Wang, J., Nie, W., Cheng, Y., Shen, Y., Chi, X., Wang, J., Huang, X., Xie, Y., Sun, P., Xu, Z., Qi, X., Su, H., and Ding, A.: Light absorption of brown carbon in eastern China based on 3-year multi-wavelength aerosol optical property observations at the SORPES station and an improved Absorption Ångstrom exponent segregation method, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-49, in review, 2018.

Replies to reviewers' comments

Overview

The authors thank the reviewers for constructive comments, they helped improving the paper. We have replied to all questions raised by the reviewer. The major changes to the paper are that the we have

- reorganized the AAE method part and revised corresponding figures
- added measurement data from single particle soot photometer (SP2) at same site to support calculation
- conducting Mie-simulation using BC size distribution and mixing state measured by SP2 to derive AAE_{BC} instead of taking a fixed typical BC core size distribution and analyzed the uncertainties of the method
- added more review on AAE segregation and BrC definition

Detailed replies to Anonymous Referee #3

General Comments This study provides an improved approach on deriving the brown carbon absorption from AE31 measurement, and highlights the importance in using the proper AAE to extrapolate the BC absorption from longer to shorter wavelength. If the technical part of this study could be more convincible, then it could be considered for publication. I would suggest to improve the technical part by considering the following points.

***Response**: Thanks for the valuable comment. To avoid the uncertainties caused by assumed BC size distribution, we will reorganized our manuscript by using BC size distribution and mixing state measured by single particle soot photometer (SP2) at the SORPES station instead of using empirical BC size distribution from previous studies. We found that our previous method is still available but we modified the method by combining SP2 data in the revision. The contribution of BrC gets larger with new parameters. Also, in the revised version we estimate the uncertainties of the calculation and update the method and result parts.*

Specific comments

1) The issue of deriving the coating content from OC/EC measurement is not just from the uncertain OM/OC ratio, but also that many of the OM may not contain BC, i.e. externally mixed, the OC/EC method would tend to largely overestimate the coating associated with BC. You could use some external results to estimate this.

***Response**: Thanks for the comment. We agree that the mixing state of BC has impact on BC optical properties. We will use available SP2 data during the study to get mixing state and size of BC and revised the calculation supported by SP2 results. OCEC data are no longer used in this study. Detailed description will be presented in the revision.*

2) The crucial results here are from the AE31. How it has been corrected is important. It is a challenge to get the proper result from this instrument (especially at shorter wavelength). Though this may have been done in your previous publications, but it's worthy to mention here, e.g. how you get the multiple scattering and scattering correction, and how you have corrected these at different wavelength, and these may substantially affect the derived AAE because these corrections rely on the AAE as well.

***Response**:*

Thanks for the comment. For Aethalometer data correction, we implemented the correction algorithm presented by Collaud Coen et al. (2010). b_{abs} at wavelength λ is corrected for filter-loading effect, scattering effect and multiple scattering effect, with equation shown as

$$b_{abs}(\lambda) = \frac{b_{ATN} - s' \cdot b_{scat}}{C_{ref} \cdot R}$$

where R is the function for filter-loading correction and s' represents the fraction of scattering coefficient resulting in ATN change (Shen et al., 2018). C_{ref} is the multiple scattering correction factor, which is set to be 4.26 according to Collaud Coen et al. (2010). Detailed calculations of R and s' can be found in Collaud Coen et al. (2010). The comparison of different Aethalometer corrections (Saturno et al., 2017) shows that AAE derived by Collaud Coen correction algorithm agrees well with that from multi-wavelength reference measurement, proving the reliable AAE values calculated from this correction. Saturno et al. (2017) also proves that Collaud Coen correction shows a good performance in obtaining absorption coefficients at 370 nm, which is the critical wavelength in BrC segregation. We will add more description of Aethalometer data correction in the revised manuscript.

3) The BC core size is not constant, the BC from open biomass burning or domestic solid fuel burning has a larger core than from traffic (Schwarz et al., 2008) (Liu et al., 2014). As you have already raised, the larger core will have a lower AAE. Some external data could be used to make more constrains for biomass burning BC.

***Response**: Thanks for the comment. We agree that BC core size can vary under influence of different dominant sources. As suggested, to derive AAE_{BC}, we will use real-time core size and coating thickness of BC measured by SP2 instead of taking a fixed typical BC core size distribution.*

4) The ideal approach would be combing with the SP2 measurement, such as a similar study (Liu et al., 2015), however the advantage of this study is the long-term measurement using the less-cost instrumentation, and different contributions such as open biomass burning or residential burning occurred in different months, which is interesting. If by somehow, this study could benefit by constraining some of the inputs from the existing information and doing sensitivity test.

***Response**: Thanks for the comment. We will combine SP2 measurement to modify the method. Since there are lots of historical observation data conducted without SP2, it is worth to try and find a way of tracing back light absorption of BrC with satisfactory uncertainty range. At this site, firstly, we will calculate AAE_{BC} at short and long wavelength range based on available SP2 data during the study period, and analyzed their variation ranges. Uncertainty and sensitivity of this method will also be added in the revised manuscript.*

5) It is better to show the location of the experimental site and the surrounding major emissions.

***Response**: Thanks for the comment. The map and emission character surrounding the site have been given in many of our previous works, e.g. (Ding et al., 2013;Ding et al., 2016). Here we would like to cite these reference and describe the character in the text. The location of the SORPES site will also be given in several figures in the revision.*

6) Equation 4 and 5 could be merged into one, it would be useful to show the Rs-1 in time series or monthly variation.

***Response**: Thanks for the suggestion. R_{AAE} will be used in revised manuscript instead of R_{s-l} for better* *understanding. We will add the definition of R_{AAE} in the text instead of showing as Eq. 4. The Eq. 5 in the original manuscript will be numbered as Eq. 6 in the revised manuscript. We will also show the seasonal variation of R_{AAE} in the revision.*

7) Please give the refractive index you used for BC, clear coatings and brown coatings in the main plot legends.

***Response**: Thanks for the suggestion. The refractive index (RI) of BC core was set to be 1.56+0.47i according to Dalzell and Sarofim (1969) and RI was 1.52+0i for clear shell (Pitchford et al., 2007). These values will also be listed in the revised manuscript and Figure 1. The brown coating case will not be discussed in our revised manuscript, due to large uncertainties on brown coating's refractive index.*

8) Cl-/EC, as an indicator of coal combustion, needs more reference, would you be able to derive the MAE of brown carbon at different months, which will be very interesting. It looks the higher Babs/K+ ratio possibly means the Dec. BrC had a higher absorption efficiency maybe?

***Response**: Thanks for the comment. After revision of the method and related results, we will also modify the source analysis part in the revised manuscript. Detailed description can be added. About MAE_{BrC}, thanks for this interesting suggestion. However, mass of BrC was not measured directly at this site and we can only use OC data to convert the mass of OC to mass of BrC. Since BrC mass fraction in OM may change over time and case due to different dominant sources, especially in China where the emission profile is complicated, it is hard to differentiate the change of MAE_{BrC} and BrC mass fraction. In the future, we will try to calculate MAE_{BrC} by using water soluble OC (WSOC) measurement and have a detail study focusing on this topic. Thanks again for your valuable suggestion.*

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