Referee comment on "Determination of the multiple-scattering correction factor and its cross-sensitivity to scattering and wavelength dependence for different AE33 Aethalometer filter tapes: A multi-instrumental approach" by Jesús Yus-Díez et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-46-RC1, 2021

Answer from the authors to referee #1

On behalf of all the authors of the manuscript, we would like to acknowledge the work done in the review as well as the suggestions and comments for improving the study.

Hereafter we will answer and resolve the comments. Any minor comment, typo or writing corrections will be directly corrected in the manuscript.

General comments

Offline absorption coefficient measurements were done using the PP_UniMI techique. However, it is not clear if the samples could have been affected between sampling time and measurement in Milan. The authors should provide more details on how the samples were handled and how long it took from sampling to analysis. One of the issues is, for example, that brown carbon could have been modified on the filter, thus affecting absorption wavelength dependence.

The measurements in Milan were done between May and July 2019 (cf. table below) whereas the MAAP measurements were performed between October 2018 and June 2019 in BCN, between June 2018 and December 2018 at MSY, and between June 2018 and November 2018 at MSA. Therefore, the time between MAAP sampling and PP_UniMI measurements is the time between the time each spot was measured at the station and the PP_UniMI measurements. The samples, once extracted from the MAAP filter roll were stored in a fridge. Each spot was separated and saved in a petri dish and sent to Milan, where measurements started as soon as the samples arrived.

Station	Measurement period
Montsec	22/05/2019 to 29/05/2019
Barcelona	20/06/2019 to 25/06/2019
Montseny	26/07/2019 to 09/07/2019

Indeed, brown carbon concentrations could have been modified on the filter during this process.

We have specified this issue in the manuscript in the lines 200-204:

"The time elapsed between the MAAP measurements and the MAAP spots analysis with the PP_UniMI in Milan varied between one year and one month. Once selected and cut, each MAAP spot was stored in a petri dish in a fridge and then sent to Milan. We assumed that there were no major particle losses affecting the measured optical properties, although some volatile compounds could have been evaporated over the period.

Section 3.1: How valid is a comparison between C values for the different tapes when the measurements were done at different times, with different aerosol

conditions/properties?

Given the length of the measurement periods (Fig. S1), the measurements performed at the three stations covered a wide range of aerosol particles properties, thus we expect that the measurements well represented the aerosol conditions typically observed at the measurement stations. Therefore, we assume that the reported C values represented well the different filter tapes characteristics with minimal influence from different aerosol conditions.

We have added a sentence at the end of the last paragraph of Section 2.3.1 (lines 286-289) to point this out:

"Given the length of the measurement periods, we assumed that the AE33 filter tapes considered here were characterized under a wide range of aerosol particle properties typically observed at the measurement stations and that the non-simultaneity of AE33 measurements with the two filter tapes did not prevent the comparison between the obtained C.

I understand the authors use a 3- or 7-wavelength log-log fit to retrieve AAE from Aethalometer measurements. However, this method is inaccurate. Please check https://doi.org/10.1140/epjb/e2004-00316-5

We have, indeed, used a 7- wavelength log-log fit to retrieve AAE from the AE33. As commented by the referee, and as shown in Goldstein et al. (2004), this method of fitting the data introduces a greater error to the fit result. However, the fitted value remains fairly similar (Table 1 in Goldstein et al., 2004). Thus, since it is common practice in the aerosol community to derive the Ångström exponents through the log-log fit (e.g. Bergstrom et al., 2007; Ealo et al., 2016, Bernardoni et al., 2017), we prefer considering this method as a valid option for our work.

One of the key arguments of the article is the wavelength dependence of the multiple scattering correction factor, C, at a remote station. Is this finding specific to remote stations? To remote stations subject to Saharan dust influence? Or only to this particular station? Please comment. It would be useful if the authors can provide other references showing similar findings.

We acknowledge the reviewer for its comment and interest on the application to other stations. We have modified the structure of the manuscript and the approach to the analysis. We have specified its limitations and the possible extrapolations to other measurement stations. The main driver of the changes in the C value is its cross-sensitivity to scattering, therefore, remote stations with high SSA values, such as MSA, not only those affected by Saharan dust intrusions, may find of interest the results. Also, other regional background stations, when SSA is high, such as MSY, may find that the cross-sensitivity to scattering is having an impact on their measurements.

As far we know, no other similar study has been previously carried out that can show similar results; but we encourage the community to do so if they have the ability to carry out such intercomparison.

We have included a few remarks at the conclusion of the manuscript at lines 564-582 with the aim of providing a clearer guide:

"In summary, based on the results herein presented, the absorption coefficients from AE33 data can be corrected with different degrees of confidence depending on the information available to estimate the multiple scattering parameter C:

- A tailored dynamic multiple scattering parameter can be obtained if on-line simultaneous reference absorption measurements are available. In this case, a dynamic C with high

temporal resolution can be obtained, allowing an in-situ correction of AE33 data and allowing studying for example diel/seasonal cycles of the multiple scattering parameter. Here we used on-line MAAP absorption measurements at one wavelength for the determination of a dynamic C at the same MAAP wavelength.

- If independent reference multi-wavelengths absorption measurements are available, then the dependence of the multiple scattering parameter with wavelengths can be studied. Here we determined the wavelength dependence of the multiple scattering parameter by using the polar photometer (PP_UniMI) off-line absorption measurements performed on the MAAP filter spots and by comparing the off-line PP_UniMI measurements with AE33 attenuation data integrated over the MAAP filter spots time stamp.
- If reference absorption measurements are not available for the experimental determination of the C, then the average values of the multiple scattering parameter provided here for three different measurement stations can be used as reference.
- If both independent reference absorption measurements and scattering measurements are available, then the cross sensitivity to scattering of AE33 data can be determined by studying the relationship between C and single scattering albedo (SSA). In this case, a parameterization can be obtained relating C and SSA.
- If SSA measurements are not available, this work provides parameterized formulas that allow calculating C over a wide range of SSA values."

Specific comments

L172: Please remove comma after "Thermo".

Done.

L174: Please avoid starting a sentence with an acronym.

The sentence now starts as: "Black carbon, eBC, […]".

L188: When were the PP_UniMI measurements done?

As stated in the previous table, PP_UniMI measurements were performed between May and July of 2019.

Station	**Measurement period**
Montsec	22/05/2019 to 29/05/2019
Barcelona	20/06/2019 to 25/06/2019
Montseny	26/07/2019 to 09/07/2019

We have included this information in the lines 200-204 of the manuscript:

"The time elapsed between the MAAP measurements and the MAAP spots analysis with the PP_UniMI in Milan varied between one year and one month. Once selected and cut, each MAAP spot was stored in a petri dish in a fridge and then sent to Milan. We assumed that there were no major particle losses affecting the measured optical properties, although some volatile compounds could have been evaporated over the period.

L214: Please remove comma after "Pty".

Done.

L275: Please detail how you calculated the AAE.

The AAE was calculated through a log-log linear fit to the 7 absorption wavelengths measured by the AE33. We include a clarification of this in the manuscript lines 294-296, in Sect. 2.3.2:

"The absorption coefficients from the PP_UniMI were inter/extrapolated to the seven AE33 wavelengths using the attenuation Ångström exponent, obtained through a log-log from the PP_UniMI absorption measurements."

Fig 2: You mentioned previously that high SSA was observed in summer season and it increases with C but here it is shown that the highest C values are reached in winter, at least for MSY and MSA.

We can see in Fig. 2 that on average the annual cycle of the C at MSY and MSA showed an increase during the summer period, corresponding with an increase of the SSA (new Fig. S7). At MSY, the annual cycle of the M8020 C was less pronounced but it still mirrored the variability of the SSA observed during the period with the M8020 filter tape. (Fig. S7**.)**

Here we present the now Fig. S7, which shows the SSA seasonal analysis.

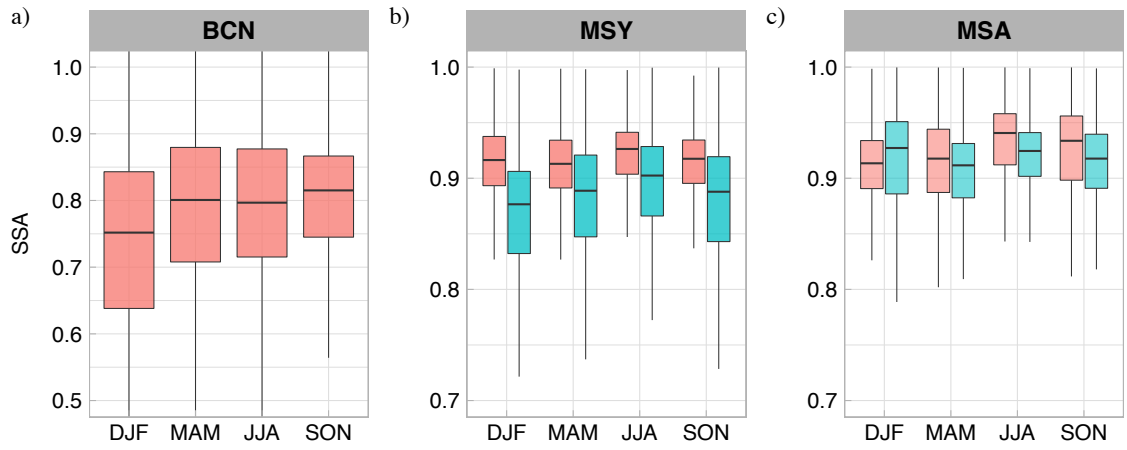

Figure S7. Seasonal evolution of the SSA at a) BCN, b) MSY and c) MSA measurement stations for both M8020 and M8060 filter tapes. The box plot boxes show the range between the first and third quartile (IQR) with the median value for each season distribution represented by the inner line; the maximum whisker length is proportional to 1.5·IQR.

L370: I guess the "3.3" is a typo.

Corrected the typo.

L423: Could you please provide more references here? Other remote sites with SSA > 0.95?

We have increased the references at the line to:

"(Collaud Coen et al., 2004; **Gyawali et al., 2009**; **Andrews et al., 2011**; Pandolfi et al., 2014a, 2018; **Schmeisser et al., 2018**; **Ferrero et al., 2019**; **Laj et al., 2020**)"

With the new references being:

1 Gyawali, M., Arnott, W. P., Lewis, K. & Moosmüller, H. In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of absorbing and non-absorbing organic coatings on spectral light absorption. *Atmos. Chem. Phys.* **9**, 8007–8015 (2009).

2 Andrews, E. *et al.* Climatology of aerosol radiative properties in the free troposphere. *Atmos. Res.* **102**, 365–393 (2011).

3 Schmeisser, L. *et al.* Seasonality of aerosol optical properties in the Arctic. *Atmos. Chem. Phys.* **18**, 11599–11622 (2018).

4 Ferrero, L. *et al.* Aerosol optical properties in the Arctic: The role of aerosol chemistry and dust composition in a closure experiment between Lidar and tethered balloon vertical profiles. *Sci. Total Environ.* **686**, 452–467 (2019).

5 Laj, P. *et al.* A global analysis of climate-relevant aerosol properties retrieved from the network of GAW near-surface observatories. *Atmos. Meas. Tech.* **13**, 4353–4392 (2020).

Bibliography.

1. Goldstein, M. L., Morris, S. A. & Yen, G. G. Problems with fitting to the power-law distribution. *Eur. Phys. J. B* **41**, 255–258 (2004).
2. Bergstrom, R. W. *et al.* Spectral absorption properties of atmospheric aerosols. *Atmos. Chem. Phys.* **7**, 5937–5943 (2007).
3. Ealo, M. *et al.* Detection of Saharan dust and biomass burning events using near-real-time intensive aerosol optical properties in the north-western Mediterranean. *Atmos. Chem. Phys.* **16**, 12567–12586 (2016).
4. Bernardoni, V., Valli, G. & Vecchi, R. Set-up of a multi wavelength polar photometer for off-line absorption coefficient measurements on 1-h resolved aerosol samples. *J. Aerosol Sci.* **107**, 84–93 (2017).