Reviewer #2

Color legend:

- Reviewer's comments in black
- *Authors' responses in red italic*

The paper presents a numerical study on the optical properties, especially absorption, of black carbon based on different assumptions on the mixing states and regional aerosol models. The AQMEII-3 results are used as the input aerosol properties, and a generalized off-line tool FlexAOD provides a unified method to give the optical properties, which are compared and evaluated by the AERONET results. By connecting the modeled aerosol properties and the observed optical properties, the manuscript presents a unique aspect to understand the absorption of BC aerosols. The paper is well designed and well organized, and I recommend it to publish in ACP after revision. The following lists my comments on the manuscript.

We would like to thank the reviewer for the insightful comments delivered on our work. We believe that addressing them made the results more robust and general with respect to the initial submission. We performed a number of additional sensitivity tests (see new section 3.1 in the manuscript) and comparison to measurements in order to clarify the detailed points, as illustrated below.

General comments:

1. As mentioned by the authors, the underestimation on total mass is the primary reason for the low AOD obtained, and this can neither be ignored on discussing the absorption properties of BC. This becomes critical because the BC concentration, which is also the primary factor for absorption estimation, may be significantly over or underestimated. Considering the completely different profile results presented in Figures 2 and 3, the relative performances of the models would definitely influence the absorptions.

In summarizing the results, we tried to limit the influence of model bias in terms of mass and size distribution filtering out scenes with a large bias in terms of effective radius and volume concentration in the final analysis. However, the suggestion of the reviewer may help to present the results in a more clear way, and we thus compiled the following table with correspondences of AERONET stations and monitoring stations providing aerosol speciation measurements in Europe and North America:

AERONET Site	Latitude	Longitude	Database	Site	Latitude	Longitude
Europe (20 sites)						
Andenes	69.28	16.01	-			
Barcelona	41.39	2.12	EMEP	Montseny	41.77	2.35
Brussels	50.78	4.35	-			
Brujassot	39.51	-0.42	-			
Ersa	43.00	9.36	-			
Huelva	37.02	-6.57	-			
Karlsruhe	49.09	8.43	-			
Kyiv	50.36	30.50	-			
Lecce University	40.36	18.11	-			
Malaga	36.72	-4.48	-			
Messina	38.20	15.57	-			
Moldova	47.00	28.82	EMEP	Leova II	46.49	28.28
Munich University	48.15	11.57	-			
OHP Observatoire	43.94	5.71	-			
Palencia	41.99	-4.52	EMEP	Campisabalos	41.28	-3.14
Salon de Provence	43.61	5.12	-			
Sevastopol	44.62	33.52	-			

Thessaloniki	40.63	22.96	-			
Toravere	58.26	26.46	-			
Toulon	43.14	6.01	-			
North America (9 sites)						
Bozeman	45.66	-111.05	-			
BSRN BAO Boulder	40.05	-105.01	IMPROVE	Rocky Mountain NP	40.28	-105.55
Chapais	49.82	-74.98	-			
Easton Airport	38.81	-76.07	IMPROVE	Washington DC	38.88	-77.03
Egbert	44.23	-79.75	IMPROVE	Egbert	44.23	-79.78
El Segundo	33.91	-118.38	IMPROVE	San Gabriel	34.30	-118.02
Halifax	44.64	-63.59	-			
Railroad Valley	38.50	-115.96	IMPROVE	Great Basin NP	39.01	-114.22
Saturn Island	48.78	-123.13	IMPROVE	Olympic	48.01	-122.97

For Europe, a reasonable correspondence is found only for 3 stations, while in North America for most stations (6 on 9). We collected the available observations for the year 2010 at these stations and carried out the comparison with concentrations of aerosol species at the first model level. The results are summarized in the supplementary revised Table S1 and the new Figure S7, and briefly discussed in section 2.2 in a new paragraph:

"Additional indications about models skills are gathered from the comparison with PM composition measurements available near the AERONET stations, for which we have stored the simulated PM speciation profiles of AQMEII models. The comparison is carried out at 3 stations over Europe and 5 stations over North America, and results summarized in Table S1 and Figure S7. Over North America, the two models have yearly average values mostly within ±1 µg m-3. Over Europe, most values are also within the same range, but there is a tendency toward overestimation of inorganic secondary species (sulfate, nitrate, ammonium) and black carbon, and underestimation of the organic carbonaceous fraction."

Given the very limited number of stations, we haven't emphasized the comparison to more than an additional indication on models skills.

We then used this, although partial, information to evaluate the impact of aerosol mass model bias on the main conclusions, as reported in a paragraph of the new section 3.1:

"We run the tests in the two extreme and more physically relevant mixing assumption adopted above, i.e. external mixing (EXT) and core-shell (CSBC). The first subset of tests is related to the influence of the model bias in terms of aerosol species mass. From Table S1, we estimate that model IT2 overestimates sulfate by a factor of 3, ammonium and BC by a factor of 2, while nitrate and organic fraction is in the range of observations. The tests 2-4 thus explore the effect of the mass adjustment on $\omega_{0,440}$ and AAE_{675}^{440}, as illustrated in the related scatterplot in ***Errore. L'origine riferimento non è stata trovata.****. The correction of secondary inorganic aerosol mass yields a negligible change in terms of calculate absorption properties, while the correction of BC mass introduces more change: the reduction of BC mass, as might be expected, reduces the absorption ($\omega_{0,440}$ increases) and makes its spectral variation more steep (AAE_{675}^{440} increases). The change is of the order of 3-4%, which is comparable to the magnitude of models' $\omega_{0,440}$ bias, but it is of the same sign and magnitude for external and core-shell mixing. The bias of BC mass is thus unlikely to alter the main conclusions regarding calculated absorption properties illustrate above."*

2. The modeled results are evaluated by comparing with the observations from AERONET measurements. However, the optical properties of the AERONET are retrieval products based on certain assumptions, and this means the results may differ if different assumptions were made for the retrieval. In other words, how would the uncertainties related to the AERONET observations themselves influence the evaluation of this study?

This is an interesting point, but difficult to address here. According to the documentation of the AERONET retrieval algorithm (https://aeronet.gsfc.nasa.gov/new_web/Documents/Inversion_products_V2.pdf) the iterative inversion for the estimation of absorption aerosol properties relies on the minimization of the difference between observed and simulated radiances. The simulated radiances are calculated from a mix of spherical and non-spherical particles, divided in two log-normally distributed modes, fine and coarse. Moreover, the vertical distribution of the aerosol layer is assumed to be homogeneous. A clean way to make the comparison of AERONET inversion products with simulations such as the ones presented here, would be to change the retrieval process itself, trying to minimize the error of the simulated radiances using underlying aerosol types more similar to those having the properties listed in Table 3 of the present manuscript. However, this is a very demanding task which is clearly beyond the scope of this study.

3. The mean radius of BC is assumed to be 11.8 nm based on Table 3, which is close to the size of monomer in BC aggregates. It is well known that the BC in the atmosphere is in the form of aggregates of those small monomers, and how would the non-spherical geometry influence the results. It would be difficult to account for the aggregation structures in such a work due to the computational burden, but it is worth to discuss the potential influences by considering previous studies. For example, Li et al. (https://doi.org/10.1002/2015JD024718) evaluated the influences of aggregation on BC optical properties especially AAE, and the effects of internal mixing was also studied by the same group (https://doi.org/10.1016/j.jqsrt.2016.10.023).

We believe this is an important point that we did not discussed properly in the first version of the manuscript. A test and a summarizing paragraph in the new section 3.1 was devoted to that:

"In the second test devoted to size distributions (BC05), we modified only the size of BC. As shown in ***Errore. L'origine riferimento non è stata trovata.****, the mean radius of the BC size distribution is assumed to be 0.0118 µm, which is comparable to the size of a single spherule (monomer) of BC. As mentioned in section 2.3, the real atmosphere observed form of BC goes from fractal aggregates of monomers to more compact forms as it ages. We thus repeated the calculations with an increased mean radius of 0.5 µm, in the middle of the range of radiuses explored by Li et al. (2016). The effect in the external mixing case is a slight increase of the* $\omega_{0,440}$ *and increased variability of the* AAE_{675}^{440}*. In the core-shell case, both* $\omega_{0,440}$ *and* AAE_{675}^{440} *decrease, implying that larger BC cores increase the absorption and flatten its spectral dependence toward values more comparable with those deduced from AERONET measurements. As a caveat, the increase in the mean BC radius is what explains the difference between the CSBC and the CSBCV cases illustrated above. However, the* E_{abs} *also increases by about 50% (not shown), thus a better simulation of* AAE_{675}^{440} *is only apparently happening for the right reason, but this is certainly a point that should be further explored in future studies."*

4. It is noticed that the primary organic aerosol is also absorptive. Is the influence also considered for estimating BC absorption? Its effects should also be removed for estimation on the absorption enhancement.

We acknowledge that the description and impact of the treatment of BrC was not properly addressed in the original manuscript. Given the many uncertainties in the absorption properties of organic matter (OM), we preferred to focus the discussion on BC absorption. Regarding OM we made one extreme choice, i.e. we treat primary OM as BrC and secondary OM as non-absorbing. From model output delivered for the intercomparison, we only have POM and SOM (when simulated) total mass, without any tracking of the sources or the aging. We acknowledge, however, that the extent to which this assumption influences the

main conclusions is not clear and we assessed it with further sensitivity tests on assumption on the treatment of OM absorption properties.

The outcome of additional tests is summarized in these paragraphs of the new section 3.1:

"The final subset of tests 7-9 are devoted at exploring the role of assumptions made on the absorption properties of BrC. In the baseline sensitivity tests presented above, we adopted the extreme choice of assigning BrC characteristics to the primary organic fraction. However, also the primary fraction is generally a mix of white and brown aerosol (e.g. Laskins et al., 2015). In test BRC0, we switch off the absorption due to BrC, setting the imaginary part of primary OC to the low value of 10^{-8}. The effect is a decreased absorption, denoted by the increase of $\omega_{0,440}$. More remarkably, there is a complete suppression of the spectral dependence of the absorption, denoted by the flattening of the simulated AAE_{675}^{440} values. In the case of external mixing, $AAE_{675}^{440} \sim 1$, with very little variability, which is consistent with the presence of only externally mixed BC as an absorber (Liu et al., 2017b, Liu and Mishchenko, 2018). In the case of core-shell, most of the variability is also suppressed, but the mean value of AAE_{675}^{440} is around 1.4, denoting the absorption amplification E_{abs} by the shell around BC. According to recent calculations reported by Luo et al. (2018), the core-shell model is expected to exaggerate this amplification especially at shorter wavelengths, thus artificially increasing the calculated AAE_{675}^{440}.

In tests 8 (BRCS), we swapped the role of primary and secondary organic carbon as radiation absorber. The results are generally similar to the reference case, but there is an increased variability in the simulated values, reflecting the secondary nature of the aerosol, which is photochemically produced down-wind of the sources, and thus generally more variable. In the last test 9 (BRCSH), we further suppressed the hygroscopic growth assumed for the secondary organic fraction, while the primary was assumed hydrophobic in all the tests. The absence of water uptake by the aerosol increases the absorption (indeed water has a refractive index of 1.32-1.35 in the visible and it does not absorb light significantly), but does not affect much the its spectral variation."

Specific comments:

1. "Grid Spacing" in Table 2 is listed as either km or degree, and it should be unified for better comparison.

The grid-cell spacings previously given at native model resolution in degrees, were converted to approximate distance in km.

2. The information in Table 4 is not well summarized, and the differences among the six models should be known with only reading the table.

We further split the relevant information of the differences among the cases in the attempt of making them more readable.

3. In Figure 5, the colormaps for small and large count numbers are close, and clearer colormap is suggested.

We reduced the number of colorscale bins in order to make the figure more readable.

4. It seems that Europe and N. America do not show too many differences on the conclusions related to the mixing state and absorption simulations. There are much less data for evaluating the N. America case, which makes the discussion less solid (e.g. Figs 5b and 8b). Why not just focus on Europe, because it will not change the conclusions of the manuscript but makes it much easier for discussion.

This intercomparison exercise is based on the collaboration of communities from both continents, thus, even if not central to the main scope of the manuscript, we prefer to keep it as it is. We believe that it is still a useful term of comparison for the reader and it thus not disturb much the presentation of the results. We added a short last paragraph to the result section 3: "Summarizing the comparison between the two continents, the selected AERONET observations generally show more absorbing (mean $\omega_{0,440}$ of 0.82 vs. 0.91) and spectrally dependent (mean AAE_{675}^{440} of 1.19 vs. 1.10) aerosol over North America than Europe. The models broadly capture this variability, but display generally a larger bias over North America. The changes induced in the calculated optical quantities by the modifications tested here on the mixing state assumptions are consistent on the two regions.". The same concepts are reiterated also in the conclusions.

5. For Figures 6 and 9, are the values in the y-axis in unit of percentage? For example, does 1.0 mean 100% or 1%?

The legend on the y-axis was wrong. 1 means 100%, not 1%. We corrected the Figures, removing the "(%)" near "NMB" on the y-axis.