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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.

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RESPONSES TO THE REFEREES AND CHANGES MADE TO THE MANUSCRIPT.

The authors would like to thank the three referees for their constructive comments which helped to make the paper clearer and easier to understand. This document presents, for each comment from the referees, a response and a note clarifying what has been changed in the manuscript. Indications of page and line numbers refer to the revised version of the manuscript (without track changes).

Anonymous review of manuscript: General remarks

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Discussion paper

This paper investigates the activation of internally mixed black carbon in fog by making use of the low supersaturations within fog to do a closure study on the droplet activation behavior of BC-containing particles. The measurements were taken during a field campaign in a residential area of Zurich in the winter, and indicate that aerosols sourced from traffic during rush hour periods are generally less hygroscopic than aerosols sourced from wood burning. The paper is well-written and uses novel methods to demonstrate good agreement between predicated and observed behavior. It is appropriate for ACP and is a useful scientific result that will help to constrain the lifetime of BC in the atmosphere, and demonstrates that simple parameterizations of hygroscopicity in terms of a kappa-Köhler parameter are in good agreement with atmospheric observations. The methods and measurements are adequately described, as are comparisons with previous atmospheric observations. There are a few minor points that should be clar-ified to make the paper clearer. The paper would also benefit from a more focused discussion on the major conclusions of the paper, as it is sometimes challenging to follow.

Specific comments from Referee #3:

Comment: “Some of the figures are hard to read (the text is very small). There are also quite a large number of figures (11) and I would suggest moving some of the less important figures (e.g. figures 4, 5, or 6) to the supplemental information to draw more attention to the other figures.”

Changes: We increased the font size in most of the figures. Figure 5 was moved to the Supplement as well as the corresponding text. However, Figure 4 gives a good overview (the only time series) of several parameters during the week of the four fog events and Figure 6 is important to describe the impact of the vehicle emissions during the “rush hours”.

Comment: “To improve the clarity of the discussion it would be useful to have a table summarizing the different variables, such as the activation diameters and supersatura-

tions."

Response: The activation diameters, supersaturations, as well as other information concerning the four fog events, are already listed in Table 2.

Changes: We added two references to Table 2 in the manuscript: "Two values of SS-peak are given for each fog event in Table 2" in Sect 3.3 (p. 16, l. 11) and "D_halfˆfog and D_50ˆfog lay in the range 320 to 380 nm and 370 to 470 nm, respectively (see Table 2)", also in Sect. 3.3 (p. 16, l. 6).

Comment: "It would be useful to clearly state the upper and lower limits for the optical size range of non-BC containing particles detected by the SP2 in the 8-channel configuration, and at what optical size the scattering detectors are saturated."

Response: Various quantities can be inferred from the data delivered by the 8 channels of the SP2 alone and also from combinations of these data, each of which has its own lower and upper limits and detection and quantification. The dynamic ranges covered by the SP2 for different parameters are directly accessible where needed, i.e. from the range of data shown in the figures: e.g. optical diameter for standard sizing and LEO-fit based sizing in Figs. 3a and 7, rBC mass equivalent core diameter in Fig. 8-10, and coating thickness in dependence of BC core diameter in Fig. 10.

Changes: Several limits of detection are now also explicitly mentioned in the methods section (Sect. 2.2.3). "The respective lower limits of quantification are $\sim$0.32 fg translating to and $\sim$70 nm (note, smaller BC core can also be detected with a detection efficiency of less than unity). At the upper end, BC size distributions are only shown up to 300 nm in diameter, due to insufficient counting statistics at larger sizes." "The peak amplitude of the elastically scattered light is used for optical sizing of BC-free particles from 130 nm to 380 nm."

Comment: "It looks like the laser power in the SP2 used to determine the optical size was only calibrated twice with PSL's, before and after the campaign; were these two

calibrations consistent?"

Changes: One sentence added to the instrumental section (Sect. 2.2.3), p. 6, l. 30: "The laser monitor did not indicate a laser power drift and the calibration coefficient for the scattering detector varied by less than 2% between the two calibrations. Therefore a constant calibration coefficient was applied for the whole campaign."

Comment: "Why was the AMS not used to estimate the index of refraction of the coatings based on the chemical composition of the bulk aerosols? Also, what is the motivation behind choosing the refractive index values for the coatings? These values were given without justification or reference. How much would the index of refraction vary based on the observed bulk aerosol chemical composition, and what is the sensitivity of the calculated kappa values for different values of index of refraction for the BC coating?"

Response: Choosing a refractive index of 1.50 + 0i at 1064 nm very often brings mobility sizing and optical diameter to close agreement for atmospheric aerosols. ACSM derived estimates of the refractive index would not provide additional benefit given the relatively large "representative diameter" of the mass based bulk measurement and the uncertainty of the actual refractive index of the organic fraction at 1064 nm.

Changes: The following addition was made in the methods section (Sect. 2.3.3), p. 6, l. 32:. "Calibrated scattering cross section measurements of BC-free particles were converted to optical diameters (Dopt) assuming spherical particles with a refractive index of 1.50+0i at 1064 nm. With this choice, the particle number size distributions measured by the SMPS and the SP2 agree well in the overlapping size range (not shown) and optical sizing is only weakly sensitive to the applied refractive index (Taylor et al., 2015)."

Comment: "Figure 9 – This size dependence could also potentially be explained by dry deposition removing larger, thickly coated BC particles more efficiently. It would be useful to estimate the relative importance of dry deposition.

Remark: Due to the move of Figure 5 to the Supplement, Figure 9 is now Figure 8. We use the latter name in the paragraph below.

Response: Figure 8c gives an activated fraction, i.e. the fraction of particles that activated to cloud droplets among airborne particles. The brown line in Figure 8c only considers the subset of BC with thick coatings, and gives the activated fraction of this group. Particles deposited to the ground are not considered in this calculation. For example, even if 60% of the thickly coated BC got deposited to the ground by dry processes, the brown line in Figure 8c gives the activated fraction of the other 40% that are still suspended in the air. However, our instrumentation does not allow us to quantify the relative importance of condensation and dry deposition time scales.

Changes: No changes to the manuscript.

Comment: “Also, are there any potential size-dependent biases in using the delay time SP2 method for separating the two populations of aerosols?”

Response: Yes, care needs to be taken with the delay time method because the “delay time” cannot be detected for “thinly coated” small BC cores nor for “thickly coated” large cores. However, we only show BC-core size segregated data and only for the core size range where lower/upper detection limits do not bias the result (see grey shadings in Fig. 8b&c).

Changes: No changes to the manuscript.

References (same for the responses to all referees):

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