The authors appreciate the detailed insights and suggestions by Dr. Dozier. We have considered all comments and have responded below.

## Jeff Dozier, reviewer #1

The manuscript describes customized instruments for experimentally depositing BC, BrC, and mineral dust on snow. The manuscript includes some cursory analysis of the results, based on measurements of the spectral HCRF (hemispherical-conical reflectance factor). However, given the wealth of the data gathered, this analysis could be more robust and help the reader appreciate the importance of the work.

## **Specifically:**

The y-axes of Figures 4-7 are labeled "directional reflectance." The caption should include the geometry, solar zenith angle, observation angle and azimuth with respect to the sun.

- The figures have been updated to reflect this information. Additionally, azimuth has been added to the table of information for each measurement (Table 1). The observation angle of the field spectrometer is nadir-looking for each measurement. This information has been added to the manuscript (p. 4, 1. 33).

Although the "H" in HCRF designates "hemispherical," most of the illumination when skies are clear is in the direction from the sun. The reflectance measurements (in the Supplement) should be sufficient to estimate the imaginary part of the refractive index. Skiles et al. (2016) have published a method to retrieve the imaginary part of the complex index of refraction from measurements of reflectance. It would be interesting to apply their method to these data. Moreover, Skiles' method could be compared to the published measurements for hematite (Scanza et al., 2015).

- While the authors agree that methods outlined in Skiles et al. (2016) are useful, they are outside of the scope of this manuscript, which describes a deposition apparatus and method. However, it is a great example of a potential application of the apparatus described here and we therefore mention it in our manuscript as such (p. 6, l. 35-37).

Knowledge of the bulk optical properties of the absorbing particulates would be needed to model snow reflectance. Also important would be the size distribution, or at least the effective spherical radius, along with the particulate concentration in the upper snowpack. The shape of the spectral reflectance between the blue and red wavelengths depends partly on the size of the contaminating particles.

- We have updated the manuscript to include the particle size distributions for the three aerosols used in this particular study by placing this information in the supplemental data section. However, these size distribution data have been produced in laboratory settings and not in situ. For BrC and BC, the size distributions in the updated manuscript represent measurements made under nearly identical combustion conditions in laboratory settings during previous, related studies. For the hematite used in this study, we used a pure, artificially manufactured powder, for which a size distribution was measured by the manufacturer. For each of the three number size distributions presented, we supply the fractional lognormal distribution and the corresponding geometric mean diameter and geometric standard deviation.

- Future versions of the apparatus described in this manuscript could add ancillary characterization (e.g., size distribution) of the aerosols during deposition, and we have included this information in the updated manuscript (p. 6, l. 7-10).

For the historical context on the experimental approach, the manuscript should address the work of Conway et al. (1996). Specifically, they examined the fate of deposited black carbon and volcanic ash, either remaining near the surface or washed downward during melt depending on size and composition. Sterle et al. (2013) also examined the fate of deposited BC, an important consideration in assessing the effect of absorbing aerosols on hydrology and climate.

- The work presented in the manuscript represents depositions of aerosol that are in the topmost layer(s) of snow; an investigation to whether the aerosol migrate further into the snowpack is not included here. For each of the three aerosols used in the study, a new study site was chosen upwind of the last, as to not contaminate reflection measurements. Aerosols found in the topmost layer, as pointed out by the reviewer, may represent 1) aerosols that have been very recently deposited, such as a recent dust storm, or 2) aerosols that remain in the uppermost layer of snow during the ablation season when water soluble species are washed out with snowmelt. We thank the reviewer for pointing out yet another application of this apparatus and have added this to our section on potential applications in our manuscript (p. 6, 1. 20-27).

## A few nits:

- "Dozier" not "Dozer" page 1, line 23.
  - Fixed. Our apologies, Dr. Dozier.
- "Sierra Nevada" means snowy mountain range, so eliminate "Mountains" on page 2, line 43.
  - o Fixed.
- How about "Inside the" instead of "Inside of the" on page 3, line 9?
  - o Fixed.
- In the Acknowledgments, you should include Ned Bair, who helped you a lot.
  - Acknowledged.

The authors appreciate the critical but helpful comments of reviewer #2. He or she has pointed to a few key missing points to be made about the performance of deposition apparatus. However, the authors feel that after making some simple clarifications in the manuscript described below, we would encourage the reviewer to reconsider their judgement of "premature" and that this work can spark inspiration and innovation for a very valuable experimental method of artificially depositing aerosol onto a snow surface to study aerosol-snow interactions for future researchers. We have outlined our responses in-line below and have updated the manuscript to reflect these changes.

### Anonymous reviewer #2

The aim of the present manuscript is to describe a portable apparatus for the generation and deposition on snow of solid aerosol. The presentation of the method and results is logic, well organized and easy to read. Nevertheless, the apparatus is still under development and potentially affected by some biases. Moreover, the scarce amount of data limits the judgment of the apparatus performances: absolute concentration of deposited aerosol, temperature enhancement of air and snow in the deposition chamber. The discussion on the change of snow optical properties is very basic and based on the simple inverse proportionality between impurities presence and snow reflectance. Without knowing the amount of deposited aerosol, the overall meaning of such results is very limited. However, the topic is of interest for the "snow community" and matches the scientific domain of AMT, the lack of investigation of the above-mentioned issues makes me judge the submission of the manuscript as premature. I thus do not recommend the publication of the present work in its current status, and I encourage the authors to perform additional measurements and resubmit their results. The comments listed below might help the authors to improve their work.

### 1) Amount of deposited material

As an apparatus for deposition, the range of aerosol in snow concentration that can be achieved should be known, this was unfortunately not quantified. Up to the reviewer, this lack affects the entire manuscript, limiting the assessment of deposition homogeneity and the understanding of radiative snow properties. First, the visual assessment of deposition is not exactly robust. From Figure 4 it is evident that there is a remarkable pattern of impurities dispersion, within the same experiment and among the different aerosol types. Second, in order to study radiative impact or potential migration of BC/BrC/dust, the operator should know the initial concentration of impurities. Here, without such information is extremely hard to contextualize the results shown in Figure 5, 6 and 7. The authors are encouraged to collect the snow and quantify the presence of particles across the deposition areas by nebulizing the snow with a pneumatic nebulizer (Lim et al., 2014) and by measuring the absorption coefficient (Ajtai et al., 2010) or refractory black carbon concentration (Katich et al., 2017). Preferably, the concentration of BC or absorption should be quantified before and after deposition.

- To the reviewer's first point, Figure 4 represents the inhomogeneity of previous implementations of the apparatus (left and middle panel) to the current one (right panel). The caption of the image and the manuscript did not reflect this, and they have been updated appropriately.
- While the deposition uniformity is not quantified for this particular study, the authors point out that the current apparatus provides a sufficient uniformity for meaningful directional reflectance measurements of the deposited area and that of unaffected snow. While it may be

desirable for some applications to quantify the three-dimensional distribution of the deposited impurities in the snow, this is beyond the scope of our manuscript. It's important to note that other users may have very different uses for this experimental setup; the updated manuscript suggests some additional examples (Section 4). The present manuscript simply describes novel approaches of depositing aerosols onto the snow surface and thereby allowing for characterization of snow properties modified by such deposition. While, we use snow reflectance modified by the deposition of absorbing aerosols as an example, we have in no way attempted to characterize all properties or consequences of aerosol depositions.

- Previous publications on artificially depositing or "doping" snow with light-absorbing impurities have shown that this is sometimes difficult, may be ineffective, and may require an overbearing amount of equipment. Here, we describe a simple apparatus for the deposition of both sub-micron combustion aerosols and super-micron mineral dust aerosols. A future study (Beres et al., in prep.) addresses the need to connect the albedo reduction with the mass of aerosol deposited.

## 2) Vertical penetration

While the vertical distribution of the impurities affects the overall light absorption through the snowpack (Tuzet et al., 2017), melting might change the vertical distribution of BC particles (Doherty et al., 2013). It is thus of extreme importance to know the exact location of the impurities layer. In the here presented setup, the particles are transported from the generator to the deposition chamber by an air stream. The authors should verify if the air flow pushes the particles within the snowpack and to which depth the penetration occurs.

- The airflow is assumed to push aerosol onto and into the snowpack as there is no additional outlet or escape for airflow inside the deposition chamber present. This design may partly reproduce the phenomenon of air or wind pumping in which there is an exchange of air within and above the snowpack. Visually, we have examined a vertical profile of the aerosol deposited snow and found that the absorbing impurities are located nearly exclusively in the top 2 cm of the snowpack, where further uncertainties of their vertical location (i.e., within the top 2 cm) have very little influence on snowpack optics.
- In the current manuscript, the verification of deposition by way of directional reflection measurements occurs immediately after the deposition; that is, the fate of the aerosol after deposition (whether any loss is due to melt scavenging, photochemical reactions, or other processes) is not considered. The authors (and others) are working to compile a manuscript that examines the fate of BrC aerosol deposited with the present apparatus and the depth at which they find deposited organic carbon, both before and after the experiment (Beres et al., 2019, in prep.).

## 3) Temperature artifacts in the deposition chamber

The authors mentioned that the temperature in the chamber might increase during the deposition process and might modify the size and optical properties of the snow grains. However, this potential bias was not quantified. I suggest the authors repeating the deposition experiments without aerosol generation and simultaneously monitoring the air and snow temperature inside and outside the deposition chamber. Ideally, the assessment should be conducted under different environmental conditions: cloudy-sunny, cold-warm temperature. This method will provide an indication of the temperature increase inside the chamber. The subsequent and potential change in the properties of the snow such as liquid water content, density, specific surface area, and reflectance should be quantified. Beside the

warming caused by the "greenhouse" effect of the chamber, I imagine that the exhaust of the combustion might contribute to the temperature enhancement. Although the long coil line (Figure 3) and the cold ambient temperatures might mitigate the heat transport, the potential warming effect should be assessed.

We agree that temperature in the chamber is affected by outside air temperature, incoming solar radiation, outgoing thermal infrared radiation, and wind speed. However, for sufficiently short deposition periods, these effects can be neglected, but should be kept in mind for both experimental design and data analysis. A thorough quantification of these effects should be interesting but, given the extensive matrix of parameters, is outside the scope of our manuscript. However, one way to gauge the change in effective grain size is to employ the methods outlined by Nolin and Dozier (2000) in which they examine the 1.03 µm absorption feature present in snow reflectance spectra. The authors have performed this calculation and find that – for this particular set of depositions – there is no consistent increase in grain size when comparing the measured directional reflectance of the natural snowpack to that of the snow infused with absorbing aerosol. However, in response to the reviewer's comment, the authors have expanded the discussion portion of Section 3.3 "Challenges and further development" to include the reviewer's concerns and how they might be mitigated.

## 4) Combustion chamber

The in-situ generation of combustion generated particles is definitely interesting but of complex deployment, especially in extreme cold conditions (here tested at air temperature above 5°C) and might contaminate the surrounding snow (Figure 5 shows that deposition is not limited to the area below the deposition chamber). Moreover, the variety of fuels, the combustion efficiency (function of relative humidity, temperature, and altitude) limits the reproducibility of the experiments. The suspension of dry black carbon powder, similar to dust, might reduce the risk of contamination, increase the reproducibility of the experiments and reduce the weight of the entire apparatus. Did the author consider this option?

The reviewer has noted some limitations of the deployment of this apparatus for in-situ deposition of combustion aerosol. The manuscript describes deposition of emissions of black and brown carbon produced on-site and this is part of the novelty of this device, which wasn't emphasized enough in the original manuscript. The authors are not aware of any similar methods to produce the albedo-reducing effect of combustion aerosols found in the atmosphere. However, there have been many attempts to reproduce this effect by using standins such as Carbon Black or other dry BC-like powders. However, combustion aerosols are typically ~100 nm in diameter, and of fractal-like nature (Chakrabarty et al., 2006); such aerosols cannot easily be reproduced by de-agglomeration of powders due to the large adhesion forces found for sub-micron particles.

### **SPECIFIC COMMENTS**

P3L22: the airflow exiting the combustion chamber is of approximately 5 L, where does the air exit the combustion chamber? At the bottom through the snow? Wouldn't this contaminate the surrounding snow? Did the authors ever consider the installation of an exhaust line with a total filter?

- The apparatus in its current form is designed to function in the very way that the reviewer is concerned about: by pumping aerosol *into* the snowpack. Indeed, the snowpack acts to filter

particulate matter (and other atmospheric constituents not discussed in this manuscript) due to air or wind pumping, a gas-exchange phenomenon between air in the snowpack and that of the atmosphere above (Kuhn, 2001). For our particular albedo-reducing verification (via directional reflectance measurements), contamination of the surrounding snow would only be an issue if the contamination was to the snow that we use as reference for "natural snow". The authors have taken care to not contaminate this snow by performing reference measurements more than 1 m away from deposition sites. For the current design of the apparatus, the installation of an exhaust line would deviate from the intended purpose of aerosol penetration into the snowpack.

## P3L30: do the authors know how much dust is actually transported to the deposition chamber?

- No, the authors do not currently have a method to access the transport efficiency from the entrainment chamber to the snowpack when depositing mineral dust. For this study of the apparatus, the application of darkening the snow surface to better understand the impacts on snow surface albedo reduction due to absorbing aerosols was qualitative, and characterization of the transport efficiency for entrained material may be of future interest.

## P3L41: would a fan (moved by the airflow or a portable battery) enhance the dispersion of the particles?

- A previous implementation of the apparatus described in the manuscript housed a single 12V DC fan at the approximate centroid of the deposition chamber, powered by portable battery. As the reviewer suggested, the authors hypothesized that a fan would help facilitate a uniform dispersion of particles throughout the deposition area. On the contrary, a fan caused the deposition to be much *less* uniform. In fact, the two leftmost panels of Figure 4 represent hematite deposition *with* a fan in use. It was only after removing the fan that hematite deposition became very uniform (far right panel of Figure 4 and Figure 7, left panel). The authors updated the manuscript to indicate the efficacy of previous iterations of development of the apparatus and the use of a fan in the production of depositions in Figure 4.

# F1-2: the schematics are basic, technical details should be added: airflow intake and output, size of combustion and deposition chambers, interested snow area.

- The authors thank the reviewer for this valuable suggestion. Updated figures have been provided in the updated manuscript to include this information.

### **References**

- Beres, N. D., Sengupta, D., Samburova, V., Painter, T. H., and Moosmüller, H.: Brown Carbon on snow: reduction of spectral albedo and its implications, Atmos. Chem. Phys., manuscript in preparation, 2019.
- Chakrabarty, R. K., Moosmüller, H., Garro, M. A., Arnott, W. P., Walker, J., Susott, R. A., Babbitt, R. E., Wold, C. E., Lincoln, E. N. and Hao, W. M.: Emissions from the laboratory combustion of widland fuels: Particle morphology and size, J. Geophys. Res. Atmos., 111(7), 1–16, doi:10.1029/2005JD006659, 2006.

- Nolin, A. W. and Dozier, J.: A Hyperspectral Method for Remotely Sensing the Grain Size of Snow, Remote Sens. Environ., 74(2), 207–216, doi:10.1016/S0034-4257(00)00111-5, 2000.
- Kuhn, M.: The nutrient cycle through snow and ice, a review, Aquat. Sci., 63(2), 150–167, doi:10.1007/PL00001348, 2001.

## List of all relevant changes:

- Briefly addressed the manuscript by Conway et al. (1996), as recommended by referee J. Dozier (p. 2, l. 36-39)
- Corrected which winter season each experiment took place during (p. 2, 1. 46-49)
- Corrected section and subsections numbering (Sections 2-3, p. 3 and 4)
- Clarified that the apparatus described does not have a dedicated exhaust outlet and indeed pumps aerosol into the snowpack, as questioned by anonymous referee #2 (p. 3 l. 43 p. 4. l. 2)
- Addressed the implementation of a fan inside the deposition chamber, as questioned by anonymous referee #2; Clarified use of a fan in a previous implementation of the apparatus and two images within Figure 4 show depositions with a fan (p. 4, 1, 3-8)
- Added a sentence referencing aerosol size distributions in supplementary material (p. 4, l. 12-13)
- Clarified observation geometry (nadir) for directional reflectance measurements (p. 4, l. 33)
- Expanded information regarding "Challenges and further development" (Section 3.3) to include limitations brought up by anonymous reviewer #2
- Expanded Section 4 regarding further discussion of possible future implementations of the deposition apparatus and the greater importance of using and further-developing this deposition apparatus, as recommended by both referees
- Added a sentence referencing a manuscript in preparation by the authors and others (Beres et al., 2019) (p. 6, l. 15-17) that addresses some of limitations of applications of the device in the present manuscript, as questioned by anonymous referee #2
- Clarified the data availability statement to include information about the aerosol size distribution in the supplemental data
- Acknowledged Edward Bair, as recommended by referee J. Dozier
- Included additional references to support the work in the manuscript:
  - Beres et al., (2019, manuscript in prep.)
  - Flannigan et al. (2009)
  - Mahowald et al. (2010)
  - Nolin and Dozier (2000)
  - Oris et al. (2014)
  - Skiles et al. (2018)
- In Table 1, clarified solar geometry information, including
  - Adding the solar azimuth during each measurement
  - Correcting solar zenith angle information due to miscalculation
- Updated Figure 1 as recommended by anonymous referee #2:
  - Added annotations to better describe airflow and chamber dimensions
  - Rewrote caption to better describe figure
- Updated Figure 2 as recommended by anonymous referee #2:
  - Included chamber dimensions and approximate deposition area
- Clarified caption for Figure 4 to better describe each image panel and reason for nonuniformity between depositions
- Updated Figures 5, 6, and 7 to include the solar zenith and azimuth angles, as recommended by referee J. Dozier

• Added additional information in the Supplementary Data to include information regarding the aerosol size distributions of the aerosols used in the manuscript, including a figure and table

## **Apparatus for Dry Deposition of Aerosols on Snow**

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Abstract. Deposition of light absorbing aerosol on snow can drastically change the albedo of the snow surface and the energy balance of the snowpack. To study these important effects experimentally and to compare with theory, it is desirable to have an apparatus for such deposition experiments. Here, we describe a simple apparatus to generate and evenly deposit light

- 10 absorbing aerosols onto a flat snow surface. Aerosols are produced (combustion aerosols) or entrained (mineral dust aerosols) and continuously transported into a deposition chamber placed on the snow surface where they deposit onto and into the snowpack, thereby modifying its surface reflectance and albedo. We demonstrate field operation of this apparatus by generating black and brown carbon combustion aerosols and entraining hematite mineral dust aerosol and depositing them on the snowpack. Changes in spectral snow reflectance is demonstrated qualitatively through pictures of snow surfaces after
  - 15 aerosol deposition and quantitatively by measuring hemispherical-conical reflectance spectra for the deposited areas and for adjacent snowpack in its natural state. Additional potential applications for this apparatus are mentioned and briefly discussed.

#### **1** Introduction

Aerosols in the Earth-atmosphere system play a critical role in radiative forcing and climate change (IPCC, 2013). However, our understanding of how they affect the cryosphere upon deposition onto snow surfaces is still limited (Qian et al., 2015),

20 particularly for aerosols other than black carbon (Skiles et al., 2018). Understanding aerosol-cryosphere interactions is important on several levels, including: (1) the radiative properties of the snowpack modified by deposited aerosols (Warren and Wiscombe, 1980; Warren 1982; Gardner and Sharp, 2010; Bond et al., 2013; Qian et al., 2015), which alter the seasonal timing of snow melt, runoff, and water management (Painter et al., 2010; Dozier, 2011), (2) the relation between deposited nutrients and the snowpack biosphere (Thomas and Duval, 1995; Jones, 1999; Kuhn, 2001; Hodson et al., 2008), and (3)

25 snowpack chemistry and photochemistry, which are influenced by the deposited chemical compounds and their interaction with solar radiation (Grannas et al, 2007). One limitation in our understanding is establishing links between theoretical and experimental results because it is difficult to experimentally characterize these interactions in a controlled manner; generally, aerosol deposition on snow is spatially and temporally very inhomogeneous, and often deposition and its immediate effects are minor. One scarcely-used but effective experimental method is to artificially deposit aerosols of interest directly onto the area many surface.

30 snow surface.

Atmospheric deposition is an important process by which an exchange of nutrients, gasses, and particles takes place between the atmosphere and land and sea surfaces. Many deposition processes are irreversible; for example, once the deposition of particles occurs, the probability of re-entrainment is low. Atmospheric constituents are removed from the atmosphere through dry and wet deposition (Seinfeld and Pandis, 2016). Dry deposition is facilitated by gravitational settling,

35 inertial impaction, and Brownian diffusion processes. Wet deposition, on the other hand, involves the scavenging of gasses and particles by clouds and precipitation through dissolution, CCN activity, and collision processes. <u>Aerosol and gasses can</u> <u>enter into the ice-grain matrix of the snowpack through different means and alter chemical, physical, and radiative properties</u> (Kuhn, 2001; Grannas et al., 2007). For snowpack radiative processes and energy balance, the deposition of light\_absorbing aerosols (i.e., black carbon (BC), brown carbon (BrC), and mineral dust, Moosmüller et al., 2009) is of special interest because

<sup>40</sup> deposition of even minute quantities of strongly light-absorbing aerosols drastically increases the co-albedo of the snow surface in the visible and near-visible spectral regions, where pure snow is "snow-white" with hardly any intrinsic absorption (Warren, 1982).

Carbonaceous aerosols in the atmosphere, including BC and BrC (Chakrabarty et al., 2010; Lack et al., 2014), are dominantly generated by incomplete combustion of fossil and biomass fuels with significant additional generation of secondary organic aerosols through oxidation of volatile precursors in the atmosphere (Bond et al., 2004; Lin et al., 2014). These aerosols are lofted into the atmosphere, where, during transport of a few days to weeks, they undergo secondary processing (Jimenez

- 5 et al., 2009) and eventually are removed from the atmosphere through wet or dry deposition (Bond et al., 2013). BC is a ubiquitous light-absorbing aerosol in the atmosphere that directly affects Earth's radiative budget (Jacobson, 2001; Bond et al., 2013) and the cryosphere (Hegg et al., 2009; Flanner et al., 2009; Hadley and Kirchstetter, 2012; Qian et al., 2015). Recently, BrC has become of interest regarding its role in atmospheric light absorption (Laskin et al., 2015) and even more recently a topic of concern for affecting snow albedo and energy balance (Dang and Hegg, 2014; Doherty et al., 2014; Lin et al., 2015; Lin et al., 2015; Lin et al., 2015; Lin et al., 2015; Lin et al., 2014; Lin et al., 2015; Lin et al., 2014; Lin et al., 2014; Lin et al., 2015; Lin et al.
- 10 al., 2014; Wu et al., 2016). The fraction of BC versus BrC mass emitted by combustion sources depends greatly on a number of factors, including fuel type, fuel moisture content, and packing density (Sumlin et al., 2018), combustion phase (Patterson and McMahon, 1984; Reid et al., 2004; Bond et al., 2004), and other elements of the system (Chen and Moosmüller, 2006). For BC, the imaginary part  $\kappa$  of the refractive index is relatively large ( $\kappa \approx 0.79$ ) and varies little over the visible and near-visible wavelength regions (Bond et al., 2006; Moosmüller et al., 2009). Because the imaginary part of the refractive index is
- 15 so high for BC relative to that of snow, a small amount of BC present in the snowpack significantly increases the co-albedo in those spectral regions (Warren and Wiscombe, 1980; Cereceda-Balic et al., 2018). For BrC, the imaginary part of the refractive index generally increases towards shorter visible and ultra-violet wavelengths (Chakrabarty et al., 2010; Moosmüller et al, 2009; Moosmüller et al., 2011). Combustion systems – typically smoldering wildland fires – that produce a significant amount of BrC will have smoke plumes that are brownish or yellowish in appearance because of this spectral dependence (Andreae
- and Gelencsér, 2006). For this study, the boreal peat fuel used is the same as used by Sumlin et al. (2018), who found a value of  $\kappa = 0.014$  at 375 nm,  $\kappa = 0.003$  at 532 nm, and  $\kappa = 0.002$  at 1047 nm. We are not aware of any published studies that explore the impact of deposition from a BrC dominated emission source on snow or identify the spectral reflectance signature of BrC particles deposited onto snow.
- In addition to the importance of carbonaceous aerosol deposition on snow, mineral dust deposition has been shown to be an important driver of early snowmelt in some mountains (e.g., Painter et al., 2007; Skiles et al., 2012; Painter et al., 2012; Painter et al., 2017). The light absorption of mineral dust is mostly caused by iron oxides, such as hematite (Fe<sub>2</sub>O<sub>3</sub>) (Moosmüller et al., 2012; Zhang et al., 2015; Engelbrecht et al., 2016). Hematite is a mineral dust component with globalscale abundance and has been found to significantly absorb light in the ultra-violet and visible wavelengths - the imaginary refractive index of hematite is larger the shorter visible wavelengths and near UV ( $\kappa \approx 0.90$  at 460 nm) and decreases with
- 30 increasing wavelength (e.g., κ ≈ 0.035 at 2500 nm) (Querry, 1985; Moosmüller et al, 2009; Dubovik et al, 2002) and may have the largest optically absorptive impact of all mineral dust components found in the cryosphere (Hegg et al, 2010). Mineral dust deposition on snow is a major concern for the albedo feedback effects in certain areas of the cryosphere (McConnell et al., 2007; Painter et al., 2007; Skiles et al., 2012; Zhou et al., 2017), and accurate experimental representation of dust depositions is critical to understanding their interaction with snow.
- 35 Previous experiments have utilized aerosol artificial deposition techniques to test non-natural snow surface albedo perturbations, but with varying success. Conway et al., (1996) manually mixed soot and volcanic ash with loose snow in a bucket before being spread over a large plot. These methods were useful in understanding the characteristics and vertical location of aerosol during conditions of melt, but they likely restructured the bulk aerosol into agglomerations larger than what can be seen from normal deposition. Brandt et al. (2011) measured the snow albedo resulting from mixing commercially-
- 40 available soot into tap water and spraying the mixture through a commercially-available snow-making machine over a field of artificial snowpack, produced with the same means. More recently, Peltoniemi et al. (2015) distributed chimney soot, glaciated silt, and volcanic sand onto the snow surface using a "salt shaker" in an attempt to measure the bidirectional reflectance factor of the resulting, contaminated snow. In addition, the snow albedo response to absorbing impurities on snow has been characterized by Singh et al. (2010) "spraying...soil equally on the surface".
- 45 Our work presented here describes a simple apparatus to evenly deposit aerosols in an artificial manner onto a flat snow surface through dry deposition for the study of snow-aerosol interactions. Tests <u>during</u> the <u>snow season of</u> 2015-2016 were conducted at the Cold Regions Research and Engineering Laboratory/UC Santa Barbara Energy Site (CUES, Bair et al, 2015) on Mammoth Mountain, California, USA. <u>During the 2016-2017</u> and <u>2017-2018 snow seasons</u>, <u>experiments were conducted</u> <u>at Tamarack Lake</u> in the Carson Range of the Sierra Nevada in Nevada, USA. Controlled deposition experiments using the
- 50 apparatus for hematite mineral dust entrainment and combustion aerosol production of BC and BrC to modify snow surface reflectance are presented as examples.

#### **2** Description and Operation of Apparatus

The deposition apparatus presented here is composed of two primary components: the aerosol production or entrainment chamber and the deposition chamber. The materials used for the air source and production or entrainment chamber depend on the type of test aerosol generated or entrained and deposited onto the snow surface. The deposition chamber is the same for any type of aerosol used.

#### 2.1 Aerosol Production Chamber (Combustion Aerosols)

A schematic diagram of the aerosol production chamber for generating combustion aerosols is shown in Figure 1 Figure 1. It consists of a flat plywood base with an area of  $\sim 0.5 \text{ m}^2$  and a  $\sim 10 \text{ L}$  near-cylindrical galvanized steel volume fitted with a  $\sim 1$ L volume cone to its top; the cone funnels aerosols towards the chamber outlet. Inside the chamber, a ~6 cm high stage is

- 10 mounted on the plywood base, directly above a perforated inlet that disperses inlet air horizontally under the combustion stage. The bottom rim of the chamber is lined with a rubber seal that, when mounted to the plywood base with clamps, becomes nearairtight. The combustion stage can be adapted to house different combusting configurations to fit the user's needs; generally, it symmetrically distributes inlet air around the stage and serves as platform for fuels to be burned. The air source used in the combustion aerosol configuration has three purposes: (1) to provide the appropriate amount of air for (incomplete) combustion
- to take place, (2) to provide continuous flow to move the combustion aerosols vertically (with assistance from flame buoyancy) 15 through the production chamber towards the outlet, and to create a pressure differential between the production chamber and the deposition chamber that ultimately moves aerosols from the production chamber to the deposition chamber and finally onto and into the snowpack. If the heat from the combustion source is too intense, a small amount of metal tubing arranged in a coil may be necessary to cool the combustion air before being transported through any conducting rubber tubing, as shown 20 in Figure 3Figure 3.

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The air source used for this configuration is a battery-powered, 12-volt air pump that provides inlet air into the entrainment chamber, with a flow rate of ~5.3 LPM at the outlet of the production chamber. This outlet is connected to the deposition chamber mostly via 6 mm OD copper tubing, as rubber or plastic tubing near the production chamber may melt from the combustion heat.

The user of this apparatus can vary the mass of combustion material deposited by limiting the amount of time the fuel is 25 combusting. Here, we purposefully produced heavy depositions for optical inspection and verification of the proper operation of the apparatus.

#### 2.2 Aerosol Entrainment Chamber (Mineral Dust Aerosols)

- For pulverized, dry solids such as mineral dusts, the entrainment chamber consists of a 1 L volume glass Büchner flask (e.g., 30 Jensen, 2006), but used in an opposing manner, providing positive pressure rather than a vacuum. Here, the flask is filled with a generous amount (~90 g) of the dust of interest, changing this amount allows for controlling the dust mass deposited onto the snow in the deposition chamber. Compressed air is introduced into the flask through the large opening at its top, sealed with a rubber stopper. The flask's short, horizontal tube acts as the outlet for entrained aerosols and is connected directly to the deposition chamber via conductive tubing. Dry solids are adequately re-suspended using short, intense bursts of air typically
- 35 between 200 and 300 kPa in pressure from a hand-operated, positive-displacement piston pump, such as a common bike floor pump. A similar setup for the entrainment of mineral dust has previously been used by Moosmüller et al. (2012), Engelbrecht et al. (2016), and Piedra et al. (2018).

#### 2.3 Deposition Chamber

The deposition chamber for all aerosols is a near-cylindrical galvanized steel volume that measures approximately 50 cm in 40 diameter at its midpoint and has a volume of  $\sim 100$  L. This piece of the apparatus is shown in Figure 2Figure 2. There is an inlet at the top of the volume that allows for aerosol flow from the production or entrainment chamber and disperses the aerosols into the volume by splitting the aerosol flow into three separate outflows symmetrical about the center of the chamber, for uniform aerosol deposition onto the snow surface. There is no exhaust or outlet for air entering the deposition chamber besides through the snowpack, and the flowrate of air entering the deposition chamber may vary. The design of the instrument

is to pump aerosol into the snow, and the depth of particle capture by the snowpack depends on a variety of issues, as discussed in Kuhn (2001) and references therein.

Previous implementations of the apparatus included a 90-mm diameter, 12V DC-powered fan that was thought to help facilitate the dispersion of particles within the deposition chamber. This battery-powered fan was mounted in the approximate centroid of the chamber and was tested in several different flow directions. The left and center panels of Figure 4 represent depositions of hematite while using this fan in the deposition chamber to test for increased deposition uniformity. However, the presence of the fan created less-uniform depositions, so the fan was removed. The right panel of Figure 4 shows a deposition of hematite with the most up-to-date configuration of the deposition apparatus, without fan.

#### **3** Characterization of Apparatus and Results

- 10 To demonstrate the effectiveness of this deposition apparatus, it was tested during the spring of the 2016, 2017, and 2018. Deposition of three different aerosol types was demonstrated: BC and BrC in the combustion aerosol production configuration and a dry, sieved mineral dust, hematite (Fe<sub>2</sub>O3), to test the entrainment configuration. <u>Information regarding the size</u> <u>distribution of each tested aerosol can be found in the Supplemental Data accompanying this manuscript</u>. Hematite deposition was tested at the Cold Regions Research and Engineering Laboratory/UC Santa Barbara Energy Site (CUES, Bair et al, 2015)
- 15 on Mammoth Mountain, California (39° 38' 34.8" N, 119° 1' 44.3994" W, 2940 masl) on 11 May 2016. BrC and BC deposition was tested in the Carson Range of the Sierra Nevada Mountains at Tamarack Lake, NV (39° 19' 2.517" N, 119° 54' 19.512" W, 2694 masl) on 02 May 2017 and 24 April 2018, respectively; the lake was frozen and covered by snow deposited on its ice surface, creating a flat, homogeneous natural snowpack that is ideal for testing this apparatus.
- To quantify the effect of aerosol deposition onto a natural snow surface, both subjective and objective measures were used. The uniformity of the aerosol deposition was inspected visually for aerosol types that are visually dark, or optically absorbing, as for the hematite deposition in <u>Figure 4Figure 4</u>. In addition, the hemispherical-conical reflectance factor (HCRF, Schaepman-Strub et al., 2006) was measured with an Analytical Spectral Devices (ASD) FieldSpec3 spectroradiometer to verify that aerosol was indeed altering the surface spectral reflectivity of the snow and to spectrally quantify that effect. Note that the measured HCRF is related to the surface albedo, controlling solar energy input to the snowpack (Schaepman-Strub et
- 25 al., 2006). After deposition of the different aerosol species, HCRF was measured for the deposition area as well as for an adjacent, untreated, natural surface that mirrored the snow properties of the deposition area prior to the experiment. This allowed the characterization and verification of the snow reflectivity reduction due to aerosol deposition. Ten measurements of HCRF were performed and averaged for each deposition area. This averaged value is presented throughout this paper with one standard deviation of the mean.
- Measurement information and environmental conditions are summarized in Table 1. Solar zenith and azimuth angles have been obtained from the date and time of the measurement using the NOAA ESRL Solar Position Calculator (https://www.esrl.noaa.gov/gmd/grad/solcalc/azel.html). The spectrometer optical input is a cone from the instrument's bare optical fiber with full-angle field of view of 25°; all observation angles are nadir within a few degrees. Meteorological information was obtained for the Tamarack Lake site from the nearby NRCS SNOTEL site at Mt Rose (2683 masl,
  - 35 https://wcc.sc.egov.usda.gov/nwcc/site?sitenum=652) and for the CUES site from its meteorological instrumentation (http://snow.ucsb.edu/).

#### 3.1 Example: Combustion Aerosol Deposition

#### 3.1.1 Black Carbon (BC) Deposition

- Generation and deposition of BC aerosol took place during the spring of 2018 at Tamarack Lake. A kerosene lamp was used to produce BC aerosol (Arnott et al., 2000; Arnold et al., 2014) to be deposited onto the snow surface. First, the 12-volt pump was started to begin moving air through the complete apparatus and provide enough air to sustain the combustion; without this forced air, the flame would quickly be extinguished due to lack of oxidant. The kerosene lamp was filled with fuel and lit before being placed onto the combustion stage and the production chamber being sealed with clamps. Then, the deposition chamber was lowered onto the desired snow surface, thereby initiating BC deposition on the snow. For this experiment, the
- 45 kerosene lamp was ignited, and aerosol emissions were pumped into the deposition chamber for 45 minutes. After this time, the flame was extinguished manually, and air was pumped through the apparatus for an additional 15 minutes to facilitate

further deposition of aerosol onto and into the snowpack. The deposition chamber was promptly removed, and spectral reflection properties of the deposited and nearby <u>unsoiled</u> snow surfaces were characterized with HCRF measurements.

This deposition of BC aerosol reduced the HCRF from  $\sim 0.87 \pm 0.003$  to  $\sim 0.58 \pm 0.006$  at 500 nm, an approximate reduction

of ~33% corresponding to a large increase (~factor of 3) of the solar energy input to the snowpack. An image of the deposition
 area is presented in <u>Figure 5</u> along with corresponding HRCF measurements made for the deposition area and for adjacent natural snow. Of the example depositions presented, this deposition was the least uniform.

#### 3.1.2 Brown Carbon (BrC) Deposition

The fuel combusted for BrC aerosol generation consisted of boreal peat samples collected from interior Alaska, USA. Details of this fuel – including its collection and preparation – have been given by Chakrabarty et al. (2016). The optical, physical, and chemical properties of aerosol emissions from combustion of this fuel have been extensively studied to evaluate the impact

- 10 and chemical properties of aerosol emissions from combustion of this fuel have been extensively studied to evaluate the impact of its combustion emissions 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; Sumlin et al., 2018). Prior to combustion, the fuel samples were placed into a round, insulated container to mimic simple, real-world conditions in which there is little lateral heat flux due to largely homogeneous horizontal conditions. The fuel samples were
- 15 burning with nearly exclusively smoldering phase combustion, producing OC-rich biomass burning aerosols. The fuel samples were smoldering for ~30-40 minutes; after this period, we continued to pump air through the apparatus for an additional 15 minutes to facilitate further deposition of aerosol onto and into the snowpack. Following the deposition, the deposition chamber was removed and HCRF measurements were made on the deposited and nearby undisturbed snow surfaces.
- BrC deposited onto the snow surface greatly reduced the measured HCRF, but only at the shorter visible and UV 20 wavelengths. The effect presented here reduced the reflectivity of a springtime snowpack from  $0.92 \pm 0.003$  for the adjacent natural snowpack to  $0.53 \pm 0.02$  at 350 nm, a reduction of ~39% for the BrC deposited snow. The resulting image and spectrum are presented in Figure 6Figure 6. The image of the deposition clearly shows a yellowish appearance, an indication of BrC's preferential absorption of blue wavelengths. This visual impression and the corresponding spectrum (Figure 6Figure 6), confirms the dominance of BrC over BC aerosol within the snowpack, as BC would have reduced snow reflectivity across the

25 visible and near-visible spectrum and resulted in a greyish or blackish appearance on the snow surface.

#### 3.2 Example: Deposition of <u>Re-suspended</u> Mineral Dust - Hematite

Mineral dust in the atmosphere – and that deposited into the cryosphere – has varying optical properties depending on its chemical and mineralogical composition. Here, synthetically-made, pure hematite (Fe<sub>2</sub>O<sub>3</sub>) was used as surrogate for strongly absorbing, red-<u>colored</u> mineral dust with high iron-oxide and hematite content. Depositions of hematite were made on a relatively level, undisturbed snow surface at the CUES research site.

A generous amount – approximately 90 g – of hematite dust was placed into the Büchner flask and was entrained and pumped into the deposition chamber as described in Section 2. The dust was allowed to gravitationally settle onto the snow surface for approximately 30 minutes, at which time the deposition chamber was removed and HCRF measurements were made for the deposited snow surface and the adjacent natural snow. The resulting image and spectrum from this test are shown

35 in <u>Figure 7</u>. The resulting HCRF was reduced by approximately 35% in the 350 – 575<u>-nm</u> wavelength range when compared to that of the nearby natural snowpack.

#### 3.3 Challenges and Further Development

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The apparatus described here is not perfect but a work in progress and will benefit from further development by us and others. Some of the limitations and potential biases are outlined below.

- 40 A substantial but not quantified fraction of the aerosols generated in the production or entrainment chamber is lost during transport, deposited on walls and tubing as evidenced by the surfaces of the apparatus darkening and acquiring a typical smell for BrC depositions. Additionally, the authors made no effort to monitor the mass of deposited material or how deep the aerosol penetrated into the snowpack; instead, they leave this issue for future development. Particle deposition onto the snow is not perfectly homogenous and this homogeneity varies from deposition to deposition. The dominant factor controlling
- 45 homogeneity of the deposit seems to be wind, which causes the deposition to favor the lee side of the deposition area due to the air's ability to permeate and travel through the snowpack (e.g. Waddington et al., 1996). Additionally, this apparatus may

alter the grain size of snow located at the top of the snowpack due to heating of the air inside the deposition <u>chamber</u>. Monitoring the grain size of surrounding natural snow and comparing to that of grains within the deposition area after the experiment can shed light on the induced effects. By conducting this analysis using the methods outlined in Nolin and Dozier (2000), we have concluded that, for this set of experiments, there is no consistent change in grain size. Temperature artifacts

5 <u>in the deposition chamber</u> could partly be mitigated by painting the <u>exterior surface</u> white to better reflect incident solar radiation.

<u>Ensuring</u> identical conditions inside and outside of the deposition chamber would be very challenging. <u>Additional</u> instrumentation within the deposition chamber can help quantify the impact that outside air temperature, incoming solar radiation, outgoing thermal infrared radiation, and wind speed could have on experimentation. Perhaps, the use of an identical deposition chamber, one with and one without introduced aerosols could minimize this problem.

#### 4 Conclusion and Discussion

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The apparatus described here provides a means to generate or entrain and to artificially deposit aerosols evenly onto a snow surface. The apparatus has been proven to efficiently deposit carbonaceous aerosols - BC and BrC - from two combustion sources as well as entrained dry mineral dust onto snow, thereby altering the surface reflectivity of snow. The reduction of

- 15 spectral surface reflectivity was verified by measuring the directional surface reflectance within the area of deposition and comparing it to the reflectance of neighboring natural snow. To the authors' knowledge, this study is the first to deposit primary aerosol from combustion sources in-situ, which provides the user of the apparatus with a novel tool to investigate the impact that these prolific snow impurities have after deposition. This investigation has proven that future applications of this apparatus are numerous.
- 20 The type of aerosol being deposited, the total mass of that aerosol, and the environmental conditions surrounding the deposition area can be adjusted by the users to suit their needs. The methods outlined by Skiles et al. (2016) to retrieve the refractive index of deposited aerosols from directional reflectance measurements can be applied to the artificial deposition methods described here with some additional radiative transfer analysis. Similarly, one can apply this apparatus to the testing of snow radiative transfer codes (e.g., SNICAR, Flanner and Zender, 2005; TARTES, Libois et al., 2013) as to their treatment
- 25 of the influence of impurities deposited onto the snowpack. Beres et al., (2019, in preparation) deposit varying concentrations of BrC onto the snow surface and verify measured total organic carbon concentrations to their albedo-reducing effect in an aerosol-snow coupled radiative transfer model.

The growing importance of understanding the link between radiative forcing by a variety of aerosols found in the atmosphere and their relationship to the change in physical, chemical, and optical properties of snow and ice are a field that

- 30 can be utilized by this apparatus in the future. BrC aerosol have an impact on the cryosphere that is still being understood based on the close proximity of BrC-rich fuel sources and the proclivity of wildfire present in the Boreal forests of the northern latitudes (Flannigan et al., 2009; Oris et al., 2014; Beres et al., 2019, in prep.). Additionally, the increased emissions of dusts across the globe (Mahowald et al., 2010) and their impact on snow radiative forcing can benefit from this device. For example, one could entrain other globally important dusts, such as those found in Engelbrecht et al. (2016), using this apparatus. While
- 35 the first implementation of any new apparatus is imperfect, the usefulness and interest to the cryosphere sciences can benefit from this work.
- *Data Availability*. The authors provide the numerical values of HCRF spectra shown in figures within the manuscript in the supplementary material. <u>Additionally, the authors have provided normalized, laboratory-measured size distributions that correspond to aerosols produced for this study under similar conditions.</u>

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

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<b>Table 1:</b> Overview of deposition and HCRF collection information
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Site	Deposited Aerosol	Altitude	Date of Experiment	SZ*	AZ**	Avg. air temp.***
		(m)		(°)	<u>(°)</u>	(°C)
CUES, CA, USA	Hematite (Fe <sub>2</sub> O <sub>3</sub> )	2940	10 May 2016	<u>21</u> .5	<u>154.3</u>	8.4
Tamarack Lake, NV, USA	Brown carbon (BrC)	2694	02 May 2017	<u>34.6</u>	<u>235.0</u>	10.4
Tamarack Lake, NV, USA	Black Carbon (BC)	2694	24 April 2018	<u>26.7</u>	<u>167.6</u>	11.2

\* SZ: solar zenith angle at time of HCRF data collection

\*\* AZ: solar azimuth angle at time of HCRF data collection

Average temperature during the HCRF data collection period. For the Tamarack Lake site, values are taken from the
 Mt Rose NRCS SNOTEL site, which lies 0.9 km southeast at 2683 m in altitude. For the CUES site, these are taken directly from the CUES meteorological instrumentation.



**Figure 1:** <u>A battery-powered pump provided combustion and transport air into</u> the aerosol <u>production chamber</u> for combustion aerosol (left), which flows to the deposition chamber at a flow rate of ~5.3 LPM. A 1-liter glass Buchner flask was used as the entrainment chamber for dry, pulverized dusts (right), which was provided short, quick bursts of air from a positive-displacement piston pump. The schematics are not drawn to scale.



**Figure 2:** The deposition chamber for all aerosol types consists of a near cylindrical volume with aerosols pumped into the inlet at the top of the chamber from the production or entrainment chamber. <u>The deposited area measures approximately 0.79 m<sup>2</sup></u>.



Figure 3: Use of the apparatus at the Tamarack Lake site for depositing BC aerosol.



**Figure 4:** Images of three hematite depositions allow for visual inspection of the <u>deposition uniformity</u>. The left and center panels represent a previous apparatus design which featured a fan mounted inside the deposition chamber, which created less-uniform depositions. The right panel represents a hematite deposition using the final configuration of the apparatus.



Figure 5: Image and HCRF spectra for a BC deposition and adjacent natural snow in May of 2018 at the Tamarack Lake site. This BC deposition drastically reduced the high natural snow reflectivity in the visible and near-visible spectral regions. <u>Solar zenith: 21.5°; solar azimuth: 154.3°</u>.



**Figure 6:** Image and HCRF spectra for a BrC deposition and adjacent natural snow in May of 2017 at the Tamarack Lake site. This BrC deposition drastically reduced the high natural snow reflectivity in the ultraviolet and short-wavelength visible (< 500 nm) spectral regions. Solar zenith: 34.6°; solar azimuth: 235.0°.



**Figure 7:** Image and HCRF spectra for a hematite deposition and adjacent natural snow in May of 2016 at the CUES research site. This hematite deposition drastically reduced the high natural snow reflectivity in the ultraviolet and short-wavelength visible (< 600 nm) spectral regions. <u>Solar zenith: 26.7°; solar azimuth: 167.6°</u>.