This manuscript concerns the optical properties of black carbon (BC) and brown carbon (BrC): significant (if not major) contributor to light-absorption by aerosols. The climate effects of these components are highly uncertain, while the optical properties are an important contributor to these uncertainties. As such, this manuscript addresses a topical subject.

The authors use a number of different, complementary techniques suitable for the purpose, and arrive at important conclusions for the field. However, the presentation of these should be condensed, emphasized and clarified.

The data is a few years old – but this is of little concern here. I like the comparison between an urban and rural location. However, I would like to see a stronger emphasis of site-specifics when comparing to other sites around the world. Further, I think a deepened discussion regarding an underlying assumption of this work: that composite aerosol absorption by different components is additive – comments below.

Below I list a number of major and minor comments. If carefully addressed I think this contribution should be suitable for publication in ACP.

Major comments:

An underlying assumption in this, and also several similar other studies (including citations in the manuscript) is that the absorption by, e.g., different BrC compones, or BrC/BC mixtures is additive. Example: isolated BC abs + isolated BrC abs = abs of mixture. This might be a fair first order approximation for many applications. Not only considering particle level effects (including scattering effects, e.g., lensing), but also properties at a molecular level. However, a complication with, e.g., BrC, may be illustrated by the almost universally observed power-law like spectrum (as for BC), which suggests that the underlying absorption mechanism is not simply the addition of a large number of different chromophores (which would produce a multi-peak spectrum), but that the absorption is also modulated by interactions of the chromophores with other components; extending to the limit of the graphene-like structure of BC (Andersson, 2017). Empirical evidence shows that this is the case: interactions of BrC chromophores with other components may significantly enhance absorption, e.g., through charge transfer complexes (Phillips and Smith, 2014a,b). Or in other words: the absorption of the mixture is more than the sum of the parts, also at a molecular level. This argument is further, at least to some extent, supported by the observation that absorbing BrC molecules in solutions are much larger (>1000 Da; e.g., increase of molecular size during transport in biomass plumes, associated with loss of absorption by smaller molecules, Di Lorenzo and Young, 2016; Di Lorenzo et al., 2017) than often explored (in laboratory and ambient studies) single-molecule chromophores such as PAHs; nitro-aromatics and similar. Along these lines, the nature of these larger molecular units (large molecules or aggregates?) suggests structural settings by which individual chromophores are likely to interact with surroundings, differently then, e.g., through free translational diffusion. Furthermore, it is not too far-fetched to consider interactions (e.g., charge transfer) between BC and BrC in mixture.

For the purpose of this paper I think the first order approximation of additive absorption is ok. However, I would like to see a discussion about potential limitations of these assumptions - ranging from source apportionment techniques to differentiation of BrC and BC in mixtures - including above references.

Overall the paper is a bit hard to read: many numbers, abbreviations and technical details. I count 135 references, suggesting a thorough literature review, but at the same time indicating a potentially too broad scope for a single paper (more like a review) – with the purpose of clarity and transparence as guiding principles. Perhaps splitting into separate BrC and BC papers would be a possibility. However, this is not a 'reviewer' recommendation, but a thought. In the meantime, I think the authors should revise the text with the goal of a more accessible presentation. Squeeze out the essence (conclusions and interpretations, rather than numerics and technical details) and put the rest into the SI.

Minor comments:

Abstract:

Along with the R&D I find this too long, with too many numbers. I think the reason why, e.g., many high impact journals (e.g., Science or Nature) promotes short abstracts is valid in general: effective and clear communication. I suggest to cut to half its length.

It is not clear to me why the authors focus on the imaginary refractive index as a measure for absorption, when most other studies on BC and BrC report mass absorption cross-section (MAC) or mass absorption efficiency (MAE; different names – same thing). It is clear that the imaginary index goes into the Mie Calculations, but I think it would be much preferred to report MACs etc, especially in Abstract to allow direct comparison with existing literature.

Tar balls appears important in certain regions, while then appears absent here. Is this a key result for the abstract? Further (R&D): what is the significance of this, besides an empirical finding? Does this change the spectral properties in any way? Why are they absent? – sources?

M&M:

Line 50: the absorption of BC is not wavelength-independent. That would suggest constant absorption over all wavelengths, meanwhile it is typically described by a power law, with a variable exponent. Remove.

Line 57: I think the mixing state for BrC (lensing etc) may be equally important for BrC.

Line 63: 'Certain studies suggest that aging leads to bleaching, whereas others suggest formation of secondary BrC'. There is no fundamental reason as to why both cannot happen, and do happen, to different extents, in different environments. I think rephrase to remove possibility of misinterpretation: there is no implicit contradiction in these results.

Line 74: What do you mean by the 'aethalometer model' here? I assume it is not the actual instrument model, but some kind of source apportionment approach. Rephrase.

Line 80: I would expect that it is quite unclear how it responds to photo-chemical aging as well (e.g., the cited Dasari et al., 2019).

Line 84: 'The first consists of a direct estimation of total BrC absorption online by subtraction.' This sentence is unclear: subtraction of what from what? From what data? Etc.

Line 91: What other non-refractory material is intended here?

Line 129: I am not familiar with the MWAA instrument. From the SI appears that this instrument is a filter-based instrument – like the aethalometer. Although probably an improved technology, being filter-based it has the same principal disadvantages regarding measurements of absorption as the aethalometer. Since the MWAA is used for calibration (of the multiple scattering parameter, C) of the aethalometer, I wonder – how is the MWAA calibrated towards non-filter-based methods, e.g., photo-acoustics? Why is this a reliable reference? I am sure this is outlined in the original reference, but it is of importance here and should be clarified.

Section 2.2:

I think this section reads much more technical than what it actually is. Aim to make clearer.

Line 182: The word matrix is mentioned three times in one sentence. But what is this matrix? Do you mean the absorption spectra (dimension 1: wavelength; dimension 2: data point)? Be more concrete.

Line 186: What is meant by: 'This approach provides MAE specific to an OA factor in hypothetical pure form.'?

Line 198: I suppose the MIE calculations are based on size distributions from DMPS? Clarify.

Section 2.3.

I wonder about the fundamental assumption here: that the absorbing species are equally distributed through the particle sizes: this topic was discussed in detail in the cited, Liu et al., 2013. I would like to see a slightly expanded discussion on this compared to the different scenarios explored for different size regimes.

R&D:

Section 3.1.

To provide context, I suppose it would be good to start out with a brief presentation of the study sites, prevailing meteorology (precipitation? Air mass origins? Seasonality? etc), topography or other relevant conditions. I find that – in general - comparisons in the BC/BrC literature tend to aim for general conclusions, while site-specific variability may play a very large role. For instance, comment on line 63.

Further, start out with discussing concentration data, rather than ratios. Even though I am familiar with BrC literature, these ratios mean little to me a priori. This also goes for Figure 1.

Line 232: The means for the winter is indeed lower compared to summer. Meanwhile, the numerical ranges are strongly overlapping. Are these differences statistically significant?

Line 250: PMF provides ‘principal components’ – which are mathematical deconvolutions. However, these then need to be interpreted in terms of different sources somehow. How was this done? How are HOA, fOOA, SOOA etc attributed to these?

Line 317: The significance of tar balls in this context is unclear.

Lines 355 and forward: Comparisons of MAC are tricky. Different methods and different parametrizations are often used: please discuss these features, and how they relate to the present methodology.

Lines 378-384: I think these important comparisons are glanced over too quickly here. Please see comments on Figure 5 and elaborate.

Summary:

Line 480: If using the word holistic, I would say this study attempts, rather than provides.

The emphasis on biomass influence here, and elsewhere, is not clear to me: do the authors believe this is a central motivation for conducting this study? Or should one interpret this as one of the central findings? The later interpretation precludes the following sentences.

Figure 1.

As a first figure I would like to see the ‘raw data’ time-series data (absorption coefficients; WSOC concentrations etc), before moving into more convoluted forms, e.g., ratios.

Figure 2.

Overall a good figure, but a bit cluttered. Are you sure all of these data points are essential, or could parts of this figure be presented in the SI?

Figure 4. Try to make the square boxes equal in size.

Figure 5.

I appreciate this comparison, based on geographical variability. Since East and South Asia are quite distinct in terms of the aerosol regime, I would separate this into two groups.

I would also like to see separation w.r.t. atmospheric transport times (e.g., photochemical aging):

For instance, differentiating between near-source (e.g., urban) vs receptor sites; can we pick up a trend?

References to add: Cui et al. (2016); Chen et al. (2017).

Figure 8.

Avoid statements like: 'a new framework'. Just write out what this figure is about.

Appendices:

Line 573: What is the origin of the equation for relative error of EC? What does the number 0.03 signify and how did you arrive at this?

Line 640: What is Zendo? Please provide a link or similar.

References:

Andersson, A (2017) A Model for the Spectral Dependence of Aerosol Sunlight Absorption. ACS Earth Space Chem. DOI: 10.1021/acsearthspacechem.7b00066

Chen et al. (2017) Light absorption enhancement of black carbon from urban haze in Northern China winter. Environ. Poll. DOI: 10.1016/j.envpol.2016.12.004

Cui et al. (2016) Radiative absorption enhancement from coatings on black carbon aerosols. STOTEN. DOI: 10.1016/j.scitotenv.2016.02.026

Di Lorenzo, RA and Young, CJ (2016) Size separation method for absorption characterization in brown carbon: Application to an aged biomass burning sample. GRL. doi:10.1002/2015GL066954.

Di Lorenzo, RA, et al. (2017) Molecular-Size-Separated Brown Carbon Absorption for Biomass-Burning Aerosol at Multiple Field Sites. ES&T. DOI: 10.1021/acs.est.6b06160

Phillips, SA and Smith, GD (2014a) Light Absorption by Charge Transfer Complexes in Brown Carbon Aerosols. ES&T. doi: 10.1021/ez500263j

Phillips, SA and Smith, GD (2014b) Further Evidence for Charge Transfer Complexes in Brown Carbon Aerosols from Excitation−Emission Matrix Fluorescence Spectroscopy. J. Phys. Chem. DOI: 10.1021/jp510709e.