Response to Anonymous Referee #2

## *Major comments:*

***1) The authors have ignored the recommendations as proposed by Petzold et al. (2013), recommendations that are generally accepted by the scientific community, on how black carbon (BC) should be reported when derived from instruments that measure light attenuation, i.e. filter based or photoacoustic sensor. BC derived from these techniques should be reported as equivalent BC, or eBC. If or when this paper is resubmitted, the title should reflect clearly that it is eBC that is being discussed, not BC.***

Response: Thanks for your recommendation. The term 'black carbon' (BC) was changed into 'equivalent BC'. As suggested by Petzold et al. (2013), equivalent BC was abbreviated to "EBC" in the revised manuscript.

***2) A large fraction of the introduction is devoted to the importance of BC for climate change due to radiative forcing. What the authors fail to understand is that in the context of their study, the corrections to the MAC that they are proposing is completely irrelevant. Sensors that measure light absorption like the Aethalometer, are already providing the necessary information that is relevant to climate change, i.e. it is not the mass concentration that is important it is the optical cross section. I will address this further below with respect to the mixing state of BC, but the primary point is that the mass concentration of BC is not important when doing radiative transfer calculations if you already have the primary measurements of the coefficients of scattering and absorption. The authors also mention that BC might be efficient CCN or IN, both true statements but again irrelevant with respect to their study. Hence, the introduction needs to be completely rewritten to explain the real relevance of the current study, and that is to set some error bounds on eBC derived from Aethalometer measurements and NOT a cutting edge, new methodology that will in any way improve the accuracy of such measurements.***

Response: Thank you for your comments. The introduction was rewritten to emphasize the importance of MAC correction when deriving EBC from light absorption based on filter-based instrument.

***3) This study should be written up as a detailed analysis of the uncertainties in the MAC related to the mixing state of BC, i.e. the refractive indices, real and imaginary, the wavelength of incident light, and the relative sizes of the core and shell. Secondly, in the introduction, it should be made quite clear how this analysis differs from the many others that have already been published.***

Response: Thank you for your comments. Detailed uncertainty analysis, including refractive indices, was input in section 5 of the revised manuscript. The discussions included uncertainties of MAC caused by using idealized core-shell model, using constant BC-containing particle fraction, and variation of RI. The influence of sizes of core and shell were discussed in the uncertainty analysis. With respect to wavelength, EBC is derived from $\sigma_{ab}$ at a specific wavelength, namely 880 nm. At 880 nm, aerosol absorption is mainly from BC (Ramachandran and Rajesh, 2007). At shorter wavelength, absorption of organic carbon is not negligible any more, leading to difficulty of extracting BC absorption from total absorption. Therefore, the wavelength dependency of MAC was not discussed since the main goal of this study was to derive EBC, and the organic component was not included in this study.

The variation of MAC due to mixing state was not considered when deriving EBC from $\sigma_{ab}$ in the previous studies, difference between this study and previous studies was input in the text. The motivation of this study was to propose a modified approach considering variation of MAC due to mixing state.

***4) The methodology that is discussed in this paper is being promoted as a way to derive a more accurate EBC but this is misleading because in order to apply this you need a lot of additional complementary information about the size distribution of the BC, the fraction of particles that are mixed with BC, etc. If you had all the necessary information to begin with, then you wouldn't even need to try and derive EBC using***

***a variable MAC because you would already have enough information to estimate BC without the light absorption instrument. This should be made quite clear in a resubmission of this paper.***

Response: Thank you and we agree with your comments. Filter based instruments such as AE33 are used in operational networks worldwide due to their advantages such as low cost, simplicity of operation, less maintenance and convenience for data processing. However, the EBC measured by AE33 is not accurate because it uses a constant MAC. The motivation of this study was to propose a method to consider the variation of MAC to make EBC measured by AE33 more accurate.

The size distribution of BC required in this study was size distribution of absorption measured by aethalometer, which could be achieve by DMA in tandem with AE51. As for the number fraction of particles mixed with BC ($N_{BC}$), it was a reference value in this study. Uncertainty analysis showed that derived EBC was not that sensitive to $N_{BC}$.

***5) It is my opinion that the modeling that is being discussed with this study has as much importance for setting the error bars on light absorption derived from the filter-based measurements as for setting error bars for deriving eBC. There are many corrections that have been proposed to adjust the light absorption measurements for the impact of overloading, filter matrix effects, etc., but perhaps the results from the current study could also be used to establish how mixed state BC leads to under/over estimates of the absorption coefficient. The authors should give this serious consideration if they want their study to have more relevancy than it does in its current state.***

Response: Thank you for your comments. As mentioned above, the filter-based instruments such as AE33 are widely used in operational networks due to their advantages. This study aimed to investigate the role of variation in MAC on the derived EBC by AE33. Besides correction to EBC, more discussions about the effect of mixing state on the absorption coefficient were input in the text.

## *Specific comments:*

***1) Line 1, "determination of black carbon mass concentration from aerosol light absorption using variable mass absorption cross-section". Here and from here on out this is to be called "equivalent black carbon".***

Response: "Black carbon" was changed into "equivalent black carbon" in the text.

***2) Line 10, "the mass absorption cross-section (MAC) is a crucial parameter for converting light absorption coefficient ($\sigma_{ab}$) to mass equivalent BC concentration ($m_{BC}$)". Here and forward, change this into eBC.***

Response: $m_{BC}$ was modified as EBC in the revised manuscript here and forward.

***3) Line 11, "traditional filter-based instrument, such as AE33, uses a constant MAC of 7.77 $m^2/g$ to derive $m_{BC}$, which may lead to uncertainty in $m_{BC}$." Add the wavelength that this is for.***

Response: Thanks for your recommendation. wavelength of 880 nm was appended to 7.77 $m^2/g$ in the text.

***4) Line 22, "because of its highly absorbing properties in the visible spectral region, BC is considered to have a significant influence on global warming." By definition, "black" means all wavelengths, not just visible.***

Response: "In the visible spectral region" was deleted in the revised manuscript.

***5) Line25, "despite the importance of BC to climate, the global mean direct radiative forcing of BC particles still spans over a poorly constrained range of 0.2 – 1 $W/m^2$." Please clarify. I don't understand what this means.***

Response: This sentence was removed from the text to avoid ambiguity.

***6) Line 29, "to fully evaluate the influences of BC particles on solar radiation or***

***precipitation, more precise measurements of BC mass loading in the atmosphere are required." This is an incorrect argument for saying that more accurate measurements of BC are needed because instrument like the aethalometer measure light absorption directly without the need for converting it to eBC. With respect to the impact on clouds, what is needed is better measurements that can show just exactly how BC does form droplets or ice. Hence, an accurate MAC is not relevant for these impacts. The only impact that BC mass has that is important is on health or damage to building surfaces.***

Response: Thanks for your comments. This sentence was deleted in the revised manuscript to make the content more relevant to the correction to EBC.

***7) Line 31, "a variety of techniques have been developed to measure real-time BC mass concentrations." None of these measure BC mass concentrations.***

Response: These absorption measurement techniques was removed and instruments measuring BC mass concentration, such as SP2, OCEC, SP-AMS, was input in the text.

***8) Line 37 and line 38, "it measures real-time BC concentrations by converting the absorption coefficient ($\sigma_{ab}$) into mass equivalent BC concentrations ($m_{BC}$) through a constant mass absorption cross-section (MAC), which provides the BC absorption per unit mass." AE33 does not measure BC concentrations and the wavelength dependency of MAC has to be discussed at the very beginning.***

Response: "BC concentrations" was changed into "BC absorption" and "880 nm" was appended to MAC.

***8) Line 53, "a wide range of MAC (2 – 25 $m^2/g$) has been reported in previous studies." This range is due to wavelength dependency. What is the range for a single frequency, especially for the one being used here?***

Response: Thanks for your comments. This sentence was changed into "A wide range of MAC has been reported in previous studies. For instance, Bond and Bergstrom (2006) reported MAC at 550 nm varying from 1.6 $m^2/g$. Sharma et al. (2002) reported MAC

at 880 nm varying from 6.4 to 28.3 $m^2/g$." to make wavelength dependency clear.

***9) Line 62 to 64, "the hypothetical BC mixing state affects the corresponding absorption properties. It is critical to propose a method to infer $m_{BC}$ from light attenuation measurements considering aerosol size and the process by which BC aerosols mix with other aerosol components." Is this being proposed, completely independent of any other information about the environment?***

Response: The mixing state of BC was one of the important factors that affect the absorption properties of BC-containing particles. The size of aerosol was required to estimate the effect of mixing state on BC absorption. It was dependent on other information, such as refractive index (RI). The influence of RI on the uncertainty of MAC was discussed in the later content. This sentence was removed to avoid ambiguity.

***10) Line 70, "this modified method measures size-resolved $m_{BC}$ accurately and improves the evaluation of BC radiative forcing." How can a theoretical model "measure" eBC?***

Response: Thanks for your comment. "Measure" was modified into "estimate".

***11) Line 77, "the DMA (Differential Mobility Analyzer)-SP2 system measurements to determine the number fraction of BC-containing aerosols and to compare <u>AE33</u> and the three-wavelength photoacoustic soot spectrometer (PASS-3) were conducted in Taizhou." What wavelength of AE33 are compared?***

Response: The wavelengths used for comparison between AE33 and PASS-3 were 405 nm, 532 nm and 781 nm. 405 nm, 532 nm and 781 nm are the wavelengths PASS-3 measures. The wavelengths AE33 measures are 370 nm, 470 nm, 520 nm, 590 nm, 660 nm, 880 nm and 950 nm. For AE33, 405 nm, 532 nm and 781 nm were calculated with wavelengths pairs of (370 nm, 470 nm), (520 nm, 590 nm) and (660 nm, 880 nm) through Ångström relationship:

$$\frac{\sigma_{\mathrm{ab}}(\lambda_1)}{\sigma_{\mathrm{ab}}(\lambda_2)} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-\alpha_{ab}},$$

$$\sigma_{\mathrm{ab}}(\lambda) = \sigma_{\mathrm{ab}}(\lambda_1)\left(\frac{\lambda}{\lambda_1}\right)^{-\alpha_{ab}}.$$

Detailed description can be found in (Zhao et al., 2020). Wavelengths (405 nm, 532 nm and 781 nm) as well as the reference was appended to the manuscript.

***12) Line 84, "Meanwhile, from March 21, 2017 to April 9, 2017 at the Peking University site, the results from simultaneous measurements from AE51 (model 51, microAeth, USA) and AE33 were compared." What wavelength?***

Response: The wavelength of AE51 was 880 nm. Wavelength of "880 nm" was appended to "AE51 and AE33".

***13) Line 99, "the dry aerosol scattering coefficients at 525 nm were measured simultaneously by an integrated nephelometer (Ecotech 100 Pty Ltd., Aurora 3000) with a flow rate of 3 L/min." How does this wavelength correspond to the Aethalometer wavelengths?***

Response: The dry scattering coefficient at 525 nm here was used as a proxy of pollution level. At a specific wavelength, higher (lower) dry scattering coefficient could indicate a relatively polluted (clean) episode. Dry scattering coefficient at 525 nm was not used for comparison with light attenuation measured by aethalometer. "As an indicator of pollution level" was appended to the sentence.

***14) Line 105, "factor k was set as 0.004 and ATN is the measured light attenuation when particles load on the fiber filter of AE51." Where does this value come from?***

Response: "k = 0.004" was from the work by Zhao et al. (2019). "(Zhao et al., 2019b)" was appended to "0.004" in the manuscript.

***15) Line 114 – 115, "according to the measurements from Taizhou, only 17% of the ambient particles that contained BC averagely for bulk aerosol populations." This is***

***an incomplete sentence.***

Response: this sentence was modified into “according to the measurements from Taizhou, only 17% of the ambient particles contained BC averagely for bulk aerosol populations.”.

***16) Line 116, “we adjusted the measured wavelengths of AE33 to the measured wavelengths of PASS-3 (405 nm, 532 nm, and 781 nm).” How the adjustment is made?***

Response: 405 nm, 532 nm and 781 nm are the wavelengths PASS-3 measures. The wavelengths AE33 measures are 370 nm, 470 nm, 520 nm, 590 nm, 660 nm, 880 nm and 950 nm. They are not consistent. For comparison, the wavelengths of AE33 were interpolated to the wavelengths of PASS-3 in this study. Specifically, For AE33, 405 nm, 532 nm and 781 nm were interpolated with wavelengths pairs of (370 nm, 470 nm), (520 nm, 590 nm) and (660 nm, 880 nm) through Ångström relationship:

$$\frac{\sigma_{\mathrm{ab}}(\lambda_1)}{\sigma_{\mathrm{ab}}(\lambda_2)} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-\alpha_{\mathrm{ab}}},$$

$$\sigma_{\mathrm{ab}}(\lambda) = \sigma_{\mathrm{ab}}(\lambda_1)\left(\frac{\lambda}{\lambda_1}\right)^{-\alpha_{\mathrm{ab}}}.$$

More detailed description could be found in (Zhao et al., 2020). The interpolation method was added to the manuscript. “Adjusted” was changed into “interpolated”.

***17) Line 182 – 183, “it should be pointed out that the retrieval algorithm of BCPMSD is based on the assumption that BC-containing particles of a fixed diameter are all core-shell mixed and the corresponding $D_{BC}$ for a specific $D_{particle}$ is same.” A major assumption. Where is the sensitivity study that evaluates this assumption? This uncertainty analysis belongs in the main text, not in a supplement.***

Response: Thanks for your comments. The sensitivity study from the supplement was moved to the section 5.1 in the revised manuscript.

**Reference**

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S. M., Baltensperger, U., Holzer-Popp,

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Ramachandran, S., and Rajesh, T. A.: Black carbon aerosol mass concentrations over Ahmedabad, an urban location in western India: Comparison with urban sites in Asia, Europe, Canada, and the United States, J. Geophys. Res.-Atmos., 112, 19, 10.1029/2006jd007488, 2007.

Zhao, G., Tao, J. C., Kuang, Y., Shen, C. Y., Yu, Y. L., and Zhao, C. S.: Role of black carbon mass size distribution in the direct aerosol radiative forcing, Atmospheric Chemistry and Physics, 19, 13175-13188, 10.5194/acp-19-13175-2019, 2019.

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