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.

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 25 in the updated manuscript) should be expanded to include more information regarding the Sievers 900 instrument, its analysis protocols, and reported measurement accuracy and precision. These items have been included in the updated manuscript, beginning on Page 6, Line 27, which reads:

> "The instrument photo-chemically oxidizes organic compounds in a liquid sample through chemical oxidation with ammonium persulfate and reactions with hydroxyl radicals produced through the photolysis of water under UV irradiance. Within the instrument, the sample stream is split in two, where one analysis path (for IC) determines the CO2 formed through interaction with the ammonium persulfate only, and the other path (for TC) determines the CO2 produced through both ammonium persulfate and UV-induced oxidation (TC). Then, the CO2 in the sample stream is measured through a patented conductivity detector. The instrument has a reported lower detection limit of 0.03 ppb TOC and accuracy of ±2% or ±0.5 ppb, whichever is greater."

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.

We have added a sentence to the end of the paragraph describing the TOC analyzer summarizing the above, which reads (Page 7, Line 11):

> "To the best of the authors' knowledge, the Sievers TOC analyzer and its methods to oxidize organic compounds in liquid samples is unable to convert any BC present to CO_2, as BC is chemically inert and resistant to oxidation (Bond and Bergstrom, 2006), especially under the limited exposure that instrument subjects the sample to (~4 minutes)."

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. This sentence now reads (Page 6, Lines 19-21):

> "Here, snow samples were processed to quantify total organic carbon (TOC) concentration using a total carbon analyzer and absorbance using a laboratory spectrophotometer at the Desert Research Institute (DRI) in Reno, NV, USA."

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 manuscript includes a comprehensive section (Section 4, Page 15) 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 updated manuscript, we also mention that the polyurethane vials used when melting the snow contributed a non-negligible amount of TOC to the overall determination of TOC in the melted snow samples (Page 7, Line 10):

> "Contamination of the polypropylene vials with TOC was measured to be 0.020 ± 0.001 g m^{-3}, which was subtracted from each of the meltwater TOC measurements."

This contamination and its uncertainty was propagated throughout the calculations involving TOC values in this study, 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 experiments to the TOC.

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.

Reply to the review by Anonymous Referee #2 for the manuscript, "Deposition of Brown Carbon onto Snow: changes of snow optical and radiative properties" by N. D. Beres et al.

The authors thank the anonymous reviewer for their comments and recommendation for publication. Below, comments by the reviewer are in blue and the responses by the manuscript authors are in black.

The natural snow albedo is not 1.0 in the visible (see Fig.3). Therefore, it is clear that the snow samples were already polluted before introduction of BrC. I think, the better idea would be to use fresh or artificial (not polluted) snow samples.

The experiments in this study were conducted using a simple and portable deposition device (Beres and Moosmüller, 2018) which can mimic real-world aerosol dry deposition processes of varying mass concentrations onto real-world surfaces, such as snow. The goal of this study was a first investigation of how brown carbon (BrC) produced through the combustion of an important fuel source, can change snow optical and radiative properties; future investigations using the deposition apparatus can benefit from varying the snow conditions (high versus low snow mass density, varying grain radii, etc.). However, for our study, to isolate the influence from BrC only, the presence and influence of light-absorbing impurities in the snowpack before the deposition experiment is negated by finding the difference in values of spectral albedo, TOC concentrations, and spectrophotometric absorption, as measured before and after the deposition of BrC. This way, the BrC influence – on the snow optical properties, primarily – is isolated and investigated. As noted in the manuscript, it will require further research to refine our methods and determine some additional BrC-related effects to snowpack chemistry and optics. Using an artificial snowpack – such as the methods described in Hadley and Kirchstetter (2012) – may reduce some uncertainty in these methods while increasing others because natural and artificial snow differ in morphology, etc., but the macroscopic BrC-related effects and key results presented in our manuscript will likely remain intact.

The snow samples used for analysis in this study were indeed already "polluted" (that is to say, not "pure" or free from *all* impurities) prior to the artificial deposition of BrC, and the authors were aware of this fact. Concentrations of BC measured in snow of the Sierra Nevada in the United States, for example, have values in the 10s or low-100s of ppb and dust concentrations may be greater (Hadley et al., 2010; Sterle, et al., 2013). It should be noted that "pure" snow spectral albedo equal to 1.0 in the visible is never found in a natural snowpack, and light-absorbing impurities will be found even in snow of the most pristine areas of the Earth's cold regions, which will reduce the measured albedos or reflectance in the visible wavelength region (e.g. Warren and Wiscombe (1982), Warren et al. (2006), and Forrström et al. (2009)).

Beres, N. D. and Moosmüller, H.: Apparatus for dry deposition of aerosols on snow, Atmos. Meas. Tech., 11(12), 6803–6813, doi:10.5194/amt-11-6803-2018, 2018.

Forsström, S., Ström, J., Pedersen, C. A., Isaksson, E. and Gerland, S.: Elemental carbon distribution in Svalbard snow, J. Geophys. Res., 114(D19), D19112, doi:10.1029/2008JD011480, 2009.

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.

Hadley, O. L. and Kirchstetter, T. W.: Black-carbon reduction of snow albedo, Nat. Clim. Chang., 2(6), 437–440, doi:10.1038/nclimate1433, 2012.

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.

Warren, S. G. and Wiscombe, W. J.: A Model for the Spectral Albedo of Snow. II: Snow Containing Atmospheric Aerosols, J. Atmos. Sci., 37(12), 2734–2745, doi:10.1175/1520-0469(1980)037<2734:AMFTSA>2.0.CO;2, 1980.

Warren, S. G., Brandt, R. E. and Grenfell, T. C.: Visible and near-ultraviolet absorption spectrum of ice from transmission of solar radiation into snow, Appl. Opt., 45(21), 5320, doi:10.1364/AO.45.005320, 2006.

Reply to the review by Anonymous Referee #3 for the manuscript, “Deposition of Brown Carbon onto Snow: changes of snow optical and radiative properties” by N. D. Beres et al.

We thank the anonymous reviewer for their thorough and thoughtful response and their recommendations to improve the manuscript. Below, questions and comments by the reviewer are in blue and the responses by the manuscript authors are in black.

Abstract:

I think a more useful result emerging from this work is the calculation of spectrally-resolved mass absorption efficiencies for TOC:absorption across 275-815nm. As noted below, the authors have not convinced me that this is purely a BrC MAE; in particular, BC is likely contributing to measured absorption. Nonetheless, given the paucity of observed values of spectral absorption by BrC, this dataset will be useful. As such, I think this aspect of the work should be highlighted in the Abstract.

The authors agree with the reviewer that the mass specific absorbance, B_{λ}, is a useful dataset for BrC-contaminated snow, though we respectfully disagree that this is “a more useful result” than the novel BrC deposition experiments and this closure study. We have included the following statement in the abstract highlighting the B_{λ} dataset on Page 1, Line 20:

> “These measurements were used to first derive a BrC (mass) specific absorption (m^2 g^{-1}) across the UV-vis spectral range.”

The wording in line 23 of the Abstract needs to be modified so the focus isn’t on the magnitude of the RF but the forcing per mass of deposited aerosol. I’d suggest editing to, e.g.: “The instantaneous radiative forcing per unit mass of total organic carbon deposited to the ambient snowpack was found to be 1.23 (+0.14/-0.11) W/me per ppm. This snowpack already contained light-absorbing impurities; in a completely lean snowpack the forcing per mass TOC deposited would have been 2.68(+0.27/- 0.22) W/m2 per ppm of BrC deposited.”

The authors thank the reviewer for this suggestion. The wording of the final lines of the abstract (Page 1, Lines 24-27) have been updated to better reflect the focus of these findings to read:

> “The instantaneous radiative forcing per unit mass of total organic carbon deposited to the ambient snowpack was found to be 1.23 (+0.14/-0.11) W m-2 per ppm. We estimate the same deposition onto a pure snowpack without light-absorbing impurities would have resulted in an instantaneous radiative forcing per unit mass of 2.68 (+0.27/-0.22) W m-2 per ppm of BrC deposited.”

Main paper:

1) The paper reads as if BrC will exist as an aerosol independent of other aerosol components (e.g. BC). In the ambient atmosphere BrC and BC pretty quickly become internally mixed. There are two aspect of this work that are likely not reflective of the ‘real world’, and which should be acknowledged: Here, the aerosol are generated in what sounds like fairly realistic peat-burning conditions (kudos to the team for this aspect of the work!). However, the aerosol are deposited to snow within minutes (or less), whereas in the real world most deposited aerosol will have had, at a minimum, hours to chemically evolve and mix. As widely documented, the chemical and optical properties of combustion aerosol – and the organics in particular – rapidly evolve in the first hour or few hours after emission. It needs to be acknowledged that real ambient aerosol that is deposited is likely internally mixed, so BrC “aerosol” doesn’t really exist, whereas the aerosol deposited in this experiment is likely closer to an external mixture (e.g. pg. 3, lines 5-8 reads as if BrC “aerosol” exist in the ambient; these are likely mostly internally mixed aerosol with both BrC and BC). Second, and more important, is that ambient deposited aerosol is likely at least hours and mostly closer to days old, so the chemical composition and therefore optical properties of combustion aerosol, and the BrC component in particular, may be substantially different from the aerosol measured in the reported experiment.

The experiment outlined in our manuscript describes a process to isolate the optical and radiative effects of BrC aerosol on snow albedo. Previous studies (e.g. Chakrabarty et al. (2016) and Sumlin et al. (2018a, 2018b)) have shown large fractions of emitted aerosol from very similar combustion conditions as to our study are primary BrC aerosol – which is to differentiate from more chemically, optically, and physically aged or secondary organics that may absorb light. Here, we demonstrate the effect of these aerosols emitted from the combustion of a fuel that is found in regions physically adjacent to snow and ice regions that may or may not undergo long-range transport before depositing and affecting the radiative and optical properties of the snow. The exact extent and impact of depositions such as this has yet to be determined or estimated, but it is not outside the realm of possibility or reason for this investigation and future ones involving deposition studies with or without BrC aerosol.

2) pg. 8, lines 16-20 and It's noted that 700-860nm absorbance was subtracted from the measured signal, as a way of normalizing for drift.
a) What is the source of this "drift"? (instrumental?)

Yes, the source of the "drift" is instrumental, by which the spectrophotometric analysis may be affected. As outlined in Hecobian et al. (2010) and Sengupta et al. (2018), we subtract the light-absorption signal measured in longer wavelengths by the spectrophotometer (where we expect no influence from the experiment). This helps to account for instrument drifts during analysis periods and in between zeroing. We have added the word "instrumental" to the manuscript to clarify this on Page 8, Line 28 and now reads as:

> "The raw absorbance measurements were baseline-corrected to account for instrumental drift by subtracting the scan average over the 700 – 860 nm wavelength range from individual wavelength absorbance values, similar to the method outlined by Sengupta et al., (2018)."

b) The authors comment that this reduces the influence of any BC particle absorption since this is "fairly independent of wavelength across the UV-vis spectrum". However, this isn't really true: As stated in the paper itself, AAE for BC is ~1.0 – a slope of 1 in log-log space, not a slope of zero. So this is a sort of partial removal of BC signal, but not total. As the authors are trying to isolate absorption by BrC, a better approach would be to use the stated AAE of 1.0 for BC to subtract the estimated BC absorbance from the measured signal (starting with the assumption that all 700-860nm absorption is due to BC). Why not do this?

First, the reviewer is right in stating that our statement was incorrectly worded: the imaginary refractive index of BC is wavelength independent and the BC aerosol absorption coefficient is inversely proportional to the wavelength over the visible and near-visible spectral regions (AAE $\approx$ 1). This has been updated in our manuscript on Page 8, Line 30 to read:

> "In addition, this reduces some influence of BC particle light absorption across the UV-vis spectrum, while only minimally affecting BrC particle absorption that is much greater at blue and near-UV wavelengths (Bahadur et al., 2012; Chakrabarty et al., 2010; Kirchstetter and Thatcher, 2012; Lu et al., 2015; Sumlin et al., 2018b)".

Our statement is correct in stating that our baseline correction "reduces" the influence of any BC present in the liquid samples, but, yes, it may not completely eliminate the influence of any BC aerosol present in the sample.

If the residual absorbance in the 700-860 nm band would be solely due to BC it would, indeed, be best to model the BC absorbance over the whole spectrum. However, the residual absorption in the 800-860 nm band is caused by both instrumental drift and by BC absorption, and for our measurements, it likely that instrumental drift is dominant because very little BC is produced by the nearly exclusive smoldering combustion of peat. Had we been able to isolate the signal from the presence of BC only, we would have taken the approach as suggested by the reviewer. However, a conservative and, in our opinion, best approach was taken to uniformly subtract the 700-860 nm absorbance signal which reduces the influence of any BC present.

3) pg. 9, lines 27-29: "In the UV, there was a stronger reduction (in albedo) than in the visible. . . At 700nm however albedo was reduced by less than 0.01, indicating very little BC added to the snowpack through the deposition experiments". Perhaps I'm misunderstanding: Won't this by definition be the case since the absorptance spectra were effectively zero'd out at 700-860n (pg 8, lines 16-20) for all spectra?

The sentence within the manuscript to which you refer (Pg. 9, lines 27-29) speaks specifically to the measured in-situ spectral albedo, which was not "effectively zero'd out"; it was in fact the table-top spectrophotometer absorbance measurements that were adjusted to account for instrument drift (see point 2b above). This sentence that you refer to attempts to convince the reader that because the measured spectral albedo of the deposited area differed from that of the natural snowpack by less than 0.01 at 700 nm, there must be only a small amount BC aerosol added through the combustion/deposition experiment, especially considering the large amounts of BrC aerosol deposited. For example, if we use the SNICAR model to simulate the snowpack and sky conditions for the natural snowpack for Experiment 1 and add 65 ng of BC to the top 2.5 cm of snow, the spectral albedo at 700 nm (average of 695 and 705 nm values) will be reduced from 0.832 to 0.823, a difference of ~0.01. While we don't quantify BC within our snow samples, the amount of BC deposited is dominated by the amount of BrC deposited (4,425 ng in the top 2.5 cm for Experiment 1!) and this can be seen optically through spectral albedo as well, mostly for values lower than 700 nm, where we expect the spectral response to be.

In general, I think the paper needs to do a clearer job of convincing me that what was measured was the spectral absorption of BrC, not a mix of BrC and BC.

While the authors don't deny that there may be very small amounts of BC aerosol mass present, there are many studies that the emission ratios of OC to BC are very large for smoldering combustion of peat fuels. We employ the same methods as previous studies that BC and BrC can be separately detected using optical methods (e.g., Bahadur et al. (2012), Kirchstetter and Thatcher (2012), Lu et al. (2015)). Additionally, our manuscript utilizes very similar combustion methods to previous studies that investigate the optical properties of emissions from the smoldering combustion of peat (e.g. Chakrabarty et al. (2016), Sumlin et al. (2018)). For example, Chakrabarty et al. (2016) list OC emission factors that are 20 to 110 times larger than those of BC. In our study, we find further evidence that there was not a great influence from any BC present in the deposited material, through the use of different optical measurements like snow reflectance and absorbance measurements of melted snow samples.

Similarly, "TOC" is used synonymously with "BrC", when some of the organics in fact may not be light-absorbing (and therefore not BrC, by definition).

The reviewer is correct: the fraction of light-absorbing organic carbon versus non-light-absorbing organic carbon is not known to us for this particular fuel and combustion scenario and, for this study, we do not attempt to resolve it. However, throughout the optical measurements (snow surface albedo and spectrophotometric absorbance of melted snow samples), we consistently measure the same organic material without filtering. This way, we consistently use an "average" absorption of all organic compounds present and isolate those from the deposition experiment by using background subtraction.

4) Section 3.2.2 "Estimating changes in snow albedo and radiative forcing by BrC deposition" should be split into two sub-sections and renamed:

Pg. 13, line 10 through pg 14 line 17 is really describing how the observations were used to optimize the model so it produces the observed albedos.

Pg. 14 line 18 through pg 15 line 2 is really about the forcing *efficiency* per mass of deposited organics in a clean snowpack versus in the specific ambient snowpack where they did the experiment. Because the experiment was not producing realistic amounts of deposited aerosol (and was not intended to) the calculated "forcing" is not itself meaningful.

The authors agree that this separation of sections could help the reader understand the manuscript better. The manuscript has been updated to reflect this change. Section 3.2.2, beginning on Page 13, Line 19, is now named "Utilizing models for comparison with observations". Following the reviewer's point (6) below, some content of Section 3.2.3 has been removed; the remaining content – along with the content as recommended here (Pg. 14 line 18 through pg 15 line 2) - has been added to a new Section 2.3.2, which is named "Estimating the radiative forcing by BrC deposition", which begins on Page 15, Line 1.

5) pg. 14, lines 9-10: It's highlighted that the difference in spectral albedo between the modeled and observed albedos was less than 0.15. However, isn't this a result of the fact that BC and mineral dust amounts used in the model were specifically specified to reduce differences in the observed and modeled spectral albedos? This seems rather problematic, to then assert good agreement when it is built in by design!

Indeed, BC and mineral dust were added to the SNICAR natural/background snowpack in order to better match the measured "natural snowpack" albedo with a modeled one. This was done only for the "natural snowpack" before the deposited BrC was added within the model for comparison purposes.

The differences seen in the values to which you refer is when only after BrC is added to snowpack within SNICAR. In order to better test the derived BrC optical properties, we must make an assumption on the amount and type of impurities present before the deposition experiment. This is also done, of course, because BC and mineral dust mass were not measured. Similarly, the measured and modeled spectral albedo of the natural snowpack would not match in the 350 and 550 nm spectral range if light-absorbing OC (i.e. BrC) was not also added (note the "dip" in the measured albedo in this spectral range in Figure 3a and 3b for the natural snowpack). But, we didn't have to make an assumption here; we measured the BrC (inferred through the TOC) in the natural snowpack and added it to the natural snowpack impurity values.

6) Section 3.2.3 Radiative forcing by impurities as a function of existing conditions. As noted above, it is not at all a new finding that forcing for a given deposited mass will depend on how dirty the snowpack was to begin with. Given that many studies have calculated forcing by assuming a totally clean ambient snowpack it is worth emphasizing this point, but it is not worth an entire page and figure (Figure 9) to show this, as it seems rather tangential to the rest of the work presented. I think it would be sufficient to reference earlier studies making this point, and to simply give the difference in forcing efficiency for the ambient snowpack versus for a clean snowpack.

The authors agree in the reviewer's recommendation and appreciate their suggestion. We have removed the suggested content in this section and the corresponding figure. The remaining content has been added to a new section titled "Estimating the radiative forcing by BrC deposition" beginning on Page 15, Line 1.

7) I think the reported experiment could be improved upon in two ways. I'd ask the authors to consider whether they agree, and if so perhaps include these as areas for future work in the discussion at the end of the paper. (Note that this is not intended as a criticism of the work presented, only as ideas that were evident to me in reading the paper):
First, conducting the same experiment on a snowpack that is at and remains below freezing for the duration of the experiment would simply things. The authors do a good job of addressing how the above-freezing temperatures affected snow grain evolution and transport of aerosol in the snowpack. However, this did complicate their analysis – and the presence of liquid water may have affected the loss of particles in the snowpack and to the Whirlpak bags post-sampling.

Second, and more substantial: As noted above, the optical properties of organic aerosol in particular may rapidly evolves over the hours to day (or more) after emission in the atmosphere and in the high-actinic-flux environment of the snowpack. While the nature of the experiment is such that it would be logistically prohibitive to try and make a mixing chamber that would allow for in-atmosphere aging of the emissions before deposition, it would be feasible to monitor the evolution in snowpack spectral albedo over the hours to day+ following deposition. Because the experiment adds such a large concentration of absorbing impurities relative to the ambient snowpack impurities and relative to the amount of additional aerosol that would be deposited to the snowpack through natural processes over, e.g., a day, if the snowpack was below freezing (so aerosol is not being transported through the snowpack), the observed evolution in albedo could be largely attributed to the evolution in the properties of the deposited aerosol.

The authors agree that to further improve the understanding of BrC in snow and the changes it may cause to the optical, chemical, and physical properties of snow, much more experimentation and analysis is needed. This study provides a first look at how BrC aerosol produced using a relevant fuel may directly change the optical and radiative impacts of snow and ice, much in the same way that experimental studies of artificial BC deposition (and comparing to modeled results) have lent themselves to understanding how solely BC changes the optical and radiative impacts of the cryosphere, although BC is never the only impurity found in a natural snowpack.

The evolution of BrC in snow over different time scales is addressed in Section 4 (Page 15, Line 26), where we discuss uncertainty in the study. In particular, the authors have conducted preliminary experiments of temporal changes to the snow reflectance and the degradation of absorption after deposition of primary BrC on snow over time, similar to studies showing photooxidation contributing to the loss of absorptivity over time of BrC aerosols in the atmosphere (e.g., Sumlin et al. (2017)). These unpublished results are interesting and require further testing to isolate the causes of this optical change over time.

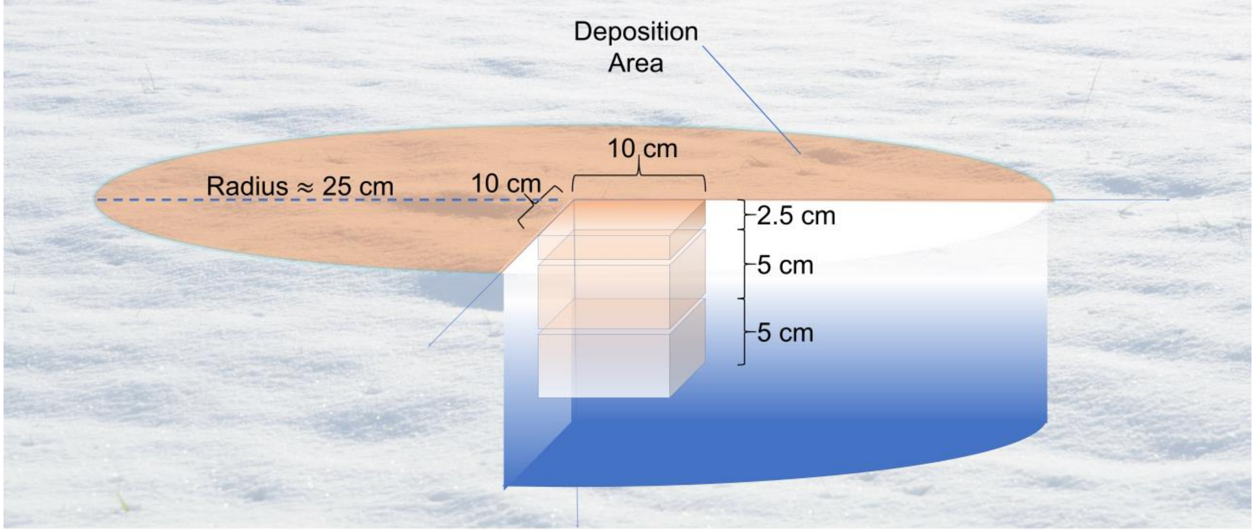
SMALLER COMMENTS:

Pg. 5, lines 10-11: What was the diameter of the cylinder used for the deposition end of the apparatus? (see comment below on addition of this to Figure 1).

The approximate areal extent of aerosols deposited on the snow surface has been added to the end of the paragraph containing a brief description of the deposition apparatus (Page 5, Line 20), which now reads:

> "The areal extent of deposited aerosol is approximately 0.20 m^2."

The radius (~25 cm) has been added to Figure 2, as seen here:



pg. 5, lines 29-30: Small question: Is there a reason for selecting specifically a 25% increase in mass?

Yes. We chose combustion conditions – such as the fuel moisture content – to be the same or as similar as possible to previous studies (i.e., Sumlin et al. (2018)) that have carried out similar experiments. This way, we would be able to recreate the combustion conditions, apply some derived values from other studies – such as the size distribution of BrC aerosol, and (hopefully) recreate similar properties of the aerosol emitted for our purposes.

Pg. 6, lines 1-4: Over what surface area was albedo measured? Were any corrections required for, e.g., the shadow of the instrument itself? Any issues with fact that the deposition area is not infinite and you were using a cosine collector?

To the best of the authors' knowledge and abilities, the in situ measurements of snow albedo were performed without shadowing the deposition area while minimizing the distance between the deposition area and the optical fiber; however, as the reviewer has pointed out, the cosine response of the fore optic attachment includes reflectance information from outside the deposition area, albeit with less weighting for the large radius areas beyond the deposition area. While we don't quantify this effect within the manuscript, the authors thank the reviewer for pointing out our oversight in addressing this uncertainty in our measurements. Our section on uncertainties of this study (Section 4.1) has been updated to include the fact that a majority of the spectral information will be from within the deposition area and much less influence will be from the surrounding ambient snowpack; specifically to this reviewer question, we've added the following sentences, beginning on Page 16, Line 6:

> "One limitation of the deposition apparatus and measuring the hemispherical spectral albedo of the subsequent deposited aerosols is the limited areal extent of deposited material when compared to the viewing angle of the cosine-corrected receptor. Because the deposition area is not infinite, there will be a fraction of reflected light from the surrounding ambient snowpack that will influence the

> measured spectral albedo; however, this fraction of information is reduced for greater angles. Great care was taken during the experiment to minimize the effect from both instrument shadowing while decreasing the distance between the optical detector and the snow surface. As such, a majority of the reflected signal is from within the deposition area and some information is from the surrounding natural snowpack."

pg. 11, lines 13-15: is there a reference to substantiate this assertion about the UV absorption of aromatic nuclei is at ~255nm?

In the updated manuscript, on Page 11, Line 24, we have included two references to support this assertion, namely Pretsch et al. (2000) and Samburova et al. (2016). The updated statement now reads:

> "The UV absorption band in the presence of an aromatic nuclei (no other functional group specified) can be around 255 nm (Pretsch et al., 2000; Samburova et al., 2016), which is very close to our observed β_{abs_snow} values."

9) pg 12, line 19: The AAEs given in Doherty et al. (2010) are for 450-600nm. AAE over this wavelength band might be quite different from that at the band of 330-400nm given here.

The values for AAE_{400}^{330} reported in our manuscript are from Zhang et al. (2019) who, in their Fig. 6, report AAE_{400}^{330} and not AAE_{600}^{450}. We assume Zhang et al. (2019) have adapted the Doherty et al. (2010) values to the wavelength range of 330-400 properly.

Figure 1: Overall this is a useful figure. I was, however, a bit confused by the box on the left side that simply says "Sumlin et al. (2018)". What is this supposed to represent? If it's the source of the BrC size distribution, just put "Sumlin et al. (2018)" inside that box.

The Sumlin et al. (2018) study lends two items to our manuscript, the size distribution of BrC aerosol particles used in this study as well as values of derived BrC imaginary refractive index, which we use only to compare against values derived through our methods; hence, there is a "T" split in the figure. There are two dashed lines in shown in the legend as well, indicating that the dashed box emphasizing that "Sumlin et al. (2018)" is from an outside study. The figure caption has been updated to clarify the figure and the use of outside studies, and now reads as:

> "Overview of methods used to derive BrC optical properties and compare measured and modeled albedo. The Sengupta et al. (2018) and Sumlin et al. (2018) studies both provide previously derived values of κ_{BrC} to compare, and Sumlin et al. (2018) provide a size distribution of BrC aerosol under similar combustion conditions to those used in this study."

Figure 2: Can you add the radius/diameter of the deposition area to this figure, please?

Figure 2 has been updated to include the approximate radius of the deposition area and has been scaled a bit to better represent relative lengths, as seen here:

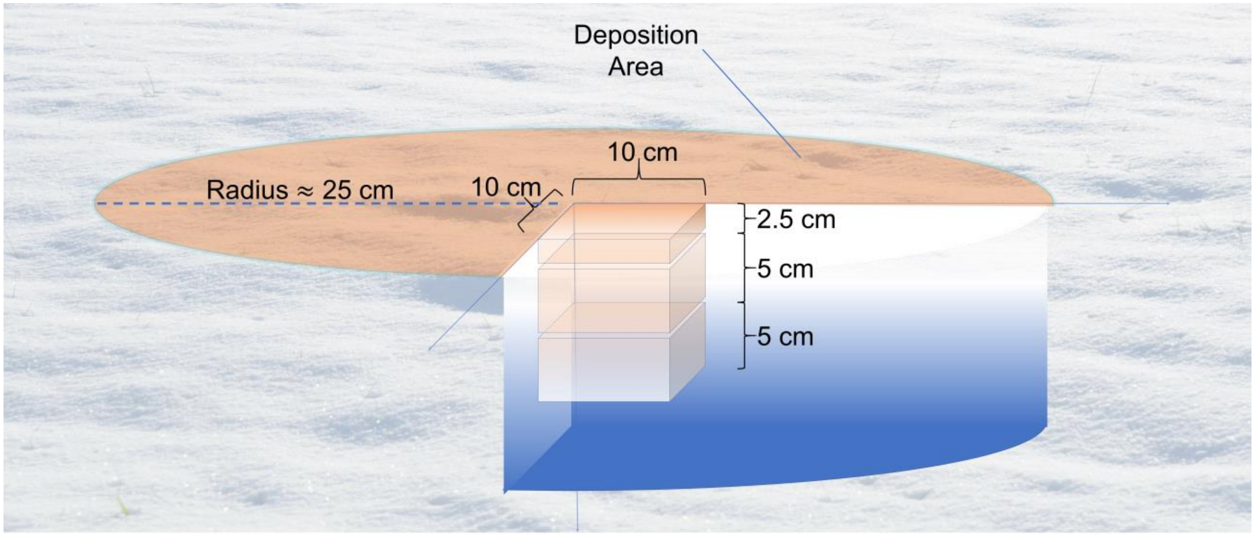


Figure 5 caption should state the R-squared at each wavelength (as shown in panel a for 275nm) is also given for each wavelength, and it should note at what wavelength resolution the mass-specific absorption is given.

Thank you for this suggestion; the figure caption has been updated and now reads:

> “Panel (**a**): Absorption coefficient at 275 nm as function of total organic carbon (TOC) concentration. The fitted linear regression slope gives the mass-specific absorption B_λ= 7.5 ± 0.3 m^2 g^{-1} with a correlation coefficient of R^2 = 0.98. Panel (**b**): Mass-specific absorption, B(λ), and R^2 for each value of B(λ) across the wavelength range 215 – 815 nm for BrC deposited experimentally and found in natural snow at Tamarack Lake together at a 1-nm resolution. These panels include absorption coefficients of all meltwater samples characterized in Fig. 2.”

Figure 7 caption should note that the real index of refraction is assumed to be a fixed value of 1.6 across all wavelengths. This is why both g and the mass extinction coefficient are so invariant (small gray area in panels b and c)!

Thank you for this suggestion; the figure caption has been updated to read:

> “Single scattering albedo (SSA, Panel **a**), asymmetry parameter (g, Panel **b**), and mass extinction coefficient (MEC, m^2 kg^{-1}, Panel **c**) of BrC derived using Mie theory. BC optical properties used in the SNICAR model are provided for reference. The grey area represents the outputs of each parameter for the range of κ_{BrC} retrievals used in the Mie code, where the largest relative discrepancy lies in the SSA. Note: the real refractive index for the calculations presented in this figure is assumed to be n_λ = 1.6 across all wavelengths.”

Table 1: The air temp is given (footnote!) at a site 0.9km SE of the sampling site and at 2683m altitude. What is the altitude of the sampling site itself? Considerably different from 2683m? (Can you use the dry adiabatic lapse rate to estimate the sampling site temp if it wasn’t measured, which would have been easy enough to do!)

The Tamarack Lake experiment site lies at an altitude of 2694m above sea level. The authors believe that the difference in altitude (~11 m) and linear distance (< 1 km) between the Mt. Rose SNOTEL site and the Tamarack Lake site does not represent a significant change in atmospheric or meteorological conditions; thus, the air temperatures provided are representative.

SUGGESTED SMALL EDITS:

Pg. 2, line 10: “cryosphere are a growing” –> “cryosphere is a growing”

Done; the statement now reads (Page 2, Lines 10-11):

> "The light-absorbing properties of BrC – an optically defined component of organic carbon (OC) aerosol – in the cryosphere is a growing topic of research."

Pg. 2, lines 20-22: Dang and Hegg (2014) should also be cited here.

Added. The statement now reads (Page 2, Lines 20-22):

> "Many studies have focused on the light absorption of the water-soluble fraction of OC, particularly that of humic-like substances (HULIS) (Dang and Hegg, 2014; Graber and Rudich, 2006; Samburova et al., 2005; Sun et al., 2007)."

Pg. 3, line 3: "toward shorter wavelength" –> "towards shorter wavelengths"

Done; the statement now reads (Page 3, Lines 2-4):

> "The imaginary part of the BrC refractive index (i.e., κ_λ) increases greatly toward shorter wavelengths in the visible and near-UV spectrum, giving BrC its colored appearance and namesake (Andreae and Gelencsér, 2006).

Pg. 3, line 25: add a comma after "(Ervens et al., 2004)"

Done; the statement now reads (Page 3, Lines 24-25):

> "Chemical transformation of BrC during atmospheric transport can involve either fragmentation processes, producing lower molecular weight compounds (Ervens et al., 2004), or oligomerization (Carlton et al., 2007)."

Pg. 4, line 26: "presented in Fig. 1 to guide through the relationships" –> "presented in Fig. 1 as a guide to the relationships"

Done; the statement now reads (Page 4, Lines 26-27):

> "An overview of methods used for this work is presented in Fig. 1 as a guide to the relationships between measurements and modeling presented in the following sections."

Pg. 7, line 24: "This method assumes there is little contribution to scattering to the overall extinction of light along the path." These samples surely include insoluble particles, so there was some contribution from particulate scattering to the signal. Was the possible magnitude of this effect estimated? Would this constitute a positive bias in derived absorption?

The authors agree that the wording of this sentence may be a bit misleading in our intentions and assumptions made during our analyses. Indeed, the contribution to the scattered signal from insoluble particles is not quantified but is assumed to be low enough and present in both the snow albedo measurements after deposition and the absorbance measurements of the melted samples. Thus, any bias that is present in the measurements derived from scattered light is (assumed to be) accounted for. Additionally, visual inspection of the melted snow samples did not show any cloudiness, which would indicate scattering by a large fraction of insoluble particles. Instead, the uncertainty pertaining to insoluble particulates is present in the TOC analysis, where some insoluble OC may be unaccounted for (addressed in Section 4). We have updated the manuscript on Page 8, Line 1 to better state this assumption; it now reads:

> "This method assumes there is little contribution of scattering to the overall extinction of light along the path (Bosch et al., 2014). This is appropriate for a suspension or solution where extinction is

> dominated by absorption caused by dissolved OC and/or by carbonaceous particles smaller than the wavelength of incident light, where extinction due to scattering can be neglected.”

Pg. 9, line 25: “. . . in Fig. 3 alongside measured” –> “. . . in Fig. 3 alongside the measured”

Fixed. The updated sentence now reads (Page 10, Lines 7-9):

> “This strong wavelength dependence was quantified by the measured spectral albedo for each deposition experiment, which is shown in Fig. 3 alongside the measured albedo of the natural snowpack upwind of the deposition.”

Pg. 9, line 27: “In the UV, there was a stronger reduction than in the visible, up to ~0.14 and ~0-.21 at 350nm for Experiment 1 and 2, respectively (Fig. 3)” –> “In the UV, there was a stronger reduction in albedo than in the visible, of ~0.14 and ~0-.21 at 350nm for Experiment 1 and 2, respectively, relative to the natural snowpack albedo (Fig. 3)”

Fixed. The updated sentence now reads (Page 10, Lines 10-12):

> “In the UV, there was a stronger reduction in albedo than in the visible, of ~0.14 and ~0.21 at 350 nm for Experiment 1 and 2, respectively, relative to the natural snowpack albedo (Fig. 3).”

Pg. 10, line 23: “snow sampling sites, L1 TOC” –> “snow sampling sites, the L1 TOC”

Added “the”. The new sentence now reads (Page 11, Lines 2-4):

> “Averaging samples collected at both natural snow sampling sites, the L1 TOC concentration was 0.579 ± 0.014 g m^{-3}, for L2 it was 0.436 ± 0.048 g m^{-3}, and for L3 it was 0.425 ± 0.008 g m^{-3}.”

Pg. 10, line 26: “air pollution from mining. . .” –> “air pollution from the mining. . .”

Done. The sentence now reads (Page 11, Lines 5-7):

> “Meinander et al. (2013) found a factor of six larger, where concentrations are heavily influenced through deposition of air pollution from the mining and refining industry.”

Pg. 10, line 30: “concentration than Experiment 1” –> “concentration as Experiment 1”

Fixed. The sentence now reads (Page 11, Lines 9-11):

> “Experiment 2 deposited approximately twice the TOC mass concentration as Experiment 1, which is evidenced not only in the measured TOC concentration for each layer, but also in a stronger decrease of measured UV albedo (Fig. 3).”

Pg. 10, lines 31-32: delete “. . .for Experiments 2 than for Experiment 1 relative to the natural snow” (it’s not needed).

Done. The new sentence now reads (Page 11, Lines 9-11):

> “Experiment 2 deposited approximately twice the TOC mass concentration as Experiment 1, which is evidenced not only in the measured TOC concentration for each layer, but also in a stronger decrease of measured UV albedo (Fig. 3).”

Pg. 11: “UV absorption band in presence of an aromatic..” –> “The UV absorption band in The presence of an aromatic..”

Added “the”, and the updated manuscript now reads (Page 11, Line 24):

> "The UV absorption band in the presence of an aromatic nuclei (no other functional group specified) can be around 255 nm (Pretsch et al., 2000; Samburova et al., 2016), which is very close to our observed β_{abs_snow} values."

pg. 11, lines 29-30: I'm not sure what you're trying to say here with ". . .together with those of the nature snowpack, TOCsnow, as one dataset." Figure 5 only shows one dataset. ?

Reference to "one dataset" refers to the fact that the TOC concentrations from the natural snow and TOC concentrations from the deposition area snow are used together in the linear regression to derive the value of the specific absorbance, B_λ, instead of deriving a value of B_λ for each individual set of TOC concentrations: one for TOC_{snow} and one for TOC_{BrC}. This strengthens the derivation of B_λ over the wavelength range to include more TOC/absorbance pairs for higher concentrations.

pg. 13, line 24: Figure 7 should be referenced at the end of the sentence at the beginning of this line.

Added. The new sentence now reads (Page 14, Lines 2-4):

> "Mie theory calculations with our range of κ_{BrC} used as input into the Mie code return a range of single particle properties for the single scattering albedo (SSA), asymmetry parameter (g), and mass extinction coefficient (MEC, m^2 kg^{-1}) (Fig. 7)."

Pg. 14, lines 9: the reference to Figure 7 should be to Figure 8.

Fixed. The new sentence now reads (Page 14, Lines 19-22):

> "Once reasonable agreement (Δalbedo ≤ 0.025) is achieved for the natural snowpack measured and average modeled albedos across the wavelength range, the solar geometry, snowpack properties, and BrC concentration are updated to reflect that of the BrC deposition; results are presented in Fig. 8."

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Reply to the review by handling editor Dr. Timothy Garrett and overview of other minor changes for the manuscript, “Deposition of Brown Carbon onto Snow: changes of snow optical and radiative properties” by N. D. Beres et al.

We thank Dr. Garrett for his suggested adjustment to the manuscript. Dr. Garrett’s comment is in blue and the responses by the manuscript authors are in black.

One adjustment I request is that a link is provided to the datasets placed a formal data repository, replacing the statement "Data will be made available upon request by the corresponding author".

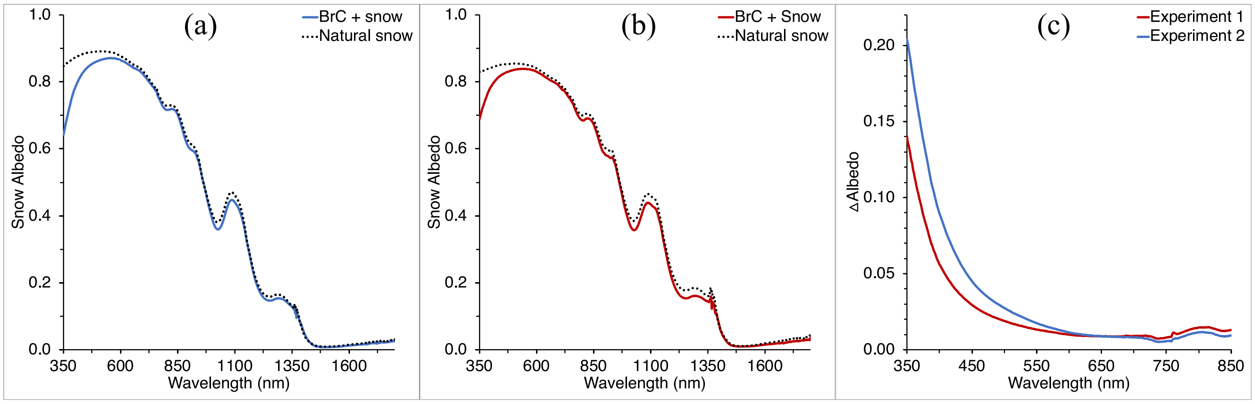
The data presented throughout the manuscript has been uploaded to the following Zenodo respository:

https://doi.org/10.5281/zenodo.3736325

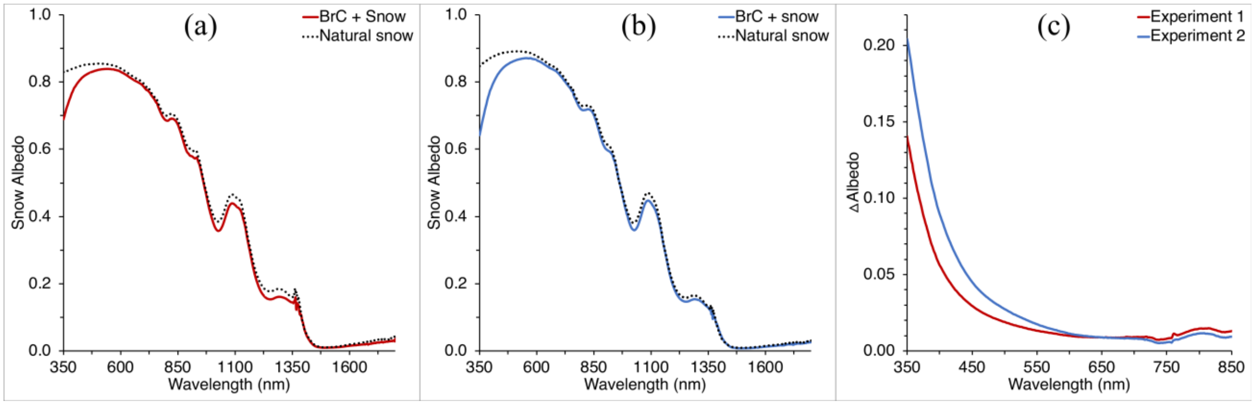
Data was uploaded in the form of an Excel spreadsheet, where each tab of the spreadsheet corresponds to the data presented in the figures and tables throughout the manuscript. Additionally, the “Data Availability” statement at the end of the manuscript was replaced to read (Page 18, Lines 25-26):

> “*Data Availability:* Data presented and used throughout this study can be accessed through the following data repository: https://doi.org/10.5281/zenodo.3736325.”

Additionally, one update to a figure was implemented. The panels (**a**) and (**b**) of Figure 3 were arranged in the wrong order. Before, the figure was shown as:



They have been updated to reflect the correct order of the experiment and to match the caption, and are now shown as:



Deposition of brown carbon onto snow: changes of snow optical and radiative properties

Nicholas D. Beres[1,2,3], Deep Sengupta[1,2], Vera Samburova[1], Andrey Y. Khlystov[1], Hans Moosmüller[1]

[1]Division of Atmospheric Sciences, Desert Research Institute, Reno, NV, ~~89512,~~ United States
[2] University of Nevada-Reno, Reno, NV, ~~89512,~~ United States
[3]Aerosol and Environmental Physics, University of Vienna, Vienna, Austria

Correspondence to: Nicholas D. Beres (~~Nic.Beres@DRI.edu~~nicholas.beres@univie.ac.at)

Abstract. Light-absorbing organic carbon aerosol – colloquially known as brown carbon (BrC) – is emitted from combustion processes and has a brownish or yellowish visual appearance, caused by enhanced light absorption at shorter visible and ultraviolet wavelengths ($0.3\ \mu m \lesssim \lambda \lesssim 0.5\ \mu m$). Recently, optical properties of atmospheric BrC aerosols have become the topic of intense research, but little is known about how BrC deposition onto snow surfaces affects the spectral snow albedo, which can alter the resulting radiative forcing and in-snow photochemistry. Wildland fires in close proximity to the cryosphere, such as peatland fires that emit large quantities of BrC, are becoming more common at high latitudes, potentially affecting nearby snow and ice surfaces.

In this study, we describe the artificial deposition of BrC aerosol with known optical, chemical, and physical properties onto the snow surface and we monitor its spectral radiative impact and compare it directly to modeled values. First, using small-scale combustion of Alaskan peat, BrC aerosols were artificially deposited onto the snow surface. UV-vis absorbance and total organic carbon (TOC) concentration of snow samples were measured for samples with and without artificial BrC deposition. These measurements were used to first derive a BrC (mass) specific absorption (m^2 g^{-1}) across the UV-vis spectral range. We then estimate the imaginary part of the refractive index of deposited BrC aerosol ~~with~~using a volume mixing rule. Single particle optical properties were calculated using Mie theory, and these values were used to show that the measured spectral snow albedo of snow with deposited BrC was in general agreement with modeled spectral snow albedo using calculated BrC optical properties. The instantaneous radiative forcing ~~my impurities present in the snow before the deposition experiments was measured~~per unit mass of total organic carbon deposited to ~~be BrC aerosol deposition~~ the ambient snowpack was found to ~~increase the instantaneous radiative forcing at the surface of the natural snow at our site by~~be 1.23 (+0.14/-0.11) W m^{-2} per ppm~~ of BrC deposited. However, we~~. We estimate ~~that~~the same deposition onto a ~~clean~~pure snowpack without light-absorbing impurities would have resulted in ~~a more than twice as large~~an instantaneous radiative forcing per unit mass of 2.68 (+0.27/-0.22) W m^{-2} per ppm of BrC deposited.

1 Introduction

Aerosol light absorption in the Earth's atmosphere lowers the planetary albedo, thereby causing radiative heating (Moosmüller et al., 2009; Satheesh and Krishna Moorthy, 2005; Stier et al., 2007; Stocker et al., 2013). Aerosols with a zero or very large imaginary part of their refractive index do not absorb light (Moosmüller and Sorensen, 2018b, 2018a, 2019; Sorensen et al., 2019). However, with the exception of metals, all atmospheric aerosols with a non-zero imaginary part of their refractive index do contribute to atmospheric light absorption. This absorption is distinct from other radiative forcing mechanisms such as aerosol scattering because light-absorbing aerosols continue to substantially contribute to radiative forcing after deposition onto high albedo surfaces such as snow and ice (Skiles and Painter, 2018; Warren, 1982). They contribute to climate forcing (Hansen and Nazarenko, 2004; Jacobson, 2004) by significantly lowering snow and ice albedo (Chýlek et al., 1983; McConnell et al., 2007; Warren and Wiscombe, 1980), thereby reducing snow cover duration and changing spring runoff timing (Déry and Brown, 2007; Painter et al., 2007; Strack et al., 2007). Most previous work discusses reduction of snow albedo due to deposited black carbon (BC) and mineral dust, but little is known about snow albedo reduction due to deposited brown carbon (BrC). The light-absorbing properties of BrC – an optically defined component of organic carbon (OC) aerosol – in the cryosphere ~~are~~is a growing topic of research. One source of atmospheric BrC is smoldering biomass combustion, which is becoming more common at high latitudes as peatlands increasingly dry out and become available as fuel for smoldering wildland fires (Brown et al., 2015; Kohlenberg et al., 2018; Turquety et al., 2007). Primary BrC aerosol deposition onto snow and ice is of particular concern in northern latitudes, due to the proximity of peat fuels to snow and ice surfaces (e.g., Evangeliou et al., 2019).

1.1 Brown Carbon Aerosol

Both BC and OC aerosols are formed by incomplete combustion of carbon containing fuels (Andreae and Gelencsér, 2006; Bond et al., 2013), and their contribution to a source's total emissions and their characteristics depends on many factors including fuel type, moisture content, packing density and source depth (Sumlin et al., 2018b), combustion phase (Bond et al., 2004; Patterson and McMahon, 1984; Reid et al., 2005), and other elements of the system (Chen et al., 2006). Many studies have focused on the light absorption of the water-soluble fraction of OC, particularly that of humic-like substances (HULIS) ~~(Graber and Rudich, 2006; Samburova et al., 2005; Sun et al., 2007)~~(Dang and Hegg, 2014; Graber and Rudich, 2006; Samburova et al., 2005; Sun et al., 2007). In addition, some recent work has focused on the light-absorption characteristics of non-water-soluble polycyclic aromatic hydrocarbons (PAHs) (Samburova et al., 2016) and other compounds derived from extractions with solvents of different polarity and pH (Sengupta et al., 2018).

OC compounds that are absorbing in the ultra-violet (UV) and short-visible wavelengths are colloquially known as BrC. The wavelength dependency of BrC aerosol absorption coefficients can be described by an absorption Ångström exponent (*AAE*) that is significantly greater than one (Chakrabarty et al., 2010; Corr et al., 2012; Kirchstetter et al., 2004; McNaughton et al., 2011; Moosmüller et al., 2011), whereas BC aerosol absorption coefficients are less wavelength-dependent (i.e., $AAE \approx 1$)

over the visible range and its periphery (Bergstrom et al., 2007; Moosmüller et al., 2009). The optical properties of BrC are described by the wavelength dependent BrC complex refractive index m_λ written as

$$m_\lambda = n_\lambda + i\kappa_\lambda, \quad (1)$$

where n_λ is the real part and κ_λ is the imaginary part of the refractive index, the latter of which provides the primary control of the absorption coefficient (Sorensen et al., 2019). The imaginary part of the BrC refractive index (i.e., κ_λ) increases greatly toward shorter ~~wavelength~~wavelengths in the visible and near-UV spectrum, giving BrC its colored appearance and namesake (Andreae and Gelencsér, 2006). BrC aerosol optical properties are determined both by BrC bulk properties discussed above and by particle size distribution and morphology. As BrC particles from smoldering combustion are mostly homogeneous (Sumlin et al., 2018a) and spherical (Chakrabarty et al., 2010), BrC aerosol optics can be described by BrC complex refractive index and size distribution using Mie theory (Mie, 1908; Moosmüller et al., 2011; Sumlin et al., 2018b).

A variety of fuels emit BrC during open combustion (Laskin et al., 2015), but peat is of particular interest for its common physical proximity to snow and ice at high latitudes of the northern hemisphere (Joosten and Clarke, 2002) and due to its strong tendency to burn in the smoldering combustion phase (Watts and Kobziar, 2013). Peatlands are a land surface primarily found in the northern hemisphere composed of organic soil and decomposing plant material. They make up a small fraction (~2-3%) of Earth's land surface but store a significant amount (~25%) of the world's soil organic carbon (Turetsky et al., 2014; Xu et al., 2018). Under a warming climate, by the end of the 21st century, a doubling of boreal area burned per year has been predicted (Flannigan et al., 2009; Oris et al., 2014). Increases in boreal peatland fires will cause increased emissions of OC and BrC aerosols at high latitudes.

Approximately 88% of all carbonaceous aerosol mass emitted globally is from wildland fires and biomass fuel combustion (Bond et al., 2004), and approximately 80% of those emissions are from smoldering combustion phase fires (Einfeld et al., 1991). OC is emitted together with BC from combustion sources, but more so in the smoldering phase (Bond et al., 2004; Patterson and McMahon, 1984; Reid et al., 2005) and it differs significantly in optical and chemical properties (Bond et al., 2013; Chakrabarty et al., 2016; Kirchstetter et al., 2004; Laskin et al., 2015; Lewis et al., 2008; Moosmüller et al., 2009).

Carbonaceous particles also often undergo continued, secondary changes – physically, chemically, and optically – during ~~transported~~transport within the atmosphere (Bhattarai et al., 2018; Jimenez et al., 2009; Sengupta et al., 2018; Sumlin et al., 2017). Chemical transformation of BrC during atmospheric transport can involve either fragmentation processes, producing lower molecular weight compounds (Ervens et al., 2004), or oligomerization (Carlton et al., 2007).

1.2 Brown carbon in the cryosphere

OC ~~present in snow can be deposited after atmospheric transport of~~from both natural and anthropogenic ~~aerosols~~sources can be deposited onto snow after transport. The presence of OC in snow and ice not only plays a critical role in the carbon cycle of the cryosphere and of waterways downstream of snowmelt, but also contributes to the darkening of the snow surface at the short-visible and UV wavelengths. While studies of impurities in snow and their reduction of snow albedo have focused on light-absorption by BC and/or mineral dust (Qian et al., 2015; Skiles et al., 2018), these impurities alone may not explain the decreased albedo at short-visible and UV wavelengths in the cryosphere.

Although only a small fraction of the total incoming solar irradiance at the snow surface is in the UV, it is a major driver of snow photochemistry that can be altered by BrC deposition near the top of the snowpack. Snow photochemistry includes nitrate and nitrite photolysis, which releases NO_x back into the atmosphere (Domine et al., 2008; Honrath et al., 1999, 2000) and mercury and bromine chemistry (Fain et al., 2006; Grannas et al., 2007). Additionally, biota in the cryosphere, such as snow algae (e.g., *Chlamydomonas nivalis*) (Thomas and Duval, 1995; Yallop et al., 2012), depend on incident light, and deposited impurities, such as BrC, can alter this interaction by absorbing light.

The influence of deposited BrC aerosols on surface and in-snow radiative properties is a recent and emerging topic. Grannas et al. (2007) provided an excellent overview of reactive organic compounds in snow and their impact on photolysis on and within the snow, but the direct impact of BrC on increased absorption and subsequent albedo effects was not addressed. Doherty et al., (2010) discovered the widespread presence of non-BC absorbing impurities (including ~25-50% contribution to total light-absorption by BrC) throughout the Arctic~~;~~, but the spectral albedo of these non-BC constituents was not reported. Similarly, Lin et al., (2014) extensively modeled the impact of BrC and other organic aerosols on global radiative forcing estimates. Wu et al., (2016) provided a nice but brief overview of BrC in the cryosphere and estimated effects on snow radiative forcing. One issue affecting ~~the~~ advances in understanding the roles ~~in which~~that BC and non-BC aerosols play for radiative forcing in and on snow is that partitioning their influence is difficult. Dang and Hegg (2014) described an extensive investigation identifying the absorbing components of non-BC materials – likely including BrC – in snow from across the Western United States using chemical extraction methods. For example, extraction of aerosols collected on filters allows for the separation of BC and non-BC constituents, but temporal resolution is lost when compared to real-time measurement methods.

Deposition of BrC onto and into snow and its radiative effects can be studied in detail and at sufficiently high BrC concentrations by artificially depositing BrC onto the snow surface (Beres and Moosmüller, 2018). Such experiments can deposit BrC from fuel sources, such as peat, that are likely to exist near snow surfaces, onto snow and quantify changes in snow composition (e.g., OC concentration) and radiative properties (e.g., spectral albedo). Here, we describe artificial deposition experiments, performed on natural snow surfaces in the Sierra Nevada, USA and report changes in snow OC content, meltwater absorption spectra, measured and modeled snow spectral albedos, and resulting radiative forcing due to the presence of impurities both naturally occurring and artificially deposited.

2 Methods

2.1 Study area and field work

An overview of methods used for this work ~~are~~is presented in Fig. 1 ~~to~~as a guide ~~through~~to the relationships between measurements and modeling presented in the following sections. Experiments for this study were conducted at a small, alpine lake, Tamarack Lake~~,~~, (2694 m a.s.l.), in the Carson Range of the Sierra Nevada in Nevada, USA. Depositions for this study took place during the winter/spring season of 2018-2019. ~~Downwind of~~Near the experiment site is a well-travelled mountain highway over Mount Rose pass, a ski resort, and the city of Reno, Nevada, USA. Sources of snow contamination include vehicle emissions along that highway, regional anthropogenic and biogenic sources in the Lake Tahoe region (Green et al., 2012; McDaniel and Zielinska, 2015), and long-range transport from California and Asia (Hadley et al., 2010). While our study site is not free from influences of natural and anthropogenic sources, it provides a flat, easy-to-access, untrodden snow field, ideal for experiments involving the artificial deposition of aerosols. Experiments were conducted on a clear-sky, windless day.

Field experiments were conducted in the following order: (1) begin combustion of fuel to generate aerosols for deposition; (2) during deposition of aerosols (~40 min.), excavate abbreviated snow pit upwind of deposition, measure snow density, and collect snow samples of "natural" snowpack; (3) upon completion of aerosol deposition, immediately remove deposition apparatus and measure spectral albedo over the deposition area; (4) measure spectral albedo of "natural" snowpack several meters upwind of deposition area; (5) excavate snow pit at deposition site and collect snow samples. A second deposition experiment was conducted on the same day, and steps (1)-(5) were repeated and results included in this study.

Aerosol depositions onto the snow were made using the apparatus previously described by Beres and Moosmüller (2018). Briefly, combustion aerosols are produced and deposited onto and into the snowpack by means of two near-cylindrical volumes. The first acts as an aerosol production chamber, where small-scale combustion of fuels produces aerosols in-situ. Using an external, battery-powered pump, aerosols are pumped from the first to a second, larger volume that is placed over the snow surface where deposition is desired. Prior to combustion, the fuels are placed into a round, insulated container (volume of this container is ~1610 cm^3) to mimic real-world conditions in which surrounding, unburned peat provide insulation for the smoldering biomass, similar to the methods of Chakrabarty et al. (2016), Sengupta et al. (2018), and Sumlin et al. (2017, 2018b, 2018a). ~~The fuels burned mostly with lower-temperature, smoldering-phase combustion producing OC-rich biomass-burning aerosols (any visible flaming combustion was quickly extinguished). Fuels burned for approximately 40 minutes, with air from the combustion volume being pumped into the deposition volume for an additional 10 minutes after the end of fuel combustion to encourage deposition of aerosols remaining in the volume. After this, the deposition volume was removed, and the experiment continued.~~The fuels burned mostly with lower-temperature, smoldering-phase combustion producing OC-rich biomass-burning aerosols, where any visible flaming combustion was quickly extinguished to ensure OC to EC emission ratios are high (Chakrabarty et al., 2016). Fuels burned for approximately 40 minutes, with air from the combustion volume being pumped into the deposition volume for an additional 10 minutes after the end of fuel combustion to encourage deposition of aerosols remaining in the volume. After this, the deposition volume was removed, and the experiment continued. The areal extent of deposited aerosol is approximately 0.20 m^2.

The fuel combusted for BrC aerosol deposition consisted of boreal peat samples collected from interior Alaska (hereafter "AK peat" or "AKP" in figures). Details of the fuel collection and preparation can be found in the supplemental information of Chakrabarty et al. (2016). The properties of this fuel's combustion emissions have been extensively studied under similar combustion conditions for their impact on air quality and radiative forcing in the atmosphere through optical, physical, and chemical characterization (Chakrabarty et al., 2016; Samburova et al., 2016; Sengupta et al., 2018; Sumlin et al., 2017, 2018a, 2018b). Peat fuels were harvested and stored under refrigeration after collection. A few days before snow deposition fieldwork, these fuels were conditioned in a heating and drying oven (Fisherbrand Isotemp, Waltham, MA) at a temperature of ~90 °C for one day in order to remove fuel moisture. Dried peat samples were weighed, and deionized water was sprayed homogeneously on those dry samples until the total mass of the wet peat samples showed a 25% increase compared to dried peat sample mass (i.e., fuel moisture content of 25%). Prior to their use in field combustion experiments, the wet peat samples remained for one day of equilibration in U-Line static shielding Ziploc bags.

Spectral albedo measurements of natural and aerosol-deposited snow surfaces were made using a fiber-coupled, high-resolution spectroradiometer (FieldSpec3, Analytical Spectral Devices, Inc., Boulder, CO, USA). This instrument measures over a wavelength range of 350-2500 nm with full-width, half maximum bandwidths of 3 nm (at 700 nm) to 10 nm (at 1400 and 2100 nm). Raw spectra are resampled and splined at 1 nm spectral resolution. Albedo was measured using a cosine-weighted optical diffuser at the fiber-end, mounted to a tripod for consistency of measurement location and angle. Reported albedo for this study is the ratio of the average of ten down-looking (upwelling radiation) measurements to the average of ten up-looking (downwelling radiation) measurements. We limit the reported albedo to the wavelength range of 350-1800 nm, due to noise being introduced further in the NIR as well as the fact that aerosol absorption in snow mostly affects the UV and visible wavelengths albedo (Skiles et al., 2018; Warren and Wiscombe, 1980), which is the focus of this study. Spectral albedo of the deposited area was measured immediately after the deposition experiment, and spectra of an adjacent, undisturbed snowfield were collected immediately after that.

Snow samples were collected at each deposition area as well as at an upwind area to provide a background for OC and other impurities present in the natural snowpack. First, samples were collected in an area of undisturbed natural snow, and later directly below the center of the deposition area at three depth ranges to gauge the depth at which BrC penetrated into the snowpack: surface (0 cm) to 2.5 cm (referred herein as "L1"), 2.5 to 7.5 cm ("L2"), and 7.5 to 12.5 cm below the surface ("L3") (Fig. 2). Each snow sample was placed into a polyethylene sampling bag (e.g., Whirlpak) and kept frozen in a freezer at -20 °C to prevent melting and minimize scavenging of OC.

2.2 Laboratory measurements

Light-absorbing impurities in snow include BC, mineral dust, and OC compounds; they are commonly quantified with optical methods (Cereceda-Balic et al., 2019; Grenfell et al., 2011; Skiles et al., 2017), X-ray fluorescence (Moosmüller et al., 2012), and chemical oxidation (Godec et al., 1990), respectively. Here, snow samples were processed to quantify total organic carbon (TOC) concentration using a total carbon analyzer and absorbance ~~in the UV and visible wavelength ranges~~using a laboratory spectrophotometer at the Desert Research Institute (DRI) in Reno, NV, USA. For analysis, snow was transferred into 50-mL volume polypropylene vials that were first pre-soaked in 18.2 MΩ ultrapure water (UPW, model Purelab Ultra, Elga Labwater, Paris, France) for over 48-hours and then thrice rinsed using UPW. Snow was allowed to melt at room temperature (~21 °C) to discourage the volatilization of organics (as may occur when melting with a microwave or other accelerated means) and then immediately analyzed for TOC.

TOC was measured using a total carbon analyzer (Sievers 900-Series, SUEZ, San Jose, CA, USA). This instrument measures total carbon (TC) and inorganic carbon (IC) concentrations and infers TOC as the difference of these two (i.e., TOC = TC – IC). The instrument photo-chemically oxidizes organic compounds in a liquid sample through chemical oxidation with ammonium persulfate and reactions with hydroxyl radicals produced through the photolysis of water under UV irradiance. Within the instrument, the sample stream is split in two, where one analysis path (for IC) determines the CO_2 formed through interaction with the ammonium persulfate only, and the other path (for TC) determines the CO_2 produced through both ammonium persulfate and UV-induced oxidation (TC). Then, the CO_2 in the sample stream is measured through a patented conductivity detector. The instrument has a reported lower detection limit of 0.03 ppb TOC and accuracy of ±2% or ±0.5 ppb, whichever is greater. TOC concentrations are reported as volume fractions (ppb) by the instrument but will be reported as OC mass concentrations herein (ppm = mg L^{-1} = g m^{-3}) unless otherwise stated. According to the instrument manufacturer, a 60-μm stainless-steel inline filter is used to restrict very large plant detritus, insects, etc. from passing through the sample inlet line into the instrument. Because surface snow spectral albedo was measured with all particles present before and after the deposition experiments, the snow meltwater samples were not filtered prior to TOC determination and therefore may contain particulate and non-particulate OC, as well as water-soluble and insoluble OC compounds. BrC aerosols generated through similar small-scale smoldering combustion experiments generally do not exceed 1 μm in diameter and have a geometric mean diameter close to 100 nm (Chakrabarty et al., 2016; Sumlin et al., 2017, 2018b) and therefore are included in the measured TOC concentration. Contribution of the Whirlpak bags to measured TOC concentration is nonzero, even for samples that remain frozen (Legrand et al., 2013); however, we expect these discrepancies to mostly subtract out between deposited and natural snow samples. Contamination of the polypropylene vials with TOC was measured to be 0.020 ± 0.001 g m^{-3}, which was subtracted from each of the meltwater TOC measurements. To the best of the authors' knowledge, the Sievers TOC analyzer and its methods to oxidize organic compounds in liquid samples is unable to convert any BC present to CO_2, as BC is chemically inert and resistant to oxidation (Bond and Bergstrom, 2006), especially under the limited exposure that instrument subjects the sample to (~4 minutes).

For this study, we assume that while the natural snowpack contributes to the measured TOC, it is free from influence of the aerosol deposition experiment. We can thus infer the background TOC concentration of both the natural snowpack located at the Tamarack Lake site as well as the direct TOC contribution of the BrC aerosol deposition experiment. That is, the OC added to the snowpack through the deposition experiment $TOC_{BrC,i}$ can be calculated for layer *i* as

$$TOC_{BrC,i} = TOC_{bulk,i} - TOC_{snow,i}, \tag{2}$$

where TOC_{bulk} is the measured TOC from the deposition site samples that include both OC already in the "natural" snowpack and artificially deposited OC and TOC_{snow} is the measured TOC from the natural snowpack. The uncertainties presented in this study for TOC concentrations represent the standard deviation of the mean for all n individual TOC determinations ($13 \leq n \leq 72$) and N replicate meltwater TOC measurements ($2 \leq N \leq 4$).

Light absorbance of the melted snow was measured using a Perkin-Elmer Lambda 1050 UV/Vis spectrophotometer (Waltham, MA, USA). Prior to measurement, the melted snow samples were sonicated for ~20 min. and stirred to ensure the solubility of water-soluble organic compounds (WSOC). However, uniformity of suspended particles cannot be controlled through sonication. Sonicated melt water samples were placed into a 1-cm-path-length cuvette. To avoid potential contaminations between samples, this cuvette was flushed with methanol and rinsed several times with UPW. Measurement scans were performed for each sample over the 200-860 nm wavelength range with 1-nm resolution and with UPW serving as reference. The instrument reports the absorbance A as the power exponent of the Beer-Lambert law, where the transmittance, T, of light through a solution is a ratio of the radiant flux transmitted Φ_e^t to the total radiant flux incident Φ_e^i on the sample. This method assumes there is little contribution of scattering to the overall extinction of light along the path (Bosch et al., 2014). This is appropriate for a suspension or solution where extinction is dominated by absorption caused by dissolved OC and/or by carbonaceous particles smaller than the wavelength of incident light, where extinction due to scattering can be neglected. Transmittance T is related to absorbance A as

$$T = \frac{\Phi_e^t}{\Phi_e^i} = 10^{-A} = e^{-\tau}, \tag{3}$$

and absorbance A is related to optical depth τ by

$$A = \frac{\tau}{\ln 10}. \tag{4}$$

If the optical attenuation is uniform along the path of light, the absorption coefficient β_{abs} is related to the optical depth τ in Eq. (4) as

$$\tau = l \cdot \beta_{abs}, \tag{5}$$

where l is the path length of light through the liquid sample. For absorbance spectra, we consider the bulk matter of melted pure ice grains and the impurities that lie both internal and external to the grain matrix and evaluate them as a bulk sample.

The bulk absorption coefficient β_{abs_bulk} is directly related to the imaginary refractive index κ_{bulk} (Moosmüller et al., 2009) as

$$\beta_{abs\_bulk} = \frac{4 \cdot \pi \cdot \kappa_{bulk}}{\lambda}, \tag{6}$$

so, the estimation of κ_{bulk} follows as

$$\kappa_{bulk} = \frac{\ln(10) \cdot \lambda \cdot A}{4 \cdot \pi \cdot l}. \tag{7}$$

The raw absorbance measurements were baseline-corrected to account for instrumental drift by subtracting the scan average over the 700 – 860 nm wavelength range from individual wavelength absorbance values, similar to the method outlined by Sengupta et al., (2018). ~~This also~~In addition, this reduces ~~the~~some influence of ~~any~~ BC particle light absorption~~ that is fairly independent of wavelength~~ across the UV-vis spectrum, while only minimally affecting BrC particle absorption that is much greater at blue and near-UV wavelengths (Bahadur et al., 2012; Chakrabarty et al., 2010; Kirchstetter and Thatcher, 2012; Lu et al., 2015; Sumlin et al., 2018b). Additionally, each spectrum was smoothed using a running average of 15 spectral data points to reduce high-frequency noise while maintaining low-frequency structure.

Similar to measurements of TOC, we consider the bulk snow-aerosol sample when estimating a value for κ – both for the natural snowpack including its naturally-occurring impurities (κ_{snow}) and for the BrC aerosol deposited snow (κ_{bulk}) – as determined through spectrophotometric absorbance measurements. We can isolate the influence of BrC absorption (via the imaginary refractive index) by incorporating the TOC concentration through a simple volume mixing approximation (Chýlek et al., 1988) as

$$\kappa_{bulk,i} = \kappa_{BrC,i} \cdot VF_{BrC,i} + \kappa_{snow,i} \cdot VF_{snow,i}, \tag{8}$$

where VF_{BrC} and VF_{snow} are the volume fractions of BrC and bulk meltwater TOC, respectively, for each layer i, such that $VF_{BrC} + VF_{snow} = 1$. Because the absorbance measurements are made against UPW, we do not include κ or the volume fraction of UPW in Eq. (8). Therefore, the average imaginary part of the refractive index of BrC, here labelled as $\kappa_{BrC,i}$ for each layer i, can be written as

$$\kappa_{BrC,i} = \frac{\kappa_{bulk,i} - (\kappa_{snow,i} \cdot VF_{snow,i})}{VF_{BrC,i}}. \tag{9}$$

Because two samples of the natural snowpack at Tamarack Lake were collected on the same day, we average values of TOC and absorbance over these two natural snowpack samples for each layer. Henceforth, the natural snowpack will be presented as one set of three depths/layers unless otherwise stated.

3 Results and Discussion

Aerosol deposition experiments were conducted during April 2019. Two separate depositions of BrC aerosols produced from the smoldering combustion of AK peat were performed (Experiment 1 and 2), and the analysis of the snow properties and spectral albedo from the field and laboratory and modeling results are discussed below.

3.1 Field work

Two aerosol depositions were performed on April 24, 2019 (approximately two weeks after the last snowfall) at Tamarack Lake. Snowpack and experimental conditions are presented in Table 1. The average snowpack depth measured on April 24, 2019 at Tamarack Lake was 180 cm. Liquid water was noted in approximately the top one cm of snow, but snow below the surface did not contain liquid water.

The first and second deposition included aerosol generation through smoldering combustion for 40 and 37 minutes, respectively, followed by 10 minutes of residual aerosol deposition. At the start of the experiments, the wet fuel mass was 55 g and 45 g for first and second deposition, respectively. The fuel packing density for both burns was ~0.03 g cm^{-3}, which is considered to be a low fuel-packing density (Sumlin et al., 2018b).

Similar to the BrC aerosol deposition experiment previously discussed by Beres and Moosmüller (2018), deposited BrC aerosol from the smoldering combustion of AK peat appeared yellowish to the eye, indicating a preferential light absorption of blue-violet wavelengths by the aerosol deposited. This strong wavelength dependence was quantified by the measured spectral albedo for each deposition experiment, which is shown in Fig. 3 alongside the measured albedo of the natural snowpack upwind of the deposition. Spectral albedo reduction due to the presence of freshly deposited BrC aerosol increased with decreasing wavelength in the UV-visible wavelength region for both deposition experiments. In the UV, there was a stronger reduction in albedo than in the visible, ~~up to~~of ~0.14 and ~0.21 at 350 nm for Experiment 1 and 2, respectively, relative to the natural snowpack albedo (Fig. 3). At 700 nm however, albedo for each deposition was reduced by less than 0.01, indicating very little BC added to the snowpack through the deposition experiments. Both depositions reduced the spectral albedo in the measured near-infrared (NIR) region as well and we hypothesize that the reduction of spectral albedo in the NIR is not due to the BrC or BC deposited but may be due to the deposition volume enclosing the snowpack and increasing temperature and accelerating metamorphism of the snow grains to larger grain sizes (Davis et al., 1993; Doherty et al., 2013), and thus increasing absorption in that spectral region (Dozier and Painter, 2004; Warren, 1982; Wiscombe and Warren, 1980). At 1030 nm, the albedo was reduced by 0.029 and 0.022 for the first and second experiments, respectively. Using SNICAR (see Sect. 3.2.2 and Flanner et al., 2007) to estimate effective grain radii by fitting modeled and measured albedos at that wavelength resulted in an increase of grain size from ~682 to ~807 µm and ~605 to ~695 µm for the deposition experiments 1 and 2, respectively.

The Sierra Nevada snowpack generally receives sufficient aerosol BC and non-BC absorbing aerosols to affect the energy balance in an appreciable manner (Dang and Hegg, 2014; Hadley et al., 2010; Sterle et al., 2013). The albedo of the natural snowpack measured during this study indicates the presence of absorbing impurities without artificial deposition of BrC: a hypothetical pure snow spectral albedo – holding other input parameters constant, modeled through SNICAR – would have an albedo near 0.98 at 350 nm. However, measured albedos at 350 nm for the natural snowpack are appreciably lower, near ~0.84. Along with BrC already present in the snow, other impurities are likely a mix of mineral dust and BC (Hadley et al., 2010; Sterle et al., 2013); however, it is BrC and mineral dust that are responsible for greater albedo decreases (compared to pure snow) at these lower wavelengths (Laskin et al., 2015; Lu et al., 2015; Moosmüller et al., 2009; Skiles et al., 2017; Warren et al., 2019; Wu et al., 2016). While we can account for the presence of BrC already in the snowpack by assigning the laboratory-measured UV-vis absorption to the measured TOC concentrations of the natural snowpack samples, dust concentrations were not measured and therefore remain unknown. However, they are subtracted out in our quantification of the imaginary part of the deposited BrC refractive index (κ_{BrC}) because we consider only the difference between natural snow with and without artificial deposition of BrC.

3.2 Analysis of snow samples

TOC concentrations in the snowpack before and after deposition are presented in Table 2. Averaging samples collected at both natural snow sampling sites, the L1 TOC concentration was 0.579 ± 0.014 g m^{-3}, for L2 it was 0.436 ± 0.048 g m^{-3}, and for L3 it was 0.425 ± 0.008 g m^{-3}. Our OC concentrations fall within the wide range of values found in literature. For example, our concentrations are similar to those reported by Zhang et al. (2019) (and references therein) for OC in snow. Meinander et al. (2013) found a factor of six larger, where concentrations are heavily influenced through deposition of air pollution from the mining and refining industry. Legrand et al. (2013) showed that values of OC found in pristine areas of the world are one or two orders of magnitude lower than our values. The deposition of BrC aerosol added at least one order of magnitude more TOC than was already in the snowpack and increased TOC concentrations below the surface layer as well. Experiment 2 deposited approximately twice the TOC mass concentration ~~than~~as Experiment 1, which is evidenced not only in the measured TOC concentration for each layer, but also in a stronger decrease of measured UV albedo ~~for Experiment 2 than for Experiment 1, relative to the natural snow (Fig.~~(Fig. 3). Therefore, deposited TOC greatly dominated that already existing in the natural snowpack. While the surface layer (L1) captured a majority of the deposited TOC, the BrC penetrated deeper into the snowpack during the deposition experiments, either through movement or “pumping” of air into the snow (Colbeck, 1997; Harder et al., 1996; Waddington et al., 1996) or through melt-induced movement of soluble material through the snowpack (Meyer and Wania, 2008, and references therein). The latter may be more likely in a natural scenario of dry deposition (Clifton et al., 2008), although under the conditions of artificial deposition in the winter-spring transition period, the result may be due to both pumping and meltwater flush.

After determining κ_{bulk}, we calculated the absorption coefficient β_{abs_bulk} following Eq. (6). The values of β_{abs} for deposited BrC, β_{abs_BrC}, are found by subtracting the natural snowpack meltwater absorption coefficient averaged over the two natural snowpack sampling sites, denoted as β_{abs_snow}, from that of the deposited BrC-snow meltwater, denoted as β_{abs_bulk}; spectra calculated for β_{abs_BrC} and β_{abs_snow} can be found in Fig. 4. Indicative of BrC in water and aerosol, absorption spectra of meltwater from all samples exhibited a general increase in absorption with decreasing wavelength through the visible wavelength range, as well as peaks in the UV. β_{abs_snow} spectra had a local absorption peak near ~258 nm. ~~UV absorption band in presence of an aromatic nuclei (no other functional group specified) can be around 255 nm,~~The UV absorption band in the presence of an aromatic nuclei (no other functional group specified) can be around 255 nm (Pretsch et al., 2000; Samburova et al., 2016), which is very close to our observed β_{abs_snow} values. The presence of aromatic nuclei in the absorption spectra may be attributed to the traffic emissions near the field site or to decomposition of indigenous biogenic compounds in the atmosphere and snow surface. With the deposition of BrC aerosol to the snowpack, the absorption increased significantly in the short-vis and UV wavelength ranges and created a more prominent local absorption maximum at 275 nm ~~for~~, with β_{abs_BrC} spectra ~~demonstrate~~demonstrating a very similar pattern to phenolic compounds, ubiquitous in biomass combustion (Yee et al., 2013), with two peaks around 210 nm and 275 nm and a shoulder around 240nm (Linstrom and Mallard, 2018). The preponderance of phenolic compounds in absorption spectra over other chemical classes make them potential markers for BrC deposition on snow. However, the fate of such phenolic compounds on snow surface, through photochemical processing, for example, is still unknown and should be further investigated.

The calculated absorption coefficients correlate positively with measured TOC mass concentration of these samples. The slope of the linear regression between absorption coefficient (m^{-1}) at wavelength λ and TOC concentrations give the (mass) specific absorbance, B_λ (m^2 g^{-1}), which is an indicator of the contribution by OC to the absorption coefficient in snow and ice (Warren et al., 2019; Zhang et al., 2019). B_λ – sometimes referred to as specific absorbance – at various wavelengths is used to describe other properties, such as aromaticity of the OC (Hansen et al., 2016), drinking water quality (Potter and Wimsatt, 2012), or used to help describe different processes in rivers, lakes, and oceans (Fichot and Benner, 2011; Twardowski et al., 2004; Yacobi et al., 2003). The B_λ spectrum over the wavelength range of 215-815 nm is presented in Fig. 5 along with the calculated correlation coefficient R^2 for each regression using TOC_{BrC} concentrations together with those of the natural snowpack, TOC_{snow}, as one dataset. The absorption is well-explained (R^2 > 0.9) by TOC in the meltwater throughout the UV, but above 353 nm, the confidence in that relationship drops quickly (R^2 < 0.5). If we consider just the natural snow samples, the calculated R^2 doesn't suggest high confidence (R^2 < 0.7 across all wavelengths), likely due the low sample size (n=6). For the natural snow samples, B_λ is nearly equivalent to that of BrC at a wavelength of 258 nm ($B_\lambda \approx 7.3$). B_λ for the natural snowpack is two orders of magnitude greater than values inferred by Warren et al. (2019) for Alaskan sea ice at 400 nm, but closer to that of snow and ice in the northern Tibetan Plateau (Yan et al., 2016).

The absorbance of BrC generally decreases with increasing wavelength, and a power-law relationship is often used to describe this wavelength dependence, as

$$p(\lambda) = c\lambda^{-AE}, \tag{10}$$

where p is the parameter exhibiting the wavelength dependence, c is a constant, *AE* is the Ångström exponent (Ångström, 1929). For absorption, p becomes the absorption coefficient β_{abs}, yielding the absorption Ångström exponent (*AAE*). The *AAE* can be written for two wavelengths as (Moosmüller et al., 2011)

$$\frac{\beta_{abs}(\lambda_1)}{\beta_{abs}(\lambda_2)} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-AAE}, \tag{11}$$

$$AAE_{\lambda_2}^{\lambda_1} = -\frac{\ln(\beta_{abs}(\lambda_2)) - \ln(\beta_{abs}(\lambda_1))}{\ln(\lambda_2) - \ln(\lambda_1)}. \tag{12}$$

Zhang et al. (2019) have compiled a graphic (their Fig. 6) showing AAE_{400}^{330} values for several studies of OC in snow and the atmosphere. Our values for AAE_{400}^{330} (2.68, 1.84, and 1.48 for L1, L2, and L3, respectively) derived from the average natural snowpack β_{abs_snow} are on the lower-end of their spectrum for *AAE*s of snow, close to Zhang et al. (2019) results of OC in snow from the Altai Mountains and to that of Arctic snow (Doherty et al., 2010). AAE_{400}^{330} derived from β_{abs_bulk} in our study has a range from 4.12 to 6.28 (mean = 5.32) for all layers; and, when considering β_{abs_BrC}, the larger AAE_{400}^{330} values are in the range of 4.86 to 9.79 (mean = 7.76). Our values for BrC-only *AAE* over this wavelength range agree well with those presented in Fig. 6 of Zhang et al. (2019) for BrC aerosol in different regions of the world.

3.2.1 Estimation of the imaginary refractive index, κ

Equation (7) was used to estimate the imaginary part of the refractive index, κ, from each absorbance spectra. The volume mixing rule described in Eqs. (8, 9) was used to normalize the calculated κ for each spectrum using the BrC volume fraction measured for each sample to obtain κ_{BrC}, the imaginary part of the BrC refractive index. The value of κ_{BrC} displayed in Fig. 6 represents that for all samples containing deposited BrC from AK peat combustion. The black dotted lines represent the range of retrievals for κ_{BrC} for each snow sample from the deposition experiments, which incorporate the uncertainty of TOC concentrations assigned to each absorption spectrum of BrC.

Comparing the values of κ_{BrC} obtained with this method to those of other selected studies for peat biomass combustion shows general agreement, with differences being greatest at different wavelength regions for different studies. Sumlin et al. (2018b) identified values of κ_{BrC} at four wavelengths (375, 405, 532, and 1047 nm) under varying conditions of combustion for different peat fuels, including Alaskan peat; they identified the fuel packing density as the parameter dominating κ_{BrC}, but found little dependence on the type of peat. Sengupta et al. (2018) estimated the refractive index of BrC from smoldering combustion of peat from Siberia (SBP, unpublished results) and Florida (FP), extracted in water. κ_{BrC} for

SBP decreased with increasing wavelength faster than that from other peat samples in Fig. 6. In the visible part of the spectrum, the values of κ_{BrC} for FP smoke are in agreement with our results for AK peat smoke, while in the UV, the largest qualitative difference is the presence of a peak at 275 nm in our spectrum; this peak is not discernible in the other spectra.

~~3.2.2 Estimating changes in snow albedo and radiative forcing by BrC deposition~~

3.2.2 Utilizing models for comparison with observations

Using a complex index of refraction and particle size distribution as an input to Mie theory, we estimate single-particle optical properties of BrC under the assumption of homogeneous, spherical particles. These properties are inputs characterizing impurities for the SNow, ICe, Aerosol, and Radiation (SNICAR) model (Flanner et al., 2007) to estimate spectral snow albedos over the wavelength range of 0.3 μm to 5.0 μm, enabling us to compare them to measured albedos of the artificially-deposited BrC discussed in Sect. 3.1. We limit the reported albedo comparison to the wavelength range of 350 – 845 nm to match measured spectral albedo using BrC optical properties derived through UV-vis spectroscopy.

Sumlin et al. (2018b) show that for BrC from combustion of AK peat, the real part of the refractive index, n, is insensitive to changes in fuel moisture content, source depth, and geographic origin, and is constrained between 1.5 and 1.7. Because of this and the fact that no information exists regarding n below 375 nm or above 1047 nm for AK peat, we use n=1.6 across the wavelength range of interest and, along with the range of values retrieved for κ_{BrC} in Sect. ~~3.3~~2.2, as input for Mie theory calculations. We also utilize the Sumlin et al. (2018b) particle size distribution that was measured under similar combustion conditions as encountered in our study for AK peat with low fuel packing density and smoldering combustion for 35-40 minutes. This lognormal distribution is described by a geometric mean diameter and standard deviation of 157 nm and 1.7, respectively. Mie theory calculations with our range of κ_{BrC} used as input into the Mie code return a range of single particle properties for the single scattering albedo (SSA), asymmetry parameter (g), and mass extinction coefficient (MEC, m^2 kg^{-1}) (Fig. 7). The average BrC SSA returned increases with increasing λ close to a value of 1.0 near λ = 800 nm. The SSA values in the UV and short visible wavelengths agree with those given by Sumlin et al. (2018b) in the UV and visible wavelength ranges, although they display the largest relative range of returned values from the Mie calculations, spanning 0.72 – 0.86 at 305 nm.

The widely-used SNICAR model (Flanner et al., 2007) uses a two-stream radiative transfer scheme to simulate the optical properties of snow and ice in the presence of different types of impurities under different solar and snowpack conditions. The model user can specify multilayer snowpack properties, including individual layers, their snow densities and effective grain radii, as well as the concentrations of impurities, such as: BC, an absorbing mineral dust in four size ranges, and/or volcanic ash. Impurity concentrations are specified for each individual layer, and impurities are treated as externally mixed with the ice grains. Optical properties (SSA, g, and MEC) for all impurities are described in lookup tables. Here, we incorporate our calculated SSA, g, and MEC for AK peat BrC into SNICAR and assign a BrC concentration in the modeled multi-layer snowpack equal to that measured by TOC analysis.

For this study, the modeled albedo was first matched to measured spectral albedo of the natural snowpack along the UV-visible wavelength range, considering both the observed snowpack properties and solar geometry for the measurement day and time. The average TOC concentration for the natural snow layers (L1, L2, and L3) is input as BrC concentration, and values for BC and mineral dust are added to match the modeled albedo to the measured albedo. Once reasonable agreement ($\Delta\text{albedo} \leq 0.025$) is achieved for the natural snowpack measured and average modeled albedos across the wavelength range, the solar geometry, snowpack properties, and BrC concentration are updated to reflect that of the BrC deposition; results are presented in Fig. ~~7~~8. Using this method, the difference in spectral albedo along the wavelength range in question was less than 0.15 for the deposited BrC for both deposition experiments. The modeled albedo for Experiment 1 generally matched the measured albedo better than Experiment 2, especially when considering the upper and lower bounds of BrC optical properties (shaded regions in figures). However, for Experiment 2, the additional deposited BrC resulted in an averaged modeled spectral albedo significantly lower (up to $\Delta\text{albedo} = 0.063$) than the measured albedo throughout most of the visible and short-visible wavelength ranges. For Experiment 1, the modeled albedo in the NIR for the natural snowpack did not match that of the measured albedo ($\Delta\text{albedo} < 0.025$) without adding more BC and removing the mineral dust concentration nearly completely, which is unrealistic for the Sierra Nevada snow during the winter-spring transition. Additionally, adding more BC would have lowered the albedo across the entire wavelength range of interest, not just in the NIR.

3.2.3 Estimating the radiative forcing by BrC deposition

Because spectral albedo was measured before and after BrC aerosol deposition, we can directly assess the enhanced absorption of solar radiation from the deposited BrC by estimating the increase in instantaneous radiative forcing (RF) due to the presence of light-absorbing impurities as

$$RF = \sum_{\lambda_1}^{\lambda_2} I \Delta a \, \Delta\lambda, \tag{13}$$

where I is the measured surface spectral irradiance and Δa is the difference between the spectral albedo of snow in the presence and absence of impurities between the wavelengths λ_1 and λ_2. For this study, λ_1 = 350 nm (the ~~lowest~~shortest wavelength used) and λ_2 = 1000 nm, where we expect very little change due to the presence of impurities (Painter et al., 2013). Pure snow albedo is ~~estimate~~estimated through SNICAR and incorporating snowpack and sky conditions. Before BrC deposition, the natural snowpack had an instantaneous radiative forcing of 51.7 W m^{-2} and 35.2 W m^{-2} for the two natural snow sites at Tamarack Lake, where the spectral albedo measured included all light-absorbing impurities inherently present. Considering the difference in measured spectral albedo of the natural snowpack and after depositing BrC aerosol from the combustion of AK peat (Fig. 3), the deposition experiment added an additional 14.0 W m^{-2} and 19.8 W m^{-2}, for a total instantaneous RF of 65.7 W m^{-2} and 55.0 W m^{-2} for Experiment 1 and 2, respectively. If we integrate the deposited BrC concentrations for L1, L2, and L3 for each deposition, the result in an average mass-weighted instantaneous RF efficiency of 1.23 (+0.14/-0.11) W m^{-2} per ppm of BrC (or OC) aerosol deposited from the combustion of AK peat.

~~3.2.3 Radiative forcing by impurities as a function of existing conditions~~

The radiative impact of the deposition of light-absorbing impurities in snow is dependent on multiple variables including snow age (i.e., grain size), existing impurity content prior to deposition, and solar/sky conditions (Skiles et al., 2018). The rate at which the albedo is lowered, and thus the RF increases, by increasing total impurity loading decreases as the impurity concentration increases, a direct result of the great differences between the absorption properties of the impurities and that of ice itself in the UV-visible wavelength ranges (Warren and Wiscombe, 1980).

~~For example, we consider consider the rate of the direct albedo effect on RF using a SNICAR modeled snowpack, with a 5-cm thick surface layer and an optically semi-infinite layer below that, both with densities of 200 kg m^{-3} and effective grain radii of 500 µm. In Fig. 9, we demonstrate the reduction of broadband albedo α_{BB} evaluated between 350 nm and 1000 nm as function of concentrations of BC in the surface layer ranging from 0 ppb to an unrealistic amount of 50 ppm As expected, α_{BB} is rapidly reduced from a pure snow value of ~0.90 until the resulting α_{BB} is completely dominated by BC and the albedo approaches reported values of the SSA of freshly generated BC (Bond et al., 2013). If we assume the sky conditions during the deposition experiments in Sect. 3, the RF can be calculated using Eq. (13), where $\Delta\alpha$ is now the idfference between the pure snow spectral albedo and that of the snow with increasing values of BC. The RF dramatically increases with small amounts of BC added to the surface layer of a pristine snowpack, and then begins to saturate as the layer becomes optically thick with BC. Similar to α_{BB}, the rate at which the RF increases with increasing BC loading is reduced until it is reduced to zero. These results are similar for the mineral dust component of SNICAR as well as for the BrC component, using optical properties from this study. That is, we can apply this same concept to the deposition of BrC on pristine snow using SNICAR and the average BrC optical properties from this study. Using the snowpack and irradiance properties from the experiments described in Sect. 3, we simulate the deposition of just BrC aerosol onto a pristine snowpack. Again, as expected, the RF values are greater than those found in the previous section, where the BrC deposited onto pure snow results in RF of 25.8 W m^{-2} and 53.3 W m^{-2} for the first and second experiment, respectively. And, the resulting~~We've calculated the RF efficiency increases from 1.23 (+0.14/-0.11) W m^{-2} per ppm of BrC deposited to 2.68 (+0.27/-0.22) W m^{-2} per ppm of BrC deposited onto a pristine snowpack. ~~While these examples don't examine the resulting effects of the grain-size feedback (lowering albedo due to grain size growth from enhanced metamorphism from light absorption of impurities heating of the snowpack), they provide insight and quantify differences between instantaneous radiative forcing of impurities deposited onto pure snow versus onto older snow with already existing impurities.~~

4 Addressing uncertainty and sources of error

This study provided an estimation of the imaginary refractive index κ of BrC artificially deposited on a snowpack. While the authors have taken care to limit the amount of uncertainty throughout the study, the analyses and calculations used in this study are not without assumptions and sources of error. These are addressed below.

4.1 Deposition experiment, spectral albedo measurement, and snow sample collection

Artificially depositing aerosols onto the snow surface is a proven method to study aerosol-cryosphere interactions (Beres and Moosmüller, 2018; Brandt et al., 2011; Conway et al., 1996; Peltoniemi et al., 2015). For this study, aerosol deposition, spectral albedo measurement, and the collection of snow samples were conducted as quickly as possible while minimizing contamination in an effort to reduce uncertainties discussed below.

One limitation of the deposition apparatus and measuring the hemispherical spectral albedo of the subsequent deposited aerosols is the limited areal extent of deposited material when compared to the viewing angle of the cosine-corrected receptor. Because the deposition area is not infinite, there will be a fraction of reflected light from the surrounding ambient snowpack that will influence the measured spectral albedo; however, this fraction of information is reduced for greater angles. Great care was taken during the experiment to minimize the effect from both instrument shadowing while decreasing the distance between the optical detector and the snow surface. As such, a majority of the reflected signal is from within the deposition area and some information is from the surrounding natural snowpack.

In this experiment, the deposition apparatus increased the snow effective grain radii at the snow surface through localized heating due to covering the snowpack with the deposition volume during ambient conditions of warm temperatures and high solar insolation. While temperature inside the deposition volume was not monitored, this effect was observed by examining the difference in measured NIR (700 – 1300 nm) albedo, where snow reflectance is most sensitive to changes in grain size. Additionally, the presence of liquid water may also have been enhanced due to this local heating of the snow surface, and the induced melt may have contributed to impurities percolating lower into the snowpack.

Another important observation made over several BrC deposition experiments is the loss of BrC absorption over time. While the spectral albedo measurements in our study were made within 5 minutes of aerosol deposition, and likely were not affected significantly by this phenomenon, unpublished data of the hemispherical-conical reflectance factor (HCRF, Schaepman-Strub et al., 2006) of the snow surface after the deposition of BrC aerosols from AK peat and another BrC aerosol-producing surrogate fuel ~~(i.e., incense, Chakrabarty et al. (2013) and Gyawali et al., 2012)~~(i.e., incense, Chakrabarty et al. (2013) and Gyawali et al. (2012)) showed that the UV-reflectance increased over some time after deposition. In one instance, the HCRF at 350 nm increased from 0.69 to 0.83 in 207 minutes, whereas the HCRF of the natural snow nearby did not exhibit any discernible change over the same time period. We hypothesize that there are a couple factors that may have contributed to this increase. First, experiments were conducted during the winter/spring seasonal transition, where warm air temperatures induced melt in the surface layer(s) of the snow (the snowpack was not isothermal). Liquid water present in the snowpack flushes water-soluble OC compounds through the snowpack, which may have occurred in the top layers of the snow. Secondly, high UV solar irradiance may photobleach and heat from absorbed radiation may volatilize chromophoric organic compounds (Bertilsson and Tranvik, 2000; Grannas et al., 2007; Laskin et al., 2015; Sumlin et al., 2017). These absorption-reducing photochemical processes may also apply to BrC aerosol deposited on snow, and the resulting change in snowpack chemistry and optics are not fully understood. Likely, the reason for the reduction in absorption is a combination of above scenarios and further work is needed to quantify these effects.

4.2 TOC concentration measurements

The concentrations of TOC presented in this paper represent those of melted snow and its impurities. However, while care was taken to minimize the contamination of snow samples collected, a small amount of TOC was likely scavenged from the Whirlpak plastic bags used to store and transport snow samples. We have derived an upper limit of this effect by measuring the TOC concentration ~~from~~of UPW shaken in new Whirlpak bags to be 0.493 ± 0.025 g m^{-3}; we consider this to be the upper-limit of contamination from liquid water interacting with the Whirlpak bags and we would expect this value to be much smaller as our samples remained frozen until TOC determination. Additionally for this study, we did not filter the samples prior to TOC determination, as many studies have done using 0.45 or 0.22 μm pore size filters, to determine "dissolved" organic carbon (DOC). Instead, we assume that the total OC measured represents water-soluble and insoluble OC as well as particulate and non-particulate OC. With regard to soluble versus insoluble OC, the manufacturer of the instrument used for this study states that the recovery of insoluble OC in the instrument is greater than 90%. There are no data from the manufacturer regarding particulate OC (POC) versus non-particulate OC measured using a TOC analyzer; however, Potter and Wimsatt, (2012) indirectly indicate that the mean DOC and TOC concentrations measured for seven unfortified source waters showed a nearly one-to-one relationship (TOC = 1.09·DOC; $R^2 = 0.997$) using a similar instrument as the one used in this study. Additionally, the fraction of light-absorbing organic carbon versus non-light-absorbing organic carbon is not known to us for this particular fuel and combustion scenario and, for this study, we do not attempt to resolve it. However, throughout the optical measurements (snow surface albedo and spectrophotometric absorbance of melted snow samples), we consistently measure the same organic material without filtering. This way, we consistently use an "average" absorption of all organic compounds present and isolate those from the deposition experiment to those in the natural snow by using background subtraction.

4.3 Absorbance and determination of κ_{BrC}

Measurement of the UV-vis absorbance is a common practice to determine the optical properties of atmospheric aerosols and impurities in rivers, lakes, oceans, and snow and ice. However, there are some uncertainties with regard to these methods.

While the authors assume that all of the measured absorption from impurities in the melted snow samples is solely due to BrC, it is likely that small fractions of BC (both from the deposition experiment as well as naturally occurring) and light-absorbing mineral dusts are present as well, and mineral dust poses a particular risk in assessing κ_{BrC}. The absorption of atmosphere mineral dusts is typically dominated by iron components (e.g., hematite, goethite), particularly in the UV and at short visible wavelengths (Engelbrecht et al., 2016; Moosmüller et al., 2009; Sokolik and Toon, 1999; Zhang et al., 2015). Skiles et al. (2017) developed a method to determine κ for dusts found in the snow of Colorado mountains in high concentrations, but there has been no similar study performed for impurities in Sierra Nevada snow, where sources are much different. Sterle et al. (2013) found dust mass concentrations in the upper 30 cm of snow in the Eastern Sierra Nevada to be between 1-44 ppm in the 2008-2009 winter season, which is not insignificant. Here, we made no attempt to measure the concentrations of mineral dust, BC, or other inorganic material in Sierra Nevada snow.

In addition, some of the raw, high-spectral-resolution absorbance data had a poor signal-to-noise ratio; we improved this by smoothing the raw absorbance spectra through a moving average smoothing algorithm of 15 data points (corresponding to 15 nm) on either side of the data point in question.

5 Summary

Light-absorbing OC (or BrC) from smoldering biomass combustion present near snow and ice environments induces UV and short-visible light-absorption that has previously been unaccounted for in snow albedo, energy balance, and radiative forcing modeling. Biomass combustion of peat at high latitudes of the Northern hemisphere has particular potential of reducing snow and ice albedo due to the close proximity of snow and ice to BrC-rich emissions from peatland wildfires at high latitudes.

This study provides a first estimation of the spectral signatures of BrC particulate matter, from peat biomass combustion, present in the snowpack. ~~Beres and Moosmüller (2018) have artificially deposited combustion aerosols directly onto and into the snowpack.~~Previously, Beres and Moosmüller (2018) have artificially deposited combustion aerosols directly onto and into the snowpack. They have shown that the deposition of emissions from small-scale smoldering combustion of Alaskan peat is effective in altering the snow surface reflectivity, especially in the UV and short-visible wavelength region. Here, we utilized this same method, together with UV-vis spectroscopy of melted snow samples, TOC concentrations in the snow before and after deposition, and Mie theory-calculated optical properties to estimate the imaginary refractive index of deposited BrC. This method has been shown to generally agree with other studies investigating these optical properties, and, by incorporating a range of derived values into SNICAR, modeled spectral albedos are shown to agree with measured albedos as well, within 5% across the UV-visible wavelength region. The instantaneous radiative forcing of BrC deposited onto the natural snowpack was shown to have a mass-weighted value of 1.23 (+0.14/-0.11) W m^{-2} per ppm of BrC (or combustion OC) deposited, while deposition onto a clean (without other light-absorbing impurities) snowpack would have resulted in a more than twice as large instantaneous radiative forcing of 2.68 (+0.27/-0.22) W m^{-2} per ppm of BrC deposited. These results can further inform the impact of deposited combustion aerosol on snow albedo and radiative forcing. In addition to the RF, BrC deposition can greatly reduce UV actinic flux, thereby reducing photochemistry in the snowpack. Further investigations are necessary to refine this method, as well as to address some uncertainty in the behavior of BrC in snow.

Data Availability: : Data presented and used throughout this study can be accessed through the following data repository: https://doi.org/10.5281/zenodo.3736325.~~Data will be made available upon request by the corresponding author.~~

Author Contributions. NDB and HM designed the experiments and NDB, HM, and DS carried them out. NDB performed measurements and analysis of data. NDB and HM prepared the manuscript with contributions from DS, VS, and AYK.

Competing Interests. The authors declare that they have no conflicts of interest.

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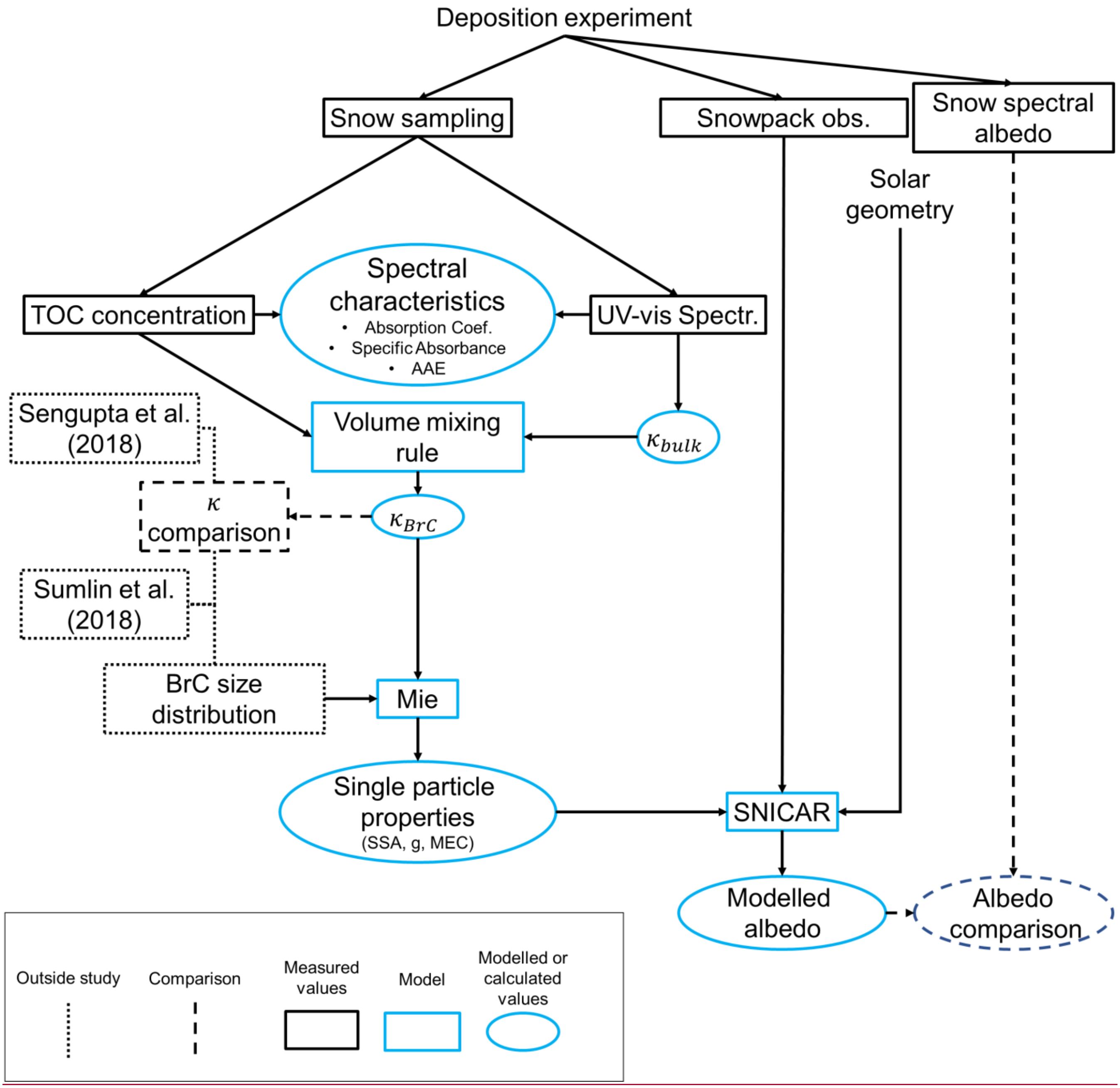


Figure 1. Overview of methods used to derive BrC optical properties and compare measured and modeled albedo. The Sengupta et al. (2018) and Sumlin et al. (2018) studies both provide previously derived values of κ_{BrC} to compare, and Sumlin et al. (2018) provide a size distribution of BrC aerosol under similar combustion conditions to those used in this study.

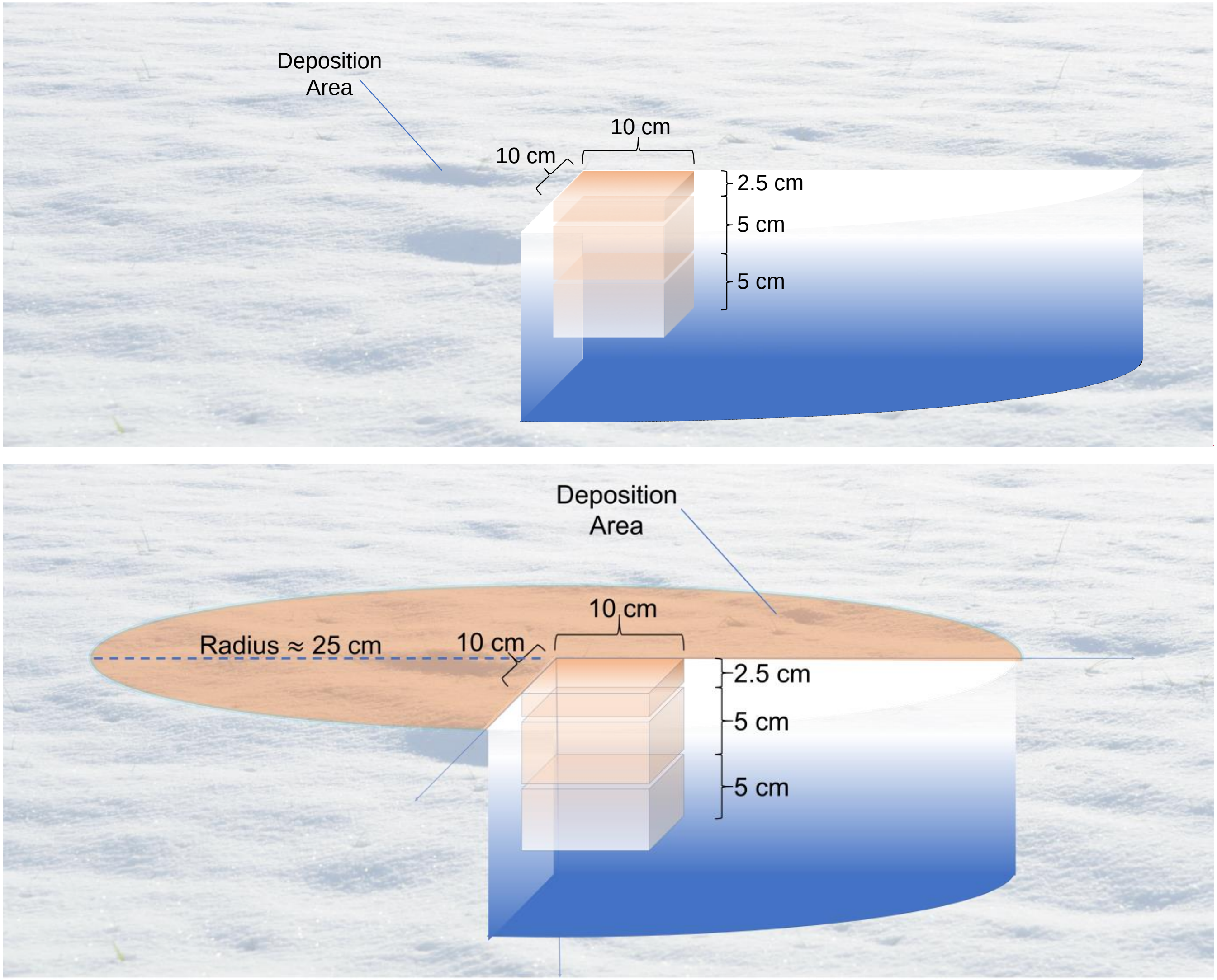
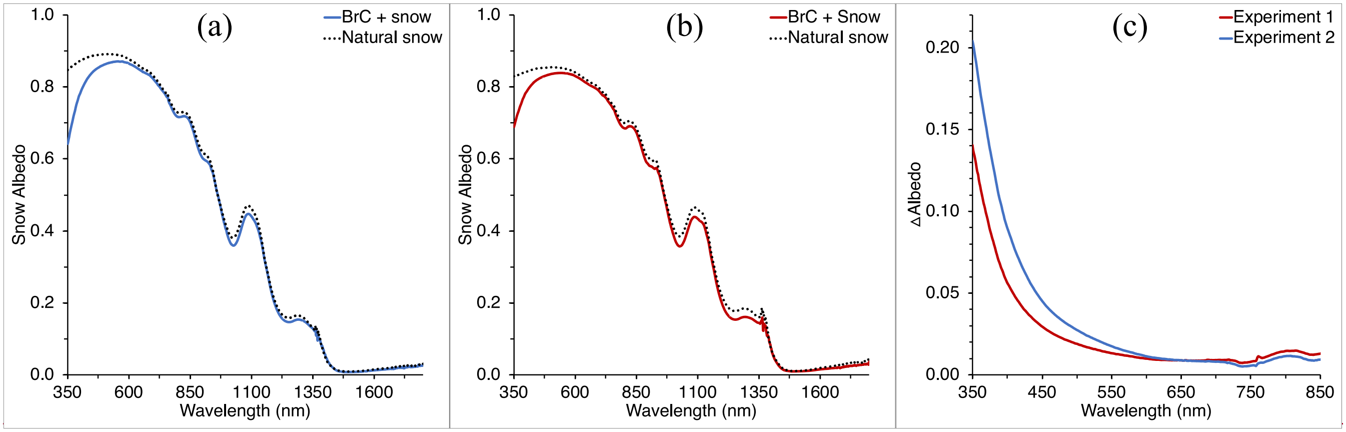


Figure 2. Samples for analysis are collected in three vertical layers. For the deposition ~~site~~area, as depicted here, samples are taken from the center of the area. Samples to represent the natural snowpack are collected similarly upwind of the deposition to minimize contamination. Note: lengths are not to scale.



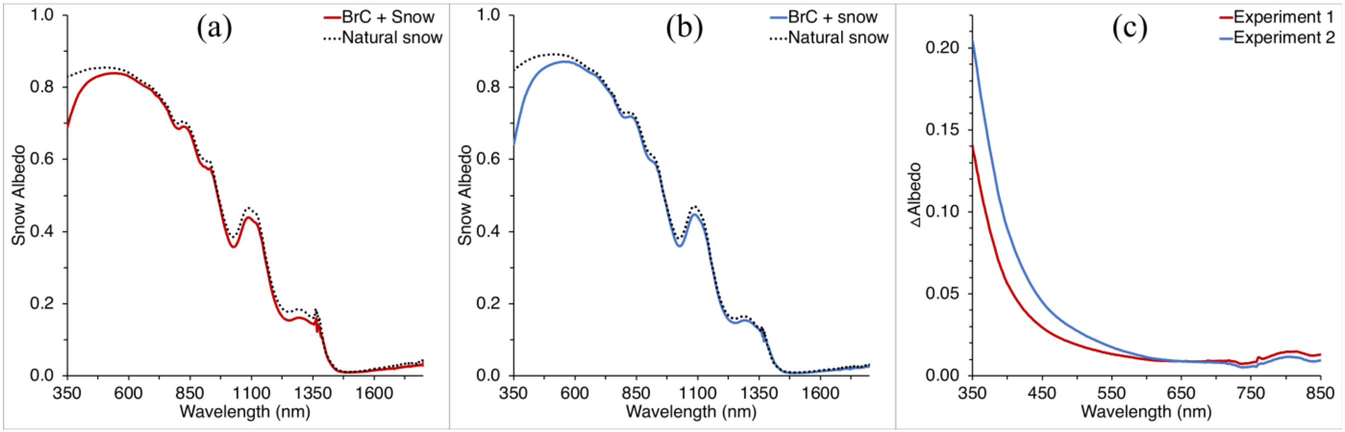


Figure 3. Panels (**a**) and (**b**) show the measured spectral snow albedo of the deposition area for Experiment 1 and Experiment 2, respectively, along with an adjacent area of natural snow. Panel (**c**) indicates the difference between the spectral albedo of the snow with and without deposited BrC in the UV-vis wavelength range, indicating a strong wavelength dependence of absorption by impurities deposited on the snow.

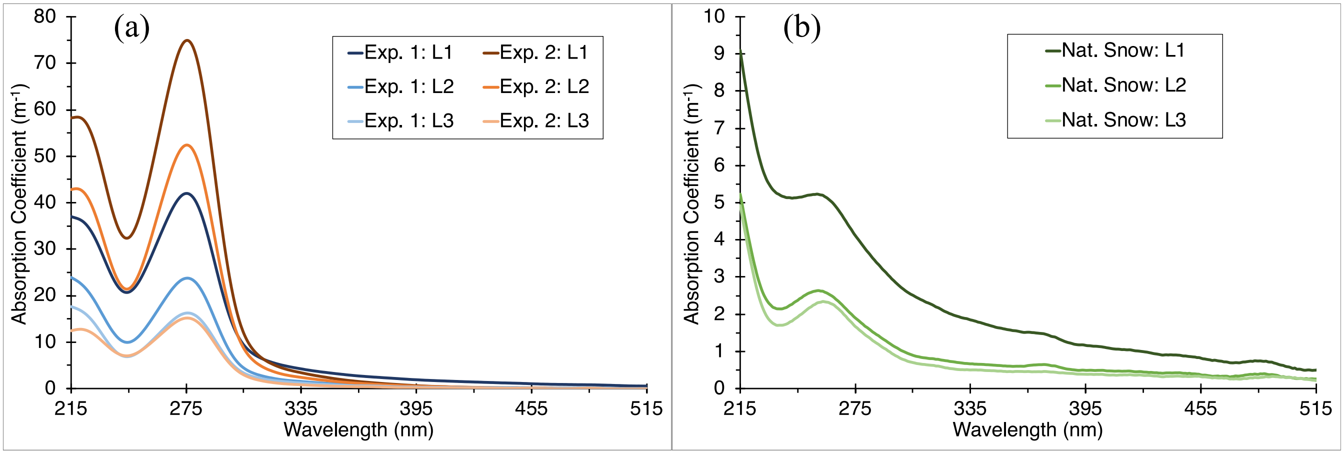


Figure 4. Absorption coefficient of snow meltwater for layers L1, L2, and L3 for deposited BrC (Panel **a**) and for background impurities occurring in the natural snowpack (Panel **b**). “Exp. 1” and “Exp. 2” refer to the first and second BrC deposition experiments, respectively. The spectra of the natural snow (Panel **b**) have been averaged for two locations at the experiment site.

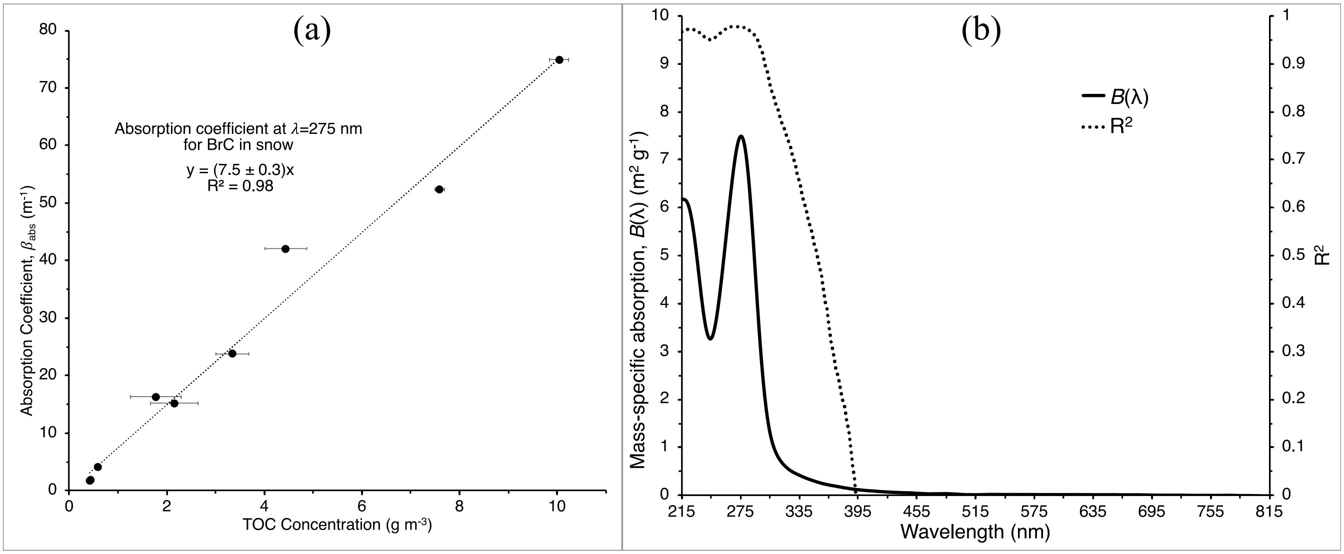


Figure 5. Panel (**a**): Absorption coefficient at 275 nm as function of total organic carbon (TOC) concentration. The fitted linear regression slope gives the mass-specific absorption $\boldsymbol{B}_{\boldsymbol{\lambda}}$ = 7.5 ± 0.3 m^2 g^{-1} with a correlation coefficient of R^2 = 0.98. Panel (**b**): Mass-specific absorption, $B(\lambda)$, and R^2 for each value of $B(\lambda)$ across the wavelength range 215 – 815 nm for BrC deposited experimentally and found in natural snow at Tamarack Lake together~~.~~ at a 1-nm resolution. These panels include absorption coefficients of all meltwater samples characterized in Fig. 2.

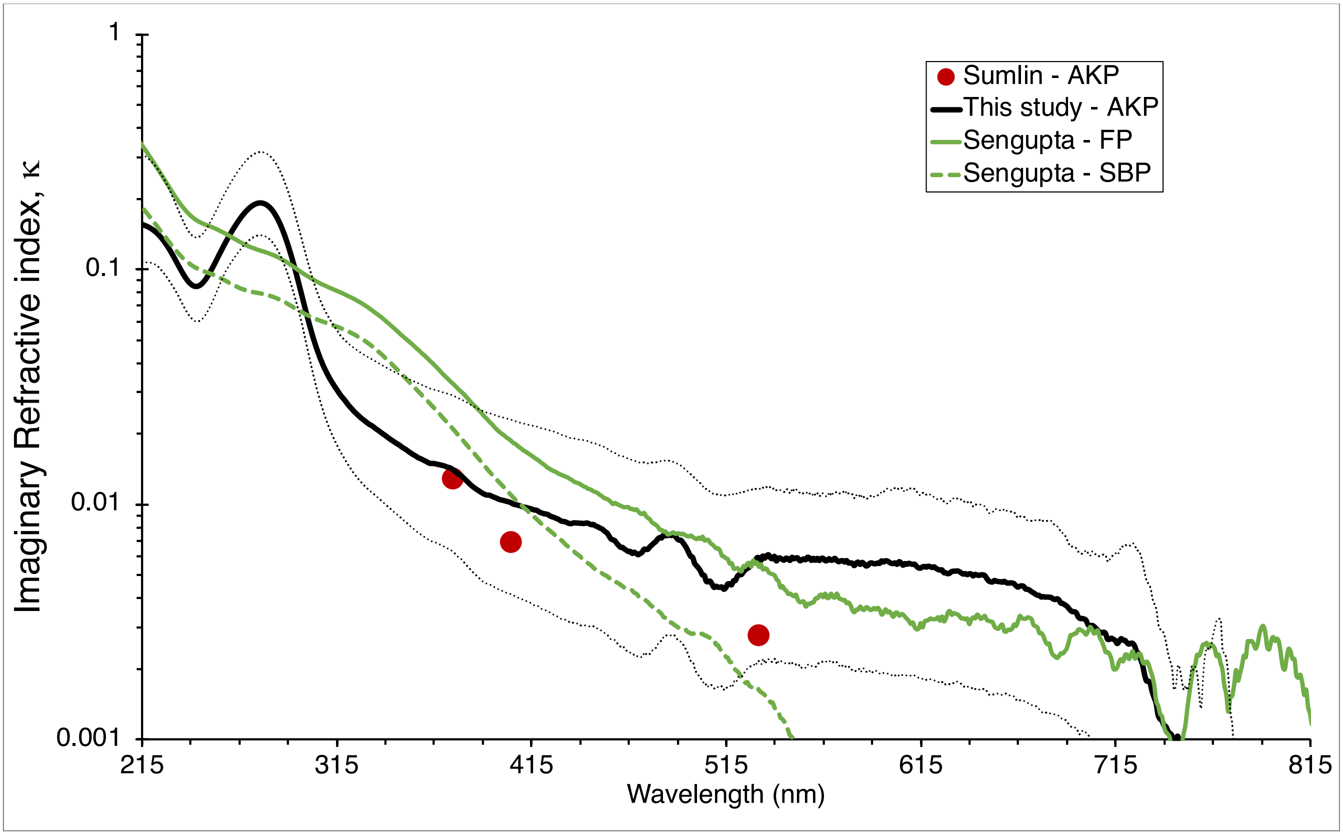


Figure 6. BrC imaginary refractive index, $\boldsymbol{\kappa_{BrC}}$, as estimated in this study and compared to values from selected other studies: Siberian peat (unpublished data) and Florida peat – SBP and FP, respectively - from Sengupta et al. (2018), and Alaskan peat (AKP) from this study and Sumlin et al. (2018b). The thin, dotted black lines indicate the upper and lower bounds of $\boldsymbol{\kappa_{BrC}}$ retrievals for this study.

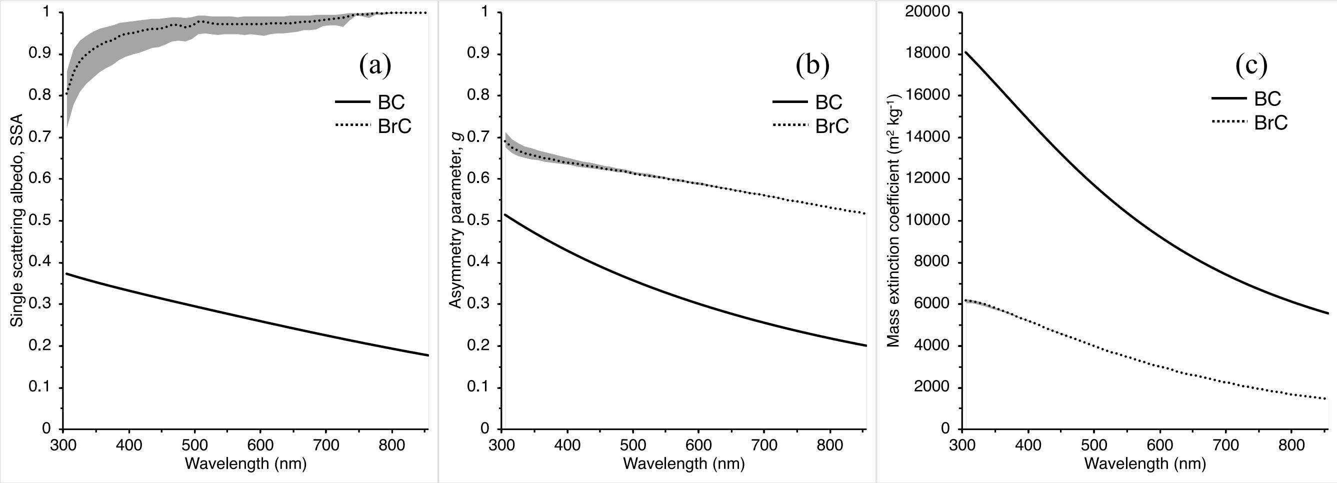


Figure 7. Single scattering albedo (SSA, Panel a), asymmetry parameter (*g*, Panel b), and mass extinction coefficient (MEC, $m^2\ kg^{-1}$, Panel c) of BrC derived using Mie theory. BC optical properties used in the SNICAR model are provided for reference. The grey area represents

the outputs of each parameter for the range of κ_{BrC} retrievals used in the Mie code, where the largest relative discrepancy lies in the SSA. Note: the real refractive index for the calculations presented in this figure is assumed to be $n_{\lambda} = 1.6$ across all wavelengths.

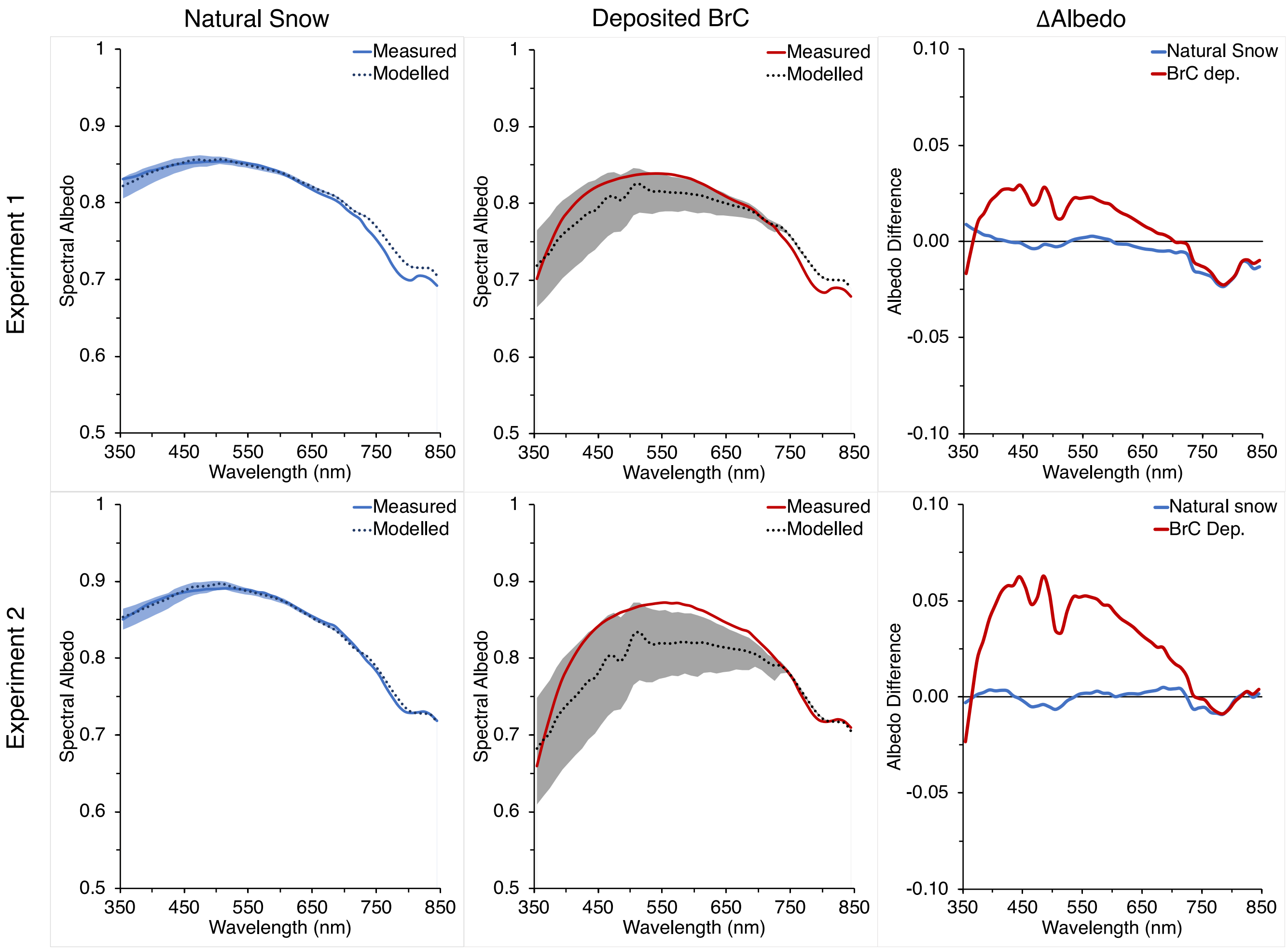
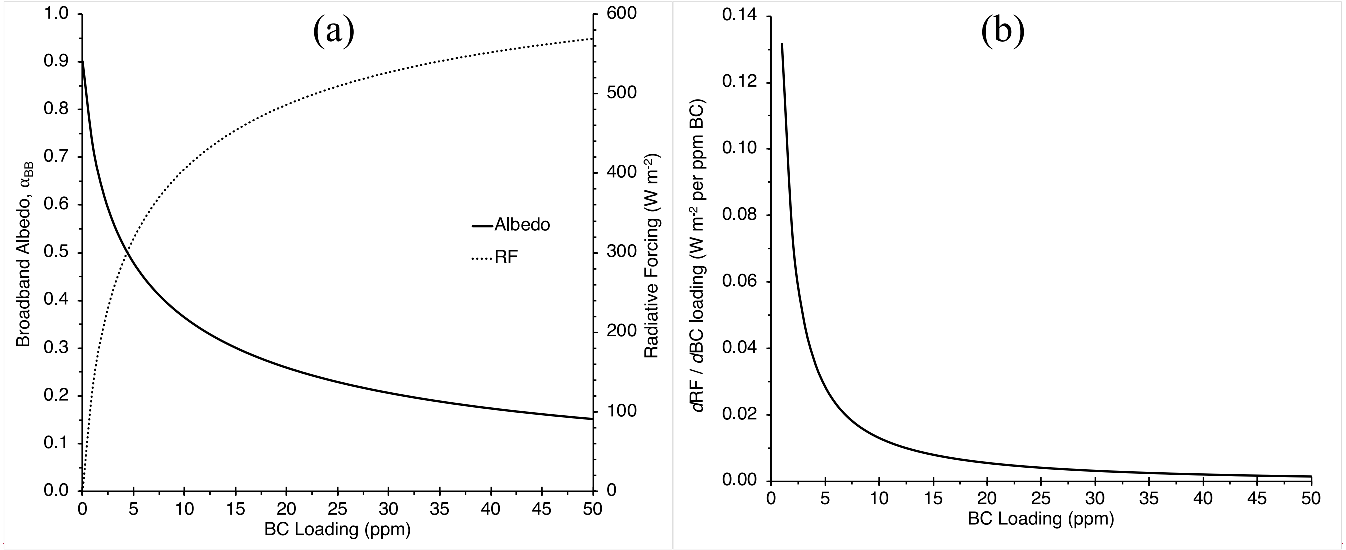


Figure 8. Comparisons of measured and modeled spectral albedos and their differences for Experiment 1 (top row) and Experiment 2 (bottom row). The left column shows modeled and measured albedo of the natural snowpack at Tamarack Lake, NV. The middle column shows the measured albedo compared to SNICAR-calculated albedo by adding concentrations of deposited BrC into the model. The right column represents the difference between the measured albedo and the mean modeled albedo for the natural snow and deposited BrC. The range of modeled albedos from the range of κ_{BrC} retrievals for BrC deposited is represented by shaded regions in the left and middle columns.



~~**Figure 9.** Panel **a**: The relationship between broadband albedo, α_{BB}, and radiative forcing as a function of increasing BC concentration for a hypothetical snowpack. Panel b: Changing radiative forcing efficiency as a function of increasing BC impurity loading in the snow, demonstrating the diminishing increase in radiative forcing with ever increasing values of impurity loading.~~

Table 1. Snowpack observations and spectral albedo properties

Site	Snow Layer (cm below surface)	Density (kg m^{-3})	Surface Eff. Grain Radii[a] (µm)	SZ[b] (°)	Avg. air temp.[c] (°C)
Natural Snow 1	0 - 2.5	480	682	39.70	12.2
	2.5 - 7.5	400			
	7.5 - 12.5	410			
Natural Snow 2	0 - 2.5	480	605	29.28	14.9
	2.5 - 7.5	440			
	7.5 - 12.5	450			
Deposition 1	0 - 2.5	480	807	39.87	12.2
	2.5 - 7.5	400			
	7.5 - 12.5	360			
Deposition 2	0 - 2.5	480	695	29.37	14.9
	2.5 - 7.5	440			
	7.5 - 12.5	350			

[a] Effective grain radii are estimated for the snow surface using SNICAR by matching the measured albedo at the 1030 nm ice absorption feature to a modeled albedo at the same wavelength (average of 1025 and 1035 nm values).

[b] Solar zenith angle at the time of albedo spectra retrieval

[c] Average air temperature during albedo spectra retrieval, as measured from the Mt. Rose SNOTEL site, which lies 0.9 km southeast at 2683 m in altitude.

Table 2. TOC concentrations for the natural snowpack and snow after deposition

Site	Snow Layer (cm below surface)	TOC (g m^{-3}) [a]	TOC Uncertainty (g m^{-3}) [a]
Natural Snow 1	0 - 2.5	0.593	0.029
	2.5 - 7.5	0.388	0.018
	7.5 - 12.5	0.433	0.080
Natural Snow 2	0 - 2.5	0.565	0.010
	2.5 - 7.5	0.485	0.055
	7.5 - 12.5	0.417	0.014
Deposition 1	0 - 2.5	5.018	0.412
	2.5 - 7.5	3.783	0.286
	7.5 - 12.5	2.200	0.519
Deposition 2	0 - 2.5	10.618	0.188
	2.5 - 7.5	8.024	0.051
	7.5 - 12.5	2.582	0.482
BrC[b] - Experiment 1	0 - 2.5	4.425	0.441
	2.5 - 7.5	3.395	0.304
	7.5 - 12.5	1.767	0.599
BrC[b] - Experiment 2	0 - 2.5	10.053	0.198
	2.5 - 7.5	7.540	0.106
	7.5 - 12.5	2.164	0.496

[a] (ppb = µg L^{-1} = mg m^{-3})

[b] BrC TOC is the difference between TOC from the deposition and the natural snow