

Reviewer #2:

We thank Reviewer #2 for taking the time to consider and review our manuscript. We respectfully disagree, however, with the reviewer on the purpose and value of the presented work. We feel there is a fundamental difference of opinion in 1) what can be considered as “text-book knowledge”; 2) the value of well-defined laboratory studies with simple model systems for evaluating theoretical approaches used to describe more complex atmospheric systems. Our point-by-point responses to the issues raised by the reviewer are below.

By reviewing the manuscript on the CCN activation properties of BC particles coated with a few organic species I feel myself rather uncomfortable. The study is a carefully planned and executed combination of experimental work and theoretical calculations aiming at providing new insights into the CCN behavior of atmospheric BC particles. However, the entire approach looks as a textbook-like routine exercise that contains no traces of novelty and innovation that would have been required by a high-standard journal like Atmospheric Chemistry and Physics. Any laboratory study in itself is free to use virtually any combinations of agents and conditions, yet it would be expected to be a quasi-realistic model of physical reality. The basic concept of the present study does not fulfil this fundamental requirement.

We are sorry to hear that the reviewer feels uncomfortable, but thank him/her for acknowledging the quality of the presented work nevertheless. However, there seems to be a fundamental misunderstanding regarding the purpose and novelty of our work. As we wrote in the “Introduction”, the CCN activation of uncoated and coated insoluble particles, such as BC-particles coated with soluble species, is usually described theoretically by multilayer adsorption models accounting for the curvature of the particles. One of these theories is adsorption activation theory which is a combination of FHH adsorption isotherms and classical Köhler theory to describe the equilibrium growth of insoluble particles. Later, Kumar et al. (2011) introduced a new framework of CCN activation of dust containing a soluble salt fraction, based on a combination of the classical Köhler and FHH adsorption theories. However, systematic experimental testing of the applicability of combined Köhler and FHH theory with agglomerated insoluble particles coated with organic species of varying solubility is lacking. Unlike what the reviewer claims, these theoretical approaches are not “well-established” in terms of systematically testing their applicability with known molecular species. This was the main motivation and novelty of our study and we will highlight this now more clearly in the Introduction of the revised manuscript.

What sort of real-life BC particles do Regal black stand for? Aged diesel soot particles or BC particles from flaming biomass combustion? Levoglucosan, which is an abundant pyrolysis product of wood combustion, is not a semi-volatile species that is available for adsorption or condensation in the global atmosphere such as PAHs or n-alkanes. It is always present internally mixed with smoke particles, not as a gaseous species. Oleic acid is also a primary tracer which—unlike levoglucosan—is present in the gas phase but on a very limited spatial scale near its sources. I would doubt that this photochemically reactive species can make it to the free troposphere to participate in cloud nucleation. I wonder if anybody has ever detected oleic acid in cloud water or precipitation. In spite of these serious limitations oleic acid experiments at least yielded some unexpected results which are not fully exploited in the manuscript. Perhaps a molecular adsorption modelling approach would have helped explain the

observations. Glutaric acid does exist as SOA product in the atmosphere, though at far lower concentrations than smaller dicarboxylic acids or other SVOC species. In addition, a very similar study was published for the CCN effect of adipic acid. I suspect that upon releasing fresh BC particles from any source there is a plethora of co-emitted semi-volatile species that are ready to be adsorbed onto their surfaces. It is strange that in the experimental section no temperature values are given for the coating procedure. These temperatures would also indicate that the atmospheric occurrence of such processes is unlikely.

We are well-aware that in the atmosphere a myriad of different organic and inorganic compounds accompany BC in the particulate phase. The main purpose of this study was to test the model frameworks used to describe these extremely complex mixtures in various atmospheric models by comparing their predictions to well-defined particles generated in the laboratory. It is a well-established approach to use laboratory measurements of model compounds representing the variation of properties relevant for the studied processes to evaluate theoretical frameworks. In fact, a major part of laboratory measurements represent this kind of approach. Hence we feel that the criticism raised by the reviewer is poorly justified and unfair.

As also pointed out in our responses to Reviewer #1, Regal black has been standardly used as 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. Canagaratna et al. (2015) have shown that regal black and flame soot appear very similar, at least from the perspective of the mass spectrometry. However, it should of course 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. We will highlight this in the revised manuscript (see also response to Reviewer #1).

We would also like to highlight that the studied organic substances were chosen based on their properties, not solely based on their atmospheric relevance. The solubility and other properties of atmospheric organic material varies considerably (e.g. Goldstein and Galbally 2007; Jimenez et al. 2009) and this variation directed our choice. We will, however, add a table detailing the coating temperatures to the revised manuscript. The oleic acid results simply highlight the need of laboratory measurements with simple model compounds. Our results show that by using the existing model frameworks, we cannot explain all of the experimental observations, but more theoretical work is needed. We will highlight this in the revised manuscript.

For lack of originality, the manuscript just declares plain trivialities such as on Page 9 Line 5-6 "As expected, the critical supersaturation is generally higher for pure BC particles than for the particles with organic coating and the pure organic particles have the lowest critical supersaturation". Overall, this manuscript presents a lab-based approach in combination with a well-established theoretical approach that has little if any atmospheric relevance. It is a textbook-like repetition of previous studies and completely lacks originality and innovation.

First, we would like to point out that the fact that the present theories are shown to work well for two of the studied organic compounds does not mean that the results are not novel. Not all novel results need to be surprising or worrying in terms of the application of the theory for atmospherically relevant calculations. The reviewer claims that our study is a repetition of previous studies, but does not provide any references to back up these claims. We are not aware of any previous studies that investigate the applicability of the adsorption-activation approaches for BC particles systematically coated with the studied organic compounds using a similar approach to the one presented here. We therefore find the criticism unjustified.

References

- Canagaratna MR, Massoli P, Browne EC, et al (2015) Chemical compositions of black carbon particle cores and coatings via soot particle aerosol mass spectrometry with photoionization and electron ionization. *J Phys Chem A* 119:4589–4599 . doi: 10.1021/jp510711u
- Corbin JC, Sierau B, Gysel M, et al (2014) Mass spectrometry of refractory black carbon particles from six sources: Carbon-cluster and oxygenated ions. *Atmos Chem Phys* 14:2591–2603 . doi: 10.5194/acp-14-2591-2014
- Goldstein AH, Galbally IE (2007) Known and unexplored organic constituents in the earth's atmosphere. *Environ Sci Technol* 41:1514–1521 . doi: 10.1021/es072476p
- Healy RM, Wang JM, Jeong C, et al (2015) Light-absorbing properties of ambient black carbon and brown carbon from fossil fuel and biomass burning sources. *J Geophys Res* 6619–6633 . doi: 10.1002/2015JD023382. Received
- Jimenez JL, Canagaratna MR, Donahue NM, et al (2009) Evolution of organic aerosols in the atmosphere. *Science* (80-) 326:1525–1529 . doi: 10.1126/science.1180353
- Onasch TB, Trimborn A, Fortner EC, et al (2012) Soot Particle Aerosol Mass Spectrometer: Development, Validation, and Initial Application. *Aerosol Sci Technol* 46:804–817 . doi: 10.1080/02786826.2012.663948
- Sedlacek AJ, Lewis ER, Onasch TB, et al (2015) Investigation of Refractory Black Carbon-Containing Particle Morphologies Using the Single-Particle Soot Photometer (SP2). *Aerosol Sci Technol* 49:872–885 . doi: 10.1080/02786826.2015.1074978