"Elemental and and water-insoluble organic carbon in Svalbard snow: A synthesis of observations during 2007-2018." by Zdanowicz *et al.*

**Authors' reply to comments by Reviewer 1**

*Major comments*

***[1]*** *The uncertainties of the present method are described separately in sections 2.2.2. and 4.3. The uncertainties should be given before the observational results are presented. Otherwise readers need to re-consider or re-interpret results that are shown before section 4.4. These sections should be combined together as much as the logical structure of the text allows.*

This is a matter of author or editorial choice. For example in their 2010 paper published in ACP, Doherty et al. placed their ~5-page long analysis of measurement uncertainties in section 6, after their presentation of results. However our own discussion of the uncertainties due to the presence of dust, is shorter, so we have now moved it into the Methods section in the revised manuscript, as suggested by the reviewer.

***[2]*** *Figure 4: Probably this figure is most important in this paper. It will be useful to show the vertical profiles with linear-scales, at least for the median values, in addition to the log-scale plots.*

Added to Fig 4, as suggested.

***[3]*** *Page 14, L409-412: It is stated "of particular interest". However, explanations of this "interesting" feature are very poor.*
***a)*** *There are no explanations why this feature is particularly interesting.*

The features in the $C_{snow}^{EC}$ and $C_{snow}^{WIOC}$ data are interesting (or noteworthy, if one prefers) in the sense that they depart from invariance or from homogeneity, and they appear to be temporarily consistent over the years. We have made this more explicit in the opening of section 4.3 in the revised text. Concerning the *plausibility* of the proposed explanations for these feaures, see below.

***b)*** *L411, Is the "gradient in EC and WIOC" in horizontal or vertical directions?*

Both are of course possible, i.e. the apparent systematic difference in the median $C_{snow}^{EC}$ and $C_{snow}^{WIOC}$ between Ny Ålesund and the Austre Broggerbreen site (which are ~5.5 km apart) might be linked to the relative horizontal distance from Ny Ålesund and/or from the coast (i.e. transport distance from local EC or WIOC sources), or to the difference in elevation. The two go together, since air moving inland from the coast necessarily rises over the interior plateau. We have nadded some nuances to the text to this effect.

***c)*** *There is no interpretation on how localized EC emissions impact EC in snowpack at Ny-Alesund. The effects of in-cloud scavenging, below cloud scavenging, and dry deposition should be discussed with some quantitative analysis.*

To perform the sort of quantitative analysis the reviewer suggests, one needs, ideally, simulteaneous measurements of BC (or eBC) in air and of BC (or EC) in falling snow, or at least in freshly deposited snow (e.g., Sinha et al., 2018). The snow samples collected near Ny Ålesund between 2007-2018 were obtained (as described in the methods) in an opportunistic way, whenever staff at the NPI station were available to do it. While on a few occasions the surface snow was sampled shortly after snowfall events, this was far from being always the case. Furthermore, it is difficult to ascertain what time interval of snow accumulation is represented by each surface snow layer: one can not assume linear accumulation over time, since snowfall is irregular and there is surface wind drift. This makes a direct comparison of airborne and snow BC (EC) data highly uncertain (see also 4th paragraph, below).

In a relevant study, Jacobi et al. (2019) combined optical measurements of eBC from Zeppelin station with snowpit measurements of rBC at high depth resolution (~3 cm intervals) made at a comparable altitudes on two glaciers (Kongsvegen and Austre Lovénbreen). They used the CROCUS snow model to estimate, indirectly, the temporal sequence of snow accumulation at the snowpit site, and from these data, they computed the predicted monthly cumulated BC deposition in snow by both and wet dry deposition, which they then compared with the rBC burden in the snowpit. However we can not reproduce such an analysis since our own snowpit data have a much coarser depth resolution (imposed by the sample volume requirements for TOT analysis) which can not easily be related to specific snowfall events.

Another alternative is to use some indirect, model-based estimates of atmospheric BC column loadings in combination with data on the vertical cloud structure to compute BC deposition in snow. During the preparation of this manuscript, we considered, at one point, using modeled vertical BC aerosol column loadings over Ny Ålesund produced by the CAMS global reanalysis (e.g., Pakszys & Zielinski, 2017) to estimate both dry and wet deposition and compare these results with the EC concentrations in snow. However we found, upon verification, that there was a poor match between the CAMS predicted near-surface BC mixing ratios at Zeppelin and the measured eBC, so we concluded that the CAMS reanalysis for BC were not sufficiently reliable at this latitude to be used.

In the first version of this manuscript (which the reviewer may not have seen), we actually made an attempt to relate eBC concentrations in air measured at the Zeppelin station to EC concentrations in surface snow on Austre Brøggerbreen (~same altitude as Zeppelin). We could not do the same at Ny Ålesund since we do not have aerosol data from this lower-altitude site. For Austre Brøggerbreen, we assumed dry deposition in snow to be minor or negligible (which is what our various glacier snowpit data suggest) and tested a range of plausible eBC scavenging ratios, combined with meteorologically-driven model estimates of surface snow accumulation rates, to find out which values yielded eBC concentrations in snow that most closely agreed with our measured EC concentrations. However, reviewers of the manuscript criticized this exercise to be too speculative and uncertain, partly owing to the fact that eBC and EC are not the same fractions of BC. We therefore chose to abstain from this sort of quantitative analysis in the revised manuscript, and we stand by this decision.

Our data are a survey of the spatial variability of EC and WIOC in snow across Svalbard. We observe, in these data, some apparent features for which we offer some possible explanations. These explanations could be put to the test in future studies. The higher median EC concentrations in snow at Ny Ålesund compared to Austre Brøggerbreen suggest an influence of BC emissions from the town on local snow (although it does not mean that all EC in snow there is from local sources). This certainly seems plausible. To rigorously test whether this is in fact the case, one would need to collect, simultaneously, both BC aerosols and falling snow (or freshly fallen snow) over at least one winter at both sites. In the concluding statement to the manuscript, we suggest this as a future task for experimenters and modellers, but we refrain from attempting such a quantative analysis ourselves, since we simply do not have adequate supporting data to do so.

***d)*** *Higher EC at Ny-Alesund is interpreted as due to localized EC emissions and lower EC at ABG is due to the gradient in EC. Then why do EC values vary synchronously? If EC at ABG is relatively free from local emissions, the EC values at these sites should vary quite differently from those at Ny-Alesund.*

This implies a false dichotomy, i.e. that EC deposited in snow at Austre Brøggerbreen is free of *any* influence from Ny Ålesund BC emissions, and, conversely, that EC in snow in Ny Ålesund is *only* from local BC emissions. We have made no such claim. Rather, we proposed that the higher median EC concentration in Ny Ålesund could reflect the influence of local BC emissions at this site, and the lower median EC concentration at Austre Brøggerbreen suggest that this influence is weaker up there due to greater inland distance and/or higher elevation. One possible explanation for this difference is that BC emitted in Ny Ålesund may tend to be trapped by low-level wintertime thermal or humidity inversions below the elevation of Zeppelin observatory. This is certainly plausible, and was in fact one of the reasons the observatory was established on Zeppelin Mountain. The effect of winter inversions might also apply to WIOC emitted from either combustion sources (in Ny Ålesund) or from marine sources (in nearby coastal waters). None of this excludes the possibility that EC and WIOC from other, distant sources (continental or marine) reach these sites, as must certainly be the case, based on previous studies of aerosol provenance (e.g., Eleftheriadis et al., 2009). If the efficiency of long-range transport (or deposition) of these aerosols varies between different seasons, then it is entirely plausible that this would affect EC and WIOC deposition in snow in Ny Ålesund and on Austre Brøggerbreen simultaneously, as the two sites are relatively close to one another. But even in this was the case, there could still exist a gradient in EC and WIOC concentrations in snow between the two sites, owing to the added contribution of local aerosol emissions from Ny Ålesund and nearby waters.

The reviewer's comment, however, did highlight flaw in part of our discussion. We had speculated that changes in the strength of winter inversions might account for the apparent synchronicity of the interannual variations in $C_{snow}^{EC}$ and $C_{snow}^{WIOC}$ in Ny Ålesund and on Austre Brøggerbreen, the argument being that changes in the mean thickness of the winter boundary layer might allow, in some years, more of the locally-emitted EC and WIOC aerosols to reach higher up and be deposited in snow on Austre Brøggerbreen. If this were the case, one might expect that the difference in $C_{snow}^{EC}$ and $C_{snow}^{WIOC}$ between Ny Ålesund and on Austre Brøggerbreen would decrease at these times. To find out if this was the case, we plotted, on our revised Fig. 5(d), the ratios of $C_{snow}^{EC}$ and $C_{snow}^{WIOC}$ between the two sites. Results show that in fact, these ratios increased in seasons when $C_{snow}^{EC}$ and $C_{snow}^{WIOC}$ were relatively higher in both Ny Ålesund and on Austre Brøggerbreen. Thus, changes in the mixed layer thickness are unlikely to explain the simultaneous interannual variations in $C_{snow}^{EC}$ and $C_{snow}^{WIOC}$ at the two sites. Instead, these variations, assuming they are real, seem more likely to reflect interannual variations in transport of removal of aerosols (irrespective of sources) that simultaneously affect both sites. Accordingly, we have modified parts of our discussion to stress this.

Note: While reviewing the paper to answer the reviewer's comments, an error was discovered. In section 3.2., L304, we had stated that *"...$\tilde{C}_{snow}^{WIOC}$ was 7 times higher, but as much as 30 times higher..."* in Ny Ålesund, when compared to ABG. These figures were actually incorrect: An offset of a year had accidentally been introduced in the calculations. Once corrrected, the magnitude of the differences in $\tilde{C}_{snow}^{WIOC}$ between Ny Ålesund and ABG turned out to be much closer to those seen in $\tilde{C}_{snow}^{EC}$, averaging ~2-3 in most years. The error did not affect the temporal pattern, nor our findings about $\tilde{C}_{snow}^{EC}$. We have corrected the relevant statements in section 3.2., in the discussion, and in the conclusions.

Note also that we have made some changes to Fig. 5: Panels (c) and (d) in the previous version have now been merged in panel (c), and a new panel has been added, which shows variations in the ratios of $\tilde{C}_{snow}^{EC}$ and of $\tilde{C}_{snow}^{WIOC}$ between Ny Ålesund and ABG. We also removed shadings from panels (c) and (e): These were included in the previous figure to indicate the possible range of variations in some of the results depending on whether data below detection limits were included or excluded. However co-authors commented that the shadings compromised the readibility of the figure, and so, on their advice, these were removed to simplify the plots and improve their overall clarity.

***e)*** *Regarding (d), it may be useful to investigate the relation of EC in snow at ABG with BC in air at Zappelin for understanding the source of EC in snowpack at ABG.*

See response to (c) above.

***f)*** *As one of possible sources of WIOC, natural source is discussed. Some more discussion on a relative contribution of natural additional source of WIOC is desirable.*

Presumably, this comment refers to page 14, L419-424, where we discuss possible marine sources of WIOC. We have stressed these particular sources because they might possibly account for the higher median $C_{snow}^{WIOC}$ observed near Ny Ålesund compared to Austre Brøggerbreen, higher up and further inland. We do not know of any other important *natural* source of WIOC emissions that would have such a localized impact in winter (tundra soil and vegetation emissions of volatile or semi-volatile OC are likely very low to nil in these months). There may, of course, be WIOC aerosols contributed from natural terrestrial emissions at lower latitudes (e.g., mainland Europe) and transported to Svalbard in winter, but it is not obvious why these should have systematically higher median concentrations in surface snow in Ny Ålesund compared to Austre Brøggerbreen, year after year. There may also be *non-natural* (i.e., anthropogenic) WIOC sources in Ny Ålesund, for e.g., as a component of vehicular exhaust emissions. We have added a few lines to section 4.3. to acknowledge this.

***g)*** *In the response [15] of the authors, it is claimed that synchronous variations support the reliability of the method to some extent. This is not logically correct. First, the uncertainty of the measurements must be established. Then, it should be judged if the observed variations are larger or smaller than the uncertainties.*

The statement in our response (paragaraph [15]) which the referee quotes was about *probability*, not accuracy. We merely pointed out that if the observed temporal variations at the two sites were only due to random errors (caused by one or more factors), it is *unlikely* that these errors would combine to produce the same pattern of temporal variations at the two sites over several years. While the similarity of temporal patterns at both sites does not *prove* that these variations are real, it makes it *more likely* that they are. On Fig. 5(c), the estimated uncertainties (2$\sigma$) on the seasonal median $C_{snow}^{EC}$ and $C_{snow}^{WIOC}$ are indicated as error bars. These show that the amplitude of the synchronous interannual variations seen in Ny Ålesund and on Austre Brøggerbreen are in fact larger than the uncertainties for more than half of the years in which we have data from both sites. Some rewording in the revised section 4.3. makes the points above more explicit. We can add nothing further on the subject.

*Specific comments*

*Pages 12, L365: “Sihna” should be “Sinha”*

Corrected.

*Page 16, L500: “other analytical methods”. The methods should be stated concretely. And their accuracies need to be compared with that of this work.*

We have now expanded this brief statement from L500 into a full paragraph where we provide the requested details.

*Figure 2: The colors for S and NE are similar. May be some symbols in the vertical profile can be changes (+ or open circles).*

On the revised figure, the color for the NE sector has been changed to a paler shade to increase the constrast with that for the S sector.

**No changes were made to the supplement.**

**References cited**

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