

## Reviewer #1:

*We thank Reviewer #1 for positive, constructive and detailed comments, which we will account for in the revised manuscript. Our point-by-point responses to the issues raised by the reviewer are below.*

### General comments

1. The BC particles were produced from a liquid suspension using an atomizer and a dryer before coating, this changed the morphological structure of the BC to a compact BC structure, as the authors discuss. It would help mentioning that the atmospheric BC can have different degrees of compaction (there are a few studies showing this for laboratory, but also for atmospheric particles), and discussing briefly how the presence of more open-structured BC particles in the atmosphere might affect the conclusions of this study, and the models applied. For example, that might be relevant for fresh vs. aged BC particles. Related to this topic, a lot of work published in the literature on BC morphology, compaction, and coating is completely neglected here (in particular several electron microscopy, SP2, and optical studies from different groups around the world). I think that mentioning a few of these studies would improve the paper.

*We agree with the referee and will add the following text to the revised manuscript and will modify the last paragraph in page 2:*

*“Laboratory measurements have indicated that by increasing the amount of the coating on the BC particles, the dynamic shape factor of these particles decreases, and fractal BC aggregates become restructured and more compact (Saathoff et al. 2003; Slowik et al. 2007; Zhang et al. 2008; Pagels et al. 2009; Tritscher et al. 2011; Ghazi and Olfert 2012). Investigating ambient BC particles have shown that BC particles coated by secondary aerosol constituents during atmospheric aging transform from a fractal to spherical and further fully compact shapes (Peng et al. 2016; Zhang et al. 2016). Furthermore, ambient BC measurements have demonstrated that aging of BC particles and coating by other material via condensation and coagulation can enhance the light absorption capability of BC particle (Khalizov et al. 2009; Moffet and Prather 2009; Chan et al. 2011; Liu et al. 2015; Zhang et al. 2017; Xu et al. 2018). Although this enhancement of light adsorption properties of BC-containing particles is still a large uncertainty in modelling direct radiative forcing of BC particles. Furthermore, there are uncertainties in modelling the indirect radiative forcing of the BC-containing particles, due to e.g. lack of knowledge about cloud interactions of BC-containing particles and the role of the co-emitted species. To overcome ....”*

*We will also add a brief discussion of the representativity of our BC particles to the revised manuscript.*

2. The BC particles were size selected with a DMA before being coated. This maintains the core constant while the coating thickness is increased. This approach is fine for the most part and produces interesting results. However, to untangle the effect of the coating hygroscopicity from that of size, it would have been interesting to size select before and after

the coating stage as well, to maintain the overall particle size constant, while changing the coating thickness; in this way, isolating the effect of the overall particle size (this would be particularly interesting for the case of the oleic acid). I am not suggesting the authors should conduct such experiments for this manuscript, as I think the results of the current study are very interesting on their own, but they could briefly discuss this possibility for future studies.

*Indeed, these kind of experiments would be an interesting topic for a future study. We will add a statement to the revised manuscript mentioning this possibility for future studies. However, they would be somewhat challenging (although perhaps not impossible) with the present setup, where the temperature of the furnace in effect determines the coating thickness. We selected the size of the BC cores first by a DMA and the coating thickness was varied by changing the furnace temperature. We then measured the size distribution of the coated particles exiting the furnace. At any given temperature, we then estimated the size of the coated particles using the peak value from the size distribution curves.*

### Specific comments

- Page 2 line 31, there are several studies, including some recent, that analyze and quantify the effect of coating, mixing and compaction for BC with the detail and unambiguity of electron microscopy, as well. Studies are available for both laboratory, as well as, ambient BC particles. It might be worth discussing some here.

*We will modify the revised manuscript by changing the sentence in question to (see also our response to general comment 1):*

*Laboratory measurements have indicated that by increasing the amount of the coating on the BC particles, the dynamic shape factor of these particles decreases, fractal BC aggregates become restructured and more compact (Saathoff et al. 2003; Slowik et al. 2007; Zhang et al. 2008; Pagels et al. 2009; Tritscher et al. 2011; Ghazi and Olfert 2012). Investigating ambient BC particles have shown that BC particles coated by secondary aerosol constituents during atmospheric aging transform from a fractal to spherical and further fully compact shapes (Peng et al. 2016; Zhang et al. 2016).*

- Page 3, line7, “of uncoated” seems not to belong here considering they are talking about multilayer models.

*The reviewer is correct. We will remove the reference to the uncoated particles from the first sentence of the paragraph. The adsorption activation model described by Sorjamaa & Laaksonen (2007) assumes that the CCN activation of insoluble but wettable compounds happens through multilayer adsorption of water molecules. This model was developed later by Kumar et al. (2011) to include the CCN activation of the insoluble particles coated by soluble salts.*

- Page 3, line 27. Please provide a sentence or two on why regal back and how does that represent (how well it acts as) a surrogate for atmospheric BC. This in addition to the compaction issue mentioned in the general comments section.

*We will add the following text to the revised manuscript:*

*Regal Black (Cabot REGAL R400 pigment black), which was provided by Cabot Corp., USA, is a surrogate for collapsed soot (Sedlacek et al. 2015) and is the recommended calibration standard for the SP-AMS (Onasch et al., 2012). This compound has been used in different studies (Onasch et al. 2012; Corbin et al. 2014; Healy et al. 2015; Sedlacek et al. 2015) as a model of refractory carbonaceous compounds to estimate the chemical and physical properties of the black carbon particles and Canagaratna et al. (2015) have shown that regal black and flame soot appear very similar, at least from the perspective of mass spectrometry. However, it should be borne in mind that in the ambient BC particles can vary significantly in terms of their physical and chemical properties, and is usually mixed with other pollutants present in the atmosphere.*

*This clarification will be added to the manuscript.*

- Figure 5: the theoretical calculation seems to perform less well for the larger core diameter; in fact, the range of the grey band does not seem to intersect with the experimental data even considering their uncertainties. Is there any reason for that? A short discussion would be interesting.

*This is a very good question, to which we do not have a definite answer. One reason might be just a larger uncertainty in the  $\kappa$  values than what is considered in the calculations. Different values have been reported for  $\kappa$  in different studies (Petters and Kreidenweis 2007; Chan et al. 2008; Petters et al. 2016). The  $\kappa$  we used for glutaric acid is from (Petters and Kreidenweis 2007) and is between 0.113-0.376 (we will correct it in the table 1).*

*We will add this clarification to the manuscript.*

- Figure 6, maybe this was mentioned and I missed it, but why are the theoretical calculations so much narrower here than in figure 5? I guess the spread reflects directly the  $\kappa$  range for the two coating materials, narrower for levoglucosan than for glutaric acid. Can the authors comment on that?

*This is correct and we have thus added the following sentence to our manuscript, first paragraph on page 9:*

*“The theoretical calculations of  $s_c$  are narrower for levoglucosan compared to glutaric acid, because the  $\kappa$  range is narrower for levoglucosan (it is between 0.193-0.223 for levoglucosan and 0.113-0.376 (we will correct it in the table 1) for glutaric acid).”*

## **Technical corrections**

- Page 3, line 13, consider adding the article “the” in front of Soot Particle Aerosol Mass Spectrometer

*We will add “the” in front of Soot Particle Aerosol Mass Spectrometer.*

- Page 10, line 7, “procedure”, should be “procedure”, probably.

*We will correct the word “procedure”.*

## **References**

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