Responses to the Reviewers' Comments

Manuscript Number: acp-2020-1190
Title: Estimates of mass absorption cross sections of black carbon for filter-based absorption photometers in the Arctic

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. In accordance with suggestion by the editor and reviewers, we have decided to re-submit the manuscript to *Atmospheric Measurement Techniques* (AMT) as a new submission. Before the new submission, the comments will be fully taken into account and the original manuscript will be revised. Responses to the comments raised by the reviewers are listed below. More detailed responses including explanations of actual changes to the original manuscript will be submitted to AMT together with the revised manuscript.

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Responses to Anonymous Referee #1

>> The work of Ohata and colleagues presents MAC values of Arctic BC aerosol calculated from absorption coefficient and BC mass of various filter based photometer. Considering the extensive use of MAC values in modelling and observational studies, this topic is of great scientific interest. The manuscript has a dominant technical imprint, Thus, I wonder if the manuscript wouldn't be more appropriate for AMT. Alternatively, in order to publish on ACP, the atmospheric processes controlling the MAC variability should be asses in more details.
The paper has a very strong technical declination. In the introduction very few words are dedicated to explain why MAC is important in climatic studies and what are the current subsequent uncertainties in RF estimations. The subsections 3.2-3.4 merely report slope and correlation coefficient of babs and Mbc. In Section 3.5, the calculated MAC values are compared to previous studies but no reasoning on the year-to-year variability, seasonality and spatial variability is presented. Considering the remarkable amount of data, this work could provide more climatic relevant information. From this specific thought I encourage the authors to develop a climatic relevant discussion before resubmission to ACP or submit the manuscript to AMT.

Answer 1-1: We acknowledge that our manuscript focuses a lot on technical side. Considering the reviewer's suggestion, we will refer to the importance of MAC in the revised manuscript and re-submit it to AMT as a new submission.

>> The whole MAC calculation is based on the fact that COSMOS provides mBC similar to the SP2. It is thus important to prove that this assumption is valid for Arctic conditions Although Mbc(COSMOS) and Mbc(SP2) nicely agreed (slope 1.02) in Alert, the COSMOS-SP2 Mbc ratio in Fukue varies from 0.92 to 1.14. The last value being very close to previous SP2-COSMOS Arctic comparison (Zanatta et al., 2018), but being 10% higher than in Alert. What is the overall uncertainty, in Arctic conditions, of assuming that mBC (COSMOS) is equal to mBC (SP2)? A second critical assumption is that the SP2 provides accurate BC measurements. The SP2 might report considerably different Mbc values compared to the thermal-optical technique, as shown in Pileci et al. (2020). As a matter of fact, mEC values were 1.3-1.7 times larger than mBC (SP2) in Alert (Sharma et al., 2017). Moreover, Mbc(COSMOS) observed at Barrow might overestimate by 5 ng/m3 Mec in summer (Sinha et al., 2017). This value might be

negligible in continental locations, but very important for pristine summer Arctic condition (mEC<20 ng/m3). Considering that this is the first time a filter-based absorption photometer is used as BC mass concentration reference to calculate MAC values on relatively long time series and at multiple Arctic sites, the above mentioned points should be addressed. This, in turns, will increase the technical nature of the manuscript and makes it more suitable for AMT.

Answer 1-2: We have shown that the overall uncertainty of M_{BC} (COSMOS) is 15% based on the present and previous analyses of M_{BC} (COSMOS) and M_{BC} (SP2). First, the accuracy of our M_{BC} (SP2) has been estimated to be about 10% by our previous studies (e,g., Kondo et al. 2011). Second, we have shown that M_{BC} (COSMOS) agreed with M_{BC} (SP2) in the Arctic and Asia (e,g., Ohata et al., 2019). We will make this point clearer in the revised manuscript. Also, previously reported agreement and discrepancy between measurements of M_{BC} (SP2 or COSMOS) and EC will be mentioned in the revised manuscript.

>> S2: A description of the station and measuring period must be presented here rather than in the intro.

Answer 1-3: Descriptions of the observation sites and periods will be given in the method section in the revised manuscript, as the reviewer suggests.

>> S2.2.2: I strongly believe that this section does not fit very well in the storyline of the manuscript. Mostly because the actual impact of iron oxides is not quantified for absorption-photometer other than the COSMOS. As summarized in the conclusion, the effect of FeOx on COSMOS babs is even negligible. Thus, I strongly suggest removing this section.

Answer 1-4: It is very important to assess possible uncertainty of M_{BC} (COSMOS) caused by FeOx, because M_{BC} (COSMOS) is used as a reference for the comparisons made throughout this paper. We need to demonstrate the accuracy M_{BC} (COSMOS) even considering the effect of FeOx. We have also discussed possible errors of other types of absorption photometers based on this result. Therefore, this part remains as it is. More detailed analyses of the effect of FeOx on these instruments are beyond the scope of this study.

>> S3.2-3.3-3.4 These sections read like a list of numbers as such could be completely replaced by a table. I suggest the authors to rearrange these subsections not as function of instrument but rather as function of location. In this way instrument-error analysis, comparison with previous observations and climatologic aspects can be nicely organized for each station. Currently, Section 3.5 is a bit self-standing and is the only non-fully-technical section of the paper.

Answer 1-5: In the revised manuscript, the sub-sections will be rearranged as function of location, as the reviewer suggested.

>> L75-78: If I understand correctly, COSMOS is a filter based instrument. Reading Section 2.2.1, it appears that MBC is calculated from babs with a MAC. As every other filer-based absorption photometer. The description given here is a bit misleading.

Answer 1-6: The COSMOS is also a filter-based instrument, but designed to measure M_{BC}, not b_{abs}. The heated inlet of the COSMOS changes the original aerosol properties and this heating treatment makes b_{abs} (COSMOS) more proportional to M_{BC} (i.e., more stable MAC), as compared to the other filter-based instruments. As a consequence, unlike the other instruments, the absorption coefficient of untreated original aerosols is not provided by COSMOS. We will clarify this point in the revised manuscript.

>> L83-101: the description of stations does not belong here, but rather to the method.

Answer 1-7: The description of stations will be given in the method section, as the reviewer suggested.

>> L145: What MAC (COSMOS) is used in the present work, Sinha et al., 2017 or Irwin et al., 2015?

Answer 1-8: Depending on the filters used (Pallflex or HEPA), either MAC value was used in this study. This point is mentioned in the text.

>> L315-316: How much mass is underestimated by the EC-SP2 ? Isn't it a larger uncertainty source compared to calibration and density issues discussed afterwards?

Answer 1-9: The undetected mass fraction of BC was estimated to be about 5% by the lognormal fitting analysis, which is smaller than the uncertainties from the calibration and assumed effective density discussed here.

>> L318-332: How do the pre- and post- campaign calibrations agree? Considering the Aquadag scaling factor to be applied. Anyhow, this are just details considering that SP2-COSMOS comparison is still very good.

Answer 1-10: The difference of Aquadag calibrations made before and after the campaign was less than about 10%. This will be mentioned in the text.

>> L357: please use one single name: Pallas or Pallastunturi. Or define this in the methodology and adjust the following text and sections title.

Answer 1-11: In the revised manuscript, we will define the location name in the method section and use it in the following sections, as the reviewer suggests.

>> L363: the slope calculated for the 1h or 24h average observations?
>> L365: is 12.1 m2/g the average between MAC calculated at Pallas and at Fukue? Why should this be relevant?
>> L368-369: I would be careful in suggesting dust as potential source of interference since there is no evidence in this case.
>> L370: low by a factor of two IN ARCTIC CONDITIONS.
>> L429-437: write down somewhere the resulting Aethalometer MAC.
>> L500-505: FeOx is irrelevant in the context of this study. Such detailed description in the conclusion is not needed.

Answer 1-12: These points will be appropriately modified in the revised manuscript, except for the FeOx result.

>> F2: This figure, despite being really interesting, does not fit in the overall discussion of MAC variability in the Arctic, like all Section 2.2.2.

Answer 1-13: We consider this section is important as mentioned earlier, and it will remain as it is in the text.

>> F3,4,5,8,9,10,11: the paper has many figures, the time series shown n these figures

are not strictly needed, since all the information of interest are already contained in the scatterplots. I suggest the authors to remove the time series panels or at least move them in the supplementary.

Answer 1-14: We will show time series at one site as an example. The rest of the time series will be shown in the Supplement. Additional panels showing the variability of MAC (i.e., histograms of b_{abs} / M_{BC} ratio) will be included in these figures in the revised manuscript.

Responses to Anonymous Referee #2

>> 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. The only major question I raise concerns overlap with journal scope: "[…]The journal scope is focused on studies with general implications for atmospheric science rather than investigations that are primarily of local or technical interest […]". This manuscript here certainly has broader and longer coverage than "just local interest", whereas is has a strong technical focus (as appropriately reflected in the title). On the one hand, it does provide 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. On the other hand, it only provides limited additional results and interpretation around these quantities. Therefore, a journal such as Atmos. Meas. Tech. would certainly be a valid if not more suitable choice. Journal choice is to be answered by the handling editor. As for myself, I can only stress that this manuscript warrants publication after addressing a couple of minor comment listed below.

Answer 2-1: We acknowledge that our manuscript has a strong technical focus. Considering the suggestions by the editor and reviewers, we will revise the manuscript and re-submit it to AMT as a new submission.

>> Minor and technical comments: I recommend to emphasize the peculiar set-up and purpose of the COSMOS even more clearly, as readers may overlook the fundamental difference between COSMOS and e.g. aethalometer. The heater included in the COSMOS setup aims at denuding particulate matter internally mixed with BC from these BC cores. The very purpose of modifying the aerosol in this manner is to stabilize the mass attenuation coefficient of the BC deposited on the filter tape at a well-defined and constant value independent of original BC particle mixing state. This makes the

instrument response proportional to BC mass, thereby preparing the ground for a BC mass measurement with good accuracy. However, a consequence is that the COSMOS does not provide information on the light absorption coefficient of the untreated aerosol. By contrast, the aethalometer provides a priori provides a signal that is proportional to the absorption coefficient of the aerosol (neglecting dependence of the C-value on BC microphysical properties), whereas proportionality to BC mass concentration typically is smaller due to variations in MAC of BC (this point is made in the introduction as very motivation behind this manuscript). The consequence of stabilized versus variable MAC for filter-based measurements of equivalent black carbon mass could also be put in context of terminology recommendations by Petzold et al., 2013. I.e., it seems justified to omit the "equivalent" in COSMOS derived BC mass, however, the reason for it should be laid down.
Strictly speaking, another consequence of the previous comment is that the detour via absorption coefficient when inferring M_BC from b0 measured by the COSMOS instrument is obsolete (Eqs. 1-3). Instead, one can directly obtain M_BC as b0 divided by the mass-specific attenuation coefficient in the filter matrix. This is another way of conveying the message provided on lines 148 through 153. The absorption coefficient of the modified aerosol is not of interest, hence no need to go through absorption coefficient.

Answer 2-2: As the reviewer pointed out, an important difference between COSMOS and the other filter-based instruments is the heating treatment to the sampled air of the COSMOS. This treatment enables accurate measurements of M_{BC} (COSMOS) and this is different from "equivalent" BC mass concentrations estimated from untreated b_{abs} measurements. We will clarify this point in the revised manuscript, as per recommendations by the reviewer.

>> L234: The MAC of 6.6 m2/g for the MAAP is the default factory setting, which doesn't necessarily equal a recommendation. It could be said that this value applies to fresh uncoated BC, in which case the default output of the MAAP for equivalent BC mass concentration agrees well with actual BC mass concentration. It is also known to be too low for internally mixed BC thereby leading to eBC mass being larger than actual BC mass in such cases.

Answer 2-3: We will modify the pertinent part in the revised manuscript, as the reviewer suggests.

>> L263: No direct corrections were applied to compensate for non-linear dependence of attenuation on filter loading. Please specify whether the correction was not applied as not required, which can well be the case for aged aerosol. Plotting the absorption coefficient as a function of attenuation directly reveals whether correction is necessary. In principle, the correction could be implicitly included in the MAC value for deriving BC mass concentration. However, this would hinder interpretation and comparability of MAC values and it would make them dependent on setting chosen for filter tape advances.

Answer 2-4: We will modify the descriptions of Aethalometer in the revised manuscript, as the reviewer suggests.

>> Eq. 7 and text around it: I almost got confused by the two symbols B_abs and b_abs. I wouldn't necessarily say that they are different (line 272), which sounds like two different physical quantities, but rather just make the point that filter-based measurements of b_abs always remain tainted with some uncertainty for the reasons laid down on line 273ff. (Equation 7 isn't really needed then, is it?).

Answer 2-5: We will remove discussion on B_{abs} and clarify the uncertainty of b_{abs} measured with filter-based instruments in the revised manuscript, as the reviewer suggests.

>> Fig. 2: I suggest to emphasize that the Mie curves shown in this figure only apply for bare BC particles (or FeOx) particles. Accordingly, I suggest to be more specific regarding the notation of the diameter, i.e. D_BC and D_FeOx instead of D_m (the latter could be misinterpreted as total particle diameter in cased of internally mixed particles).

Answer 2-6: In the figure caption, we will clarify that this Mie calculation is made for bare BC and bare FeOx particles. We prefer to use D_m in this figure because it is clearly defined as mass equivalent diameter for bare BC particles in the SP2 Method section.

>> Fig. 4c: There seems to be a systematic high bias of the data points relative to the fit line in the range of lower concentrations (however, it is hard to just from a graph which heavily overlapping data points; please consider alternative visualizations). The slope of

a line fitted through the origin is always much more sensitive to data points at high concentrations than to those at low concentrations. Therefore, I suggest as alternative to calculate the ratio between the two measurements for every data point and then run stats on these ratios (and show them as histogram). Comparing the resulting mean ratio with the fitted slope gives an idea on the robustness of the analysis (note: some pre-averaging before taking the ratio may be required in case of poor signal-to-noise ratio). And the width of the histogram of ratios carries information that is easier to interpret than the correlation coefficient, which is a blend of variations in ratio and variations in absolute concentration. The same should be done for the analysis of other "mean ratios".

Answer 2-7: Additional analysis suggested by the reviewer will be done for all the data presented in this study. In the revised manuscript, histograms of the ratio will be included in each figure and most of the time series panels will be moved to the Supplement. We will discuss the variabilities of MAC, based on the median ratio and 25-75 percentile ranges of the ratio.

>> Figures 5a, 9a etc: please find better ways to visualize these data. As is, the black traces simply obscure the red traces.
>> Table 1 caption: Looks like a type for the MAAP wavelength.

Answer 2-8: These points will be appropriately modified in the revised manuscript.

>> Table 8: This table shows MAC values across the wavelength spectrum covered by the aethalometer. Such values must only be presented with very clear caveats on how to use and interpret them. The light absorption coefficient at near UV wavelength is, simply speaking, the sum contributions from black carbon and brown carbon. Therefore, the MAC reported here at e.g. 370 nm is not a property of the BC particles, instead it simply is a conversion factor that provides correct BC mass concentration "on average". By contrast, the MAC reported at red and near IR wavelength, where BC dominates absorption, is both a property of the BC particles and a conversion factor to provide correct BC mass concentration. Or in other words, the apparent MAC at near IR should not change when a biomass burning plume passes by, whereas the apparent MAC (absorption divided by BC mass) at near UV wavelengths should increase. Given this, it is actually surprising that the correlation coefficients are found to be comparable in these two wavelength ranges. Does this indicate that brown carbon plays a minor role or that it makes a rather constant relative contribution? I also suggest that longer

wavelength are preferentially to be used to infer eBC mass concentrations whenever possible, whereas the measurements at shorter wavelength should only be used to infer the spectral dependence of the absorption coefficient, which gives hints on presence of brown carbon.

Answer 2-9: In the revised manuscript, we will clarify that these wavelength-dependent MAC values are the simple conversion factors to obtain average M_{BC} from b_{abs} (Aethalometer). As the reviewer points out, it is a notable feature at Alert that the impact of the other light-absorbing aerosols such as brown carbon seems to be very small. We will add these descriptions in the revised manuscript.
