Atmos. Chem. Phys. Discuss., 15, 28793–28813, 2015
www.atmos-chem-phys-discuss.net/15/28793/2015/
doi:10.5194/acpd-15-28793-2015

Atmospheric Chemistry and Physics Discussions

Dominance of brown carbon in aerosol emissions from burning of boreal peatlands

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Received: 31 August 2015 – Accepted: 29 September 2015 – Published: 23 October 2015

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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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 report the microphysical properties of aerosols emitted from controlled laboratory combustion of Alaskan and Siberian peatland samples. The emitted aerosols are brown carbon with negligible amount of black carbon content. Their mass absorption efficiencies lie in the range of 0.2–0.8 $m^2 g^{-1}$ at 405 nm and drop sharply to 0.03–0.07 $m^2 g^{-1}$ at 532 nm, characterized by a mean Ångström exponent of ≈ 9. Their top-of-atmosphere direct radiative forcing over bright surfaces is positive (warming), and their presence in the troposphere may influence photolysis driven chemistry.

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 (Turetsky et al., 2015; Gorham, 1994). 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

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global rate (Pearson et al., 2013; Lawrence and Slater, 2005; Jorgenson et al., 2001; Hu et al., 2010).

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 (Stocks et al., 1998; Sheng et al., 2004; Conard and Ivanova, 1997). Siberia is home to about 50 % 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 (Balshi et al., 2009; Bachelet et al., 2005; Flannigan et al., 2005).

Past studies have estimated that carbon released from boreal forest fires is mostly composed of greenhouse gasses – CO_2, CO, and CH_4 (Simpson et al., 2011; Oris et al., 2013). 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 $17 \pm 30\,W\,m^{-2}$ after a flaming boreal fire (Randerson et al., 2006; Oris et al., 2013). 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 (Turetsky et al., 2015; Eck et al., 2009). 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 combustion of peatland samples collected from interior Alaska and western Siberia. State-of-the-art, first-principle measurement techniques were used to make these measurements. With the knowledge of their optical properties, the potential im-

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pact of aerosols from peat fires on direct radiative forcing and atmospheric processes is estimated.

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

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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 (405, 532 and 781 nm) and a single wavelength (870 nm) integrating photoacoustic-nephelometer (IPN) (Arnott et al., 1999; Abu-Rahmah et al., 2006; Lewis et al., 2008), a Scanning Mobility Particle Analyzer (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 $\sim 4\pi$) 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^{-1}; aerosol flow = 0.3 L min^{-1}), 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 min 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 Elemental Carbon (EC) and OC mass fractions and concentrations using the IMPROVE-A TOR and TOT analyses method (Chow et al., 2011, 2007) implemented on a DRI Model 2001 thermal/optical carbon analyzer (Atmoslytic, Inc., Calabasas, CA, USA). In this study, the estimated EC mass in aerosol samples has been approximated to be the BC mass (Venkataraman et al., 2005). The fuel-based emission factor (see details in the Supplement), defined as the mass of a compound released per mass of

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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 (Solomon et al., 2007; Gustafsson et al., 2009; Stocker et al., 2013; Chung et al., 2012).

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 (Iinuma et al., 2007; Christian et al., 2003) 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 amounts 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 ≤ 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 and 9.2 to 16.6 g kg^{-1},

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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 near the lower end of 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 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^{-1} and CO EF of 113 ± 72 g kg^{-1} 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" (Laskin et al., 2015; Chakrabarty et al., 2010). 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 / O ratio ranging between 6 and 7. This ratio is consistent with those reported by previous studies on tar balls (Pósfai et al., 2003; Chakrabarty et al., 2006). 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 particles were agglomerates of tar ball spheres, which suggest that weak diffusion

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limited collisional growth mechanism was involved in their formation process in the smoldering fire.

Figure 2 shows the study-averaged mobility diameter number size distribution for the two fuels as measured by SMPS. For each fuel, it was observed that with increasing moisture content the total number concentration of the emitted particles decreased. Further, the median particle diameter for both fuel types was observed to decrease with increasing moisture content. For Alaskan peat burns, the study-averaged median particle diameters were 91 and 76 nm at 25 and 50 % fuel moisture content, respectively, while for Siberian peat, the median diameters were 136 and 109 nm at 25 and 50 % fuel moisture content, respectively.

Figure 3 shows the wavelength dependence of the measured MAE values, connected by best-fit curves (cubic spline), for the emitted aerosols. For Siberian peat samples, MAE values lie in the range of 0.5–0.8 $m^2\ g^{-1}$ at 405 nm and drop rapidly to 0.03–0.07 $m^2\ g^{-1}$ at 532 nm. The MAE values at 405 nm for Alaskan peat are slightly lower, in the range of 0.2–0.5 $m^2\ g^{-1}$, and exhibit a similar rapid decline at 532 nm. The observed wavelength-varying MAE trends for both fuels are consistent with those observed for brown carbon (BrC) aerosols – a class of OC aerosols absorbing strongly in the near-UV wavelengths – emitted from biomass combustion burning (Kirchstetter and Thatcher, 2012; Chakrabarty et al., 2010; Hoffer et al., 2006). The low MAE values at 532 nm for both peat types compare well with those of Indonesian peat (Chand et al., 2005).

Fitting power-law functions to our measured MAE spectra between wavelengths λ = 405 and 532 nm yielded mean Absorption Ångström exponent α values of 8.7 for both Siberian 25 and 50 % fuel moisture content peat burns, and 7.7 and 10.8 for Alaskan 25 and 50 % fuel moisture 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 (Moosmüller et al., 2009; Chakrabarty et al., 2010). Compared with previously reported α values for emissions from laboratory-combusted wildland fuels,

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emissions from peat burning characterized in this study displayed substantially higher values (Lewis et al., 2008; Gyawali et al., 2009; Lack et al., 2012). 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).

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 emitted mass of 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:

$$\frac{\mathrm{dSFE}}{\mathrm{d}\lambda} = -\frac{1}{4}\frac{\mathrm{d}S(\lambda)}{\mathrm{d}\lambda}\tau^2(\lambda)(1-F_\mathrm{c})\left[2(1-a_\mathrm{s})^2\beta(\lambda)\cdot\mathrm{MSE}(\lambda) - 4a_\mathrm{s}\cdot\mathrm{MAE}(\lambda)\right] \quad (1)$$

where $\mathrm{d}S(\lambda)/\mathrm{d}\lambda$ is the solar irradiance, τ is the atmospheric transmission (0.79), F_c is the cloud fraction (0.6), a is the surface albedo (average 0.19 for land and 0.8 for snow), 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. Note that this equation does not account for hygroscopicity, which could affect SFE. Net forcing in the 405–880 nm

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spectral range was calculated by integrating the SFE equation using the tropospheric solar spectrum (Levinson et al., 2010).

Figure 4a and b show forcing efficiencies at each wavelength over a bright surface (surface albedo of 0.8), which is representative 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 (land). 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.

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 for this region, especially since 47 % of incoming solar energy is distributed between 400 and 700 nm. 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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The Supplement related to this article is available online at doi:10.5194/acpd-15-28793-2015-supplement.

Acknowledgement. This material is based upon work supported by the National Science Foundation under Grant Nos. AGS1455215, DEB1342094, and DEB1354482; NASA ROSES under Grant Nos. NNX15AI66G and NNX15AI48G; NASA EPSCoR under Cooperative Agreement No. NNX14AN24A; and the Desert Research Institute's EDGES program.

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, Can. J. Forest Res., 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 21 century, Glob. Change Biol., 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. Tech., 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., 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, doi:10.1029/2005JD006659, 2006.

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Chakrabarty, R. K., Moosmüller, H., Chen, L.-W. A., Lewis, K., Arnott, W. P., Mazzoleni, C., Dubey, M. K., Wold, C. E., Hao, W. M., and Kreidenweis, S. M.: Brown carbon in tar balls from smoldering biomass combustion, Atmos. Chem. Phys., 10, 6363–6370, doi:10.5194/acp-10-6363-2010, 2010.

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.

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., 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, J. Geophys. Res.-Atmos., 108, 4719, doi:10.1029/2003JD003704, 2003.

Chung, C. E., Ramanathan, V., and Decremer, D.: Observationally constrained estimates of carbonaceous aerosol radiative forcing, P. Natl. Acad. Sci. USA, 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, J. Geophys. Res.-Atmos., 114, D11201, doi:10.1029/2008JD010870, 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.

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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, doi:10.1126/science.1164857, 2009.

Gyawali, M., Arnott, W. P., 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, doi:10.5194/acp-9-8007-2009, 2009.

Hoffer, A., Gelencsér, A., Guyon, P., Kiss, G., Schmid, O., Frank, G. P., Artaxo, P., and Andreae, M. O.: Optical properties of humic-like substances (HULIS) in biomass-burning aerosols, Atmos. Chem. Phys., 6, 3563–3570, doi:10.5194/acp-6-3563-2006, 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, J. Geophys. Res.-Biogeo., 115, G04002, doi:10.1029/2009JG001270, 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, J. Geophys. Res.-Atmos., 112, D08209, doi:10.1029/2006JD007120, 2007.

Jacobson, M. Z.: Studying the effects of aerosols on vertical photolysis rate coefficient and temperature profiles over an urban airshed, J. Geophys. Res.-Atmos., 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. W. and Thatcher, T. L.: Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation, Atmos. Chem. Phys., 12, 6067–6072, doi:10.5194/acp-12-6067-2012, 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, P. Natl. Acad. Sci. USA, 109, 14802–14807, 2012.

Laskin, A., Laskin, J., and Nizkorodov, S. A.: Chemistry of atmospheric brown carbon, Chem. Rev., 115, 4335–4382, 2015.

Lawrence, D. M. and Slater, A. G.: A projection of severe near-surface permafrost degradation during the 21 century, Geophys. Res. Lett., 32, L24401, doi:10.1029/2005GL025080, 2005.

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15, 28793–28813, 2015

Levinson, R., Akbari, H., and Berdahl, P.: Measuring solar reflectance – Part I: Defining a metric that accurately predicts solar heat gain, Sol. 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, D16203, doi:10.1029/2007JD009699, 2008a.
Moosmüller, H., Chakrabarty, R. K., and Arnott, W. P.: Aerosol light absorption and its measurement: a review, J. Quant. Spectrosc. Ra., 110, 844–878, doi:10.1016/j.jqsrt.2009.02.035, 2009.
Moosmüller, H., Chakrabarty, R. K., Ehlers, K. M., and Arnott, W. P.: Absorption Ångström coefficient, brown carbon, and aerosols: basic concepts, bulk matter, and spherical particles, Atmos. Chem. Phys., 11, 1217–1225, doi:10.5194/acp-11-1217-2011, 2011.
Oris, F., Asselin, H., Ali, A. A., Finsinger, W., and Bergeron, Y.: Effect of increased fire activity on global warming in the boreal forest, Environ. Rev., 22, 206–219, 2013.
Page, S., Wüst, 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, J. Quaternary Sci., 19, 625–635, 2004.
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, J. Geophys. Res., 108, 8484, 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.
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. Cy., 18, GB3004, doi:10.1029/2003GB002190, 2004.

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Simpson, I. J., Akagi, S. K., Barletta, B., Blake, N. J., Choi, Y., Diskin, G. S., Fried, A., Fuelberg, H. E., Meinardi, S., Rowland, F. S., Vay, S. A., Weinheimer, A. J., Wennberg, P. O., Wiebring, P., Wisthaler, A., Yang, M., Yokelson, R. J., and Blake, D. R.: 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_3CN, Atmos. Chem. Phys., 11, 6445–6463, doi:10.5194/acp-11-6445-2011, 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, Cambridge University Press, Cambridge, UK, 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), Cambridge University Press, Cambridge, UK, 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.

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 Air Qual. Res., 15, 2104–2114, doi:10.4209/aaqr.2015.03.0167, 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.

Venkataraman, C., Habib, G., Eiguren-Fernandez, A., Miguel, A., and Friedlander, S.: Residential biofuels in South Asia: carbonaceous aerosol emissions and climate impacts, Science, 307, 1454–1456, 2005.

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. Pyrol., 91, 263–272, 2011.

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Table 1. Mean fuel-based emission factors (rounded to nearest integer) for carbonaceous gases and aerosols for combustion of Alaskan and Siberian peat in this study.

Fuel Type	Fuel Moisture Content	Mean 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

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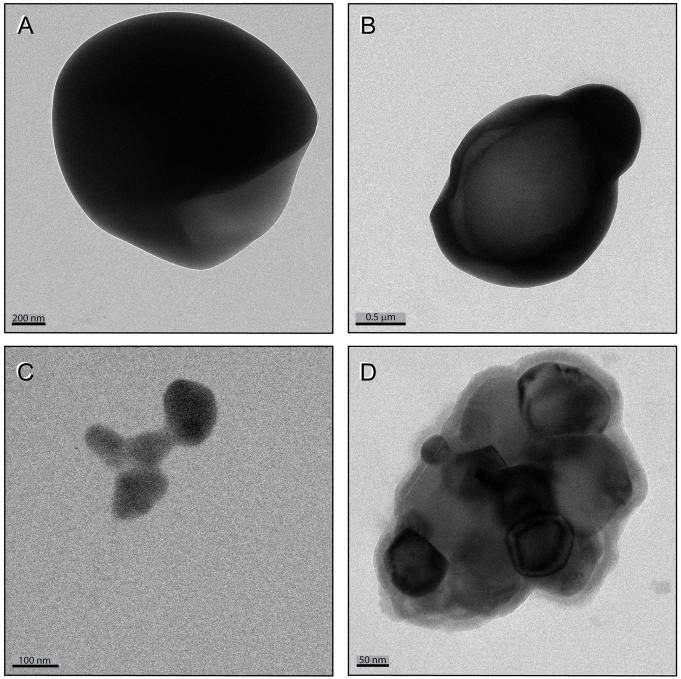

Figure 1. Transmission electron microscopy (TEM) images of typical organic carbon "tar balls", occurring as spheres and agglomerates, emitted from smoldering combustion of Alaskan and Siberian peat samples. The internal structure of these particles was amorphous in nature. Electron dispersive spectroscopy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

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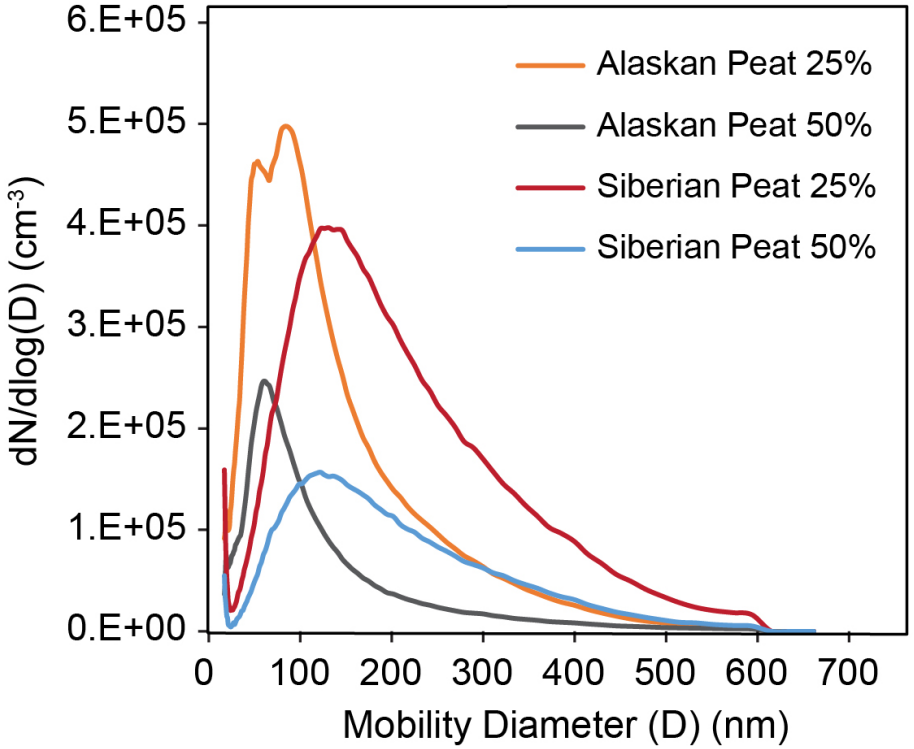

Figure 2. Study-averaged mobility number size distribution of aerosols from combustion of Alaskan and Siberian peat samples.

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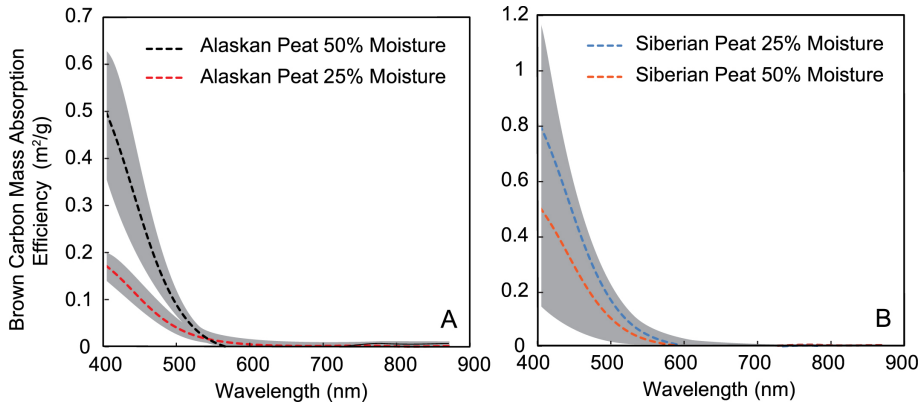

Figure 3. Wavelength-dependent mass absorption efficiency (MAE) of the sampled carbonaceous (brown) aerosols from **(a)** Alaskan and **(b)** Siberian peat smoldering combustion. The dashed lines show study-averaged values connected by best-fit curves. The shaded bands correspond to error bars measured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

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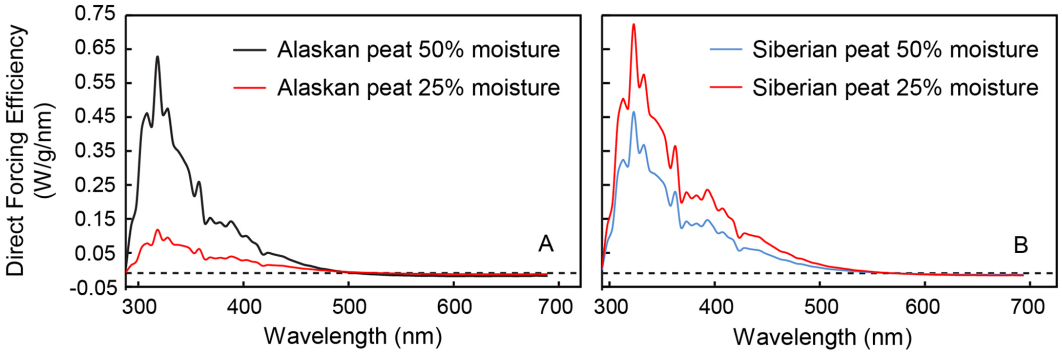

Figure 4. Direct forcing efficiency of brown carbon aerosols above a bright surface (surface albedo = 0.8). Integrated mean forcing is 20 and 38 $W g^{-1}$ for Alaskan and Siberian peat smoke aerosols, respectively.

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