Comment

This manuscript is generally well written, but there seems to be one important piece missing, which is related to BC particle structures. This study assumed spheres for externally mixed BC and core-shell coating structure for internally mixed BC. However, recent observations (e.g., China et al., 2015; Wang et al., 2017) have shown irregular fractal aggregates for externally mixed BC and various non-core-shell coating structures for internally mixed BC. 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 fractal and coating structures. Thus, the assumptions in the current manuscript may lead to uncertainty in the calculations of BC optical properties. Thus, I suggest that the authors include these recent studies and add some discussions on this important issue.

We thank Dr. He for this useful comment on our manuscript. We acknowledge that the point raised is relevant and not properly illustrated in our first version of the paper. In section 2.3, where we explain details on the calculations carried out, we added a paragraph on the issue:

"Specifically regarding the modelling of BC shape and mixing state, here we adopt the simplified approach widely used in regional and global models of assuming spherical particles and centred core-shell arrangement for internal mixing calculations, which makes the computation fast enough for 3-D applications in year-long simulations. However, observations show that BC in the real atmosphere displays a wide variety of shapes: freshly emitted hydrophobic fractal aggregates, consisting of hundreds of spherules having diameters of a few tens of nm (e.g. Posfai et al., 2003, Adachi and Buseck, 2013), typically evolve in the atmosphere assuming more compact structures, and internally or semi-internally coating with hydrophilic material (e.g. Adachi et al., 2010, China et al., 2015, Wang et al., 2017). These transformations affect the variability of the absorption properties of BC, as illustrated in several numerical studies that include detailed description of the shapes and mixing state of BC and that use advanced algorithms, such as the multiple-sphere T-matrix (MSTM) and the discrete dipole approximation (DDA), to compute the optical properties (Scarnato et al., 2013, He et al., 2015, He et al., 2016, Li et al., 2016, Kahnert, 2017, Liu et al., 2017b, Liu et al., 2018, Liu and Mishchenko, 2018). Moreover, also the shapes of BrC may vary in the real atmosphere, but their classification and investigation of numerical aspects in the calculation of optical properties is still at its beginning (Laskin et al., 2015, Liu and Mishchenko. 2018)."

Moreover, this comment, together with related comments from the anonymous reviewers, suggested a new sensitivity tests on assumptions specifically related to BC size distribution, as illustrated in a paragraph of the new section 3.1:

"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."*

References

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