Response to He's comments

The authors conducted direct field measurement of size-resolved mixing state, absorption enhancement, and single scattering albedo for BC particles during photochemical aging. It could help to advance the current understanding in the large variation of BC absorption during atmospheric aging processes. I have one short comment on the uncertainty associated with the calculation of BC optical properties. Thank you for your helpful comments. Point-by-point responses to the comments are attached below.

The computation of BC optical properties in this study assumed the core-shell coating structure. However, more and more observations (e.g., China et al., 2015; Wang et al., 2017) have shown various BC coating structures/morphology during aging processes, which are not core-shell. Further modeling studies (e.g., Scarnato et al., 2013; He et al., 2015, 2016) have indicated a large variation in BC optical properties due to the observed complex coating morphology. Thus, assuming a core-shell structure may lead to uncertainty in the estimate of BC optical properties.

This is a valuable clarification and limits of our study. Accordingly, we added some discussion of the effect of BC morphology on absorption enhancement in the revised manuscript. The morphologies of BC coating are complex and diverse (China et al., 2015; Wang et al., 2017), which also leads to a large variation in BC optical properties (He et al., 2015; He et al., 2016; Scarnato et al., 2013; Wu et al., 2018). We agree with He that novel models will improve our understanding of BC optical properties, but our study did not have particle size distribution information or morphological information and can therefore not perform this type of complex computation.

Mie theory remains a powerful tool for optical data interpretation (Lack et al., 2012) due to its computational efficiency and applicability to radiative transfer models. Morever, the reliability of the core-shell model has been verified in many optical closure studies (Lack et al., 2012; Ma et al., 2012; S. Liu et al., 2015; Wu et al., 2018). In this study, the core-shell calculation has been demonstrated to be useful to interpret the observation data albeit with the limitations that He points out.

Besides, He et al. (2015) also proposed a BC optics-aging mechanism with three evolutional aging stages, which may be useful for the authors' analysis. I suggest that the authors include these recent studies and add some discussions on this important issue.

Done. We added recent studies in the revised manuscript.

Theoretical and experiment results shown that aging causes the dramatic

changes of BC particle morphology (China et al., 2015; He et al., 2015; He et al., 2016; Scarnato et al., 2013; Wang et al., 2017) and leads to more compact black carbon with higher scattering cross section (Peng et al., 2016; Y. Wu et al., 2018), ...

References:

- China, S., Scarnato, B., Owen, R. C., Zhang, B., Ampadu, M. T., Kumar, S., Dzepina, K., Dziobak, M. P., Fialho, P., Perlinger, J. A., Hueber, J., Helmig, D., Mazzoleni, L. R., and Mazzoleni, C.: Morphology and mixing state of aged soot particles at a remote marine free troposphere site: Implications for optical properties, Geophys. Res. Lett., 42, 1243–1250, doi:10.1002/2014gl062404, 2015.
- He, C., Liou, K.-N., Takano, Y., Zhang, R., Levy Zamora, M., Yang, P., Li, Q., and Leung, L. R.: Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison, Atmos. Chem. Phys., 15, 11967-11980, doi:10.5194/acp-15-11967-2015, 2015.
- He, C., Takano, Y., Liou, K.-N., Yang, P., Li, Q., and Mackowski, D. W.: Intercomparison of the GOS approach, superposition T matrix method, and laboratory measurements for black carbon optical properties during aging, J J. Quant. Spectrosc. Radiat. Transf., 184, 287–296, doi:10.1016/j.jqsrt.2016.08.004, 2016.
- Scarnato, B. V., Vahidinia, S., Richard, D. T., and Kirchstetter, T. W.: Effects of internal mixing and aggregate morphology on optical properties of black carbon using a discrete dipole approximation model, Atmos. Chem. Phys., 13, 5089– 5101, doi:10.5194/acp-13-5089-2013, 2013.
- Wang, Y., Liu, F., He, C., Bi, L., Cheng, T., Wang, Z., Zhang, H., Zhang, X., Shi, Z., and Li, W.: Fractal dimensions and mixing structures of soot particles during atmospheric processing, Environ. Sci. Tech. Let., 4, 487–493, doi:10.1021/acs.estlett.7b00418, 2017.
- Wu, Y., Cheng, T., Liu, D., Allan, J. D., Zheng, L., and Chen, H.: Light absorption enhancement of black carbon constrained by particle morphology, Environ. Sci. Technol, 52, 6912–6919, doi:10.1021/acs.est.8b00636, 2018.

References

China, S., et al.: Morphology and mixing state of aged soot particles at a remote marine free troposphere site: Implications for optical properties, Geophys. Res. Lett., 42, 1243–1250, doi:10.1002/2014gl062404, 2015.

He, C., et al.: Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison, Atmos. Chem. Phys., 15, 11967-11980,

doi:10.5194/acp-15-11967-2015, 2015.

He, C., et al.: Intercomparison of the GOS approach, superposition T-matrix method, and laboratory measurements for black carbon optical properties during aging, J. Quant. Spectrosc. Radiat. Transf., 184, 287–296, doi:10.1016/j.jqsrt.2016.08.004, 2016.

Scarnato, B. V., et al.: Effects of internal mixing and aggregate morphology on optical properties of black carbon using a discrete dipole approximation model, Atmos. Chem. Phys., 13, 5089–5101, doi:10.5194/acp-13-5089-2013, 2013.

Wang, Y., et al.: Fractal dimensions and mixing structures of soot particles during atmospheric processing, Environ. Sci. Technol. Lett., 4, 487-493, doi:10.1021/acs.estlett.7b00418, 2017.