

**Reply to the review by Anonymous Referee #1 for the manuscript, “Deposition of Brown Carbon onto Snow: changes of snow optical and radiative properties” by N. D. Beres et al.**

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We thank the anonymous reviewer for their thoughtful response and recommendations to improve the manuscript through clarification of the analysis, particularly of total organic carbon determination of the melted snow samples. Below, questions and comments by the reviewer are in blue and the responses by the manuscript authors are in black.

The authors admitted that BC is a light-absorbing particle in snow, while how can their instrument manage only to measure TOC but avoid BC? If BC was mixingly measured, the total organic carbon could be overestimated. Section 2.2 does not introduce the instrument in detail and needs to strengthen the method’s introduction, including the principle, accuracy and precision of the Sievers 900 measuring TOC.

The authors agree with the reviewer’s latter suggestion that the methods section concerning TOC determination (beginning on Page 6, Line 24) should be expanded to include more information regarding the Sievers 900 instrument, its analysis protocols, and reported measurement accuracy and precision. These have been included in the updated manuscript, beginning on page 6, line 26.

With respect to the reviewer’s former suggestion – the instrument’s ability to determine TOC from indirect/unintentional oxidation of BC in liquid samples during its normal operation – the instrument is not believed to be able to convert BC to measurable TOC through the photo-chemical oxidation methods utilized. BC is insoluble and chemically inert and recalcitrant. One supporting publication is Peltier et al. (2007), who employ a Sievers 800-series TOC analyzer (utilizing the same oxidation methods as the 900-series instrument used in our study) and were unable to detect elemental carbon (EC) in aqueous solutions.

In addition, the concentrations of BC found in the snow, both before the deposition and even after are much smaller than those of deposited TOC. Concentrations of BC measured in snow of the Sierra Nevada in the United States, for example, has concentrations in the 10s or low-100s of ppb (Hadley et al., 2010; Sterle, et al., 2013). Even visibly dirty snow may only contain ~100 ppb of BC (Gleason et al., 2019). Additionally, the mass-fraction of EC compared to that of organic carbon (OC) is very small for emissions from smoldering combustion of Siberian and Alaskan peat. For example, Chakrabarty et al. (2016) report the OC:EC mass ratio (based on emission factors) as 70:1 for Alaskan peat combustion under very similar combustion and fuel conditions to our experiment. Thus, the light-absorbing OC produced through smoldering combustion of peat (i.e. BrC) dominates the optical, physical, and chemical presence of carbonaceous particulate matter reported in this study.

**Minor**

1. Page 6 Line 19. ...(TOC) concentration and absorbance in the UV and visible wavelength ranges, respectively, at the Desert Research Institute (DRI)...

The authors feel that this suggested change – adding the word “respectively” – is incorrect and unnecessary. The UV wavelength range does not refer to only the TOC determination; similarly, the visible wavelength range does not refer specifically to the absorbance determination. While both instruments utilize UV wavelengths of light during their operation, only the spectrophotometer uses both UV and visible light to determine absorbance. These are two separate measurements presented and utilized in this work. Therefore, “respectively” should not be added to the statement to which referee #1 refers to. However, we clarified this in the manuscript to better express what was intended, namely that total organic carbon (TOC) concentration measurements and UV-visible spectroscopy were carried out [separately] at the Desert Research Institute.

2. Page 6 Line 22. The organic carbon is very illusive to capture. In our previous work focusing on BC in ice, we completely excluded OC just for the same reason (Refer to 2.3 of Ming et al, 2008). Could you please present an estimation of the uncertainty, regarding the way of melting at room temperature?

The present manuscript includes a comprehensive section (Section 4, Page 16, Line 1) discussing possible uncertainty and sources of error throughout the study presented. This includes a statement in which the TOC concentration of ultra-pure water (UPW) shaken in Whirlpak bags (the same used for snow sample collection and frozen storage) was measured and determined to be an upper limit of contamination possible; however, this could only possibly happen if the collected snow samples were melted and shaken in the Whirlpak bags prior to TOC determination. Earlier in the

manuscript (Page 7, Line 3), we also mention that the polyurethane vials used when melting the snow contribute a non-negligible amount of TOC to the overall determination of TOC in the melted snow samples. Indeed, this contamination and its uncertainty was propagated throughout the calculations involving TOC values, including the derivation of the imaginary part of the refractive index of brown carbon deposited on the snow surface.

By the way, do you consider the newly generated bacteria inside the sample, which could, in turn, contribute some possible OC?

We do not consider any bacteria, specifically, in the contribution to measured OC of the melted snow sample, before or after collection. That is to say, we do not partition the different species of OC within the “natural” snow samples; the goal of this work is to only consider the difference in *total* organic carbon before and after deposition of BrC, thereby isolating the contribution made by the deposition experience to the TOC. However, the OC determination is not exclusive to non-biological organic material and can indeed include bacteria that contribute to the TOC determination presented in the manuscript. We clarify this by adding a statement with regard to the possible OC sources present in the collected snow samples.

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Chakrabarty, R. K., Gyawali, M., Yatavelli, R. L. N. N., Pandey, A., Watts, A. C., Knue, J., Chen, L.-W. A. W. A., Pattison, R. R., Tsibart, A., Samburova, V. and Moosmüller, H.: Brown carbon aerosols from burning of boreal peatlands: microphysical properties, emission factors, and implications for direct radiative forcing, *Atmos. Chem. Phys.*, 16(5), 3033–3040, doi:10.5194/acp-16-3033-2016, 2016.

Gleason, K. E., McConnell, J. R., Arienzo, M. M., Chellman, N. and Calvin, W. M.: Four-fold increase in solar forcing on snow in western U.S. burned forests since 1999, *Nat. Commun.*, 10(1), 2026, doi:10.1038/s41467-019-09935-y, 2019.

Hadley, O. L., Corrigan, C. E., Kirchstetter, T. W., Cliff, S. S. and Ramanathan, V.: Measured black carbon deposition on the Sierra Nevada snow pack and implication for snow pack retreat, *Atmos. Chem. Phys.*, 10(15), 7505–7513, doi:10.5194/acp-10-7505-2010, 2010.

Peltier, R. E., Weber, R. J. and Sullivan, A. P.: Investigating a Liquid-Based Method for Online Organic Carbon Detection in Atmospheric Particles, *Aerosol Sci. Technol.*, 41(12), 1117–1127, doi:10.1080/02786820701777465, 2007.

Sterle, K. M., McConnell, J. R., Dozier, J., Edwards, R. and Flanner, M. G.: Retention and radiative forcing of black carbon in eastern Sierra Nevada snow, *Cryosphere*, 7(1), 365–374, doi:10.5194/tc-7-365-2013, 2013.