consistent with the previously reported difference between M_{BC} (COSMOS) and M_{EC} at Barrow (Sinha et al., 2017). We have added these descriptions in Sect. 3.1.1.
Furthermore, the same analysis has been made for COSMOS-SP2 comparison at Fukue

in Sect. 3.5.1 (Fig. S1b in the Supplement). At Fukue, when M_{BC} (SP2) is lower than ~70 ng m^{-3}, M_{BC} (COSMOS) tended to be slightly higher than M_{BC} (SP2). A similar feature was previously reported at Cape Hedo in Japan (Ohata et al., 2019). The Cape Hedo site is located near the coast (i.e., the distance of this site to the coast is ~0.2 km) and the interference of submicron sea salt particles might contribute to this feature (Ohata et al., 2019). At Fukue, when maritime air mass is transported to the site, the relative abundance of sea salt particles to BC might be also enhanced possibly affecting the COSMOS measurements, although the distance from the site to the coast (~1.5 km) is slightly farther than for Cape Hedo. This feature was not clearly observed by a previous study at Fukue (Miyakawa et al., 2017). We have added these descriptions in Sect. 3.5.1.

>> Section 3.1.1: please put emphasis on the fact that quoted diameter values are BC mass equivalent diameter.

Answer 1-6: We have added the explanation that the diameter is reported in mass-equivalent diameter in Sect. 3.1.1, as the reviewer suggested.

>> Line 358: "These results show that on average, the agreement between MBC (COSMOS) and MBC (SP2) at Alert was within 10 %." – What exactly means "on average within 10%"?

Answer 1-7: The descriptions "on average" was not the statistically correct expression here. In Sect. 3.1.1, we have modified this part as follows: "Based on the slope value of the regression for whole M_{BC} ranges observed, the agreement between M_{BC} (COSMOS) and M_{BC} (SP2) at Alert was generally within 10 %."

>> Figure 4: It is very good that the instrument comparison is presented in different ways. Furthermore, distinction of performance in different concentration ranges as done in panels 4e and 4f is valuable. Having said so, the threshold is chosen at a very low level (2 ng/m3), which separates the data set somewhere in the single digit percentile range. I'd rather suggest to split somewhere between 15th and 25th percentile. In any case, the histogram of the low concentration data only should also be added (besides all data and high concentration data, which are already shown). This comment also applies to several other figures).

Answer 1-8: The threshold value of 2 ng m^{-3} actually corresponds to ~20th percentile at Alert (2018-2019) and at Ny-Ålesund for 1-h data. Instead of splitting somewhere between 15th and 25th percentile (reviewer's suggestion), we have split the histograms at 10 ng m^{-3} to focus on the low-concentration data, as shown in Fig. S2 in the Supplement. While the similar $\mathrm{MAC_{med}}$ values were obtained for data with M_{BC} (COSMOS) < 10 ng m^{-3} and for all dataset, the interquartile ranges of the b_{abs} (PSAP) / M_{BC} (COSMOS) ratios are larger for M_{BC} (COSMOS) < 10 ng m^{-3}. This result shows that the relative uncertainty becomes higher (lower) in summer (winter/spring) when the M_{BC} values tend to be low (high). We have added these descriptions in Sect. 3.1.2. Furthermore, the same analysis was made for all the other comparisons at Arctic sites, as shown in Figs. S3–8 in the supplement.

>> Line 393: "The babs, and therefore the MAC, for the PSAP and the two CLAP instruments (CLAP1, CLAP2) agree to within 8%". – Based on the values reported in Table 2, the difference appears to be somewhat larger?

Answer 1-9: In the revised manuscript, we have modified this part in Sect. 3.1.2 as follows: "The b_{abs}, and therefore the MAC, for the PSAP and the two CLAP instruments (CLAP1, CLAP2) agree to within 13 % at λ = 550 nm, indicating a small difference in the performance of these instruments." Thank you for pointing it out.

>> Caption of Table 3: please explain the variable "V" in the caption (i.e. inter-quartile range in relative terms). This comment may apply to other table captions too.

Answer 1-10: We have added the explanation of V_{MAC} in the caption of Table 3.

>> Line 462: check consistency of quoted V-value with Table 3.

Answer 1-11: We have corrected the V_{MAC} values in Sect. 3.2.1.

>> Line 475: PSAP derived absorption coefficients are around a factor of 1.5 larger than aethalometer derived absorption coefficients at Ny Alesund. Zanatta et al. (2016) reported a systematic difference the commercial and ITM variants of the PSAP, which is approximately in this range (Sect. 2.3.2 and Table 3 in their manuscript). Which variant of the PSAP is operated at the Ny Alesund site?

Answer 1-12: A custom-built PSAP (Krecl et al., 2007) was used at Ny-Ålesund and commercial ones were used at Alert and Barrow. We have clarified this point in Sect. 2.2.3.1.

It seems that Zanatta et al. (2016) reported correction factors for the custom-built PSAP through comparison with MAAP at 3 European background sites. In our revised manuscript, we have added analysis of COSMOS-MAAP comparison at Ny-Ålesund during 2017–2020 in Sect. 3.2.3, as described above in the "Major revisions". We have found that MAC_{cor} (PSAP) values are 17 % and 20 % larger than MAC_{cor} (MAAP) values for 1-h and 24-h data, respectively. The systematic difference of b_{abs} measured by the custom-built PSAP and MAAP was also reported by Zanatta et al., 2016, although their difference was much larger (more than 59 %) than that of our measurements at Ny-Ålesund. We have added these descriptions in Sect. 3.2.3.

>> Line 586: repetition of "at Alert".

Answer 1-13: We have corrected it in Sect. 3.6.

>> The authors discussion that BC mass from MAAP default instrument output systematically differs from their direct measurements due to a difference in factory default and actual MAC value. A brief discussion for BC mass from aethalometer default instrument output compared to their direct measurements should also be included, e.g. in Sect. 4.

Answer 1-14: In the revised manuscript, we have mentioned a manufacturer's suggested MAC value for Aethalometer in Sect. 2.2.3.2, which is given by 14625 / (λ [nm] $\times C_0$) [$m^2\ g^{-1}$]. The suggested MAC value is 7.1 $m^2\ g^{-1}$ for λ = 590 nm and C_0 = 3.5. This MAC value is considerably lower than the MAC_{cor} (Aethalometer; λ = 590 nm) values estimated in our study (12.5 $m^2\ g^{-1}$ at Alert and 10.2 $m^2\ g^{-1}$ at Ny-Ålesund for 1-h data). We have added these descriptions in Sect. 4.

Responses to Anonymous Referee #2

>> The presented article is an important contribution to the understanding of the relationship between black carbon and light absorption in Arctic regions. The methods used are appropriate for the available data and the results are well supported with figures and tables. In general, the manuscript is well written.
However, the reviewer disagrees with the authors in one particular point in the interpretation of the results. The reviewer recommends the manuscript for publication after considering the comments.

Answer 2-1: Thank you for the constructive comments. In response to the disagreed point, we try to make our assertion clear in the following answers.

>> General comment:
The authors use a filter-based absorption photometer to determine the mass concentration of black carbon. In line 136, the authors write: "We critically re-examine the concepts underpinning the use of filter-based instruments to estimate M_{BC}." This approach should have been discussed more critically.

The COSMOS is basically a filter-based absorption photometer that evaporates the volatile components by heating, so that only the absorption of a soot core is measured. This device was "calibrated" (compare lines 641 to 644) using an SP2 with one type of aerosol. This is critical because two different properties of the soot were compared. However, the determination of a value for MAC(COSMOS) for this aerosol type is correct. In this study, an application to arctic aerosols was tested. Although comparisons of M_{BC} with COSMOS and SP2 for Alert and Fukue Island stations agreed within the uncertainties, the conclusions should not be that the COSMOS using a universal MAC value is a "secondary reference device" for M_{BC}. As with any filter-based absorption photometer, the result should be labelled as "equivalent mass concentration" (see Petzold et al. 2013) using a specific MAC.

The comparison of two filter-based absorption photometers, where the volatile shell is removed in one device, thus shows the light absorption enhancement factor, which is an important component of the MAC. Furthermore, like all other filter-based absorption photometers, the COSMOS is subject to certain errors such as uncertainty due to particle penetration depth (see Nakayama et al. 2010).

In the view of the reviewer, in this study only the comparisons of a light absorption photometer with an SP2 strictly fulfil the conditions to derive MAC values. The other comparisons of COSMOS with other filter-based absorption photometers are rather studies on the enhancement factor.

The reviewer does not want to criticise the quality and overall results of this study, but to point out that soot in particular is a sensitive issue due to different metrics and the common nomenclature should therefore be followed very strictly. The reviewer proposes to refer to the facts described above and to denote the BC mass concentrations derived from COSMOS as "equivalent" mass concentrations.

Answer 2-2: We understand the importance of terminology recommended by Petzold et al. (2013) that clarifies the methods of BC measurements. However, considering the fundamental difference between COSMOS and the other filter-based absorption photometers, we would like to avoid using the term “equivalent black carbon (EBC)” for COSMOS measurements.

The COSMOS, which always uses a heated inlet and a PM_1 cyclone, is designed to measure BC mass concentrations (M_{BC}), while the other filter-based absorption photometers are designed to measure aerosol absorption coefficient (b_{abs}).

The present and previous studies (e.g., Kondo et al., 2011; Ohata et al., 2019) show that M_{BC} measured by COSMOS and SP2 generally agree to within 15 % in various environments including urban and remote atmospheres. These previous studies also evaluated the effect of particle penetration depth within a filter on COSMOS measurements and revealed that this effect is not significant under the typical variations of ambient BC size distributions. Furthermore, the previous studies show that variations of the mixing states of BC also have little effect on the COSMOS measurements. These results demonstrate that M_{BC} measured by COSMOS is generally traceable to M_{BC} measured by SP2 (i.e., “refractory BC mass concentrations” in the terminology by Petzold et al. (2013)) in various environments (not only the environment in Tokyo, where the standard COSMOS is “calibrated”).

These points are essentially different from the b_{abs} measurements by the other filter-based absorption photometers. The b_{abs} values measured by the other filter-based absorption photometers may be related to M_{BC}, but the potential contributions from brown carbon, mineral dust, and lensing effect of BC coatings are included in b_{abs} values. Therefore, linearity of the conversion from b_{abs} to M_{BC} can be much more uncertain for these instruments. This also leads to a difficulty for simply interpreting b_{abs} values reported by two filter-based absorption photometers with and without heated inlets as "enhancement factor" from lensing effects. For example, the time variations of yearly MAC_{cor} values for different filter-based instruments were not correlated at Ny-Ålesund (Fig. 7a), which means it would be difficult to discuss the enhancement factor in our study. This result is attributed to the inherent uncertainties of b_{abs} measurements for each instrument and different particle size cuts for each instrument (e.g., different contributions possibly from mineral dust).

For these reasons, we would like to avoid using the term "EBC" for COSMOS measurements, even though the COSMOS is also a filter-based absorption photometer. Our focus is not on the enhancement factor and we believe that MAC values estimated in this study can be used to obtain harmonized M_{BC} values from b_{abs} measured at Arctic sites.

>> Further comments:
Line 177: better use attenuation coefficient instead of extinction coefficient

Answer 2-3: We have modified it in Sect. 2.2.2.1, as the reviewer suggested.

>> Line 188: give values for f_{fil}

Answer 2-4: Following the reviewer's suggestion, we have modified this part in Sect. 2.2.2.1. Please see the Answer 1-2.

>> Lines 191 – 207: It is common for filter based instruments to report equivalent black carbon assuming an MAC. But in this case it is unfortunate to have to different MAC values when changing the filter type. The MAC is a property of the particle and not of the instrument. The reviewer suggests to attribute the changes of the sensitivity to the value of f_{fil} instead of MAC.

Answer 2-5: We agree with the reviewer's suggestion. In the revised manuscript, changes in the sensitivity due to filter types have been attributed to f_{fil} (i.e., B value) in Sect. 2.2.2.1.

>> Line 271: Magnetite is not a good proxy for iron oxides to the reviewer's knowledge. The imaginary part of magnetite seems to be very high. This value cannot explain the reddish colour of many minerals. Alternatively, the refractive indices of hematite could be considered (e.g. Sokolik et al. 1999).

Answer 2-6: Although hematite and magnetite are the major light-absorbing iron species, the FeO_x particles detected by SP2 in East Asia and in the Arctic were mainly magnetite (Moteki et al., 2017; Ohata et al., 2018; Yoshida et al., 2018; 2020). Therefore, we assume the refractive index of magnetite for Mie calculation in Sect. 2.2.2.2. We have clarified this point in the revised manuscript.

>> Lines 315–320: How large are the uncertainties of the correction value C0?

Answer 2-7: The uncertainty of C_0 is approximately 25 % (WMO/GAW, 2016). We have added this statement in Sect. 2.2.3.2.

>> Line 335: The MAAP was originally calibrated for the wavelength 670 nm. Was the correction factor of 1.05 taken into account to adjust the wavelength (see Müller et al. 2011)?

Answer 2-8: A correction factor of 1.05 due to the wavelength shift from the nominal value (Müller et al., 2011) was applied in this study. We have added this statement in Sect. 2.2.3.3.

>> Line 877: Line break between Petzold (2004) und Petzold (2005)

Answer 2-9: We have corrected it. Thank you for pointing it out.
