Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-811-AC1, 2019

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.

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Received and published: 23 January 2019

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).

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

Comment 3)

Anonymous review of manuscript: General remarks "This study presents the measurement of BC activation by droplet in real world, the topic is within the scope of ACP. I think there are a few places needing to be addressed before it can be accepted." Specific comments from Referee #1:

Comment: "Firstly as there is no page number, it is hard to make specific comment."

Response: This issue was already fixed in version2 of the manuscript i.e. as part of the technical correction after the "QuickAccess-Review" stage.

Comment: "The abstract is too long, I would say maximum 2 paragraphs or better with 1 paragraph."

Response: Agreed by the authors.

Changes: The abstract now reads: Among the variety of particle types present in the atmosphere, black carbon (BC), emitted by combustion processes, is uniquely associated with harmful effects to the human body and substantial radiative forcing of the Earth. Pure BC is known to be non-hygroscopic, but its ability to acquire a coating of hygroscopic organic and inorganic material leads to increased diameter and hygroscopicity, facilitating droplet activation. This affects BC radiative forcing through aerosol-cloud interactions (aci) and BC life cycle. To gain insights into these processes, we performed a field campaign in winter 2015/16 in a residential area of Zurich which aimed at establishing relations between the size and mixing state of BC particles and their activation to form droplets in fog. This was achieved by operating a CCN counter (CCNC), a scanning mobility particle sizer (SMPS), a single particle soot photometer (SP2) and an aerosol chemical speciation monitor (ACSM) behind a combination of a total- and an interstitial-aerosol inlet. Our results indicate that in the morning hours of weekdays, the enhanced traffic emissions caused peaks in the number fraction of externally mixed BC particles, which do not act as CCN within the CCNC; compared

to nighttime associated to heavily aged internally mixed BC from background air advected to the site. The very low effective peak supersaturations (SSpeak) occurring in fog (between approximately 0.03 and 0.06% during this campaign) restrict droplet activation to a minor fraction of the aerosol burden (around 0.5 to 1% of total particle number concentration between 20 and 593 nm) leading to very selective criteria on diameter and chemical composition. We show that bare BC cores are unable to activate to fog droplets at such low SSpeak, while BC particles surrounded by thick coating have very similar activation behaviour as BC-free particles. Using simplified κ-Köhler theory combined with the ZSR mixing rule assuming spherical core-shell particle geometry constrained with single particle measurements of respective volumes, we found good agreement between the predicted and the directly observed size and mixing state resolved droplet activation behaviour of BC-containing particles in fog. This successful closure demonstrates the predictability of their droplet activation in fog with a simplified theoretical model only requiring size and mixing state information, which can also be applied in a consistent manner in model simulations.

Comment: "It is recommended to include the previous studies in the introduction on BC heating on clouds, reducing cloud cover, decreasing cloud albedo."

Response: This is a legitimate request from the referee. Nevertheless, we decided to keep these additions short mainly for two reasons: First, the introduction is already quite comprehensive and our study is quite specific to fog. Second, we have a second manuscript meanwhile submitted to ACPD about BC activation in liquid clouds at a high altitude site (https://www.atmos-chem-phys-discuss.net/acp-2018-1054/), in which such previous studies on the abovementioned BC effects are more directly relevant and therefore also included in the introduction.

Changes: We added the following paragraph concerning fog lifetime to the introduction (p. 3, l. 19), as also suggested by Referee #2: "Although BC can dissipate fog through the semi-direct effect (evaporation of fog droplets due to absorption of solar radiation by BC particles and subsequent droplet evaporation), high concentrations of

other CCN were shown to influence fog lifetime in a stronger manner (Maalick et al., 2016). Because these CCN form droplets more efficiently, they lead to increased radiative cooling and decreased droplet removal through sedimentation, thus enhancing fog lifetime."

Comment: "a) what is the collection efficiency of the total inlet on collecting droplet, i.e. what is the 50% size cut-off for the droplets, some large droplets may be missed?

Response: Hammer et al. (2014) use the same inlet for measurements in fog and looked at the influence of sedimentation (SI of their paper). They found that potential systematic bias in the observed activation cut-off diameter remains below 10%.

Changes: The following statement has been included on p. 4, l. 28: "Hammer et al. (2014; Supplement) showed that systematic biases in the observed activation cut-off diameter potentially resulting from incomplete collection efficiency of fog droplets in the total inlet remains below 10%".

Comment: "b) Will the heating of inlet affect the coating amount of coating compositions of BC."

Changes: The following statement has been included in the experimental section, p. 5, l. 17: "...The temperature increase from outside ($\sim$0 °C) to inside ($\sim$25 °C) the trailer also contributed to the drying of the sample air and thus evaporation of fog droplet water. Some evaporation artefacts of other semi-volatile aerosol components cannot be excluded. However, they are not expected to be excessive for particles in the upper accumulation mode size range, based on results by Nessler et al. (2003) for comparable temperature difference but at a different location.

Comment: "c) A clear plot is needed to show how the comparison looks between total and interstitial concentration for non-fog period. From the description in the text, this scaling varied from time to time, you may need to show a time series of this scaling ratio, and how this scaling ratio was affecting the results, and why."

Response: As mentioned in the manuscript p.5, l. 7, the scaling factor used to correct for line losses in the interstitial inlet was kept at 1.16 for SP2 data before 17 January 2014 and 1.03 afterwards. The initial bias was caused by a pressure drop in the interstitial inlet line, which was fixed on 17 December.

Changes: Figure S2a (Fig.1 in this Author comment), which shows the effect of line losses on SP2 data and the corrections applied to correct for them, was added to the Supplement. The reference "see Figure S2a" was added p. 5, l. 10.

Comment: "d) Also as stated: "For the scanning mobility particle sizer instruments, size-dependent scaling factors were calculated for each fog event in order to take into account both the different line losses behind each inlet and the internal measurement errors of each SMPS." This should be clearly shown by figure."

Changes: We added Figure S2b (Fig.2 in this Author comment) to the Supplement, which shows the scaling factor applied to SMPS data for each fog event. The reference "See Figure S2b" was added p. 5, l. 12.

Comment: "More explicit definition of internally or externally mixed BC is needed".

Changes: One sentence added in the introduction (p. 2, l. 14): "Throughout this study, we refer to BC mixing state in relation to coatings, i.e. a strong degree of internal mixing is associated with thick coatings whereas externally mixed BC is associated with no or very thin coatings."

Comment: "Could you also give the scavenging mass fraction of BC or non-BC particles".

Changes: One paragraph added to Sect 3.1, p. 13, l. 1: "The scavenged mass fractions of BC and the total aerosol, i.e. the mass fraction incorporated into fog droplets, were calculated using the SP2 and the two SMPS assemblies, respectively. The scavenged mass fraction varied between 6% and 12% for BC during the four fog events, and between 15% and 20% for the total aerosol. These results are in close agreement with

the fog studies of Hallberg et al. (1992), who reported 6% for elemental carbon (EC) and 18% for sulfate, and somewhat lower than the scavenged fractions of 26% for EC and 38-94% for various inorganic species as reported by Gundel et al. (1994) ."

Comment: "What is the black colour in Fig. 11".

Response: This is explained in the caption of Fig. 11: "Black pixels in the image indicate 2D-bins for which no particle was found in the total inlet data while at least one particle appeared in the interstitial inlet data, thus leading to a negative fog-activated fraction"

Changes: No changes to the manuscript.

Comment: "A plot showing how the LWC of fog has been associated with SS and related scavenging fraction".

Response: LWC is not associated with SS, it was only used to identify fog presence.

Changes: We added two sentences in Sect. 2.3.1 (p. 8, l. 23) to clarify this: "We used a minimum LWC of 100 mg m-3 measured by the PVM during at least one hour as threshold to define fog events. Note, the LWC was not used to infer fog peak supersaturation (see Sect. 2.3.5). Throughout the field campaign, four fog events were retained in the analysis of the present study, all of them between 14 and 20 December 2015 (Table 2)."

Comment: "What is the source origin of the particles, backtrajectory analysis? A map of the site will help a lot".

Response: The diurnal patterns of BC and particle concentrations as well as spectral dependence of aerosol light absorption were used to show the influence of local traffic emissions to the air sampled at the site. This is discussed in Sect. 3.2. Back trajectory analysis would not add further relevant information to this.

Changes: We added a map of the measurement site to the Supplement (Figure S1 but

Fig. 3 in this author comment) and references to Figure S1 p. 4, l. 10 and p. 4, l. 15.

Comment: “How is ACSM used?” Response: The ACSM was used downstream the total inlet during the campaign. ACSM data were only used for time series of species concentrations in Figure 4.

Changes: No changes to the manuscript.

Comment: “However, Figure 9c clearly shows that droplet activation of BC-containing particles is the mechanism that explains the incorporation of BC cores into fog droplets in the present study: if coagulation between BC particles and fog droplets was giving a dominant contribution, then the fog-activated fraction of BC particles would exhibit much less size and coating dependence and rather with opposite trends.” This discussion is not clear at all, so have you observed the coagulation of the BC with droplet? what “opposite trends” are they?”

Changes: The following sentences are added to Sect. 3.4, p. 17, l. 31: “Coagulation scavenging efficiency decreases with increasing particle size, as shown by e.g. Ohata et al. (2016). Therefore, the BC core size and coating thickness dependence of coagulation scavenging would be opposite to the observed relationship. By contrast, the observation is consistent with expectations for nucleation scavenging.

Comment: “Six calibrations were performed, including pre and post campaign, and standard data analysis procedures using the Tofwerk “IgorDAQ” software package (Tofwerk AG, Thun, BE, Switzerland) were applied (reference).” What reference is it¿‘

Changes: This has been corrected.

Comment: “The key conclusion is to say the model combing ZSR and Kohler theory could well predict the BC activation, but there is no clear plot to show this.”

Response: This is the purpose of Figure 10. And it is also shown in Figure 11. Detailed explanations are provided in Sect. 3.5 (copied here): “For each fog event [in Figure 10],

50% fog-activated fraction is reached at an SScrit very close to the SSpeak derived from D_50ˆfog. This agreement confirms that observed activation of BC particles in the fog matches the expected droplet activation behaviour of BC-containing particles as theoretically predicted from independently measured BC-particle properties (size, BC volume fraction and coating hygroscopicity). This demonstrates that closure is successfully achieved, i.e. SP2-based characterization of BC-containing particle properties combined with κ-Köhler theory is sufficient to accurately describe the activation behaviour of BC-containing particles in fog, despite the fact that either of them are based on the simplifying assumption of spherical core-shell morphology.” And: “Figure 10b-e also contains the fog-activated fraction of BC-free particles detected by the SP2, for which SScrit was calculated using κ-Köhler theory with κmedian and optical diameter from the SP2. 50% activation is by definition reached by those particles with SScrit equal to SSpeak inferred from D_50ˆfog (small deviations are explained by binning the fog-activated fraction data in supersaturation rather than diameter space). The fact that the activation curves of BC-containing particles in Figure 10b-e agree well with the activation curves of BC-free particles implies the following: the activation of BC-containing particles to fog droplets can be described identical to the activation of BC-free particles but for adjusting the κ-value with the ZSR-rule to account for the volume fraction of insoluble BC. This is an alternative but equivalent view of how closure is achieved for the activation of BC to fog droplets.”

Changes: No changes to the manuscript.

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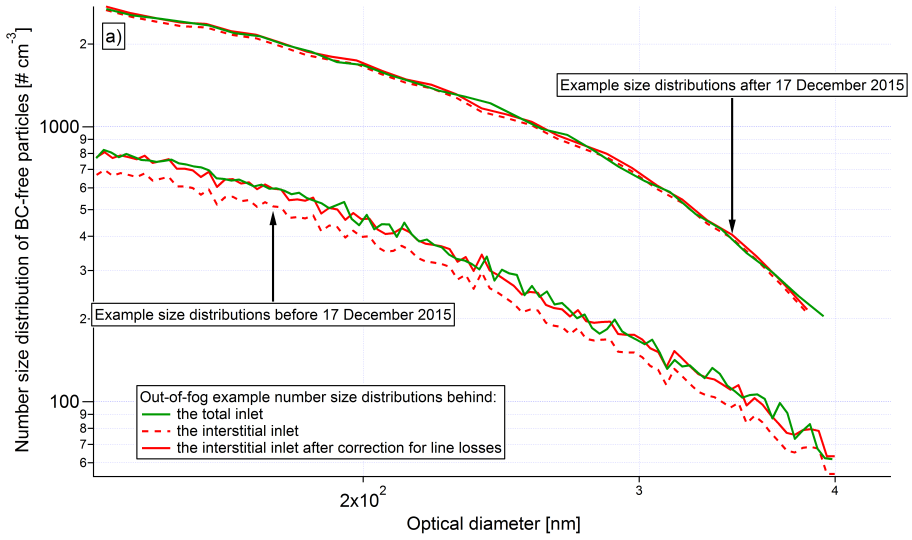

Fig. 1. Figure S2: (a) Example SP2-derived particle number size distributions during out-of-cloud conditions showing the corrections made on the interstitial inlet data by the use of scaling factors. A scalin

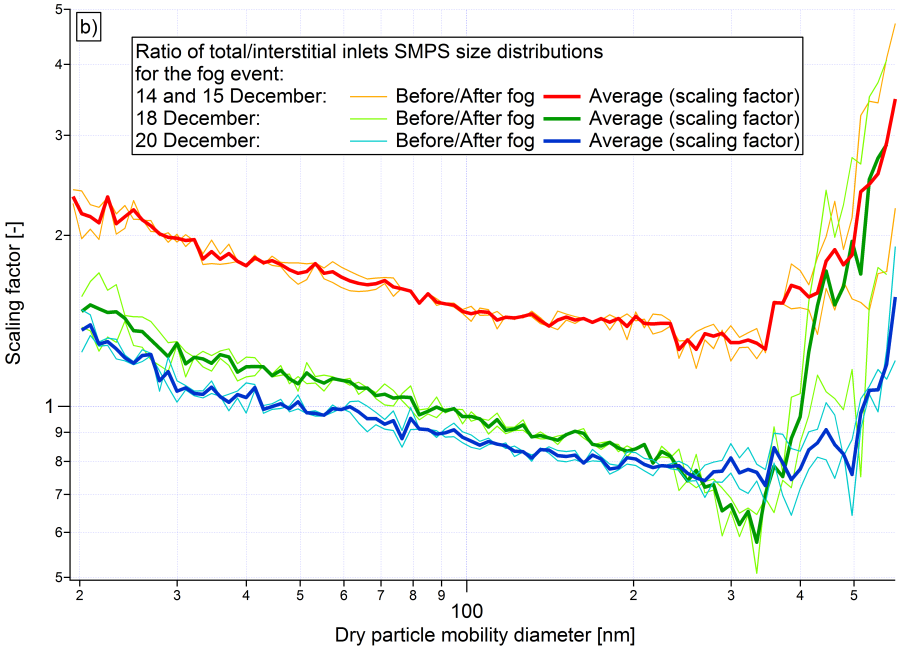

Fig. 2. Figure S2: (b) Size-dependent scaling factors for correcting SMPS data based on averaged out-of-cloud SMPS measurements before and after each fog event analysed in this study. The replacement of a con

Fig. 3. Figure S1: Satellite picture of the Irchel campus and its surroundings. The red cross denotes the location of the measurement site. Map data: ©2018 Google Earth – © 2009 GeoBasis-DE/BKG.