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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Droplet activation behaviour of atmospheric black carbon particles in fog as a function of their size and mixing state" *by* Ghislain Motos et al.

Anonymous Referee #2

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General comments:

The authors report results from a case study comprising four separate fog events observed in an urban environment in Zurich. Overall, the manuscript is well written and the data analysis has been conducted with great care. The results show that soluble coating on top of an insoluble black carbon (BC) cores indeed increases their ability to serve as condensation nuclei for fog droplets, and the threshold coating thickness decreases with increasing BC core size. Furthermore, the authors demonstrate that a simple κ-Köhler model can be used to predict the fog droplet activation when the particle size, coating thickness and hygroscopicity of the coating material are known.

Understanding the mixing state of ambient BC and its impact and fate in the atmosphere has been of great interest to aerosol community, and thus, the manuscript by Motos et al. is well within the scope of ACP. That said, the main findings of this study are more incremental rather than novel and (as such) provide a little new insight into the studied topic. Therefore, I would like to see more discussion concentrating on the implications of the results, e.g., how black carbon and its aging are currently treated in particle-resolved models (that were also mentioned in the conclusions) and how these new results could possibly improve these aspects. In other words, there is definitely no need to shift the focus of the paper from experimental research into modelling, but instead, highlight the importance of the results and point out more concretely how aerosol community could benefit from them. In my opinion, this would improve the impact of the paper substantially. Otherwise, I only have a few minor comments and suggestions to be considered by the authors.

Specific comments:

Page 3, Line 21: A relatively recent paper by Maalick et al. (2016) presents results from LEM simulations concentrating on the effect of BC on the evolution and lifetime of radiation fog. Although this specific paper does not directly deal with BC mixing state, it points out an important aspect of BC in aerosol-cloud/fog interactions and could be cited in this paragraph (if the authors wish).

Page 3, Line 35: The study by Dalirian et al. (2018) has been conducted by atomizing BC particles from aqueous solutions and then coating them with organics by using a tube furnace. Therefore, it should be referred to as laboratory study rather than a conventional chamber measurement.

Page 5, Line 26: Later in the paper, the authors are referring to uncertainties in CCN calibration (Sect. 3.1). Therefore, it would be good to briefly describe how the instrument was actually calibrated and how the possible instrumental limitations are affecting the measurement uncertainties especially at the lowest and highest supersaturations.

Page 8, Line 24: Here, the authors define that the hygroscopicity of the soluble coating $\kappa_{coating}$ is equal to κ_{median}, which according to Sect. 2.3.4 is directly inferred from CCNC measurements. To my understanding, the κ value obtained from CCNC data is representative for all particles of equal size, and thus, reflects the possible presence of non-hygroscopic black carbon. This would mean that $\kappa_{median} \rightarrow \kappa_{coating}$ only when the fraction of BC containing particles $\rightarrow$ 0.

According to the manuscript BC-free particles "represent majority of the particles" (Page 14, Line 15), and therefore, the definition of $\kappa_{coating} := \kappa_{median}$ would be justified. Is this rationale correct or have I misunderstood the applied notation? In any case, I'd like to ask the authors to describe the reasoning behind $\kappa_{coating} := \kappa_{median}$ more carefully to improve readability and to avoid any danger of misunderstanding.

This leads me to another question: can you quantify "majority of the particles"? For example, would it be useful/possible to have a plot estimating the number or volume fraction of particles with BC core as a function of dry particle size (e.g. in supplementary material)?

Page 11, Line 15: The authors state that the anomalies in the size-dependence of κ are likely due to the increased uncertainties in CCNC calibration at the lowest and highest supersaturation. In the next two paragraphs, however, the results from these two supersaturations are being discussed more detailed and the authors even use the measured value of κ_{median} = 0.6 (at SS = 1.33%) to support their hypothesis on nighttime accommodation of ammonium nitrate. Frankly, this would not make much sense if the anomalies in the size dependence of κ were solely due to calibration uncertainties. It should be addressed more carefully how the CCNC calibration uncertainties effect the data and data interpretation.

Page 11, Line 36: The authors have done great job assessing the contribution of different sources (traffic and wood burning) on the mixing state and presence of non-hygroscopic particles. However, it feels that such a comprehensive analysis and pre-

sentation shifts the attention away from the focal points of the manuscript. I would like to ask the authors to consider condensing this part of the manuscript by moving “less important” parts and maybe some of the figures to the supplementary material and to concentrate especially on those periods relevant for analyzed fog events.

Page 15, Line 6: According to Fig. 3, the range between the 95% confidence intervals also illustrates the range of variation during the fog events. Therefore, the derived uncertainty of SS_{peak} (Table 2) could be somewhat interpreted as an indicator of temporal variation. In my opinion, these uncertainty estimates should be discussed, or at the very least, mentioned in this paragraph.

Page 37, Figure 10: The figure caption says, “The variability in the fog-activated fraction induced by the choice of $\kappa_{coating}$ (retrieved $\kappa_{median} \pm 0.05$) is represented by horizontal bars“. Why is an arbitrary (?) uncertainty of 0.05 used and not the uncertainty indicated by the 95% confidence intervals like in Table 2?

Technical comments:

Page 5, Line 17: This sentence needs some minor rephrasing as something seems to be lacking, e.g., “...from 20 to 593 nm in 5.5 min, after which the monodisperse aerosol...”

Page 5, Line 30: “...was used behind the total inlet...” Should this say interstitial inlet instead of total inlet?

Page 16, Line 5: The sentence starting as “The BC cores with...” is not easy to understand and could be rephrased to improve readability.

Figures: Is it possible to increase the font sizes especially in Figures 3, 5, 7 and 11.

References:

Z. Maalick, T. Kühn, H. Korhonen, H. Kokkola, A. Laaksonen, S. Romakkaniemi, Effect of aerosol concentration and absorbing aerosol on the radiation fog life cycle, Atmo-

spheric Environment, 133, pp. 26-33, https://doi.org/10.1016/j.atmosenv.2016.03.018, 2016.

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