Brown carbon aerosols from burning of boreal peatlands: Microphysical properties, emission factors, and implications for direct radiative forcing

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Abstract

The surface air warming over the Arctic has been almost twice as much as the global average in recent decades. In this region, unprecedented amount of smoldering peat fires have been

identified as a major emission source of climate-warming agents. While much is known about greenhouse gas emissions from these fires, there is a knowledge gap on the nature of particulate emissions and their potential role in atmospheric warming. Here, we show that aerosols emitted from burning of Alaskan and Siberian peatlands are predominantly brown carbon (BrC)–a class of visible light absorbing organic carbon (OC)–with negligible amount of black carbon content. The average fuel-based emission factors for OC aerosols ranged from 3.8 to 16.6 $g.kg^{-1}$. Their mass absorption efficiencies were in the range of 0.2-0.8 m^2g^{-1} at 405 nm (violet) and dropped sharply to 0.03-0.07 m^2g^{-1} at 532 nm (green), characterized by a mean Ångström exponent of ≈9. Electron microscopy images of the particles revealed their morphologies to be either single sphere or agglomerated "tar balls". The shortwave top-of-atmosphere aerosol radiative forcing per unit optical depth under clear-sky condition was estimated as a function of surface albedo. Only over surfaces with albedo greater than 0.6, such as snow cover and low-level clouds, the emitted aerosols could result in a net warming (positive forcing) of the atmosphere.

1 Introduction

Boreal and Arctic ecosystems store large amounts of carbon, between one-fifth and one-third of the planet's terrestrial organic carbon, in peatlands, moss, and litter (Gorham, 1994; Turetsky et al., 2015). Carbon accumulation in this ground-layer biomass has been occurring over hundreds to thousands of years, and plays an important role in regulating the planetary carbon cycle and climate. These ecosystems act as a sink for carbon emissions from natural and human activities (Bonan, 2008). However, during the past several decades, substantial smoldering combustion of this ground-layer biomass has caused positive climate feedback by releasing stored carbon into the atmosphere as greenhouse gases and particulate matter (Oris et al., 2013; Turetsky et al., 2015). These low-temperature fires have contributed to changes in the quantity of seasonal snow

cover, ice and permafrost, and vegetation productivity in the Arctic Tundra, which has seen a rise in surface air temperatures at approximately twice the global rate (Hu et al., 2010; Jorgenson et al., 2001; Lawrence and Slater, 2005; Pearson et al., 2013).

In continental North America Boreal regions, the mean annual burn area has more than doubled in the past several decades (Oris et al., 2013). In Siberia, an average of 4 million hectares of peatlands burned annually between 1975 and 2005, with the frequency of fires doubling since the 1990s (Conard and Ivanova, 1997; Sheng et al., 2004; Stocks et al., 1998). Siberia is home to about 50 percent of world's peatlands; it is anticipated that burning of these peatlands will increase by as much as 100% in the coming years in response to climate change (Bachelet et al., 2005; Balshi et al., 2009; Flannigan et al., 2005). Climate change would result in drying and lowering of the water table in peat lands, which in turn would increase the frequency and intensity of peat fires (Turetsky et al., 2015)

Past studies have estimated that carbon released from boreal forest fires is mostly composed of greenhouse gasses–CO_2, CO, and CH_4 (Oris et al., 2013; Simpson et al., 2011). Past field observations and laboratory studies have also shown burning of peat lands–both tropical and boreal–to emit large quantities of greenhouse gases (Christian et al., 2003; Iinuma et al., 2007; Page et al., 2002; Stockwell et al., 2014; Turetsky et al., 2015). While much is known about gaseous emissions, properties and climatic impacts of particulate matter (or, aerosol) from these fires are poorly quantified. Black carbon (BC) aerosol has been identified as the major light-absorbing and warming agent, influencing direct radiative forcing by as much as 17 ± 30 W/m^2 after a flaming boreal fire (Oris et al., 2013; Randerson et al., 2006). Emitted organic carbon (OC) aerosols from these fires have, until recently, been assumed to be purely scattering in the visible spectrum. Very little is known about the radiative effects of aerosols emitted from

smoldering combustion, which is the more dominant and long-lasting fire phase for boreal peatlands (Eck et al., 2009; Turetsky et al., 2015). Smoldering combustion of peatlands is an important emission source as it may emit up to six times more aerosol mass concentration per unit carbon combusted compared to flaming grassland fires (Page et al., 2004).

The objective of this laboratory study is to address this knowledge gap by reporting the physical, chemical, and spectrally-resolved optical properties of aerosols emitted from the laboratory combustion of peatland samples collected from interior Alaska and western Siberia. The emitted smoke aerosols were analyzed *in situ* for their spectral optical properties using multi-wavelength integrating photoacoustic-nephelometers (IPNs) and size and morphology using a scanning mobility particle analyzer (SMPS) and electron microscopy, respectively. The aerosols were simultaneously collected on quartz-fiber filters for the quantification of mass by gravimetry and carbon mass fractions using a thermal/optical carbon analyzer. With the knowledge of their optical properties, the potential warming impacts of the emitted smoke aerosols on the atmosphere were estimated using a simple forcing efficiency model integrated over the tropospheric solar spectrum.

2 Methods

Experiments were conducted during summer 2014 in the biomass combustion chamber of the Desert Research Institute (Tian et al., 2015). This aluminum chamber measures 1.83 m by 1.83 m by 2.06 m high and facilitates burning of up to 50 g of solid biomass fuels under controlled conditions of temperature, dilution, and relative humidity. For this study, samples of black spruce peatlands, collected from the closed-crown boreal forests of interior Alaska and west Siberia (see details in the Supplement), were burned at two moisture content levels–25 and 50%. Previous studies have reported that peat mass loss upon ignition is highest for moisture content

levels below 100% (Rein et al., 2008). Prior to burning, organic soil samples were analyzed using the Flash EA 1110 analyzer (Thermo Nicolet Corporation, Waltham, USA) (Xu et al., 2011) for their carbon (C), hydrogen (H), nitrogen (N), sulphur (S), and oxygen (O) content. Based on the dynamic flash combustion method, this instrument utilizes two reaction chambers, gas chromatographic column, and thermal conductivity detector to quantify the mass fraction of C, H, N, S, and O. The fuel moisture content of the burned samples was determined by measuring the mass loss after maintaining the sample at a temperature of 90°C overnight. Fuels were prepared for combustion by arranging them in a round "pie" shape in an insulated containers to simulate "real world" conditions in which surrounding unburned peat soils provide insulation near the burn location.

Multiple runs (three per fuel per moisture content) of smoldering combustion of approximately 20 g of Alaskan and Siberian peatland samples were conducted on a continuously weighed flat fuel bed located in the chamber. Each run lasted for about an hour. Aerosol from the smoke-filled chamber was sampled through a $PM_{2.5}$ (particulate matter less than 2.5 μm aerodynamic diameter) inlet and distributed via a manifold to a suite of instruments, namely a sampling unit for collecting particles onto pre-baked 47-mm diameter quartz fiber filters (Whatman, USA), a sampling unit for collecting particles for electron microscopy and analysis (Ted Pella Inc.), a 3-wavelength (405nm, 532nm, and 781 nm) IPN (Droplet Measurements Inc.) and a custom-made single wavelength (870 nm) IPN (Abu-Rahmah et al., 2006; Arnott et al., 1999; Lewis et al., 2008a), a SMPS (TSI Inc.), a non-dispersive infrared CO gas analyzer (Testo Inc.), a NO_x analyzer (2B Technologies Inc.), and a CO_2 gas analyzer (SBA-5; PP Systems Inc.). Conductive tubing was used to transport the particles to the various instruments in order to minimize particle losses.

An IPN consists of a wavelength-specific laser module and a reciprocal integrating nephelometer aligned in an acoustic resonator. The instrument measures particle light absorption coefficient (β_{abs}) using the photoacoustic effect (Arnott et al., 1999), while the reciprocal integrating nephelometer measures the integrated (over ~ 4π) scattering from the sample volume yielding the scattering coefficient (β_{sca}) (Abu-Rahmah et al., 2006). The four wavelength IPNs used in this study facilitated simultaneous measurement, with two-second time resolution, of spectrally-varying β_{abs} and β_{sca} in addition to intensive aerosol optical properties such as single scattering albedo (SSA) and Absorption Ångström Exponent (α). The SMPS was operated with a sheath/aerosol flow ratio of 10:1 (sheath flow = 3 L/min; aerosol flow = 0.3 L/min), yielding a differential mobility analyzer size transmission width of approximately ±10%. The CO and CO_2 gas concentrations were continuously measured and the data were averaged over 5-minute intervals.

For each run, aerosols were collected on 47 mm quartz-fiber filters at 10 l min^{-1} flow rate. Immediately after sampling, filters were stored in a refrigerator and later analyzed for BC and OC mass fractions and concentrations using the IMPROVE-A TOR and TOT analyses method (Chow et al., 2007; Chow et al., 2011) implemented on a DRI Model 2001 thermal/optical carbon analyzers (Atmoslytic, Inc., Calabasas, CA, USA). The fuel-based emission factor (see details in the Supplement), defined as the mass of a compound released per mass of fuel consumed (Chen et al., 2007), of BC and OC corresponding to each sampled filter were determined using the procedure described by Chen *et al.* (Chen et al., 2007). With the knowledge of OC mass concentrations and β_{abs}, the OC mass absorption efficiency (MAE, also referred to as mass absorption cross section) was calculated in order to highlight the mass absorption

contribution by OC, a parameter often ignored in aerosol forcing calculations by climate models (Chung et al., 2012; Gustafsson et al., 2009; Solomon et al., 2007; Stocker et al., 2013).

3 Results and Discussion

The mean carbon (C) dry mass fractions of the Alaskan and Siberian peat samples were estimated at 38.1 ± 1% and 49.6 ± 0.2%, respectively. This carbon mass predominantly converts to CO_2, CO, and carbon aerosol upon combustion, thereby allowing the estimation of fuel-based EFs for BC and OC. Previous studies (Christian et al., 2003; Iinuma et al., 2007) measured slightly higher C mass fractions at 44 - 54.7% and 50.7%, respectively, for peat collected from the Sumatran region of Indonesia and the Neustädter Moor, Germany. One could qualitatively reason that past fire history and depth of sample collection may have caused this spread in C mass fractions values.

Table 1 summarizes the study-averaged, fuel-based EF values of CO_2, CO, BC, and OC emitted from the combustion of two types of peatland samples at 25 and 50% moisture content levels. Inter-sample variability of measured EF values was small, owing mainly to use of standard amount of fuels and the nearly identical, smoldering-dominated fire patterns. The fuels burned with a modified combustion efficiency (MCE)–defined as the amount of carbon released as CO_2 divided by the amount of C released as CO_2 plus CO (Ward et al., 1996)–of MCE $\leq$ 0.7, indicating pure smoldering combustion. The particulate matter mass emissions during all peat burns were completely dominated by OC. Visually the smoke appeared whitish in color with no tinge of blackness (blackness would be indicative of flaming phase). The average OC EFs (per fuel mass) for Alaskan and Siberian peats ranged from 3.8 to 7 g kg^{-1} and 9.2 to 16.6 g kg^{-1}, respectively. This range of values is consistent with values measured for German and Indonesian peat burns, 6- 12.8 g kg^{-1} (Iinuma et al., 2007). The average OC/BC mass ratios ranged between 70 and 85 for combustion of Siberian peat and between 23 and 72 for Alaskan peat. These values

are much higher than the average mass ratios of 14 and 13 for combustion of Indonesian and German peat, respectively. The EF values for BC emitted from combustion of Alaskan peat ranged from 0.09 to 0.16 g kg^{-1}, while those from Siberian peat were 0.09 to 0.23 g kg^{-1}. This range of values is lower than previous findings of 0.04 – 1 g kg^{-1} for BC EFs measured for combustion of Indonesian and German peat. The CO_2 and CO EFs were in the range of 1432 – 1700 g kg^{-1} and 50 – 204 g kg^{-1}, respectively. The observed range is in line with previous estimates of mean CO_2 EF of 1616±180 g/kg and CO EF of 113±72 g/kg from boreal forest fires (Oris et al., 2013). In our study, the effects of fuel moisture on OC and BC EFs were inconclusive. For the Siberian peat samples, the OC/EC ratios were observed to increase with increasing moisture content, while for Alaskan peat samples, the opposite trend was observed. With increasing fuel moisture, OC EFs were observed to increase, while BC EF increases for Alaskan but decreases for Siberian.

Figure 1 shows transmission electron microscopy (TEM) images of typical particles emitted from the combustion of Alaskan and Siberian peat samples. Two basic particle shapes that were identified are: spherical and agglomerates of spherical shapes. The internal structure of the particles was amorphous in nature, which suggested that they belong to the category of "tar balls" (Chakrabarty et al., 2010; Laskin et al., 2015). This was further corroborated by the semi-quantitative Electron Dispersive Spectroscopy (EDS) analysis results of these particles, which showed a very high molar fraction of C and an average molar C-to-O ratio ranging between 6 and 7. This ratio is consistent with those reported by previous studies on tar balls (Chakrabarty et al., 2006; Pósfai et al., 2003). Carbon molar fractions were larger than 80% in over 90% of the particles analyzed. It is interesting to note that a significant fraction (~60%) of the analyzed

183 particles were agglomerates of tar ball spheres, which suggest that weak diffusion limited
184 collisional growth mechanism was involved in their formation process in the smoldering fire.
185 Figure 2 shows the study-averaged mobility diameter number size distribution for the two fuels
186 as measured by SMPS. For each fuel, it was observed that with increasing moisture content the
187 total number concentration of the emitted particles decreased. Further, the median particle
188 diameter for both fuel types was observed to decrease with increasing moisture content. For
189 Alaskan peat burns, the study-averaged median particle diameters were 91 nm and 76 nm at 25%
190 and 50% fuel moisture content, respectively, while for Siberian peat, the median diameters were
191 136 nm and 109 nm at 25% and 50% fuel moisture content, respectively.
192 Figure 3 shows the wavelength dependence of the measured MAE values, connected by best-fit
193 curves (cubic spline), for the emitted aerosols. For Siberian peat samples, MAE values lie in the
194 range of 0.5-0.8 m^2g^{-1} at 405 nm and drop rapidly to 0.03-0.07 m^2g^{-1} at 532 nm. The MAE values
195 at 405 nm for Alaskan peat are slightly lower, in the range of 0.2-0.5 m^2g^{-1}, and exhibit a similar
196 rapid decline at 532 nm. The observed wavelength-varying MAE trends for both fuels are
197 consistent with those observed for brown carbon (BrC) aerosols–a class of OC aerosols
198 absorbing strongly in the near-UV wavelengths–emitted from biomass combustion burning
199 (Chakrabarty et al., 2010; Hoffer et al., 2006; Kirchstetter and Thatcher, 2012). The low MAE
200 values at 532 nm for both peat types compare well with those of Indonesian peat [*Chand et al.*,
201 2005].
202 Fitting power-law functions to our measured MAE spectra between wavelengths $\lambda = 405$ and 532
203 nm yielded mean Absorption Ångström exponent α values of 8.7 for both Siberian 25 and 50%
204 fuel moisture content peat burns, and 7.7 and 10.8 for Alaskan 25% and 50% fuel moisture
205 content peat burns, respectively. α is an intensive optical property that characterizes the inherent

material property. For BC particles, typical values of $\alpha \approx 1$ have been observed, while for BrC aerosols, α ranges from 2 to higher values (Chakrabarty et al., 2010; Moosmuller et al., 2009). Compared with previously reported α values for emissions from laboratory-combusted wildland fuels, emissions from peat burning characterized in this study displayed substantially higher values (Gyawali et al., 2009; Lack et al., 2012; Lewis et al., 2008b). It was also observed that with decreasing moisture content in the peat samples, the emitted aerosols exhibited higher α values. Over the 405-870 nm spectra, the average α for both peat types were in the range of 4.9 (±0.75) - 7.13 (±0.88). However, the trend for MAE values with varying levels of fuel moisture content was not very clear. With increasing moisture content, the MAE values of aerosols from Alaskan peat samples increased; while a decreasing trend was observed for aerosols from Siberian peat samples. A more detailed study on the optical characteristics of chromophores constituting both aerosol types might be necessary toward explanation this trend (Laskin et al., 2015). Such a study is beyond the scope of this current work.

The SSA values of the aerosol spanned a range of 0.92–1. They were consistently higher (0.99-1.00) at 532 and 781 nm than that at 405 nm for all peat samples irrespective of moisture content. This is likely due to the large proportion of BrC in all peat smoke aerosols that preferentially absorbs in the UV region thereby lowering SSA at 405 nm. The calculated SSA values compare well with previous laboratory studies for combustion of Indonesian peat samples [*Chand et al.*, 2005] and from previous field measurements of peat smoke over Moldova [*Eck et al.*, 2003].

4 Impact on Direct Radiative Forcing

We estimate the clear-sky direct radiative forcing per unit optical depth by the emitted BrC aerosols with the help of the “simple forcing efficiency” (SFE, W g^{-1}) concept (Bond and Bergstrom, 2006). Most models assume that OC emitted from biomass combustion has net

negative forcing per gram of emitted aerosol (Bond et al., 2013). The wavelength-dependent SFE equation is given as:

$$\frac{dSFE}{d\lambda} = -\frac{1}{4}\frac{dS(\lambda)}{d\lambda}\tau^2(\lambda)(1-F_c)[2(1-a_s)^2\beta(\lambda) * MSE(\lambda) - 4a_s * MAE(\lambda)] \quad (1)$$

where $dS(\lambda)/d\lambda$ is the solar irradiance, τ is the atmospheric transmission (0.79), F_c is the cloud fraction (0.6), a is the surface albedo (0.19 for earth average and 0.8 for snow (Chen, 2011; Chen and Bond, 2010)), β is the fraction of scattered sunlight that is scattered into the upward hemisphere (≈0.17 for biomass burning BrC aerosols), and MSE and MAE are the mass scattering and absorption efficiencies per gram, respectively (Chen and Bond, 2010; Griggs and Noguer, 2002; Saleh et al., 2014). Note that this equation doesn't account for hygroscopicity, which could affect SFE. Net forcing in the 405-880 nm spectral range was calculated by integrating the SFE equation using the tropospheric solar spectrum (Levinson et al., 2010).

Figure 4 a and b show forcing efficiencies at each wavelength over a bright surface (surface albedo of 0.8), which is characteristic of the snow-covered Arctic landscape and low-level clouds over which smoke plume typically moves. Integrated mean forcing over the solar spectrum is 20 and 38 W g^{-1} for BrC aerosols from Alaskan and Siberian peat burns, respectively. By assuming no absorption for the emitted aerosols, a convention often adopted by climate modelers while representing OC, we get a mean negative forcing of -3.7 and -5 W g^{-1} for smoke from Alaskan and Siberian peat samples, respectively. These calculations were repeated for a surface albedo of 0.19 (earth average (Chen and Bond, 2010)). The integrated forcing was negative in the visible wavelengths with mean values of -70 and -81 W g^{-1} from Alaskan and Siberian peat samples, respectively. It is interesting to contrast and compare the extremely high integrated forcing value for BC over land, which is around 210 W/g (Chen, 2011).

Figure 5 a and b show net forcing efficiencies, integrated over the tropospheric solar spectrum, as a function of surface albedo (a_s) for aerosols emitted from both fuel types. For Siberian peat samples, the forcing efficiency crosses over from negative (cooling) to positive (warming) values at $a_s \approx 0.5$. The cross-over points are nearly identical for varying fuel moisture content. However, for Alaskan peat with 50% moisture content, the cross-over takes place at a lower value ($a_s \approx 0.57$) compared to $a_s \approx 0.61$ for 25% moisture content. Overall, these results suggest that BrC aerosols from boreal peatland burning could result in a net warming effect of the atmosphere only if they reside over bright surfaces with albedo greater than 0.6.

5 Conclusions

Our findings show that BrC aerosols from peatland fires in the Boreal region may give rise to significant absorption in the shorter visible wavelengths and the ultraviolet regions of the solar spectrum. This strong absorptivity may result in the positive net forcing (warming) over bright surfaces. The common understanding has been that BC constitutes the light-absorbing aerosol type from boreal forest fires (Randerson et al., 2006), while OC is light scattering in nature and helps offset the BC warming effects. However, our results show that aerosols containing BrC, which is a class of OC, could further amplify the warming effects of BC in this region, especially since 47% of incoming solar energy is distributed between 400 nm and 700 nm wavelengths. Additionally, absorption in the ultraviolet range by BrC aerosols could affect photolysis-driven atmospheric chemistry and consequently reduce tropospheric ozone concentration (Jacobson, 1998).

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Table 1: Mean mass-based emission factors (rounded to nearest integer) of carbonaceous gases and aerosols from emissions of Alaskan and Siberian peat in this study.

Fuel Type	**Fuel Moisture Content**	**Mean Fuel-Based Emission Factors (g kg^{-1} fuel)**			
		CO_2	**CO**	**OC**	**BC**
Alaskan Peat	25%	1238	83	7	0.1
Alaskan Peat	50%	1598	128	4	0.2
Siberian Peat	25%	1432	204	17	0.2
Siberian Peat	50%	1698	49	11	0.1

Figure 1: Transmission electron microscopy (TEM) images of typical organic carbon "tar balls", occurring as spheres and agglomerates, emitted from smoldering combustion of Alaskan and Siberian peat samples. The internal structure of these particles were was amorphous in nature. Electron dispersive spectroscopy (EDX) of tar balls shows that these particles consist primarily of carbon and oxygen with an average molar ratio of ranging between 6-7.

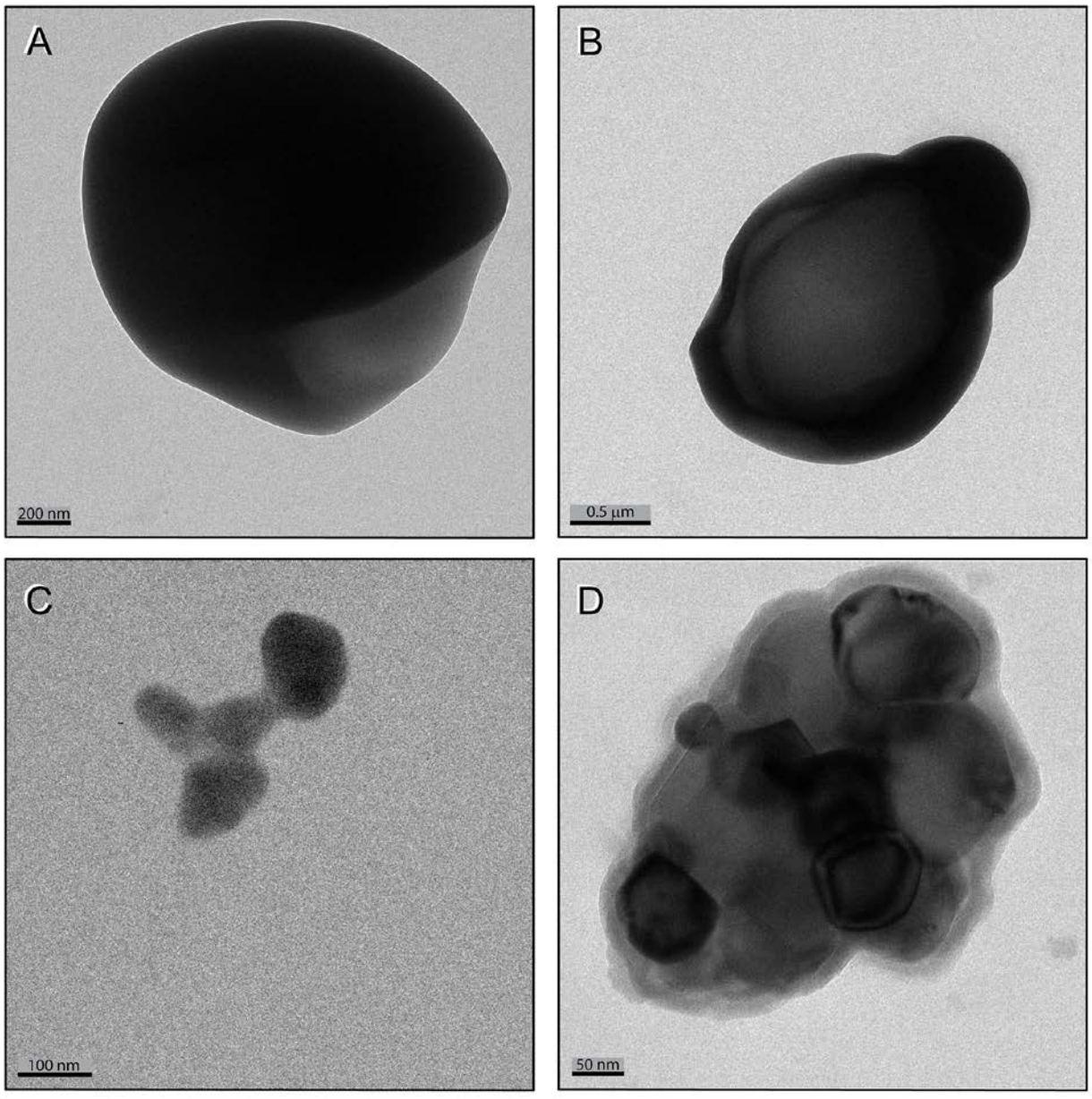

Figure 2: Study-averaged mobility number size distribution of aerosols from Alaskan and Siberian peat samples.

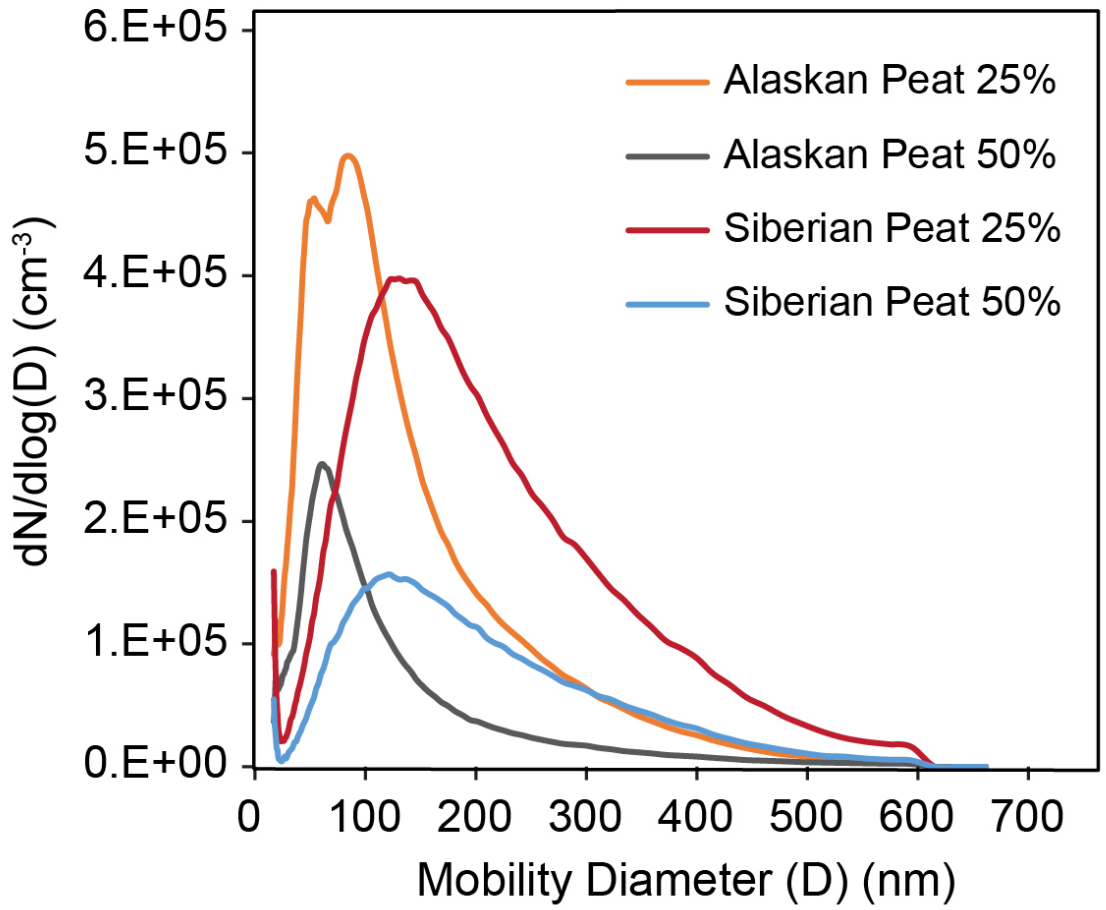

Figure 3: Wavelength-dependent mass absorption efficiency (MAE) of the sampled carbonaceous (brown) aerosols from (A) Alaskan and (B) Siberian peat smoldering combustion. The dashed lines show study-averaged values. The shaded bands correspond to error bars measured at 405, 532, 781, and 870 nm, and connected by best-fit curves.

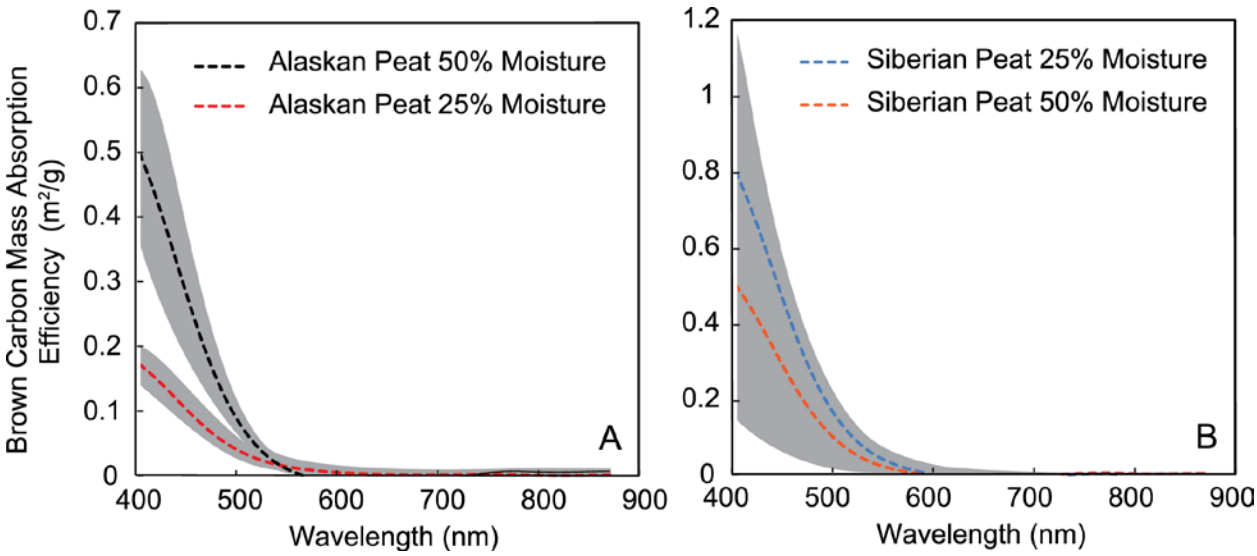

Figure 4: Direct forcing efficiency of brown carbon aerosols above snow (surface albedo = 0.8). Integrated mean forcing is 20 and 38 W g^{-1} for Alaskan and Siberian peat smoke aerosols, respectively.

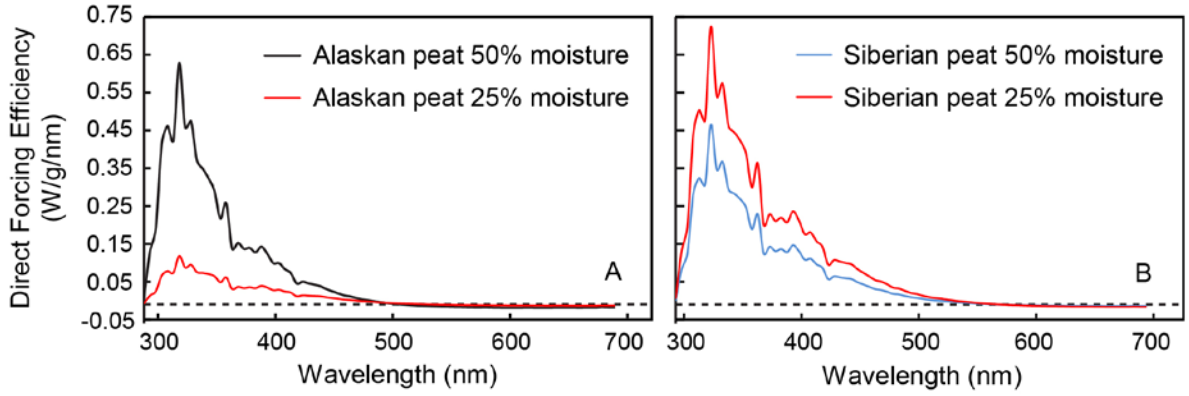

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Figure 5: Net forcing efficiencies as a function of surface albedo for Siberian and Alaskan peat smoke aerosols. The cross-over from negative (cooling) to positive (warming) values takes place between surface albedo values of 0.55 – 0.6.