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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud droplet activation properties and scavenged fraction of black carbon in liquid-phase clouds at the high-alpine research station Jungfraujoch (3580 m a.s.l.)" *by* Ghislain Motos et al.

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We thank the two referees for their constructive comments, definitely helpful to clarify certain points of the paper.

The page and line numbers enumerated in this document refer to the corrected maunuscript (without track changes).

Answers of the authors to the interactive comment of Anonymous Referee #1 (Referee Comment 1)

Anonymous review of manuscript: General remarks

The present manuscript describes the use of a sophisticated aerosol sampling system at a mountain site to investigate cloud droplet activation of both BC-free and BC-containing particles. The authors present unique data set of the in-situ CCN properties and scavenged fractions of these aerosols on a number and mass basis, which are useful for elucidating the parameters (water vapor supersaturation and aerosol microphysical properties) that control activation. Through the data analysis, they demonstrate that the simple k-Köhler theory under the assumptions of spherical core-shell structure of BC-containing particles can reasonably predict their cloud droplet activation, despite their complex morphology and non-ideal properties of coating materials in the real world. This is an important observational evidence that justifies the simple treatment of BC activation in regional and global models. Although I think some points in the manuscript need clarification, but overall I recommend this manuscript for publication in ACP after minor modification.

Specific comments from Referee #1:

P12, Line 30–31 and P13, Line 6–8: In these parts, the authors state that "Such variations in SS_peak are driven by variations in atmospheric dynamics (i.e., updraft)..." and "variations in D_half were mainly driven by variations in updraft velocities and resulting supersaturations,...", respectively. However, in addition to the updraft velocity, the SS_peak can also depend on total aerosol number concentrations (i.e., higher number concentrations of aerosols can lead to lower SS_peak). Therefore, the relationship between the absolute number concentrations of aerosols (total inlet) and SS_peak should be mentioned somewhere in the paper and the aerosol concentration data can be included in Table 1. Furthermore, to characterize cloud properties discussed in this paper, please consider including the LWC data in Table 1. The difference of the SMPS data between total and interstitial inlets may indicate the cloud droplet number concentrations, which might be also useful for characterizing cloud properties.

Remark: A very similar comment was posted by the Anonymous Referee #2.

Response: The relationship between potential CCN, cloud droplet number, updraft velocity and cloud peak supersaturation, i.e. cloud formation in CCN-limited or updraft limited regimes, was extensively studied at the Jungfraujoch site by Hoyle et al. (2016).

Changes: We modified the paragraph mentioned by the Referee (p.12, l.34) to: "Such variations in SSpeak are primarily driven by variations in atmospheric dynamics (i.e. updraft) at the cloud base and to a lesser extent by the number concentration of potential CCN, as demonstrated for the Jungfraujoch site by Hoyle et al. (2016)." We included the median liquid water content (LWC), median particle number concentration of potential CCN (i.e. particles larger than 90 nm in diameter) and the median droplet number concentration during each cloud event in Table 1. Two new references to Table 1 were added, p.12, l.37 and p.13, l.14: "Variations in effective cloud peak supersaturation are a priori unrelated to the cloud LWC (Fig. 5a and Table 1)", "The key parameters for each selected cloud period are summarized in Table 1, including the droplet number concentration inferred from the difference in particle number concentration between the total and the interstitial inlet, and the median number concentration of potential CCN, i.e. particles with a mobility diameter larger than 90 nm (N90; e.g. Hammer et al.; 2014a)."

P14, Line 18–22: If the deviations of several data points in Figure 8a from 1:1 line are greater than measurement uncertainty, some data would indicate that BC particles were scavenged more efficiently than total aerosols. I do not understand the reason for that.

Response: This is an important observation, which we did not address. We have therefore introduced the following changes in the manuscript:

Changes: The last sentence of the following paragraph (p.14, l.25) has been added in the revised manuscript: "The scavenged fraction of BC mass is only expected to be equal to the total aerosol volume scavenged fraction for all peak supersaturations, if BC

contributes an equal fraction to the aerosol volume at any particle size and if the critical activation diameters of the BC-containing particles and total aerosol are equal. While the latter condition is closely fulfilled if BC is internally mixed with substantial coatings, size-independent BC volume fractions are a priori not expected. Nevertheless, the scavenged fractions of total aerosol volume and BC mass are essentially equal on average. However, deviations of several data points in Figure 8a from the "1:1"-line are greater than measurement uncertainty, indicating that even at remote locations the BC scavenged fraction can differ from the total aerosol volume scavenged fraction in individual cloud events, likely due to some size dependence of the contribution of BC to the aerosol volume and/or disagreement between the critical activation diameters of BC-free and BC-containing particles. For example new particle formation events followed by growth to sizes below the droplet activation cut-off diameter is one possible mechanism that can result in the BC scavenged fraction becoming greater than that of the total aerosol volume."

P15, Line 10–14: What refractive index values are assumed for BC core and BC-free aerosols for the SP2 data analysis?

Changes: We included the following paragraph into the experimental section (Sect. 2.2.2), p.6, l.17: "A refractive index of 1.50+0i was chosen to convert the scattering cross section measurements of BC-free particles to optical diameters, which brought the SP2 and SMPS derived size distributions in agreement in the overlapping size range (the optical sizing is only weakly sensitive to the choice of refractive index as shown by Taylor et al., 2015). For BC-containing particles, the same refractive index was used for the coatings, while 2.00+1.00i was chosen for the BC cores. This choice resulted in agreement between the optical diameters of the bare BC cores measured just before incandescence onset and the rBC mass equivalent diameters."

Figure 9b and 9c: Which inlet for these SP2 data? Total inlet?

Response: This is all behind the total inlet.

Changes: The caption of Figure 9 was adapted.

Figure 9c: Is the y-axis number fraction of thickly coated BC? The caption is not clear.

Changes: The legend and labels of the figure were modified to make it clear.

P16, Line 6–8: As the authors mentioned in P16, Line 1–4, the SS_peak values may be underestimated due to increased interstitial aerosol number concentrations due to WBF process in mixed phase clouds. Therefore, comparing the activated fraction for T < -5 C case and warm cloud case with "comparable SS_peak" looks logically inconsistent.

Changes: This paragraph now reads (p.16, l. 16): Mixed-phase or even completely glaciated clouds may occur at lower temperatures. Mixed-phase clouds may result in the conversion of particles from droplets (activated particles) to interstitial aerosol through the Wegener-Bergeron-Findeisen process (e.g. Cozic et al., 2007), thereby potentially obscuring the causal relationship between SSpeak and droplet activation. However, Verheggen et al. (2007) showed that D_halfˆcloud remains well-defined and that only small differences in average D_halfˆcloud exist between mixed-phase and liquid clouds. This suggests that the Wegener-Bergeron-Findeisen process does not affect the inferred SSpeak. The fact that the Wegener-Bergeron-Findeisen process evaporates some cloud droplets, whereby the droplet nuclei are released back to the interstitial aerosol, explains that the BC activated fraction was lower in most clouds at temperatures below 5 °C compared to that in warm clouds at comparable peak supersaturation (see the dashed lines in Figure S3a).

Figure 11: In this figure, coating thickness is indicated by color scale for rBC with mass equivalent diameter of about 50–300 nm. However, if the small BC (D_rBC 50 nm) has thin coatings (i.e., total optical diameter < 180 nm), the SP2 cannot quantify the coating thickness?

Response: Indeed, the LEO-fit technique cannot be applied to particles with an optical

diameter smaller than around 180 nm, whether they contain BC or not.

Changes: The text "particles larger than around 180 nm only" was added to the sentence "The reconstructed scattering amplitude is then used to infer the total particle optical diameter, for particles larger than around 180 nm only", in Sect. 2.2.2, p.6, l.14. The caption of Figure 11 was rewritten and now reads: Figure 11. Activation of BC-containing and BC-free particles during the 25 June (a) and 26-27a June (b) stable cloud periods. Panels (a1) and (b1) show the critical supersaturation of individual BC-containing particles as a function of rBC mass equivalent diameter coloured by coating thickness for the interstitial inlet and in grey for the total inlet. Note that the coating thickness can only be determined for BC-containing particles with an overall diameter greater than 180 nm, which explains the missing data points in the top left part of panels (a1) and (b1). Panels (a2) and (b2) depict the corresponding activated fraction of BC-containing particles as well as that of BC-free particles (SP2-derived) and bulk aerosol (SMPS-derived). Only one fourth of data points are shown in panel (a1) in order to visualize the fraction of points originating from interstitial inlet data compared to points from the total inlet data. Horizontal light blue lines indicate the value of SS-peak retrieved using D_halfˆcloud (method explained in Sect. 3.5). Note that values of SSpeak for both cases (a) and (b) are at a level above which the SP2 detects only almost bare BC because small cores with substantial coating are outside the detection limits of the SP2.

P19, Line 28, "lower" should be "higher"?

Changes: Manuscript corrected.

Minor corrections:

P3, Line 35: "(2016))" should be "(2016)".

P5, Line 20: "(Very. . .(2000)" should be "(Very. . .(2000))".

P17, Line 22: "DrBC" should be "D_rBC"

Changes: Thank you; Manuscript corrected.

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