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.

115 An IPN consists of a wavelength-specific laser module and a reciprocal integrating nephelometer
116 aligned in an acoustic resonator. The instrument measures particle light absorption coefficient
117 (β_{abs}) using the photoacoustic effect (Arnott et al., 1999), while the reciprocal integrating
118 nephelometer measures the integrated (over ~ 4π) scattering from the sample volume yielding
119 the scattering coefficient (β_{sca}) (Abu-Rahmah et al., 2006). The four wavelength IPNs used in
120 this study facilitated simultaneous measurement, with two-second time resolution, of spectrally-
121 varying β_{abs} and β_{sca} in addition to intensive aerosol optical properties such as single scattering
122 albedo (SSA) and Absorption Ångström Exponent (α). The SMPS was operated with a
123 sheath/aerosol flow ratio of 10:1 (sheath flow = 3 L/min; aerosol flow = 0.3 L/min), yielding a
124 differential mobility analyzer size transmission width of approximately ±10%. The CO and CO_2
125 gas concentrations were continuously measured and the data were averaged over 5-minute
126 intervals.
127 For each run, aerosols were collected on 47 mm quartz-fiber filters at 10 l min^{-1} flow rate.
128 Immediately after sampling, filters were stored in a refrigerator and later analyzed for BC and
129 OC mass fractions and concentrations using the IMPROVE-A TOR and TOT analyses method
130 (Chow et al., 2007; Chow et al., 2011) implemented on a DRI Model 2001 thermal/optical
131 carbon analyzers (Atmoslytic, Inc., Calabasas, CA, USA). The fuel-based emission factor (see
132 details in the Supplement), defined as the mass of a compound released per mass of fuel
133 consumed (Chen et al., 2007), of BC and OC corresponding to each sampled filter were
134 determined using the procedure described by Chen *et al.* (Chen et al., 2007). With the knowledge
135 of OC mass concentrations and β_{abs}, the OC mass absorption efficiency (MAE, also referred to as
136 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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References

Abu-Rahmah, A., Arnott, W. P., and Moosmüller , H.: Integrating Nephelometer with a Low Truncation Angle and an Extended Calibration Scheme, Meas. Sci. Technol., 17, 1723-1732, 2006.

Arnott, W. P., Moosmüller, H., Rogers, C. F., Jin, T., and Bruch, R.: Photoacoustic Spectrometer for Measuring Light Absorption by Aerosol: Instrument Description, Atmos. Environ., 33, 2845-2852, 1999.

Bachelet, D., Lenihan, J., Neilson, R., Drapek, R., and Kittel, T.: Simulating the response of natural ecosystems and their fire regimes to climatic variability in Alaska, Canadian Journal of Forest Research, 35, 2244-2257, 2005.

Balshi, M. S., McGuire, A. D., Duffy, P., Flannigan, M., Kicklighter, D. W., and Melillo, J.: Vulnerability of carbon storage in North American boreal forests to wildfires during the 21st century, Global Change Biology, 15, 1491-1510, 2009.

Bonan, G. B.: Forests and climate change: forcings, feedbacks, and the climate benefits of forests, Science, 320, 1444-1449, 2008.

Bond, T. and Bergstrom, R.: Light Absorption by Carbonaceous Particles: An Investigative Review, Aerosol Sci. Technol., 40, 27-67, 2006.

Bond, T., Doherty, S., Fahey, D., Forster, P., Berntsen, T., DeAngelo, B., Flanner, M., Ghan, S., Kärcher, B., and Koch, D.: Bounding the role of black carbon in the climate system: a scientific assessment, J. Geophys. Res.,, 118, 5380–5552, 2013.

Chakrabarty, R., Moosmüller, H., Chen, L., Lewis, K., Arnott, W., Mazzoleni, C., Dubey, M., Wold, C., Hao, W., and Kreidenweis, S.: Brown carbon in tar balls from smoldering biomass combustion, Atmos. Chem. Phys, 10, 6363-6370, 2010.

Chakrabarty, R., 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 wildland fuels: Particle morphology and size, J. Geophys. Res, 111, D07204, 2006.

Chen, L. W. A., Moosmüller, H., Arnott, W. P., Chow, J. C., Watson, J. G., Susott, R. A., Babbitt, R. E., Wold, C. E., Lincoln, E. N., and Hao, W. M.: Emissions from laboratory combustion of wildland fuels: Emission factors and source profiles, Environ. Sci. Technol., 41, 4317-4325, 2007.

Chen, Y.: Characterization of carbonaceous aerosols from biofuel combustion: emissions and climate relevant properties, Doctoral Dissertation, University of Illinois Urbana Champaign, Illinois Digital Environment for Access to Learning and Scholarship, 215 pp., 2011.

Chen, Y. and Bond, T.: Light absorption by organic carbon from wood combustion, Atmos. Chem. Phys., 10, 1773-1787, 2010.

Chow, J. C., Watson, J. G., Chen, L.-W. A., Chang, M. O., Robinson, N. F., Trimble, D., and Kohl, S.: The IMPROVE_A temperature protocol for thermal/optical carbon analysis: Maintaining consistency with a long-term database, J. Air Waste Manage. Assoc., 57, 1014-1023, 2007.

Chow, J. C., Watson, J. G., Robles, J., Wang, X., Chen, L.-W. A., Trimble, D. L., Kohl, S. D., Tropp, R. J., and Fung, K. K.: Quality assurance and quality control for thermal/optical analysis of aerosol samples for organic and elemental carbon, Anal. Bioanal. Chem., 401, 3141-3152, 2011.

Christian, T. J., Kleiss, B., Yokelson, R. J., Holzinger, R., Crutzen, P., Hao, W. M., Saharjo, B., and Ward, D. E.: Comprehensive laboratory measurements of biomass-burning emissions: 1. Emissions from Indonesian, African, and other fuels, Journal of Geophysical Research: Atmospheres (1984–2012), 108, 2003.

Chung, C. E., Ramanathan, V., and Decremer, D.: Observationally constrained estimates of carbonaceous aerosol radiative forcing, Proceedings of the National Academy of Sciences, 109, 11624-11629, 2012.

Conard, S. G. and Ivanova, G. A.: Wildfire in Russian boreal forests—Potential impacts of fire regime characteristics on emissions and global carbon balance estimates, Environ. Pollut., 98, 305-313, 1997.

Eck, T., Holben, B., Reid, J., Sinyuk, A., Hyer, E., O'Neill, N., Shaw, G., Vande Castle, J., Chapin, F., and Dubovik, O.: Optical properties of boreal region biomass burning aerosols in central Alaska and seasonal variation of aerosol optical depth at an Arctic coastal site, Journal of Geophysical Research: Atmospheres (1984–2012), 114, 2009.

Flannigan, M. D., Logan, K. A., Amiro, B. D., Skinner, W. R., and Stocks, B.: Future area burned in Canada, Climatic change, 72, 1-16, 2005.

Gorham, E.: The future of research in Canadian peatlands: a brief survey with particular reference to global change, Wetlands, 14, 206-215, 1994.

Griggs, D. J. and Noguer, M.: Climate change 2001: the scientific basis. Contribution of working group I to the third assessment report of the intergovernmental panel on climate change, Weather, 57, 267-269, 2002.

Gustafsson, O., Krusa, M., Zencak, Z., Sheesley, R. J., Granat, L., Engstrom, E., Praveen, P. S., Rao, P. S. P., Leck, C., and Rodhe, H.: Brown Clouds over South Asia: Biomass or Fossil Fuel Combustion?, Science, 323, 495-498, 2009.

Gyawali, M., Arnott, W., Lewis, K., and Moosmüller, H.: In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of absorbing and non-absorbing organic coatings on spectral light absorption, Atmos. Chem. Phys., 9, 8007-8015, 2009.

Hoffer, A., Gelencsér, A., Guyon, P., Kiss, G., Schmid, O., Frank, G., Artaxo, P., and Andreae, M.: Optical properties of humic-like substances (HULIS) in biomass-burning aerosols, Atmos. Chem. Phys., 6, 3563-3570, 2006.

Hu, F. S., Higuera, P. E., Walsh, J. E., Chapman, W. L., Duffy, P. A., Brubaker, L. B., and Chipman, M. L.: Tundra burning in Alaska: linkages to climatic change and sea ice retreat, Journal of Geophysical Research: Biogeosciences (2005–2012), 115, 2010.

Iinuma, Y., Brüggemann, E., Gnauk, T., Müller, K., Andreae, M., Helas, G., Parmar, R., and Herrmann, H.: Source characterization of biomass burning particles: The combustion of selected

European conifers, African hardwood, savanna grass, and German and Indonesian peat, Journal of Geophysical Research: Atmospheres (1984–2012), 112, 2007.

Jacobson, M. Z.: Studying the effects of aerosols on vertical photolysis rate coefficient and temperature profiles over an urban airshed, Journal of Geophysical Research: Atmospheres (1984–2012), 103, 10593-10604, 1998.

Jorgenson, M. T., Racine, C. H., Walters, J. C., and Osterkamp, T. E.: Permafrost degradation and ecological changes associated with a warmingclimate in central Alaska, Climatic change, 48, 551-579, 2001.

Kirchstetter, T. and Thatcher, T.: Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation, Atmos. Chem. Phys, 12, 6067-6072, 2012.

Lack, D. A., Langridge, J. M., Bahreini, R., Cappa, C. D., Middlebrook, A. M., and Schwarz, J. P.: Brown carbon and internal mixing in biomass burning particles, Proc. Natl. Acad. Sci. U. S. A., 109, 14802-14807, 2012.

Laskin, A., Laskin, J., and Nizkorodov, S. A.: Chemistry of Atmospheric Brown Carbon, Chem. Rev. (Washington, DC, U. S.), 115, 4335-4382, 2015.

Lawrence, D. M. and Slater, A. G.: A projection of severe near-surface permafrost degradation during the 21st century, Geophys. Res. Lett., 32, 2005.

Levinson, R., Akbari, H., and Berdahl, P.: Measuring solar reflectance—Part I: Defining a metric that accurately predicts solar heat gain, Solar Energy, 84, 1717-1744, 2010.

Lewis, K., Arnott, W. P., Moosmuller, H., and Wold, C. E.: Strong spectral variation of biomass smoke light absorption and single scattering albedo observed with a novel dual-wavelength photoacoustic instrument, J. Geophys. Res., 113, doi:10.1029/2007JD009699, 2008a.

Lewis, K., Arnott, W. P., Moosmüller, H., and Wold, C. E.: Strong spectral variation of biomass smoke light absorption and single scattering albedo observed with a novel dual-wavelength photoacoustic instrument, Journal of Geophysical Research: Atmospheres (1984–2012), 113, 2008b.

Moosmuller, H., Chakrabarty, R. K., and Arnott, W. P.: Aerosol light absorption and its measurement: A review, J. Quant. Spectrosc. Radiat. Transfer, 110, 844-878, 2009.

Oris, F., Asselin, H., Ali, A. A., Finsinger, W., and Bergeron, Y.: Effect of increased fire activity on global warming in the boreal forest, Environmental Reviews, 22, 206-219, 2013.

Page, S., Wűst, R., Weiss, D., Rieley, J., Shotyk, W., and Limin, S. H.: A record of Late Pleistocene and Holocene carbon accumulation and climate change from an equatorial peat bog (Kalimantan, Indonesia): implications for past, present and future carbon dynamics, Journal of Quaternary Science, 19, 625-635, 2004.

Page, S. E., Siegert, F., Rieley, J. O., Boehm, H. D. V., Jaya, A., and Limin, S.: The amount of carbon released from peat and forest fires in Indonesia during 1997, Nature, 420, 61-65, 2002.

Pearson, R. G., Phillips, S. J., Loranty, M. M., Beck, P. S., Damoulas, T., Knight, S. J., and Goetz, S. J.: Shifts in Arctic vegetation and associated feedbacks under climate change, Nature Climate Change, 3, 673-677, 2013.

Pósfai, M., Simonics, R., Li, J., Hobbs, P. V., and Buseck, P. R.: Individual Aerosol Particles from Biomass Burning in Southern Africa: 1. Compositions and Size Distributions of Carbonaceous Particles, Journal of Geophysical Research, 108, DOI:10.1029/2002JD002291, 2003.

Randerson, J. T., Liu, H., Flanner, M. G., Chambers, S. D., Jin, Y., Hess, P. G., Pfister, G., Mack, M., Treseder, K., and Welp, L.: The impact of boreal forest fire on climate warming, Science, 314, 1130-1132, 2006.

Rein, G., Cleaver, N., Ashton, C., Pironi, P., and Torero, J. L.: The severity of smouldering peat fires and damage to the forest soil, Catena, 74, 304-309, 2008.

Saleh, R., Robinson, E. S., Tkacik, D. S., Ahern, A. T., Liu, S., Aiken, A. C., Sullivan, R. C., Presto, A. A., Dubey, M. K., and Yokelson, R. J.: Brownness of organics in aerosols from biomass burning linked to their black carbon content, Nat. Geosci., 2014. 2014.

Sheng, Y., Smith, L. C., MacDonald, G. M., Kremenetski, K. V., Frey, K. E., Velichko, A. A., Lee, M., Beilman, D. W., and Dubinin, P.: A high-resolution GIS-based inventory of the west Siberian peat carbon pool, Global Biogeochem. Cycles, 18, 2004.

Simpson, I. J., Akagi, S., Barletta, B., Blake, N., Choi, Y., Diskin, G., Fried, A., Fuelberg, H., Meinardi, S., and Rowland, F.: Boreal forest fire emissions in fresh Canadian smoke plumes: C 1-C 10 volatile organic compounds (VOCs), CO 2, CO, NO 2, NO, HCN and CH 3 CN, Atmos. Chem. Phys., 11, 6445-6463, 2011.

Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K., Tignor, M., and Miller, H.: The physical science basis, Contribution of working group I to the fourth assessment report of the intergovernmental panel on climate change, 2007. 235-337, 2007.

Stocker, T. F., Dahe, Q., and Plattner, G.-K.: Climate Change 2013: The Physical Science Basis, Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Summary for Policymakers (IPCC, 2013), 2013. 2013.

Stocks, B. J., Fosberg, M., Lynham, T., Mearns, L., Wotton, B., Yang, Q., Jin, J., Lawrence, K., Hartley, G., and Mason, J.: Climate change and forest fire potential in Russian and Canadian boreal forests, Climatic change, 38, 1-13, 1998.

Stockwell, C., Yokelson, R., Kreidenweis, S., Robinson, A., DeMott, P., Sullivan, R., Reardon, J., Ryan, K., Griffith, D. W., and Stevens, L.: Trace gas emissions from combustion of peat, crop residue, domestic biofuels, grasses, and other fuels: configuration and Fourier transform infrared (FTIR) component of the fourth Fire Lab at Missoula Experiment (FLAME-4), 2014. 2014.

Tian, J., Chow, J. C., Cao, J., Han, Y., Ni, H., Chen, L.-W. A., Wang, X., Huang, R., Moosmüller, H., and Watson, J. G.: A Biomass Combustion Chamber: Design, Evaluation, and a Case Study of Wheat Straw Combustion Emission Tests, Aerosol and Air Quality Research, 15, 2104-2114, 2015.

Turetsky, M. R., Benscoter, B., Page, S., Rein, G., van der Werf, G. R., and Watts, A.: Global vulnerability of peatlands to fire and carbon loss, Nat. Geosci., 8, 11-14, 2015.

Ward, D. E., Hao, W. M., Susott, R. A., Babbitt, R. E., Shea, R. W., Kauffman, J. B., and Justice, C. O.: Effect of fuel composition on combustion efficiency and emission factors for African savanna ecosystems, J. Geophys. Res., 101, 23569-23576, 1996.

Xu, R., Ferrante, L., Briens, C., and Berruti, F.: Bio-oil production by flash pyrolysis of sugarcane residues and post treatments of the aqueous phase, J. Anal. Appl. Pyrolysis, 91, 263-272, 2011.

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.

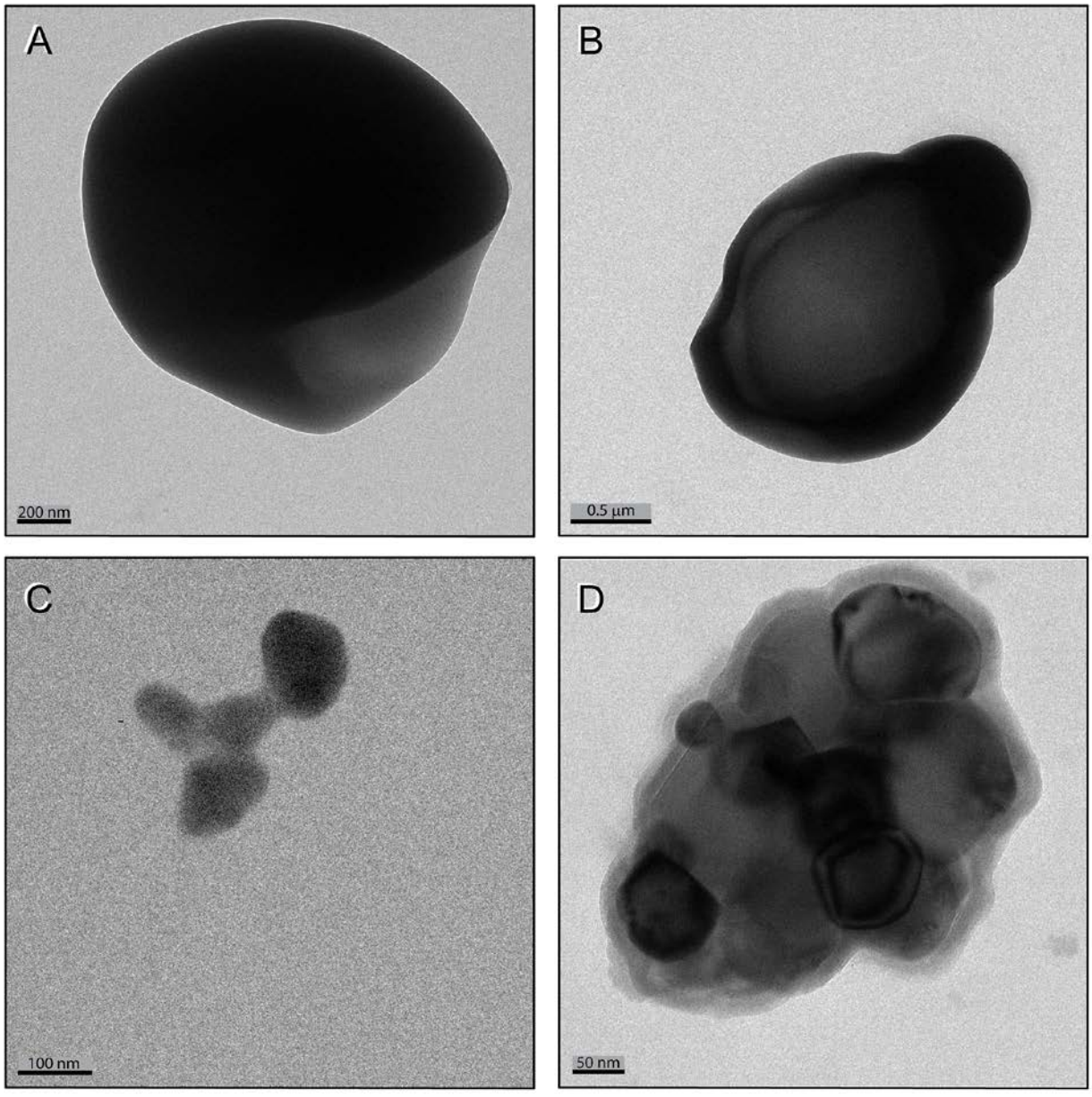


Figure 2: Study-averaged mobility number size distribution of aerosols from Alaskan and Siberian peat samples.

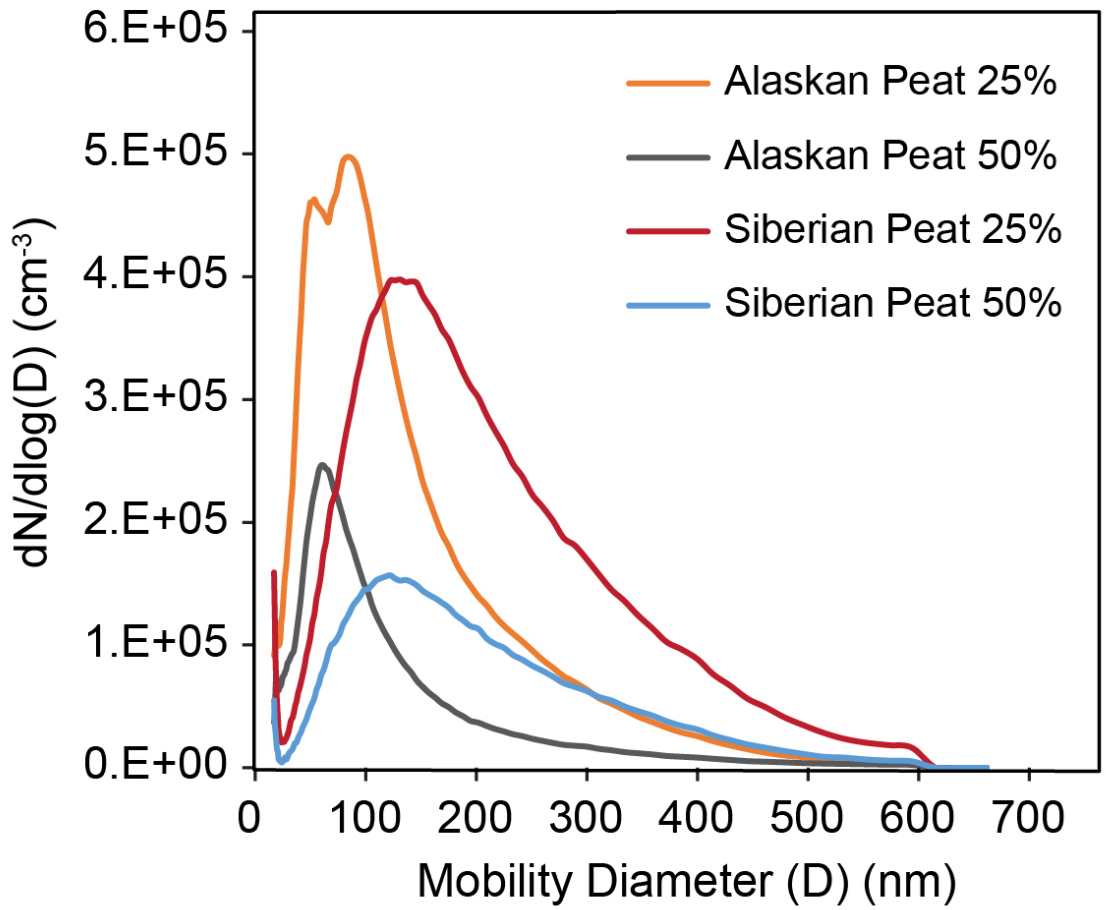


Figure 3: Wavelength-dependent mass absorption efficiency (MAE) of the sampled carbonaceous (brown) aerosols from (A) Alaskan and (B) Siberian peat smoldering combustion. The dashed lines show study-averaged values. The shaded bands correspond to error bars measured at 405, 532, 781, and 870 nm, and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

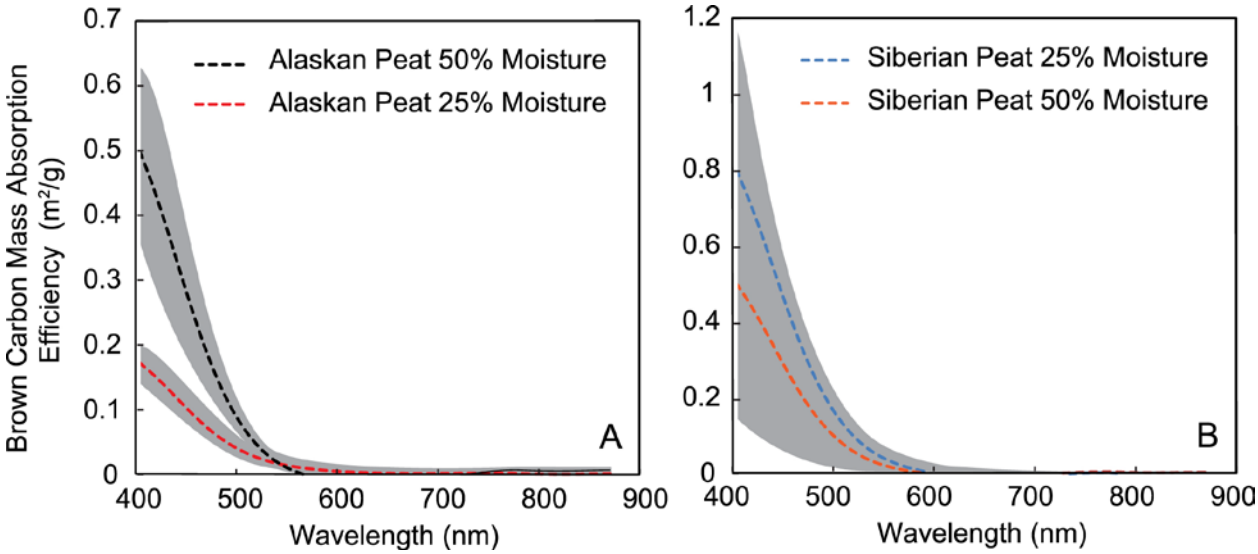
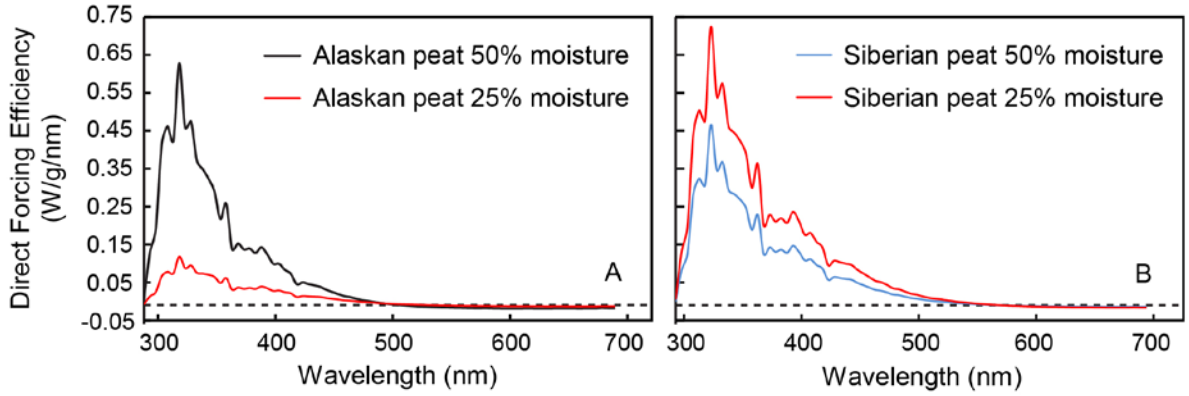
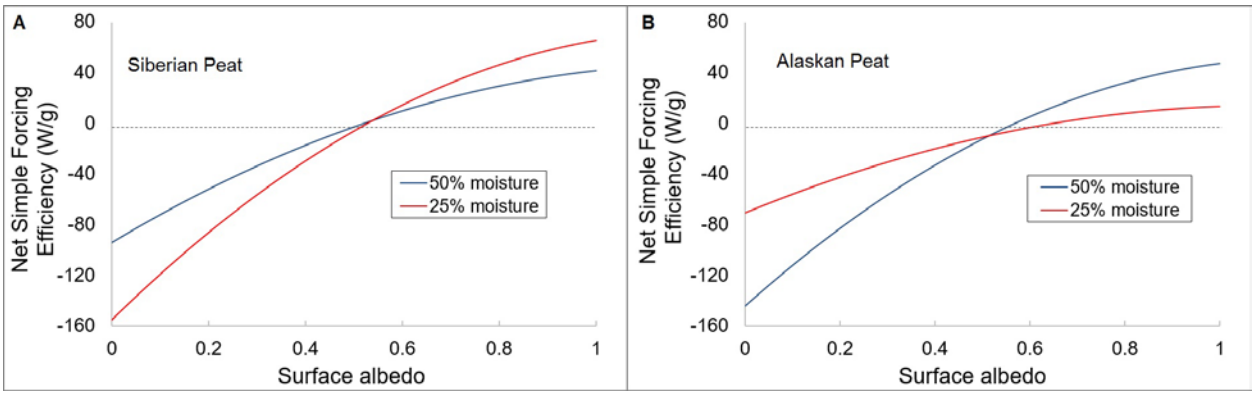


Figure 4: Direct forcing efficiency of brown carbon aerosols above snow (surface albedo = 0.8). Integrated mean forcing is 20 and 38 W g^{-1} for Alaskan and Siberian peat smoke aerosols, respectively.



443

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.



Dear Dr. Bertram,

We thank you for your thorough evaluation of our manuscript titled "*Dominance of brown carbon in aerosol emissions from burning of boreal peatlands*" which has been published in Atmospheric Chemistry and Physics Discussions. Below please find our point-by-point response to all reviewer remarks as well as a version of our revised manuscript.

The main corrections in the revised manuscript are: (a) Revision of the title to better reflect the scope and findings of our paper; (b) Elaboration of the abstract to make key findings inclusive in it; and (c) Inclusion of a new figure on the sensitivity of direct forcing efficiency of brown carbon aerosols on surface albedo. We very much look forward to a positive response from you and, ultimately, to publication of this manuscript in Atmospheric Chemistry and Physics.

Thank you.

Sincerely,

Rajan K. Chakrabarty and co-authors

(ALL AUTHOR REPLIES ARE MARKED IN RED)

Anonymous Referee #1

General

This is a very interesting study on results from biomass burning of boreal peat material from Alaska and Siberia with emphasis on brown carbon. It is very knowledgeable as for the derived radiative quantities and a possible contribution of carbonaceous particles to surface temperature increase at high northern latitudes.

The manuscript title does not make fully clear that this study is indeed concerned with peat burning. The authors may consider to make this more clear in their manuscript title. I have a number of specific points to be addressed, see below. I think the manuscript should be revised, at a level somewhere between minor and major revision.

Details

Abstract: I find the abstract quite condensed. Is this really all which should be mentioned here?

Response: The abstract has now been revised and elaborated in scope. It also highlights the major finding regarding the sensitivity of forcing as a function of surface albedo.

P 28796, l 8: Why will the Siberian peatland burning increase so much 'in response to climate change' - a few words to explain would be good.

Response: Thanks for pointing this out. The reasoning behind this statement is that drying as a result of climate change lowers the water table in peatlands and increases the frequency and extent of peat fires. A new sentence has been added to the end of this paragraph "*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"

P 28796, l 11 ff: This paragraph very well addreses past studies of boreal forest fires. Because the study really is about peat burning, please reference former such studies right here in the section - this is fully missing now.

Response: The following sentence with appropriate references have now been added about peat burning in the revised manuscript "*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)*"

P 28798, l 27: This estimate must be rough. It means BrC from this study is an upper limit because EC has to be subtracted. This should be mentioned everywhere and an estimate of the size of this error should be given.

Response: While we agree that approximating EC with BC might involve a conversion factor, this shouldn't affect the findings of this study. This is because EC determined in this study via the IMPROVE-A TOR and TOR analyses method (industry standard) in our filter samples were trace in amount. For most of the samples, EC was less than 1% by mass. So, one could safely attribute the absorption coefficients measured by multi-wavelength photoacoustic spectrometers to only the OC mass content of the particle. That is what we did. Further, it would be erroneous to say that BrC mass absorption efficiencies (MAE) calculated in this study is an upper limit. Since we didn't have to subtracted any BC mass and that we used first-principle aerosol absorption technique, we feel the BrC MAE values are not upper limits as one would expect from a filter based absorption measurement approach.

P28799,l 12: Results of previous studies could be incorporated into Table 1.

Response: We feel that incorporating emission factor measurements of peat fires from lower-latitude peatlands, like those found in Indonesia and Germany, into Table 1 might lead to confusion among the readers. The focus of this paper is peat land fires from higher-latitude Boreal region, and there is a lack of data on primary greenhouse gases and particulate matter. On the contrary, there is substantial amount of research conducted on the nature of pollutants from Indonesian and other low-latitude peat fires. Additionally, the composition of peat lands could vary significantly between the low and high latitudes. Change in composition impacts the pollutant profile as well. Hence, we are not of the opinion to include emission factors of gaseous and particulate pollutants from low-latitude peat burning in Table 1.

P 28800, l12ff: Can you nevertheless document the results of the varying humidity experiments ? What could be a possible explanation of the differing trends?

Response: We do not have a convincing and straight forward explanation to explain the effects of humidity on the particle optical properties. With increasing fuel moisture content, the BrC mass absorption efficiency of Alaskan peat increased; while for the Siberian peat, the opposite trend was observed. A detailed understanding of this phenomenon might involve investigating

the nature of the chromophores constituting the particles, which is beyond the scope of this study.

In the revised manuscript, we have added the following statement to address this point "*However, the trend for MAE values with varying levels of fuel moisture content was not very clear. With increasing moisture content, the MAEs of aerosols from Alaskan peat samples increased; while the opposite 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.*"

Anonymous Referee #2

General Comments

This paper reports the absorption properties of laboratory combusted peat samples in order to address the accelerated warming of the Artic as it relates to absorbing aerosol particles. It specifically address the smoldering phase of peat, which is known to produce brown carbon compounds very efficiently. These compounds have appreciable visible absorption and plausibly pose a threat to the Artic in terms of positive radiative forcing.

The paper is clear and well-written, with minor exceptions outlined below. Figures are easy to follow. The inclusion of the direct radiative forcing calculation strengthens the impact of this paper, as it provides a means to compare other brown carbon measurements.

The main shortcoming of the manuscript as written is the absence of a sensitivity study on the surface albedo underlying the aerosol plume. Clearly that has a significant impact on the calculated forcing but it has not been done, or has not been included. The main result of the paper is hidden before the Conclusion section and should be brought explicitly into the abstract and introduction sections. I recommend this paper for publication with these revisions. I consider them minor.

Specific Comments

The abstract would benefit from additional quantitative results, especially with respect to radiative forcing and photochemistry.

Response: The abstract has now been revised and elaborated in scope. It highlights the major finding regarding the sensitivity of forcing as a function of surface albedo. For sake of clarity to the readers, the statement on photochemistry has been omitted. This manuscript doesn't report any quantitative measurement of the impact of brown carbon aerosols on photochemistry.

pg 28796 - Are these fires burned intentionally? The statement that the burn area will increase "in response to climate change" indicates that there is some natural connection between temperature and burn area but that is not obvious to me as a reader.

Response: No, these fires are not intentional. What was implied by our statement is that drying as a result of climate change would lower the water table in peatlands and increase the frequency

and extent of peat fires. A new sentence has been now added to the end of the introductory paragraph to further clarify this point: *"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*"

The end of the introduction would benefit from the inclusion of the authors approach (in more detail) and findings, to help guide the reader as they follow the methods section. Specifically, what kinds of measurements were conduction (briefly) and what were the key findings?

Response: This suggestion is well taken. The end of the introduction section now bears a few sentences on the instruments used to carry out the specific measurements of aerosol properties. We have not introduced the key findings in the introduction section as we feel it might interfere with the linear flow of the manuscript contents.

It would be helpful for the authors to include an explanation of the atmsopheric transmission (0.79), beta (0.17), and cloud fraction (0.6) chosen for their estimation. If other studies wish to compare their results with these findings, they will need to understand the justification for those choices.

Response: The values adopted for atmospheric transmission, backscatter fraction (beta), and cloud fraction are for clear-sky conditions. The choice of these values originates from the IPCC 2001 report (https://www.ipcc.ch/ipccreports/tar/wg1/197.htm). Several researchers have adopted these values to calculate clear-sky radiative forcing estimates in the recent years. Appropriate citations of these studies have now been included in the revised manuscript.

What kind of landscape has an albedo of 0.19?

Response: 0.19 is the average earth albedo, as suggested by Chen and Bond (2010, Atmospheric Chemistry and Physics). This citation is now included in the revised manuscript for the purpose of clarity.

The authors show that by including the observed absorption from these peat smoke aerosol particles, the net forcing over snow and low level clouds shifts from small, but negative, to significantly positive. This seems like a major result, but is hiding buried in the paper. It should be in the abstract and in the end of the introduction.

Response: Thank you for this suggestion. In the revised manuscript, we have now added this finding in the abstract. In addition, we have added a new figure 5 showing the plots of integrated (net) forcing efficiency as a function of surface albedo for both fuel types. A paragraph to explain this plot has also been added to the manuscript.

Further, the title of the article does not describe the new and important findings of this work. It is doubtful that readers will be surprised that brown carbon dominates (predominates is the correct term) peatland smoke. However, readers may be surprised to find out the degree to which brown carbon compounds in the smoldering peat impact the radiative forcing of the aerosol in the Arctic region. I suggest the authors consider finding a higher profile title to represent their work.

Response: This suggestion is well taken. Accordingly, the title of the revised manuscript has been revised.

The integrated forcing appears to be incredibly sensitive to the albedo of the surface below it. It would be incredibly useful to know the albedo at which the forcing goes from positive (as over ice and cloud) to negative (as over darker land surfaces). Further, it would be useful to know what fraction of the Arctic includes surfaces above which the smoke has a positive forcing (more than half?).

Response: This is a very good suggestion. We have now added a new figure 5 to the revised manuscript. This figure shows the plots of integrated (net) forcing efficiency as a function of surface albedo for both fuel types. The forcing changes from negative to positive between a surface albedo of 0.55 - 0.6 for both fuel types. Hence, it would be safe to say that for a surface albedo greater than ~0.6, brown carbon aerosols from peat fires would give rise to a warming effect. Speculating the fraction of arctic with surface albedo greater than 0.6 is beyond the the scope of this manuscript.

In addition to a new figure, a new paragraph has been added at the end of section 4: "*Figure 5 a,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, it could be said that brown carbon aerosols from boreal peat fires would result in a net warming effect under clear-sky conditions over surfaces with albedo greater than 0.6.*"

What is SFE of soot over those same surfaces? Comparing these numbers to soot particles would help readers put the particles into perspective.

Response: Dr. Tami Bond's group calculated fresh BC SFE by using the backscattering fraction = 0.17, MAE = 7.5 m^2 /g at 550nm, and MAC at other wavelengths are calculated by assuming it is depending inversely on wavelength, i.e., AAE = 1. MSC is calculated from MAC with single scattering albedo of 0.25. They estimated BC SFE is 210 W/g over land surface (albedo = 0.19). They also speculated that with increasing surface albedo, the SFE would keep on increasing. In the revised manuscript, we have added the following sentence "*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)*".

Technical Comments

What was the fuel moisture content of the particles in Figure 1?

Response: The relative humidity (RH) of the particles were below 40%. This is because photoacoustic spectrometers work best and most reliably below a threshold particle RH of 40%. If increased beyond this RH threshold, then there could be interference from water mass transfer in the signal.