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 anonymous Reviewer#2

SUMMARY:
… I would like to see the authors to be more constrained with the ambition and focus on what can be said about rBC in Arctic could drops given the present data. I encourage the authors to leave out the sometimes-lengthy speculative parts of the manuscript; i.e. lengthy speculations about processes that cannot be addressed with these data. The manuscript could use more cohesion. It feels like the introduction is lists studies where particular phenomena have been studied and those studies have come to the listed conclusions. I would expect the authors to connect these studies in an easier way so that it is easy to follow and would subsequently open up to the reader why the authors chose to cite those articles; and why the chosen articles are relevant for the present study. Throughout the manuscript, references to previous work is done in a fashion that the authors expect the reader to know them by hearth, and sometimes mid-sentence without an obvious connection to the present work. I would encourage the authors to consider which references they want to refer to, and only refer to previous work when they are obviously relevant. That would draw more attention to your work and why it is important instead of stating that someone already did something similar in another part of the world.
The manuscript was reworked considering the reviewer’s comments, putting particular care in harmonizing the scientific message and removing the speculative parts. The objectives are now clearly listed in the introduction and include:

1. Quantify the presence and properties of cloud-active BC particles
2. Identify the main scavenging mechanism
3. Understand the impact of cloud processing on BC vertical distribution

Former Section 4 was fully removed together with former Figure 6 and Figure 9. The cloud microphysics data, formerly very detailed, are now reduced to a minimum leaving more space to the discussion and interpretation of black carbon data. The narrower focus on BC also allowed reducing the number of citations and address them in more detail.

GENERAL COMMENTS:
The nomenclature is not always consistent in the manuscript which sometimes makes the manuscript unnecessarily hard to read. Please make sure that the same terminology is used throughout the manuscript. E.g. use either CCN or cloud drop residual, not both. Use wet scavenging or cloud scavenging, or make a clear distinction what you mean if you refer to below-cloud or in-cloud scavenging or something else.
The nomenclature was thoughtfully controlled and harmonized throughout the manuscript. Table 1 was modified accordingly.

Please check the use of “this”, “those” and “these” in the text. It can be hard to follow to what the word is referring to. Please, repeat the statement or the results (in brief) instead of referring loosely to what was said before. That would make for easier reading. Examples P11L343 “Under this cloud regime…”, P14L442 “Under this very complex...”
The issue was addresses.

For the above stated reasons, I suggest that the manuscript bee distilled down to comprise results and discussion on Figures 1-5, 7.

The structure of the manuscript was modified substantially by reducing the overall length, by rearranging the structure to discuss scavenging mechanisms (Section 3.4 and subsectons) and vertical variability (Section 3.6 and subsections), and by removing the lengthy discussion on cloud processing.

The conclusions section summarizes the present study well and is a good reference for how the work should be shortened.

This comment was particularly helpful, thanks.

SPECIFIC COMMENTS

Section 2: It is never mentioned how one single SP2 measures both "rBC cloud residuals" and rBC particles outside of clouds behind the CVI. Two inlets are mentioned but only one inlet is described in detail.

The SP2 and UHSAS were sampling through a line "shared" by both the CVI and total inlet. In normal operation outside clouds, both instruments were sampling exclusively though the total inlet. During in-cloud transects, the SP2 and UHSAS were manually connected to the CVI inlet by a valve switch. A short description was added at the end of Section 2.3 and now reads:

…"SP2 and UHSAS were operated in parallel and shared a sampling line which was alternatively connected to the total inlet or the CVI inlet. Outside cloud (N_{Dro} = 0 cm^{-3} and LWC = 0 g m^{-3}), SP2 and UHSAS measurements were performed at the aerosol inlet. Inside cloud (N_{Dro} ≥ 1 cm^{-3} and LWC ≥ 0.01 g m^{-3}), the SP2 and UHSAS were sampling throughout the CVI inlet line."…

Section 3.5 and Figure 8: Different low-level clouds have been scaled to the same thickness (rage 0 – 1 where 0 is the cloud bottom and 1 is the cloud top). These data comprise low exclusively level clouds, right? I think it is misleading to talk about cloud layers inside the "ensemble" cloud. A cloud layer is a set of clouds at some altitude and makes a cloud layer when it is distinguishable from another cloud layer with cloud free skies in between. To talk about a cloud layer inside a cloud does not make sense to me. How about talking about e.g. the lowermost 20% of the "ensemble cloud" or uppermost 60% etc? Or something better that you come up with

We agree with the reviewer. Note that following reviwer#1's comment, the vertical profiles were arranged in 4 and not 5 vertical sections. The word "layer" was replaced with "quartile" or "vertical section". In Section 3.6.3, the top quartile and bottom quartile are more simply referred as "cloud-top" and cloud-bottom", respectively.

I would suggest removing the Discussion section (section 4) and compress the results contents to one short paragraph and leave out the speculation.

This comment was shared by reviewer#1. Former Section 4, Figure 6 and Figure 9 were removed from the manuscript. Some elements of Section 4 were briefly elaborated in the introduction and Section 3.4.2 and Section 3.6.3.

When talking about the size of the rBC particles in the text, make sure you state if you are talking about the mode of the size distribution, mass or geometric mean diameter or something else. It is not always clear what size is meant.

Following the reviewer's suggestion, the very generic "rBC diameter" term was replaced with the more specific "rBC mass-equivalent diameter" term (D_{rBC}). The change includes all the text and Figure 2,3,5,7. We also introduced two new acronyms: $D_{rBC\text{-}mod}$ refers to the mode diameter of the rBC size distribution; $D_{rBC\text{-}geo}$ refers to the geometric mean of the rBC size distribution. Table 1 was modified accordingly. A short statement was added in section 2.2.3 and reads:

… "The geometric mean and modal diameter of the mass size distribution will be abbreviated as $D_{rBC\text{-}Geo}$ and $D_{rBC\text{-}Mod}$, respectively." …

P1L25 “might suggest” is not that intriguing. Rephrase to raise interest.

The abstract was considerable reworked, and the statement in question modified as:

… “The vertical evolution of rBC properties from inside-cloud and below-cloud indicated an efficient aerosol exchange at cloud-bottom, which might include activation, cloud processing, and sub-cloud release of processed rBC agglomerates.” …

P1L26 It would be interesting to give the reader a hint of the evidence for this kind of processing already here.

In the abstract, we now summarized the evidences in support to our conclusion.

P2L46-47 rBC after carbon dioxide and methane makes rBC 3rd. Please rephrase or clarify.

Corrected:

… “making BC the third atmospheric Arctic warmer only after the trace gases carbon dioxide and methane” …

P2L49-50: “Precipitation occurring during long-range transport influences the seasonal cycle of BC” I suggest saying something about the sources of rBC in the Arctic somewhere close to this text. Otherwise it is not that clear why deposition during long range transport has such an impact on rBC concentrations in the Artic.

The sentence was modified as:

… “The seasonality of BC concentration at the Arctic surface is characterized by a maximum in the early spring and a minimum in summer (Quinn et al., 2015). A similar seasonality was also recently reported on the vertical scale (Jurányi et al., 2023). Due to the scarcity of BC sources within the Arctic, most of BC mass reaches the Arctic via long range transport (Xu et al., 2017). Hence, the seasonal cycle is mostly controlled by the circulation of air masses between the Arctic and southern latitudes (Bozem et al., 2019), and precipitation intensity during long-range transport (Croft et al., 2016).” …

P2L54-55 “...might easily be the limiting process…” is too vague.

The introduction was substantially reworked. This specific section now reads:

… “Overall, cloud scavenging is responsible of 90% of BC mass deposition in the Arctic (Dou and Xiao, 2016), with the highest precipitation rate in summer contributing to a decline of BC burden from late spring to autumn (Garrett et al., 2011; Mori et al., 2020).”…

P2L62-P3L65 Unclear and too long sentence. Too many references to too many studies makes for a difficult sentence to follow. “No observations… aforementioned studies … indicating … size distribution of (cloud drop size? Aerosol size?) … degree of internal mixing...”

See answer to following comment.

P3L65 A bit unclear sentence and could be made easier to follow. As it is now, the reader must know that CCN and hygroscopicity are connected, that fresh soot is small in size.

The introduction was substantially reworked. This specific section now reads:

… “The ability of BC particles to promote droplet formation (hygroscopicity) is one of the most complex parametrisations in global model schemes. In fact, the cloud nucleation ability of BC depends on fundamental particle properties such as diameter and mixing state, which change during atmospheric ageing due to condensation and coagulation processes. If fresh BC particles are not hygroscopic, aged BC particles show an increase of hygroscopicity (Schwarz et al., 2015; Ohata et al., 2016) connected with the particle diameter (Motos et al., 2019a) and with the formation of inorganic and organic coatings (Dalirian et al., 2018; Motos et al., 2019b).” …

P3L78 Suggestion to change order of words to “unprecedented vertically resolved airborne measurements”

Changed following the reviewer suggestion.

P16L484 NrBC-res is not in the figure. Did you mean NDro?
Former Section 4 and former Figure 9 were removed.

FIGURES
Figure 2b: The contribution to total mass of the largest size bins is not commented on in the text. Is this rBC or something else? How much of the mass do these particles represent?
Regarding the larger bins of the size distribution.

- The last bin of the size distribution includes all particles showing an incandescence signal larger than saturation point of the detector. Since the diameter of particles associated with a saturated signal can not be quantified, it was set to be equal to the largest quantifiable diameter, in this case 575 nm.
- More details are given about the contribution of larger rBC particles to the total mass shown in Figure 2b and in Section 3.1:
  … "These larger particles (mass geometric mean diameter above 400 nm) accounted for less than 5% of the total number concentration along the full altitude range. Nonetheless, they represented 37% of the total rBC mass observed in the lowest 500 m asl, and 17% in atmospheric layers aloft." …
- Considering the size distributions shown in former Figure 4, the rBC particles contained in the last bin of the size distribution (501-575 nm) represented between 28-35% of the total rBC mass below-cloud and in-cloud, and less than 5% of the total rBC mass above-cloud. First, a note was added in the technical Section 2.2.3 to better explain the concept of signal saturation:
  … "The rBC particles associated with a saturated incandesce signal were included in the largest bin of the size distribution (468 nm < D_{rBC} ≤ 575 nm) and attributed with the maximum quantifiable mass-equivalent diameter (575 nm) and mass (178 fg)." …
- A note was added in Section 3.3 to quantify the contribution of saturated signals to the total mass for the selected events above-cloud and below-cloud:
  … "Compared to above-cloud, and increase of rBC particles larger than saturation-diameter (575 nm) was observed below-cloud. These larger particles represented less than 5% of M_{rBC} above-cloud, and 32% of M_{rBC} below-cloud." …
- A note was added in Section 3.5.2 to quantify the contribution of saturated signals to the total mass for the selected events inside-cloud:
  … "(rBC cores larger than 575 nm of mass-equivalent diameter, representing 28% of the $M_{rBC\text{-}res}$)" …

Regarding the nature of the large particles. 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 A). 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 B). 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." …

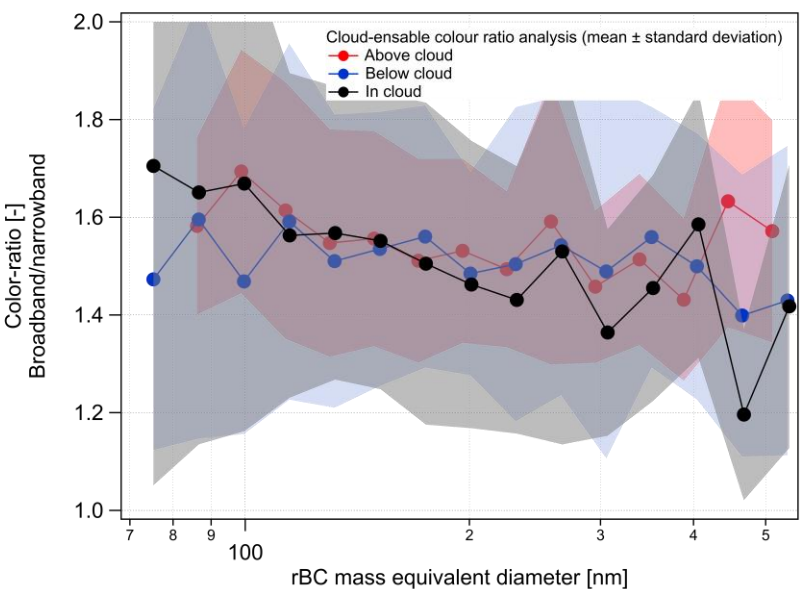

Figure A Diameter dependency of the color-ratio calculated for all incandesce signal observed above-cloud, inside-cloud and below-cloud with the SP2.

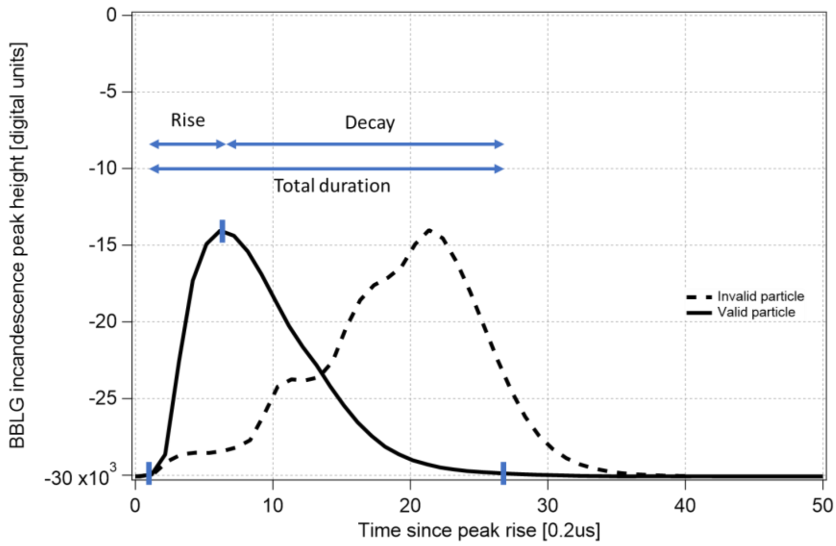

Figure B Incandescence signal of "valid" and "invalid" particle.

State in the figure caption that the in-cloud measurements were excluded.

The caption was modified and now reads:

… "rBC particles sampled behind the aerosol inlet outside clouds only and measured with the SP2" …

Figure 3: Add to the text that this figure comprises "warm period" data only.

The caption was modified and now reads:

… "Atmospheric and cloud characterisation of four flights occurred on 2, 4, 5 and 8 June 2017 (warm-period) north-west of Svalbard." …

Figure 4. rBC size range does not match Table 1.

We apologize for the mistake. The diameter quantification range of our SP2 during ACLOUD was 73-575 nm. The error was corrected in Table 1, in the caption of Figure 2,4,5,7,8, and everywhere else in the text. As a note, considering the scarcity of rBC particles inside and outside clouds, the size distribution presented in Figure 4 and 7 is built on a reduced number of bins (15), resulting in wider size-bins with minimum and maximum mid-bin diameter of 78-537nm.

Figure 5. N_rBC-res is likely to be much higher since the SP2 does not measure particles below ~75? nm. Showing the mean/median number size distribution would help understand the degree of underestimation.

- The reviewer can find the number size distribution of rBC particles in Figure C. From the figure is easy to understand that the number concentration of rBC particles presented in our work is clearly underestimated under every condition encountered during ACLOUD. In order to keep the manuscript short, Figure C was not included in the manuscript. Please note the change in the nomenclature for droplet number concentration from "N_{Dro}" to "N_{Dro10}" A clarification about $N_{\mathrm{rBC\text{-}res}}/N_{\mathrm{Dro10}}$ was added in Section 3.4.1:

  … "Considering that the number size distribution culminated at the low quantification limit of the SP2; $N_{\mathrm{rBC\text{-}res}}$ and, as a consequence, $N_{\mathrm{rBC\text{-}res}}/N_{\mathrm{Dro10}}$ were most certainly underestimated." …
- The number concentration of rBC particles below the quantification limit of the SP2 could be estimated by fitting a lognormal function to the measured number size distribution. Due to the absence of a peak in the number size distribution, the estimated number concentration below the SP2's size-cut mostly depended on manual and arbitrary tweaking of the lognormal function constants. For this reason, a lognormal fit was not applied to extrapolate the number concentration below 73 nm. This technical aspect was addressed in the methodology Section 2.2.3:

.. "**Previous works estimated the rBC concentration below the SP2's detection limit by fitting the measured size distribution with a lognormal function (e.g. Laborde et al., 2013; Zanatta et al., 2018). This correction was not applied to the ACLOUD data since a clear peak in the number size distribution was rarely resolved and the mass size distribution often culminated at the SP2's upper quantification limit. Hence, all the rBC properties reported in the present study are solely representative of rBC particles detected in the diameter range of the SP2 (73 – 575 nm).**" …

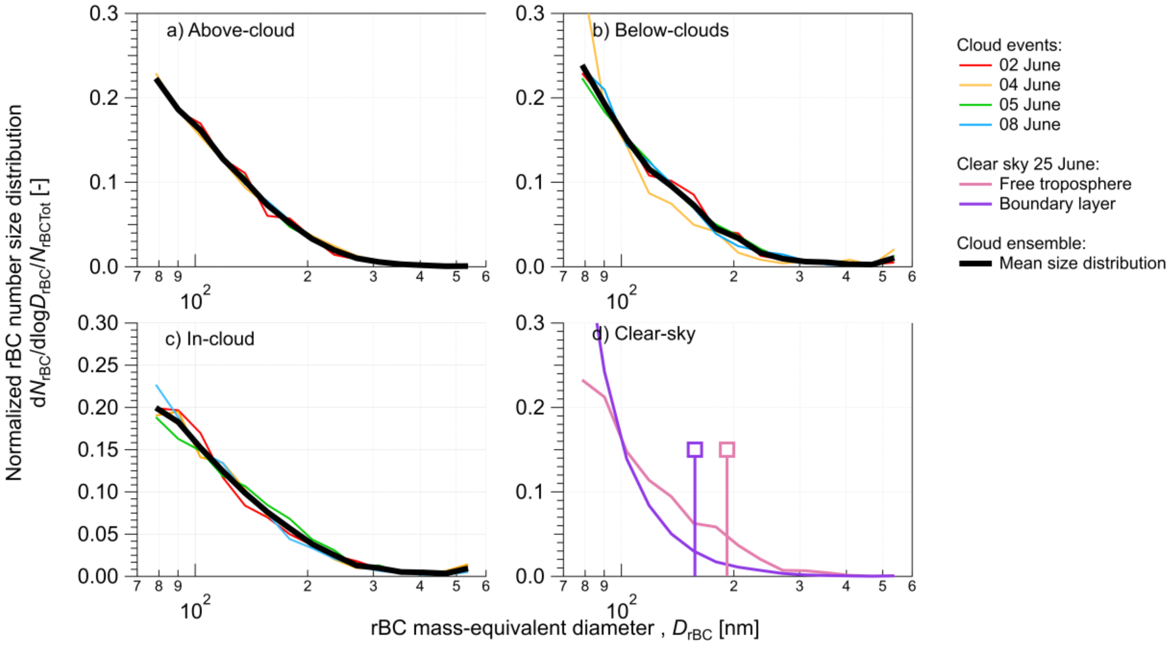

Figure C rBC normalized number size distribution observed in: a) above cloud; b) below clouds; c) in cloud; d) clear sky. rBC particles measured in the 73-575 nm diameter range with the SP2. rBC in cloud sampled behind the CVI inlet.

Figure 6. Although this is a nice picture, I would remove it since it is related to the speculative parts of the manuscript which I would like to see shortened significantly or removed altogether.

Former Figure 6 was removed.

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