

Interactive comment on “Deposition of brown carbon onto snow: changes of snow optical and radiative properties” by Nicholas D. Beres et al.

Anonymous Referee #3

Received and published: 25 December 2019

This paper describes and experiment whereby smoldering combustion is used to add light-absorbing aerosol (BC, BrC) to a controlled area of ambient snowpack in the Sierra Nevada mountains. The total organic carbon in three layers and the snowpack albedo are measured for ambient snow in an adjacent area unaffected by the intentional addition of aerosol and in the affected area. Spectral absorption by soluble and insoluble components in each of the three layers in both snowpacks was measured in the lab, using a spectrophotometric measurements of transmission of light through melted snow samples. From this, bulk absorption properties of snow impurities was measured, and spectral mass absorption efficiencies (absorption per mass of total organic carbon, TOC) and imaginary index of refraction was calculated for 215-815nm wavelengths. Assuming absorption was due to spherical particles BrC (where “BrC”

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mass is the TOC mass and the aerosol real index of refraction is fixed at 1.6) Mie code is used to calculate the BrC single scatter albedo, asymmetry parameter and mass extinction coefficient. These are used as inputs to SNICAR code to derive modeled snow albedo, which is compared to the measured spectral albedos. Radiative forcing is calculated in SNICAR and is used to derive forcing per mass of combustion aerosol (as measured by total organic carbon mass) for the ambient snowpack and for a pure snowpack.

The analysis presented is largely robust and appears to be based on a carefully executed experiment. Overall, I think the paper is excellent and contributes useful information on the spectral optical properties of BrC in snow – and also possibly in the atmosphere. There are a few issues I'd like to see addressed.

The paper is well-organized and well-written. Below my main comments I suggest a number of small edits.

MAIN COMMENTS:

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 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

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clean snowpack the forcing per mass TOC deposited would have been $2.68(+0.27/-0.22)$ W/m² 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.

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?)

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?

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?

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.

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).

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.

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

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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!

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.

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 simplify 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 evolve 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

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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.

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).

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

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?

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

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.

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.

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

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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.

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)!

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!)

SUGGESTED SMALL EDITS:

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

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

Pg. 3, line 3: “toward shorter wavelength” → “towards shorter wavelengths”

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

Pg. 4, line 9: spectral albedo of the non-BC components was not measured/derived, but they did report an assumed value of $AAE=5$ for the 450-600nm wavelength band.

Pg. 4, line 26: “presented in Fig. 1 to guide through the relationships” → “presented in Fig. 1 as a guide to the relationships”

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?

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Pg. 9, line 25: "... in Fig. 3 alongside measured" → "... in Fig. 3 alongside the measured"

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)"

Pg. 10, line 23: "snow sampling sites, L1 TOC" → "snow sampling sites, the L1 TOC"

Pg. 10, line 26: "air pollution from mining..." → "air pollution from the mining..."

Pg. 10, line 30: "concentration than Experiment 1" → "concentration as Experiment 1"

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

Pg. 11: "UV absorption band in presence of an aromatic.." → "The UV absorption band in The presence of an aromatic.."

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. ?

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

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

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2019-761>, 2019.

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