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Reply on RC2

Michele Bertò et al.

Author comment on "Variability in black carbon mass concentration in surface snow at Svalbard" by Michele Bertò et al., Atmos. Chem. Phys. Discuss.,
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Reply to Anonymous Referee #2

The manuscript 'Variability of Black Carbon mass concentration in surface snow at Svalbard' by Michele Bertò et al. reports the cumulative processes (such as atmospheric, snowpack and meteorological conditions) in governing the refractory Black Carbon (rBC) mass concentrations in the upper snow layers at Svalbard. The database (85 days in 2014, 1 Apr-24 Jun and 3-days in 2015, 28 Apr- 1 May) in this study is useful to the characterization of aerosol-cryosphere interaction over the Arctic. However, there are many weak elements and lack of clarity in several aspects. I recommend publication of this manuscript in the Atmospheric Chemistry and Physics after my comments have been addressed.

We appreciate the overall positive recommendation expressed by the reviewer. We wish also to thank the reviewer for her/his instructive comments and suggestions. Please find below our point-by-point reply.

The major concern is the main outcome of the study. Authors state that 'precipitation events were the main drivers of the BC variability (line-33)'. However, the snow precipitation amount is negatively associated with the rBC mass concentration during 3-days experiment, as against the positive association during 85-days experiment. How do authors explain these contrasting effects of precipitation on BC concentration in snow? In line 353-367, authors try to connect atmospheric eBC concentration with the wet scavenging processes, which requires better investigation. Do authors want to highlight (quantify?) how effective the wash out processes in compared to various other factors considered in this study?

Thanks for the comments. The main difference can be explained from the different sampling strategy. During the 85-days experiment we sample at daily time resolution collecting the upper 10 cm. Sampling at daily resolution means that we are observing\measure the resulting of 24 hours of deposition (and removal) without evaluate how the rBC concentration evolve during a snow deposition event. With this strategy we will observe the final effect of a snow deposition event. The 3-days experiment is designed differently with the aim to evaluate the change at hour resolution. If we consider a snow fall event that last for 10 hours most of rBC suspended in the atmosphere is deposited in the first hours. Thereafter, the subsequent snow deposition will result in a depleted

concentration of rBC since the atmosphere becomes “cleaner”. Furthermore, since we are sampling the upper 3 cm, we collect only the last snow deposit and not the entire snow accumulated during the event. With the daily resolution (the 85-days experiment) we evaluate the net effect of a snow fall while the 3-days experiment with hourly resolution allows us to monitor the evolution of rBC concentrations in the deposited snow.

During the 85-days experiment we determine a significant correlation between the occurrence of snow deposition and rBC. This suggest that the wet deposition contributes to the rBC transfer from the atmosphere to the snow although the occurrence of a snow fall event is not always associated with an increase of rBC in the snow surface. For example, the snow event occurred around the 7th of May (Figure 2) is not associated to an increase of rBC.

The difference between the two experiments could be also found in the natural variability associated to the type of atmospheric transport\snow events. If a snow event is associated to air mass transport from the Arctic ocean, then the amount of rBC will be relatively low compared to the snow concentration. Vice versa if a snow event is associated to a transport from mid latitudes (or Siberia during the fire season), then the rBC is expected to be higher.

The aim of the paper is not to estimate or quantify the wash out for rBC, as it will be not be possible following our strategy, but rather to investigate which variable could affect the daily surface concentration. Accordingly, in the paper we have not stated any intent to calculate the wash out but we only describe the possible role that wet deposition could have in modify the rBC in the upper 10 cm.

The interpretation of the standardized estimated coefficients derived from multiple linear regression models amongst rBC (in snow) and water conductivity, coarse mode particles number concentration, equivalent black carbon atmospheric concentration, snow precipitation episodes, solar radiation and the snow temperature during 85- and 3-days experiments lacks coherent interpretation as well as proper explanation of the physical processes involved. The positive and negative associations seen in the case of eBC, fresh snow and SWR during 85 days and 3-days experiments must be clearly described.

Following the comments of the reviewer, we carefully revised:

- the discussion of the selected variable in Section 2.6 of the revised paper;
- the statistical model description in Section 2.7 of the revised paper;
- the interpretation of the estimated model coefficients in Sections 3.1.3 and 3.2.2 of the revised paper.

We think part of the apparent incoherent results were mostly due to the presence of the trigonometric terms in the regression model for the 3-days experiment. These terms were included in the model to capture the diurnal cycle. However, the statistical significance of the diurnal cycle is somehow weak and based on a relatively short experiment (three days). Accordingly, we revised the statistical analyses finding that a multiple regression model without the diurnal cycle gives a more robust summary of the 3-days experiment data. The revised statistical results indicate no significant association between rBC and SWR or temp, that is the two parameters that undergo to the diurnal cycle. Accordingly, we completely rewrite Section 3.2.2 of the revised paper on the basis of the new statistical findings. We also revised the interpretation of the remaining parameters in order to make the presentation of the statistical results clearer.

Information about both dry and wet deposition processes are randomly put in different context, this could be avoided.

The discussion about the wet and dry depositions is repeated in the paper with the precise aim to explain specific condition of rBC change during the two experiments. Wet and dry depositions are the main drivers of the transfer of eBC from the atmosphere to the snow surface and these two processes should be carefully considered at any time.

The BC in the upper snowpack affects the snow Albedo. There are multi-layer approaches to understand the effect of vertical distribution of BC in the snowpack (e.g., Dang et al., 2017). In this study, what is the criterion of selecting "upper snowpack" as 10 cm in 85-days and 3-cm in 3-days experiments?

The two experiments were intentionally designed to describe the evolution of the rBC mass concentration in the surface snow in Svalbard. During the 85-days experiment we focused on the seasonal variability and we strategically adopted a daily resolution sampling frequency of the snow layer that are controlling the albedo (the uppermost 10 cm of the snowpack), at the same time limiting the rapid oscillation\change that could occur in the upper layer (3 cm) has seen for the the 3-days experiment. The light reflection and transmission occur mainly in the upper centimetre of the snow pack (line 146). On the other side, the 3-days evolution experiment was focused on the changes connected to daily variations induced by light and temperature; in order to maximize the measured impacts, we sampled only the first 3 cm of snow. Sampling the upper 3 cm for the 80-days experiment would have exposed the samples to oscillations we determined during the 3-days experiment due to physical snow processes (sublimation, hoar formation, etc.). On the contrary, by sampling the upper 10 cm we wanted to smooth out these effects while being still able to evaluate the changes occurring in the active surface snow. For instance, sampling 20 cm of snow would have cause a too drastic smoothing of the surface signal since the lower and more stable snow layers would have mainly driven the resulting concentration variability.

If SZA is the primary driver of the diurnal variation of BC in snow, what about the cloud cover? SZA is important while estimating snow albedo change, but does it really influence (under the given circumstances) the BC concentration on a diurnal scale in 3 cm snow? How magnificent is snow metamorphism when the diurnal temperature remains below -6 C?

Thanks for the comment: here the usage of SZA was a refuse that we amend in the revised paper as explained also to the other reviewer. We fully agreed that SZA cannot influence the snow metamorphism during the 3-days experiment. The snow metamorphism is not only dependent on the temperature but also on the gradient of the snow pack, maximised at the upper 20 cm of the snow pack. We use "snow metamorphism" not only to refer to the melting episode but also to all the other physical processes (sublimation\hoar formation) driven by temperature variations. We also underline that the discussion on the diurnal rBC diurnal cycle was fully based on the statistical results. However, as also suggested by the reviewer 1, the diurnal cycle is not that evident. In fact, the statistical significance of the diurnal cycle was weak and based on a relatively short series(3-days). Accordingly, we carefully revised the statistical model removing the diurnal cycle. The new results obtained from the revised statistical analysis indicate no significant correlation between rBC and SWR or temperature, the two parameters that undergo to the diurnal cycle. Accordingly, we completely rewrote Section 3.2.2. with the revised statistical findinds.

Line 35-36: "The statistical analysis suggests that the BC content in the snow is decoupled from the atmospheric BC load." This is not clear.

Thanks: we modify the text to make it clearer.

Line 82-85: The citation about the dry deposition parameters is not suitable in the context of present study

We remove the sentence "Emerson et al. (2018) empirically evaluated the in situ rBC deposition velocities over a grassland ($0.3 \pm 0.2 \text{ mm s}^{-1}$), suggesting eddy covariance as the main deposition driver".

Line 98: seasonal > intra-seasonal variability

Thanks for point-out this typo: the text has been modified accordingly.

Line 121/ 99: Sampling period is contradicting; is it May end or till June 24?

Thanks for point-out this typo: the text has been modified accordingly.

Section 2.3.1: In general, filter loading effect is negligible at Arctic due to very low BC concentrations. Why MAC at 530 nm is used? Do aethalometer derived absorption coefficients agree well with PSAP measurements? Why MAC = $7.25 \text{ m}^2\text{g}^{-1}$ is considered for PSAP estimates of eBC? Virkkula (2010) is more appropriate for PSAP data correction.

The AE-31 and the PSAP used in the present work operate at slightly different wavelengths (530 and 520 nm, respectively) which may have caused some confusion, although this is stated in the text. The instruments also use different filter materials. The absorption coefficient of the PSAP operated at the Gruebadet lab have been tested in various campaigns with several different instruments (AE-33, AE-31 and PAX) and also during a yearly-long campaign with a MAAAP and thus they could be considered reliable. The AE-31 eBC values have been carefully tested in a previous Arctic campaign (Markowicz et al. 2017). For the present work, we used the AE-31 as the reference for the eBC absolute value. The PSAP data are fully consistent with those of the AE-31, in the period of the operational overlap, using a MAC of 7.25 at 530 nm which is well in the range (7.5 ± 1.2 , Bond and Bergstrom 2006) suggested at this wavelength (see also Backman et al. 2017).

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Section 2.4.2: This section is very important, which explains the estimates of rBC in snow. However, the methodology using SP2 (please expand) for rBC (in snow)

estimation is not clear. The following statement needs to be refined:

“The nebulization efficiency was evaluated daily by injecting Aquadag® solutions with different mass concentrations, ranging from 0.1 to 100 ng g⁻¹, obtaining an average value of 61%, that was used to correct all the BC mass concentrations reported in this manuscript.”

We implement the section 2.4.2 in particular at line 216 – 226. Additional information about the method are reported in the supplementary material.

Line 345-347: Please include the values of R²?

The R² indices for the models fitted on the two experiments indicate an overall satisfactory fitting quality (R²=0.69 for the 85-days experiment; R² =0.78 for the 3-days experiment). These values are reported in the paper and also in Table 1. Please notice that we fit a multiple linear regression thus we have a single R² index for each model. Each parameter has an associated estimated coefficient in the regression model whose statistical significance is summarized with the corresponding p-value given in the paper and also in Table 1.

Line 381: “.... as residuals of carbon extraction mining activities”, add reference to this statement

Thanks for the suggestion: we included the reference Vecchiato et al. 2018.

Line 432: Why average coarse mode number concentrations are significantly higher in 3-days experiment ($\sim 26642 \pm 9261 \text{ ml}^{-1}$) than that in 85 days experiment ($\sim 4914 \pm 4109 \text{ ml}^{-1}$)?

Fig-3: Why rBC concentration is relatively higher in 3 days experiment (> 10 ppb) as compared to the values in the corresponding period of 85 days experiment (< 6 ppb)?

We can only propose two hypotheses to explain the differences in the rBC concentrations (see the lines below) and this is why we do not investigate this aspect in details in the paper.

One explanation regards the different sampling depths: In the 85 days experiment we sampled the upper 10 cm where the rBC can be more diluted in the snow mass collected compared to the 3 days experiment. This suggests that the rBC tends to accumulate in the upper layer, but this conclusion (???) is strongly dependent on the meteorological conditions. Higher accumulation of rBC in the upper layer requires a relative long period of absence of snow fall and strong wind that favours the dry deposition and the rBC accumulation in the upper snowpack. However, we cannot prove this argument since the 3 days experiment was designed for another scope.

The second possible explanation is the interannual variability of rBC for the selected site and the influence of a specific atmospheric deposition event before the 3-days experiment. The surface snow samples collected during the 3-days experiment could be affected by a single deposition event able to increase the rBC concentration. However, linking a specific atmospheric event (reconstructed with the back-trajectory approach) to the explanation of the hourly surface snow rBC variability in Ny-Alesund is rather speculative mostly because of the orography around the experimental site. The samples collected during the 3-days experiment are, most likely, not representative for the site but just a snapshot during the 3 days of the experiment.

Fig-4: 80 or 85 days? In the regression model, how the values of SWIR are considered? Diurnal average or normal incident condition?

We modify and correct in 85-days. The SWR data for the regression model are daily average during the 85-days experiment and hourly during the 3-days experiment.

Lines 465-469: "... low mass concentrations when the solar radiation is high and vice versa. The BC particles are known to be non-volatile and not photochemically active, therefore the decrease in their concentration observed when the solar radiation is higher could not be explained as a re-emission process from the snowpack into the atmosphere as observed for other aerosol species". The sentence is not clear. Please rewrite.

Thanks for the comment: the text has been modified to make the sentence clearer. Please notice that the statistical analyses have been revised by removing the trigonometric terms describing diurnal cycle from the statistical model fitted to the three-days experiment. See also the discussed made some lines before in this point-by-point reply.