# Acp-2023-30: "Airborne investigation of black carbon interaction with low-level, persistent, mixed-phase clouds in the Arctic summer"

We would like to thank the referees for their detailed and constructive comments, which helped us to improve our manuscript. While the reviewer's comments are given in black bold, our answers are given below in grey letters. Additionally, we added the **changes made in the revised manuscript in grey bold letters**.

#### Answers of the authors to Reviwer#1

#### **GENERAL COMMENTS:**

The primary objective of this study is to evaluate rBC properties in cloud hydrometeor residuals and document how these compare under varying synoptic patterns and cloud conditions. A lot of text is created documenting these patterns and conditions, and the authors are quite thorough in the detail that they include describing these patterns and conditions; however, by the end of the manuscript, it becomes quite clear that they are unable to link differences in the observed rBC mass concentrations, size distributions or mixing state to any atmospherically relevant parameter. Put a different way, neither the synoptic patterns or microphysical properties of the clouds are obviously correlated with the rBC properties.

Hence, a major portion of the manuscript that describes these patterns and cloud could either be greatly abbreviated or be relegated to the supplement. This includes much of the effort that seems to have gone into describing the SID, SIP, Nevzorov and PHIPS and how they are processed to derive size, shape and concentration since neither LWC, IWC or cloud hydrometeor size distributions are convincingly link to cloud processing of the rBC. In fact, the only features of importance are the regions below, within and above clouds, regardless of their meteorological states. I strongly recommend that the paper be greatly shortened and focus primarily on the possible mechanisms by which the rBC became part of a cloud hydrometeor and only keep those meteorological (T, RH, winds) or cloud (size distributions) parameters in the manuscript that can make the case for or against how these parameters influenced how ambient rBC is processed by the cloud.

The manuscript was modified according to the reviewer's suggestion:

- The cloud-microphysics data were reduced to a minimum. The liquid water content derived from the SID-3 and the ice water content derived from the CIP are now used to generally describe the dominant phase of the cloud. As a consequence, the technical description provided in Section 2.2.2 was also shortened.
- Inertial scavenging was investigated with the analytical approach of Baumgardner et al. (2008) and discussed in Section 3.4.1. Most likely due to the different nature of clouds observed in this study compared to Baumgardner et al. (2008), we concluded that rBC particles and the bulk aerosol were activated following a similar activation process.
- The mostly speculative Section 4 was removed, while the discussion on the in-cloud vertical variability now focusses, mostly, on cloud-top and cloud-bottom sections.

With these major changes we gave more space to the discussion of : 1) presence and properties of cloud-active BC particles; 2) scavenging mechanisms; 3) BC residuals vertical distribution.

#### **INSTRUMENTATION:**

1. Why isn't ice being distinguished in the Small Ice Detector? The high resolution CCD array is specifically designed to allow distinguishing liquid droplets from ice crystals. Why isn't this done? Why make the assumption that the SID only is measuring liquid droplets?

In the updated ACLOUD dataset, the two dimensional scattering signal of the SID-3 probe was analysed for particles sphericity (Vochezer et al., 2016) and crystal complexity (Schnaiter et al., 2016). More

specific details can be found in Järvinen et al. (2023). Now,  $N_{\text{Dro}}$  refers only to liquid droplets quantified in the 5-45 µm diameter range (See more details on the size segregation of liquid droplets in the answer to comment number 6). Due to the low number concentration of ice crystals detected by the SID-3 (Figure A), the statistics of  $N_{\text{Dro}}$  did not change significantly in the latest version of the manuscript. More details on the SID-3 data treatment and definition of cloud boundaries are given in the answers to comments number 6 and number 16. In the description of the SID-3 in Section 2.2.2 the comparison with the Nevzorow was removed and the text modified as:

... "The Small Ice Detector Mark 3 (SID-3; Hirst et al., 2001; Vochezer et al., 2016) allows deriving the cloud particle number size distribution in the 5-45 diameter μm range (https://doi.org/10.1594/PANGAEA.900261; Schnaiter and Järvinen, 2019b). Liquid droplets and ice crystals were distinguished based on analysing the SID-3 two-dimensional (2-D) scattering patterns for the particle sphericity as described in Vochezer et al., 2016. A more detailed description of processing the SID-3 data for ACLOUD including correction of coincidence artefacts can be found in Järvinen et al. (2023). In the present work, the number concentration ( $N_{\text{Dro}}$ ) and diameter ( $D_{\text{Dro}}$ ) of liquid droplets was calculated in the 10 - 45 µm diameter range to match the low size cut-off of the CVI inlet (10 µm). The liquid water content (LWC) was calculated from the size distribution assuming spherical particles and a particle density of 1 g cm<sup>-3</sup>. Due to the scarcity of ice in the 10 - 45  $\mu$ m diameter range, ice crystals measured by the SID-3 are not discussed in the present work." ...



Figure A Comparison between droplet number concentration ( $N_{Dro}$ ) and total cloud particle concentration ( $N_{Dro+lce}$ ; droplet and ice) for the cloud events presented in the manuscript (02-08 June 2017).

### 2. Why aren't you looking at both Nevzorov sensing elements to get ice fraction? The Nevzorov consists of a cylindrical LWC hot wire and a heated cup for TWC.

Unfortunately, due to the uncertainties in total water content and liquid water content measured with the Nevzorov probe, the difference method could not be used to calculate ice water content, which had the same order of magnitude than the uncertainties (Ehrlich et al., 2019). In view of this aspect and considering that Nevzorov data were not used in the results section, the Nevzorov probe is not described any longer in the method section, while the comparison with the SID-3 LWC was removed.

### 3. Throwing out the first two channels of the CIP is not a generally agreed upon action and doing so is ignoring an important size interval for drizzle.

The particle concentrations for sizes below 100  $\mu$ m are highly uncertain due to the presence of larger particles which are out of focus. Also, out-of-focus image corrections are more uncertain the lower the number of pixels. Due to this uncertainty in the first classes, we have decided to remove the first two channels of the CIP instrument.

## 4. How were ice crystals distinguished in the CIP? It is difficult to determine circularity until you have at least 6 – 8 pixels, i.e. > 200 $\mu$ m.

The validation criterium of circularity was based on area rather than diameter. We followed Crosier et al. (2011) using Particle Size Distribution (maximum diameter – all in focus) and a circularity > 1.25 only for area larger than 16 pixels. Obviously, that does not include particles with sizes smaller than 125  $\mu$ m.

#### 5. Why isn't Phips being used to fill in 45-75 fraction?

Following the comments of both reviewers, the discussion about cloud microphysics was simplified to focus more on rBC activation processes. Considering that the larger ice crystals detected by the CIP (75-1550  $\mu$ m) would have a major contribution to ice water content compared to smaller ice crystals (D<75 $\mu$ m), the CIP data provided sufficient information to generally investigate the cloud-phase (ice water content) in the new Section 3.6.2. In order to shorten the manuscript PHIPS data were removed entirely from the manuscript since they only contained non-essential information.

## 6. If the cloud concentration/thresholding is from SID, and smallest size is 10 um, you are likely rejecting a lot of cloudy air in you analysis.

The treatment of SID-3 data was significantly modified.

- Phase discrimination was applied. Now, *N*<sub>Dro</sub> refers only to liquid droplets.
- For the identification of cloud boundaries and the characterization of cloud phase, the size-cut of 10 mm was removed. Now, *N*<sub>Dro</sub> and LWC were derived from the SID-3 probe in the 5-45 μm range.
- For residuals measurements, the number concentration of liquid droplets was calculated between 10 μm and 45 μm to match the low size-cut of the CVI (as in the previous version of the manuscript). To avoid confusion, the previous nomenclature "N<sub>Dro</sub>" was modified into "N<sub>Dro10</sub>".

## 7. The SP2 also provides a way to derive the temperature of incandescence to separate light absorbing rBC from other types of material that can absorb at 1024 nm and incandesce. Have the data been properly screened to be sure the incandescing particles are only rBC?

Since both reviewers raised this point, we address it in full details as it follows.

- COLOUR-RATIO: Different black carbon or soot types are characterized by a different boiling temperature, which can be derived from the ratio of the intensity of the incandescence signal detected by the broad-band detector (wavelength detection range of 300-800 nm) over the narrow-band detector (wavelength detection range of 630-800 nm). This ratio is commonly called "colour-ratio". Schwarz et al. (2006) showed how different soot types are characterized by different boiling-point temperatures. Dahlkötter (2014) showed an increase of the colour ratio from background conditions (values around 1.6-1.0) to biomass burning conditions (values around 2.0-1.5). So, the SP2 can be used to discriminate between different types of rBC.
- MINERAL-DUST INTERFERENCE: As shown by Schwarz et al. (2006), the SP2 is also sensitive to
  metal containing particles. This feature is nowadays exploited to derive the concentration of iron
  oxides with a modified SP2, based on non-overlapping detection range of two incandescence
  detectors (blue wavelength detection range of 300-550 nm; red wavelength detection range of
  580-710 nm; Yoshida et al., 2016). According to the latter, hematite and magnetite show a distinct
  colour ratio (~1.5) compared to rBC standard material (fullerene; 2.4). Considering the different
  wavelength detection range, the values presented by Yoshida et al. (2016) cannot be directly
  compared with ACLOUD or Dahlkötter (2014) measurements.
- COLOUR-RATIO DURING ACLOUD: The colour-ratio was calculated for the ACLOUD campaign: above-cloud median=1.49 (IQR=1.38-1.62), inside-cloud median=1.45 (IQR=1.31-1.66) and below-

cloud median = 1.44 (IQR=1.30-1.66) cloud. The colour-ratio analysis was also performed as function of mas equivalent diameter (Figure B). Similar to Dahlkötter (2014), we observed a decrease of the colour-ratio with increasing rBC mass equivalent diameter. However, we did not observe a clear difference between the above-cloud, inside-cloud and below-cloud measurements, which allowed for some clear screening of incandescing but yet non-rBC particles such as iron oxides as in Yoshida et al. (2016, 2020).

- SCREENING OF INVALID PARTICLES: Following the analysis performed in Yoshida et al. (2016), we tried to identify and remove those large particles (DrBC>300nm) showing prolonged incandescence signals, which might be related to mineral dust (Figure C). A signal was considered "valid" only if its duration was shorter than 30 us and the rise-time was shorter than decay time. Overall, a minor fraction of the total particles larger than 300 nm were considered to be invalid. The invalid particles did not show a peculiar colour-ratio which could be related to mineral dust and had a negligible effect on the size distributions presented in the manuscript. Nonetheless, all the invalid particles were removed.
- TEXT CHANGES: A short statement was added to Section 2.2.3 and reads:

... "Previous studies showed that the SP2 is sensitive to metal containing particles such as hematite and magnetite, which might lead to an overestimation of rBC particle concentration (Schwarz et al., 2006; Yoshida et al., 2016). These particles are characterized by lower boiling point and colour-ratio (the ratio of thermal emission in the blue and red spectrum) but also by slow heating-rate in the laser beam of the SP2. During ACLOUD, particles associated with slow rise-time of the incandesce signal were removed. The colour-ratio analysis did not show any clear evidence of the presence of non-rBC yet incandescing particles." ...



*Figure B Diameter dependency of the color-ratio calculated for all incandesce signal observed above-cloud, inside-cloud and below-cloud with the SP2.* 



Figure C Incandescence signal of "valid" and "invalid" particle.

8. The references to Moteki and Laborde for calibration standards and procedures are not the best since Schwarz et al. (2006) was the original publication to which all other subsequent paper refers. Please update.

The reference was updated.

**9.** Line 204 "Hence, cloud particle residuals were representative of cloud condensation nuclei and/or ice nucleating particles (Mertes et al., 2005, 2007)." No, this ignores inertial scavenging (see below). We implemented the data analysis suggested by the reviewer and concluded that inertial scavenging was not relevant for the ACLOUD cases. None the less, inertial scavenging and its importance in the Arctic is now better described in the introduction and in the result section. Please find more details in the answers to comment number 21.

#### DATA EVALUATION AND PRESENTATION:

10. Show distribution by mass not by derived size. The process of deriving a mass equivalent diameter is highly uncertain because it assumes that all particles with rBC are homogeneous, spherical and constant density. Clearly this is not what in situ samples, captured on filters, reveal. Everywhere that the rBC distribution by equivalent diameter are shown should be number concentrations or mass concentrations by the measured rBC mass. The same trends and patterns should be seen as are seen with diameter, they will just have more of a physical meaning.

In the SP2-specific literature, there is an overall cohesion on the use of a fixed bulk density of 1800 kg/m<sup>3</sup> presented by Moteki et al. (2010) to convert mass into mass equivalent diameter. Bearing in mind the general consistency in SP2 literature, changing the size distribution into mass distribution would not modify the results of our study nor improve its understanding; but, on the contrary, limit the comparability with previous and future studies. Hence, the size distributions presented in the manuscript were not modified.

11. The current study looks at ratios of the rBC residual concentrations to the droplet concentrations and rBC residual concentrations to out of cloud rBC concentrations; however, a very important comparison here that is missing should be the comparison of the rBC residual concentrations ratios to the total number of residuals compared with the rBC concentrations out of cloud to the total aerosol concentrations below cloud and above cloud. Baumgardner et al. (2008) used this approach to argue that inertial scavenging was the most likely mechanism for putting rBC in cloud hydrometeor residuals (See below), a mechanism that is almost completely ignored in this manuscript.

See answer to comment number 21.

#### 12. Where are the representative size distributions from the SID and CIP?

Considering that the discussion of SID and CIP data was drastically reduced, the size distribution at cloud-top and cloud-bottom are briefly discussed in Section 3.6.3 and shown in Figure S5.

## 13. These clouds, based on how they are being defined, were between 300 and 400 m in depth, so why did the analysis divide them into 5 layers? I stably stratified clouds like these, is it physically reasonable to think that the properties will have such fine structure?

The vertical profiles presented in former Figure 8 are now presented into 4 vertical sections. Former Figure 8 was also greatly reduced, while more emphasis is now given to the differences between cloud top and cloud bottom in Section3.6.3.

## 14. Where are the wind data, vertical and horizontal? Was there vertical shear that would lead to entrainment and mixing? The authors elude to entrainment as a possible mechanism introducing the rBC to the cloud but what drives this entrainment?

We agree with the reviewer that wind velocity would help to understand the activation mechanism. As described in Ehrlich et al. (2019), the vertical wind can only be analysed as the deviation from the average for flight sections of at least several kilometres long, ideally for straight and levelled flight sections. The saw-tooth profile often adopted during the 4 flights presented here did not provide enough horizontal flight-time for a statistically robust analysis. Unfortunately, due to this technical limitation, wind data could not be included into the analysis.

#### **CLOUD DEFINITION**

15. This study defines a cloud as NDro > 10 cm-3 and LWC > 0.01 g m-3. How was this this threshold arrived at and how sensitive are the results to this definition? See answer to comment number 16.

## 16. Since NDro is derived from the SID, and since the lower threshold of the SID is cutoff at 10 $\mu$ m, in Arctic stratus clouds, this is likely eliminating a lot of cloud data and likely biases measurement depending if they are at the botton half or top half of cloud. This could be part of the reason that you see differences in the rBC properties at the bottom or top of the cloud.

As a follow up of reviewer's comment number 6, the screening of SID-3 data was modified by removing the 10  $\mu$ m cut-off. Now, the full detection range of the SID-3 (5-45  $\mu$ m) was used to quantify  $N_{\text{Dro}}$  and LWC, and to identify the cloud boundaries. In the updated version of the manuscript, the cloud LWC-threshold was kept to 0.01 g m<sup>-3</sup> (Järvinen et al., 2023), while the cloud  $N_{\text{Dro}}$ -threshold was set to 1 cm<sup>-3</sup>. These new cloud-thresholds did not impact, significantly the time of valid in-cloud measurements (difference of less than 2 minutes on the cloud ensemble) and the overall rBC variability observed during ACLOUD. Results in the figures and in the text were updated accordingly. Note that the threshold for outside-cloud measurements was not modified.

#### 17. Where are the size distributions in cloud, compare cloud base to cloud top?

Former section 3.5 was modified following the comments of both reviewers. It now includes a short description of potential temperature, liquid water content, ice water content and rBC-residuals number concentration. In the second part of the section, we compare in more details the properties of rBC-residuals and cloud particles observed at cloud-top and cloud-bottom. Size distribution of rBC-residuals are shown in the manuscript, while size distribution of liquid droplets and ice crystals are shown in the supplementary.

18. I strongly recommend that the authors read the paper by Baumgardner et al. (2008), who measured rBC in cirrus crystals over the Pacific Ocean, and concluded "Comparison of BC properties in the crystal residuals and cloud-free particles show that BC is scavenged by the crystals with an efficiency of as much as 44 ng of carbon per gram of ice water. The average number fraction of BC in

crystal residuals was 40% greater than that of the particles in the nearby, cloud-free regions and, on average, the mass equivalent diameter of BC was 10% larger. An average of 24% of the ice crystal residuals contained BC compared to 17% in cloud-free air. The large, in-cloud variability of BC properties prevents a more rigorous proof of our hypothesis that inertial scavenging is a dominant mechanism for removal of BC by clouds. The lack of correlation between the inside and out of cloud concentrations, coupled with the differences between the number fraction and MED in the crystal residuals and the background aerosol, is consistent with the inertial scavenging process and the evidence is sufficiently compelling to warrant further study." This study, while sampling a very different type of cloud than the current one, makes the case that all the data points to inertial scavenging as the primary mechanism by which the rBC ends up in cloud hydrometeors. The same arguments can be made in this study.

We carefully read Baumgardner et al. (2008), and applied their analysis to investigate inertial scavenging. The reviewer can find more details in the answer to comment number 21.

### 19. The very limited analysis of the "coating" suggests that these are uncoated or very lighty coated (read "lightly mixed") rBC particles.

The coating thickness and shell-to-core diameter ratio fell in a similar range of previous Arctic studies (Raatikainen et al., 2015; Zanatta et al., 2018; Schulz et al., 2019; Yoshida et al., 2020; Ohata et al., 2021). To the other end, freshly emitted rBC-containing particles are associated with shell-to-core around 1.1 (Laborde et al., 2013; Yoshida et al., 2020). As shown in Figure D (now included in the supplementary), coating thickness up to140 nm were observed inside and below cloud. By comparing ACLOUD data with the results of the studies presented above, rBC particles observed during ACLOUD could be classified as "medium-coated" to "thickly-coated" but not as "uncoated" or "very lightly coated" as stated by the reviewer.



Figure D Coating thickness distribution calculated for rBC particles with a mass equivalent diameter in the 200-250 nm range. Coating thickness data available only for the flight occurred on 02 June 2017. Added to the supplementary material.

20. Laboratory studies, although admittedly sparse and with varying conclusions, generally conclude that fresh, or aged yet lightly coated, rBC are not hygroscopic, i.e. not good CCN; hence, their presence in cloud residual does not imply that they were the nuclei of the droplet. In addition, vertical motion by adiabatic lifting is unlikely to lead to supersaturations large enough to activate these particles, even if slight hygroscopic.

Works performed by Schwarz et al. (2015) and Ohata et al. (2016) clearly show, as the reviewer suggest, that uncoated rBC particles (shell-to-core ratio <1.1) are not hygroscopic. Formation of coatings may, however, increase the hygroscopicity of rBC particles. In fact, 70% rBC particles with coating thickness of 40 nm may be activated in liquid droplets already at a peak SS of 0.15-0.20% (Motos et al., 2019a), while Dalirian et al. (2018), demonstrated that formation of relatively thin organic coatings (shell-to-core ratio between 1.15 and 1.30) drastically decreases the critical supersaturation. Hence coating thickness above 30 nm are sufficient to increase the hygroscopicity of rBC particles and promote nucleation scavenging. The rBC sampled during ACLOUD showed coating thickness up to 140 nm with median between 30-45 nm, enough to be classified as aged and hygroscopic. Furthermore, the number fraction analysis, suggested by the reviewer, indicated that the bulk aerosol and rBC particles were activated following the same scavenging mechanism (see below for more details).

- To better contextualize the importance of coating thickness, the introduction was modified as:
- ... "The ability of BC particles to promote droplet formation (nucleation scavenging) is one of the most complex parametrisations in global model schemes. If fresh BC particles are not hygroscopic, aged BC particles shows an enhanced hygroscopicity (Schwarz et al., 2015; Ohata et al., 2016). In fact, the nucleation ability of BC depends on fundamental particle properties such as diameter and mixing state, which change due to intra-coagulation, and to condensation and coagulation with other atmospheric species. Hygroscopicity increases with particles size (Motos et al., 2019a) and the formation of inorganic and organic coatings (Dalirian et al., 2018; Motos et al., 2019b)." ...
- Section 3.5.1 was fully updated to better present the coating distribution and its potential impact on the hygroscopicity during ACLOUD:
  - ... "First it must be noted that the analysis of the coating thickness includes only rBC particles in the 200-250 nm range of mass equivalent diameter. rBC particles in this diameter range were ubiquitously found above-cloud, inside-cloud and below-cloud (Figure 3). The distribution of coating-thickness is presented in Figure S2 in the supplementary material. The thinnest coatings were observed above clouds, where the coating thickness median was 30 nm (IQR = 23 - 48 nm) and median shell-to-core ratio was 1.51 (IQR = 1.38 - 1.8). The thickest coatings were observed below clouds, where the median coating thickness was 43 nm (IQR = 25 - 58 nm), and median shell-to-core diameter ratio was 1.67 (IQR = 1.43 - 1.98). The rBC cloud residuals showed medium coating thickness (median = 38 nm, IQR = 25 - 59 nm) and shell-to-core ratio (median = 1.58, IQR = 1.39 – 1.92 nm) respect to above-cloud and coatings to below-cloud. The coating thickness values presented here are similar to previous Arctic ground (Raatikainen et al., 2015; Zanatta et al., 2018) and airborne (Kodros et al., 2018; Ohata et al., 2021) observation, and are substantially higher than urban observations (Laborde et al., 2013; Yoshida et al., 2020). Even though thicker coatings can be found in aged continental air masses, the presence of 30-40 nm thick coatings is sufficient to significantly increase the hygroscopicity of otherwise hydrophobic uncoated BC particles in laboratory experiments (Dalirian et al., 2018) and filed observations (Motos et al., 2019a). Keeping in mind the low counting statistics of the coating analysis, we can conclude that rBC particles sampled during ACLOUD were presentative of aged and hygroscopic rBC particles which could be efficiently activated via nucleation scavenging. However, the reduced temporal coverage and the uncertainty of coating thickness quantification (17%; Laborde et al., 2012) did not allow identifying a significant change in the degree of internal mixing between rBC residuals and rBC particles sampled outside cloud."...

## 21. The study by Baumgardner et al (2008) claims that that the higher concentrations of larger rBC, seen in their studies and in the present one, is most easily explained by inertial scavenging. If the authors of the current study followed the same methodology as Baumgardner et al., I think that they would like reach a similar conclusion.

Interstitial or inertial scavenging was clearly overlooked in our work. Hence, following the reviewer's suggestion we calculated the rBC number fraction ( $F_{rBC}$ ) by combining the rBC number concentration

(SP2 measurement) and total aerosol number concentration (UHSAS measurements). The number fraction of rBC particles was extremely low below-cloud (mean=1.4%), inside-cloud (mean=1.1%) and above-cloud (mean=2.5%). These values being 1 order of magnitude lower than the values reported in Baumgardner et al. (2008) (mean in-cloud=24%, mean outside-cloud=17%). During ACLOUD, the fraction of rBC particles was not enriched in clod residuals, as in in Baumgardner et al. (2008), but remained rather constant. By remaining substantially constant between outside and below-cloud,  $F_{rBC}$  indicates that rBC is activated in the same ratio as other particles not containing rBC cores, thus sharing a similar activation pattern and, perhaps, hygroscopicity.

• To better present interstitial scavenging, a new paragraph was added to the introduction section: ... "Moreover, other in-cloud processes might compete with nucleation scavenging. In fact, interstitial BC particles (BC particles present in the cloud volume but not activated into cloud particles) can be efficiently captured by pre-existing cloud particles via interstitial scavenging (Baumgardner et al., 2008). Despite being often ignored, the number concentration of Arctic aerosol is highly sensitive to interstitial scavenging occurring during long-range transport (Croft et al., 2016)." ...

• The analysis of rBC number fraction is now presented in the new Section 3.4.2:

... "Other activation mechanisms might occur and contribute to the  $N_{rBC-res}/N_{rBC-blw}$  values above unity. First, interstitial aerosol particles may be scavenged via impaction with existing droplets (Croft et al., 2016), potentially enriching the number concentration of rBC-residuals (Baumgardner et al., 2008). We thus compared the fraction of rBC particles measured outside clouds ( $F_{rBC}=N_{rBC}/N_{AP}$ ) and inside clouds ( $F_{rBC-res}=N_{rBC-res}/N_{AP-res}$ ). An increase of  $F_{rBC-res}$  compared to outside cloud, might indicate the preponderant activation of rBC particles via interstitial scavenging (Baumgardner et al., 2008). During the ACLOUD cases, we found slightly smaller  $F_{rBC-res}$  (1.0%) than  $F_{rBC}$  above-cloud (2.3%) and below-cloud (1.2%). Similar  $F_{rBC-res}$  and  $F_{rBC}$  below-cloud suggested that rBC was activated via the same pathway of the bulk aerosol, that rBC and other aerosol particles shared similar hygroscopicity, and that rBC particles were not preferentially activated by interstitial scavenging. "...

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