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Reply on RC1

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 #1

While this paper was submitted as a new paper, it is a revision of a paper previously submitted. I am reviewing it as I would a revised paper since I also reviewed the original version. The paper is significantly improved from the earlier version. It still suffers from some of the difficulties of the original, but I will recommend publication with revision.

We thank the referee for having appreciated our efforts to improve the manuscript. We also thank the referee for her\his work in revising again the manuscript with instructive comments and suggestions. Please find below our point-by-point reply.

As noted in my review of the original paper, the difficulty is that the sampling and analysis wasn't really designed to answer the questions being posed by this analysis. (Most specifically the snow sampling was done to a fixed depth in both experiments, rather than being designed to isolate, e.g., the influence of newly fallen snow, melt layers, hoar frost, etc.) As noted by the other reviewer of the original paper, correlative analysis is also not really the right tool to be using here for processes-level insight (e.g. see comment 1 below), and that is still the approach taken. There isn't anything that can be done at this point about the sampling, and the more focused analysis presented in this revised paper is an improvement. The data collected a useful contribution to the quantification of snow BC concentrations in the Arctic so with correction of the issues below I recommend publication.

We are convinced that the sampling strategy we propose is correct for the aim of our study since we are focusing on the variability in the BC concentrations in the upper snow layer (10 cm) where these impurities can have the higher impact in the albedo reduction\change. This aim is now clearly stated in the new title of the manuscript. We cannot state that our approach is the best but we believe it is the appropriate solution for obtaining a general overview of the processes that could affect the BC concentration in the surface snow pack. We fully agree with the reviewer that if we had investigated the effect of deposition, then we would have needed to sample only the fresh snowfall at consequently different depth resolutions. However, such a study is not the goal of our project.

Our statistical analysis is not just a study of the pairwise correlations between the available parameters. Indeed, we consider a multiple linear regression model to account for the *joint effect* of different meteorological and physicochemical parameters on the BC concentrations in the surface snow pack. We acknowledge that the previous version of the paper lacked the necessary details to understand our statistical analyses. Section 2.7 of the revised paper now provides more details about our statistical modelling approach. Moreover, following the reviewer's criticism about the presence of a diurnal cycle, we revised the statistical analysis of the three-days experiment as discussed later in this point-by-point reply.

1) There is still some difficulty with the explanations for the selected variables. See my comments below about SZA as a driving variable for snow rBC concentrations. The reasoning behind expecting surface snow temperature to correlate with daily variations in surface snow rBC concentrations is unclear. Once melting commences, any time the temperature goes above freezing surface rBC will continue to increase. Based on physical understanding of process driving surface snow concentration changes you don't expect there to be a correlation between air temperature and surface snow concentration because the process of surface snow BC concentrations increasing with melt is cumulative; when temperatures go back down, there's no physical reason why rBC would then decrease. Therefore applying correlation at the hourly or daily timescale doesn't make sense.

We corrected the referee about SZA that, indeed, does not affect the rBC surface snow mass concentration. In the revised paper we replace SZA with SWI.

The snow temperature is a fundamental parameter necessary to understand the physical process of the snowpack, of the upper snow layer and the meteorological condition. We understand the point of the reviewer, in particular during the 85-days experiment. When the snow melting began, the snow temperature stayed around zero and presented no significant oscillations suggesting that the change in the BC is dependent on other variables. However, the snow temperature could be relevant during the entire experiment indicating the possible snow metamorphism, the response of the upper snowpack to meteorological conditions, including spring warming events ($T > 0^{\circ}\text{C}$), and the begin of the snowpack melting. The snow temperature is not driving the rBC during the melting phase, but the statistical analysis should be equally carried out during the entire experiment and not only during the "cold" phase of the experiment. Instead in the case of the 3-days experiment, the snow temperature can be used as an indication of the response of the upper snowpack to the change induced by the SWR. Correlation between rBC with SWR and snow temp could indicate that process with a daily scale frequency, could affect the upper snow rBC concentration. Essentially the temperature in the 3 days is a tracer of the daily oscillation. Therefore, we have improved the description in the Section 2.6 of the revised paper (line 260-266).

2) The one major issue with the paper that still needs to be addressed is that it argues there is a diurnal cycle in rBC observed during the 3-days experiment (lines 420-421 and 465, referencing Figure 3), then hypothesizes that the formation and evaporation of hoar frost is driving this cycle. However:

i.) Most problematic of all is that I don't see the purported diurnal cycle in rBC that is referenced as existing in Figure 3. As noted in my review of the original paper, the dark blue line in the bottom panel simply looks like smoothed random variations, superimposed on an overall decreasing trend. If the authors are going to argue there was a diurnal cycle and then explain why this diurnal cycle exists, they first need to show that there actually IS a diurnal cycle with some sort of statistical analysis. Simply asserting it's there is not sufficient.

- **ii) No observations are presented to support that hoar frost formed as hypothesized. Was hoar frost observed during the 3-days experiment measurements? Were the weather conditions (e.g. clear skies and low winds) conducive to hoar frost formation?**

Thanks for the sensible comments. Our previous discussion on the rBC diurnal cycle was based on the statistical analyses. However, as observed by the reviewer, the diurnal cycle is not that evident. Indeed, the statistical significance of the diurnal cycle was 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 (i.e. removing the two trigonometric terms that described the diurnal cycle in the statistical model) 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.

Smaller comments:

3) Line 149: Solar zenith angle is cited here as a likely primary driver of snow BC concentrations. It's really solar insolation that is relevant, and then really only as it drives temperature and therefore snow metamorphism changes (e.g. melting and hoar frost formation). Indeed, "Solar radiation" (W/m²) is what's shown in Figures 2 and 3, along with temperature. This text should be edited to reflect this.

Thanks for pointing out this wrong sentence; the revised paper has been modified accordingly (line 154).

4) lines 253-255 then 256-258: I don't understand this explanation given that RH is an observational variable of interest since "high RH might favour the deposition of BC suspended by the formation of water droplets through the cloud condensation nuclei." It's then stated that precipitation amount is monitored, so that would be a more direct measure of wet deposition (if that's indeed the type of deposition monitoring RH was supposed to reveal).

RH is a common meteorological parameter. High values of RH are not always associated to wet precipitation (for example, foggy conditions occurring in the Arctic during Spring). However, high values of RH could favour the formation and the grown of atmospheric water droplet in this way favouring the dry deposition. The amount of precipitations, in terms of water mass, caused by this process is negligible compared to a proper snowfall, but the grown of the water droplet around the cloud condensation nuclei can favour the deposition of the rBC in the surface snowpack. There is no direct link between RH and rBC but might an indirect relationship since it can be used to better understand the meteorological condition. For example, low RH conditions; clear sky and absence of cloud cover (for several days) can favour the sublimation of the snow from the surface. RH, similar to temperature, is not a primary driver of rBC deposition but it could promote\ favour the dry deposition. This experiment is unique and it has the aim to explore possible environmental parameters able to influence the rBC concentration in the upper snow pack. We revised the lines 272-275 of the paper to make this point clearer.

5) Lines 301-302 vs lines 320-322: The same result is expressed two different ways in two different places. Why? Lines 301-302 express this as "variability", then notes it declined through the campaign; lines 320-322 simply expresses it as a trend. The partitioning of information into Sections 3.1.1 (atmospheric eBC) and Section 3.1.2 (atmospheric conditions) is a bit odd.

Thanks for pointing out this issue. The sentence at line 301-302 has been modified from "During the experimental period, the atmospheric eBC concentration shows a noticeable variability ranging from 80 ng m⁻³ to < 5 ng m⁻³ (Figure 2)" to "During the experimental period, the atmospheric eBC concentration range between from 80 ng m⁻³ to < 5 ng m⁻³ (Figure 2) with an average of 34 ± 23 ng m⁻³" and we remove the sentence at line 302-322 since is just a repetition. We also re arrange the section 3.1.1. and 3.1.2. as suggested.

6) "coarse mode particles" (used in text) and "dust particles" (Figures 2 and 3) are used interchangeably. What's really shown in the figure is coarse mode particles. In much literature "dust" has a fairly specific meaning (mineral dust). Here, the coarse mode particle composition isn't determined so it would be best to stick with terminology that reflects what is actually measured. In the text you could then note that the coarse mode particles are likely a mix of soil, mineral dust and (as noted) possibly coal dust.

We agree with the referee and we modified accordingly the corresponding text in the revised paper. In both figures we we use the acronymous Cmp (coarse mode particles).

7) Lines 308-311: The number concentration of coarse mode particles in snow, as noted, is lower during the first half of the 85-days campaign, then increases during the second half of the campaign (Figure 2). On line 355 an 'average concentration' of 4914+/-4109 per ml is given but it's not clear whether this is across the full duration of the campaign (in which case it's not a very meaningful number, given the clear difference between the first and second half of the campaign) or it's only over the second half of the campaign (in which case the time period used for this statistic should be given).

We improved the corresponding section in the revised paper as suggested (lines 352-356).

8) lines 337 and 341: BC should be rBC

Thanks for pointing out the error, now corrected in the revised paper.

9) lines 373-375: "Dry deposition is the main depositional process for the coarse mode particles. Recently it has been suggested to have a significant contribution to the BC surface content (up to 50-60%; Liu et al., 2011; Jacobi et al., 2019)." This needs to be explained or written differently: Atmospheric BC is not a coarse-mode particle so dry deposition of coarse-mode particles as a significant source of BC to surface snow is a confusing statement.

We agree with the referee and thus we rewrote the sentence in the revised paper to make it clearer (lines 391-394).

10) Lines 379-382: "Our data support the hypothesis related to local sources' activation in enhancing the dry deposition impacts in an old mining town as Ny-Alesund. Especially during poor snow cover conditions, as during the snow-melting season, dust particles as residuals of carbon extraction mining activities are available for wind lift\suspension." It's argued that this is a significant source of BC to snow. Yet on lines 536-537 it's stated that: "We believe our results to be representative at least of the Arctic coastal areas, characterized by similar processes and seasonality." Svalbard is fairly unusual for the Arctic in the degree to which past coal mining is likely influencing the addition of rBC to the snowpack from local surface sources (lines 379-358), so how can you assert that you think the results here are generalizable to the Arctic coastal areas overall

(lines 536-537)?

Thanks for the reasonable comment. The Ny-Alesund area is ideal to perform snow research studies since the large amount of monitoring programs on-going and the supporting datasets available. However, there are some limitations at Ny-Alesund derived from the fact that it was a coalmine till the 60s. At the beginning and at the end of the snow season, the rBC concentration in the surface snow can be influenced by the characteristics of the experiment site. During the winter and spring periods the snow cover is homogeneous (with the notable exception of extraordinary dry years) and thus the upper snowpack is mainly driven by the atmospheric deposition. We improved the conclusions of the revised paper as suggested by the reviewer (see lines 399 and 544-548).

11) lines 393-395: The impact of surface snow melt on surface snow concentrations of particulates is presented too much as a hypothesized process; in fact there is significant support for this in the literatures. In addition to the modeling that simulates this and in addition to Aamaas et al 2011 there are at least three other studies showing this in observational data:

Xu, B., T. Yao, X. Liu, and N. Wang, Elemental and organic carbon measurements with a two-step heating gas chromatography system in snow samples from the Tibetan Plateau, *Ann. Glaciol.*, 43, 257–262, doi:10.3189/172756406781812122, 2006.

Doherty, S. J., T. C. Grenfell, S. Forsström, D. L. Hegg, S. G. Warren and R. Brandt, Observed vertical redistribution of black carbon and other light-absorbing particles in melting snow, *J. Geophys. Res.*, 118(11), 5553-5569, doi:10.1002/jgrd.50235, 2013.

Doherty, S. J., D. A. Hegg, P. K. Quinn, J. E. Johnson, J. P. Schwarz, C. Dang and S. G. Warren, Causes of variability in light absorption by particles in snow at sites in Idaho and Utah, *J. Geophys. Res. - Atmos.*, 121, doi:10.1002/2015JD024375, 2016.

Thanks: we added the suggest references\papers at line 420.

12) lines 424-425: It's noted that the concentration of rBC in snow is 6x higher in the 3-days experiment than in the 85-days experiment. Earlier it's pointed out that the concentrations during the 85-days experiment were consistent with those found in previous studies. Any thoughts on why the very large increase in snow rBC?

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 suggest 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 were designed for another scope.

The second possible explanation is the interannual variability of rBC for the selcted site

and the influence of a specific atmospheric deposition event before the 3-days experiment. The surface snow samples collect during the 3-days experiment could be affected by a single deposition event able to increase the rBC concentration. However, link 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.

13) line 427: Bond et al. 2013 didn't give snow rBC sized so this isn't an appropriate citation. The work of Schwartz et al. could be cited instead.

Thanks: we modify the revised paper accordingly (line 445).

14) line 435: "All the measured snow impurities time series show two common features...: Which variables are your referring to here? The description that follows matches that for rBC but not for e.g. dust/coarse mode particle concentration. What do you mean by "all the measured snow impurities time series"?

We Thanks: we added the reference to the supplementary material: Figure S4 and Section 4.

15) line 520: Remaining "unaffected" is different from "not being a primary driver of variations in surface snow rBC over XX timescales". What you've shown supports the latter statement but not the former, and only sort of, since the observational period did not include any highly elevated atmospheric eBC periods.

We agree with the referee and modified the revised paper accordingly (line 531).