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Fire in ice: two millennia of Northern Hemisphere fire history from the Greenland NEEM ice core

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Abstract

Biomass burning is a major source of greenhouse gases and influences regional to global climate. Pre-industrial fire-history records from black carbon, charcoal and other proxies provide baseline estimates of biomass burning at local to global scales, but there remains a need for broad-scale fire proxies that span millennia in order to understand the role of fire in the carbon cycle and climate system. We use the specific biomarker levoglucosan, and multi-source black carbon and ammonium concentrations to reconstruct fire activity from the North Greenland Eemian (NEEM) ice cores (77.49° N; 51.2° W, 2480 m a.s.l.) over the past 2000 years. Increases in boreal fire activity (1000–1300 CE and 1500–1700 CE) over multi-decadal timescales coincide with the most extensive central and northern Asian droughts of the past two millennia. The NEEM biomass burning tracers coincide with temperature changes throughout much of the past 2000 years except for during the extreme droughts, when precipitation changes are the dominant factor. Many of these multi-annual droughts are caused by monsoon failures, thus suggesting a connection between low and high latitude climate processes. North America is a primary source of biomass burning aerosols due to its relative proximity to the NEEM camp. During major fire events, however, isotopic analyses of dust, back-trajectories and links with levoglucosan peaks and regional drought reconstructions suggest that Siberia is also an important source of pyrogenic aerosols to Greenland.

1 Introduction

Fire influences regional and global climate through the emission of greenhouse gases and particulates that reflect and absorb incoming solar radiation (Ramanathan and Carmichael, 2008; Bowman et al., 2009; IPCC, 2007). Biomass burning emits up to 50 % as much CO_2 as fossil fuel combustion (Bowman et al., 2009) thereby affecting the climate system. Fire products such as black carbon (BC) have a radiative

absorption forcing up to 55 % that of CO_2 and a greater influence than other greenhouse gas forcings including methane (CH_4), chlorofluorocarbons, nitrous oxide and tropospheric ozone (Ramanathan and Carmichael, 2008; Jacobson, 2004; van der Werf et al., 2004; Running, 2006; McConnell et al., 2007). Combined direct and indirect effects rank BC as the second-largest contributor to globally averaged positive radiative forcing since the pre-industrial period (Ramanathan and Carmichael, 2008; Randerson et al., 2006). Estimates of the radiative forcing of combined biomass burning aerosols are still not well defined but are in the range of $+0.03 \pm 0.12\,W\,m^{-2}$ (IPCC, 2007). Human activities may have changed the net negative radiative forcing of pre-industrial fires ($-1.0\,W\,m^{-2}$) to approximately $-0.5\,W\,m^{-2}$ (from 1850 to 2000 CE) and potentially to $-0.8\,W\,m^{-2}$ (from 1850 to 2100 CE) (Ward et al., 2012).

Fire influences the climate system, but in turn centennial-scale Holocene fire variations are influenced by climate (Marlon et al., 2013; Power et al., 2008). Precipitation affects fuel flammability, where conditions must be wet enough to allow biomass to grow, and dry enough to allow combustion (Pyne, 2001). Increased temperatures and atmospheric CO_2 permit greater plant productivity and could result in greater fuel availability and hence increased fire activity (Marlon et al., 2008; Daniau et al., 2010). Human activities may also be a potential influence on trends in biomass burning (Marlon et al., 2008). Fire ignition is caused by natural sources such as lightning but human activities, including household fires and slash-and-burn agricultural practices, are increasingly altering global fire activity (FAO, 2007). The observable impact of humans on fire regimes differs by geographic region (McWethy et al., 2009; Marlon et al., 2012; Power et al., 2008) but over the course of the 20th Century human activity began to influence the global fire regime more than natural causes (Marlon et al., 2008; Pechony and Shindell, 2010).

There are many potential indicators or proxies of past fire activity (Conedera et al., 2009). Some proxies are produced solely from biomass burning, like charcoal, but most others, such as lightweight carboxylic acids, lignin and resin pyrolysis products, and major ions (K^+, NH_4^+ and NO_3^-) are have multiple potential sources other than wildfires

(e.g. coal burning, volcanic eruptions or biogenic emissions) (Eichler et al., 2011; Yalcin et al., 2006). Potassium (K^+) and ammonium (NH_4^+), for example, have traditionally been used in ice core studies as tracers of past fire events, but these ions are also emitted from numerous other sources (Eichler et al., 2011; Yalcin et al., 2006) and thus require to be corrected accordingly in order to provide source-specific information. NH_4^+, nitrate (NO_3^-), H_2O_2, and organic acids in Arctic ice are often associated with biomass burning events, where NO_3^- in particular tends to increase in samples with higher NH_4^+ concentrations (Fuhrer et al., 1996; Whitlow et al., 1994), however these indicators are also a result of complex chemical reactions that are not necessarily a direct reflection of variations in biomass burning.

Black carbon (BC) is another common fire proxy that occurs during both natural and anthropogenic processes and is formed during incomplete combustion of fossil and biofuels (Preston and Schmidt, 2006; McConnell et al., 2007). Black carbon is not a single chemical compound, however, and lacks well-defined characteristics (Goldberg, 1985; Masiello, 2004). The isotopic ratio of atmospheric gases in ice cores has been used as fire proxy, but such ratios are also not solely products of biomass burning (Sapart et al., 2012; Wang et al., 2012).

Monosaccharide anhydrides are one of the few fire proxies that have specific sources (Gambaro et al., 2008). Levoglucosan (1,6-anhydro-β-D-glucopyranose) is a monosaccharide anhydride released during biomass burning when cellulose combustion occurs at temperatures $> 300\,^{\circ}C$ (Gambaro et al., 2008; Simoneit, 2002). As with BC and NH_4^+, levoglucosan is injected into the atmosphere in convective smoke plumes, deposited on glacier surfaces through wet and dry deposition, and is preserved in snow and ice (Gambaro et al., 2008; Kehrwald et al., 2012; McConnell et al., 2007; Fuhrer and Legrand, 1997).

Unlike greenhouse gases, levoglucosan, NH_4^+ and BC are not homogeneously distributed in the atmosphere due to their short lifetimes (on the order of days to weeks) (Hennigan et al., 2010; Fraser and Lakshmanan, 2000; Ramanathan and Carmichael, 2008; Fuhrer and Legrand, 1997) and efficient removal processes from

the atmosphere. Although levoglucosan is oxidized by OH radicals in the gas phase (Hennigan et al., 2010) and in atmospheric water droplets (Hoffmann et al., 2010), its high concentration in biomass emissions implies that levoglucosan is a strong potential tracer for broad-scale fire activity (Fraser and Lakshmanan, 2000; Holmes and Petrucci, 2006, 2007; Kehrwald et al., 2012).

Here, we use a multiproxy approach to reconstructing biomass burning in the northern [high latitudes/hemisphere] during the past 2000 years. Specifically, we report levoglucosan and ammonium concentrations in the upper part of the deep North Greenland Eemian (NEEM) ice core as well as BC measured in the 410 m NEEM-2011-S1 core (Fig. 1) which was collected adjacent to the deep core in 2011 (Sigl et al., 2012) to reconstruct fire activity for the past 2000 years. We compare our results with Northern Hemisphere fire records, climate conditions obtained from historical records and paleoarchives to identify sources of and controls on fire emissions registered in NEEM. Our approach acknowledges the weaknesses inherent in most fire proxies used in ice core studies and remedies this by integrating the results from multiple fire proxies to identify the robust variations in past biomass burning during the past 2000 years.

2 Methods

2.1 Ice core sampling and analyses

The international North Greenland Eemian (NEEM) ice core drilling site (Fig. 1) is located in northwest Greenland (77.49° N; 51.2° W, 2480 m a.s.l., mean annual temperature $-29\,^{\circ}C$, accumulation 0.22 m ice equivalent $year^{-1}$, 2540 m final depth). The deep NEEM ice core (Fig. 1) was drilled from 2008 to 2012. Ice core analyses were partly performed in the field and partly in the laboratories of the participating nations. Our levoglucosan dataset consists of 273 samples from 4.95–602.25 m depth (1999 CE–1036 BCE), where each sample is a 1.10 m inner core section collected using the Continuous Flow Analysis (CFA) (Kaufmann et al., 2008) system at the NEEM camp. Samples

for levoglucosan determination were transported in a frozen state from the field to Ca' Foscari University of Venice laboratory where they were stored in a −20 °C cold room until analysis. We slightly modified the analytical procedure for determining levoglucosan (Supplement) using liquid chromatography/negative ion electrospray ionization – tandem mass spectrometry (HPLC/(−)ESI-MS/MS) at picogram per milliliter reported by Gambaro et al. (2008) (Supplement). This analytical method has the advantage of allowing the direct determination of levoglucosan by introducing the melted sample and $^{13}C_6$ labeled internal standard into the HPLC instrument. Preanalytical procedures (analyte extraction and purification) are avoided and sample contamination minimized. NH_4^+ and the other major ions were measured by CFA on the deep NEEM core directly in the field during season 2009 (Kaufmann et al., 2008). All the major ions are available at mm resolution, and we calculated 1.1 m mean values in order to directly compare with levoglucosan.

The upper 1419 m of deep NEEM core $d^{18}O$, volcanic fingerprints, Electrical Conductivity Measurement, Dielectrical Profiling, and impurity records can be matched to the NGRIP GICC05 extended time scale (Wolff et al., 2010b; Rasmussen et al., 2013). Due to the ice compression and differences in Greenland snow accumulation, each 1.10 m sample covers 1 to 5 year-long temporal periods. The temporal interval has an associated age error of ±1 year from the top of the ice core to the depth of 294.25 m (698 CE) and from 338.25 to 536.8 m (468 CE–645 BCE). An error of ±2 years exists from 295.35 to 336.05 m depth (692–480 CE) and from 541.20 to 602.25 m (671–1036 BCE). No official age model is currently available for the upper section (0.00–19.80 m) of the NEEM ice core. The uppermost 20 m of the age scale was calculated using a Herron–Langway firn densification model fit to the NEEM-NGRIP tie points for the 20–80 m long firn column (Herron, 1980).

Continuous flow analysis allows sub-seasonal reconstructions of BC in the NEEM-2001-S1 core (Sigl et al., 2012) and other Greenland ice cores (McConnell et al., 2007; McConnell and Edwards, 2008; McConnell, 2010). The NEEM-2011-S1 core was collected adjacent to the deep NEEM core (Fig. 1) in summer 2011 and the 410 m long

core was transported to the Desert Research Institute in Reno, NV, USA. The core was cut into ∼ 1.1 m long samples with cross section of ∼ 0.033 m by 0.033 m. These longitudinal samples were analyzed in early autumn 2011 using a well-established continuous ice core analytical system (Banta et al., 2008; McConnell, 2002) including determination of BC concentrations using methods described in McConnell et al. (2007) and McConnell and Edwards (2008). Annual layer counting based on seasonal variations in a number of elements and chemical species, constrained by the known volcanic markers in the deep NEEM and other ice cores, was used to determine the age model (Sigl et al., 2012). BC was measured using a continuous ice core melter system (McConnell and Edwards, 2008; McConnell et al., 2007) in the 410 m NEEM-2011-S1 core collected adjacent to the deep NEEM core in 2011 (Fig. 1). We present 1921 BC concentrations from the NEEM-2011-S1 cores, spanning the ages 78–1997 CE. A number of seasonally varying elements and chemical species were used to count annual layers in the NEEM-2011-S1 core, resulting in minimal dating uncertainty (Sigl et al., 2012) as for the deep NEEM core dating (Supplement).

As the NEEM-2011-S1 and deep NEEM ice cores have different sampling schemes, this difference means that BC samples and levoglucosan samples represent similar, but not equal, ages at the same depth ranges. BC is sampled at a higher resolution (annual resolution) than the levoglucosan (one sample continuously covering 1.10 m of the deep NEEM ice core). In order to avoid temporal resolution loss and the introduction of errors, we chose not to calculate BC averages from NEEM-2011-S1 core over the same temporal interval covered by the deep NEEM core when seeking to compare pyro-proxies from the two cores. We therefore only present annual BC concentrations.

2.2 Statistics

One of the first challenges to understanding that levoglucosan reflects/responds to variations in fire activity, is to assess its relationship with transport variability. Dust and Ca^{2+} concentrations can be used as crustal particulate tracers in ice cores, provide clues into the relative strength of their source regions, transport variability and transport strength

(Fischer et al., 2007; Obrien et al., 1995; Wolff et al., 2010a). The closest source regions for the Greenland dust are boreal Canada and boreal Russia, which are similar landscapes with similar availability of entrainable dust. The Holocene contribution of sea salts to total Greenland Ca^{2+} ice concentrations is estimated to be in the order of 10 % (Fischer et al., 2007). In order to keep the data as close to its original form as possible, and therefore to avoid including possible error due to data transformation, we use Ca^{2+} as a dust tracer without correcting for sea salt contribution.

2.3 Short- to long-term fire variation

For punctual analysis, we consider *megafires* as levoglucosan peaks with a Z-score ≥ 1 [Z-score = $(x_i - \overline{x})/\sigma$)], corresponding to a concentration ≥ 245 pg mL^{-1} (Table 1, Fig. S5 in the Supplement). Multi-decadal fire activity was extracted from the levoglucosan and BC concentration profiles by analyzing smoothed data after removing spikes. We tested different approaches in order to determine multi-decadal fire activity from levoglucosan concentrations. The challenge was to find a suitable statistical tool capable of identifying temporal trends that are not overly affected by the high peaks of anomalous events. The Global Charcoal Database (GCD) archives hundreds of sedimentary records of fire (Power et al., 2010). Regional and global synthesis allows the examination of broad-scale patterns in paleofire activity (Marlon et al., 2013). In order to compare levoglucosan data with decadal to centennial trends in other paleoclimate records, we first applied standardized statistical procedures based on those used to analyze the Global Charcoal Database (GCD) (Marlon et al., 2008; Power et al., 2008), which is a robust method for summarizing different datasets from various environmental archives. We modified this method for the NEEM data as NEEM is a single site and incorporating the outlying values results in artifacts in the LOWESS smoothing. A common approach to isolate the influence of peaks is to fix a threshold and separately study those values. We differ from the GCD procedure in our treatment of individual spikes, as these strongly affect multi-decadal trends, even when using a LOWESS regression model (details are included in the Supplement).

3 Results

Pearson and Spearman correlations (Supplement) demonstrate that, of the possible biomass burning-related products measured in the deep NEEM core, levoglucosan concentrations only slightly correlate with ammonium. No correlation is noted between levoglucosan and crustal marker Ca^{2+} or dust. Preliminary results of ICP-MS analyses on the deep NEEM ice core support the lack of correlation between other crustal markers (i.e. Ti and Ba) and levoglucosan (J. Gabrieli, personal communication, 2014). A similar correlation analysis of BC and other elements and chemical species (data not shown) measured in the NEEM-2011-S1 core indicates that during pre-industrial periods BC concentrations generally do not correlate with crustal particulate tracers but do with NH_4^+ and NO_3^-.

Megafires (levoglucosan peaks with Z-scores > 1) are inferred at 1973, 1789, 1704, 1620, 1039, 976, 922, 628, 342 CE and at 199, 274, 330, 392 and 540 BCE (Fig. 2). Other major peaks are reported in Table 1. Distinct levoglucosan spikes are also synchronous with major peaks in the BC record (BC Z-scores > 3) at 1704, 922, 628 and 342 CE and minor peaks (with BC Z-scores > 1) at 1973, 1789, 1620 and 976 CE, respectively. The levoglucosan peak at 342 CE (Z-score = 10.9) also occurs in the BC data, where the Z-score is 19.0 (Table 1).

Strong fluctuations in the temporal profile dominate the record from decadal to centennial perspectives. Relative maxima in the levoglucosan profile are evident between 1000–1300 CE and 1500–1700 CE, and with a lesser extent around 500 CE and 100 CE, while the lowest fire activity is evident around 700–900 CE and with a lesser extent around 1300–1500 CE (Fig. 3a). Smoothed BC concentration is high from 100 to 700 CE, and after an abrupt decrease it offers the lowest values between 700 and 900 CE. Between 1000–1600 CE and 1600–1800 CE the BC profile show a relative maximum and minimum, respectively, when the absolute maximum in BC concentration peaks around 1900 CE.

4 Discussion and interpretation

4.1 Biomass burning tracers in the NEEM ice core

4.1.1 Correlation with major ions

Dust, major ions and levoglucosan may not have the same source regions and/or transport history, but comparing these proxies may provide insight into common or differing air masses affecting the NEEM site. Increased wind speeds result in greater dust concentrations and larger particles in ice cores (CITE). Stronger winds may be expected to also increase levoglucosan concentrations as the increased wind strength could transport more biomass burning products in air plumes. However, the lack of correlation between Ca^{2+}, dust and levoglucosan suggests that levoglucosan variability is not dominated by changes in transport or in changes in wind strength. This evidence differs from Kawamura et al.'s (2012) conclusion, which speculated that enhanced atmospheric transport rather than an increase in fire activity may affect levoglucosan concentrations.

Ammonium has multiple anthropogenic and natural sources, mainly emissions from soil and vegetation and thus, background ammonium values in polar ice are strongly linked to temperature changes (Fuhrer et al., 1993, 1996; Legrand et al., 1992; Fuhrer and Legrand, 1997). Ammonia (NH_3), is the most abundant gaseous base in the atmosphere and can be incorporated into acidic aerosols and converted to NH_4^+. During volcanic eruptions, when vast amount of sulfate is emitted to the atmosphere, NH_4^+ will irreversibly form NH_4HSO_4 and $(NH_4)_2SO_4$ which is then deposited on the ice. Therefore, peak concentrations in NH_4^+ measured during volcanic eruptions are most likely not directly linked to biomass burning. Basing NEEM fire reconstructions only on ammonium concentrations that are not corrected for the natural variability and volcanic eruptions could potentially overestimate major fire events, however, comparing various fire proxies from the same core supplements biomass burning interpretations. The literature uses Principal Component Analysis for pyro-reconstructions (Yalcin et al., 2006;

Eichler et al., 2011). Incorporating as many fire proxies as possible within the same ice core allows a more robust reconstruction of past burning, but eigenvectors incorporate other chemical species that, even if they are fire-related compounds, are not fire tracers (NO_3^-, H_2O_2). We therefore compare individual fire tracers in NEEM rather than using Principal Component Analysis as a biomass burning reconstruction.

4.1.2 Extreme fire events

Levoglucosan data exhibit high variability (Fig. 2) due to the absence of background atmospheric levoglucosan values. Levoglucosan is only injected into the atmosphere during biomass burning events; no continuous emission processes exist. Individual levoglucosan spikes are therefore likely generated either by short "megafires" or by localized fire events rather than by long-term increases in fire activity (Figs. 2 and S5 in the Supplement. Furthermore, the mean accumulation rate in north central Greenland can be considered constant over the past 3000 years (Andersen et al., 2006), so we present levoglucosan and BC concentrations rather than fluxes.

Many levoglucosan spikes correspond with documented fire activity in Greenland or in other Northern Hemisphere ice cores, and are generally supported by the other pyro-proxies measured in the NEEM ice core (Table 1). The majority of fire events inferred from the NEEM ice core were simultaneously recorded by levoglucosan and BC. NH_4^+ spikes replicate many of the levoglucosan spikes (8 of the 14 levoglucosan spikes with Z-score > 1). Only the 1039 CE levoglucosan peak is not duplicated by any of the other proxies (Table 1). The peak at 342 CE contains by far the highest concentrations of both levoglucosan and BC present in our datasets, and corresponds to the largest fire event in the past 2000 years recorded by several charcoal records in the Northern US and Canadian Rockies (Brunelle et al., 2005; Hallett et al., 2003). Thousands of Russian firemen, soldiers and farmers were mobilized to suppress a fire raging for several weeks in August 1972 in a bog-land east of Moscow (The Palm Beach Post, 9 August 1972; The New York Times, 9 August 1972), and during the same week more than 1000 km^2 of forest burned in Central Alaska (The Milwaukee Journal, 9

August 1972). The 1972 CE event in Russia is synchronous with and probably related to an anomalous heat wave and severe droughts (Dronin and Bellinger, 2005; Golubev and Dronin, 2004).

We cannot exclude the possibility that local phenomena in Greenland, rather than extensive events, may be responsible for some of the large levoglucosan spikes. Several levoglucosan spikes were recorded in a relatively brief period between ~ 920 and 1110 CE. This period coincides with the foundation of Viking settlements in Greenland beginning in 982 CE in southwest Greenland, relatively close to the NEEM site. Historical records show that Greenland Viking settlements burned vast quantities of wood for extracting iron from bogs for tool production and these local, not necessarily extensive, fires may account for the levoglucosan peaks during this time period (Diamond, 2005). Future trace metal analyses of the NEEM ice core can help demonstrate if relatively local metal production may have caused these levoglucosan peaks.

Other ice core records provide comparisons of fire events that allow an examination of the spatial extent of past short-lived fire activity. A prominent 1617–1622 CE levoglucosan peak coincides with high fire activity inferred from oxalic acid analyses recorded in the Greenland Site J ice core (Kawamura et al., 2001). No historical documentation is available for the 1787–1791 CE peak but contemporaneous fire signatures are reported in Greenland (GISP2, G 20) and Canadian ice cores (Eclipse Icefield and Mt. Logan) (Yalcin et al., 2006; Whitlow et al., 1994). The strongest fire activity recorded in the Ushkovsky (Kamchatka) ice core occurs in 1705, 1759, 1883, 1915, 1949 and 1972 CE (Kawamura et al., 2012). Megaevents are present as both levoglucosan and BC peaks in NEEM in 1704 and 1973 CE and as a BC peak in 1914.5 and 1947.5 CE (Fig. 2).

4.1.3 Decadal to centennial fire activity variability

The NEEM levoglucosan maximum (1500–1700 CE, Fig. 3a), the highest of the past two millennia, coincides with data from the Belukha ice core (Eichler et al., 2011) that documents a period of increased fire activity between 1550–1700 CE where

1600–1680 CE records the highest forest fire activity over the last 750 years (Fig. 3d). High fire activity between 1500–1600 CE was also inferred from formate and ammonium analyses on the EUROCORE, Greenland ice core, when "high frequency and low intensity of fires" were synchronous with dry climate conditions (Savarino and Legrand, 1998).

Due to multiple anthropogenic BC sources after the Industrial Revolution, we cannot directly compare BC and levoglucosan after 1850 CE but we can compare BC between NEEM and other Arctic sites and use our data to discriminate fire, volcanic or anthropogenic emissions. The maximum BC concentration (16 ng g^{-1}) in the entire Greenland D4 ice core occurred in 1908 CE (McConnell et al., 2007). The second-highest BC peak (15 ng g^{-1}) over the past millennium in the NEEM-2011-S1 is dated to 1909.5 CE (Fig. 2b). This timing is relatively coincident with the Tunguska Event, a bolide impact in western Siberia occurring in June 1908. An ammonium spike in 1910 CE in the GISP2 ice core (Taylor et al., 1996) was attributed to large conflagrations in the northern United States and southern Canada, supported by a synchronous extremely limited growth of regional trees (drought proxy) in the Northwestern Ontario (St. George et al., 2009). The absence of levoglucosan and ammonium spikes in the NEEM ice core or vanillic acid peaks, a source-specific tracer for conifers (Simoneit, 2002), in the D4 ice core supports the hypothesis that these BC peaks were not caused by forest fires. Rather, comparisons of BC and toxic heavy metals in the ACT2 core from southern Greenland (McConnell and Edwards, 2008), as well as BC and non-sea-salt sulfur in the D4 ice core from central Greenland (McConnell et al., 2007), indicate that > 80 % of the BC during this period is associated with industrial emissions (primarily coal burning). The D4 site demonstrates a gradual rise in BC concentrations after 1850 CE and a rapid increase after 1888 CE, before beginning a gradual decline through the late 1940s followed by a clear drop in 1952 CE (McConnell et al., 2007). This trend is generally confirmed by BC in the NEEM-2011-S1 ice core, where BC shows a peak that slowly increases from ~ 1750 CE, followed by a sharp enhancement during the late 1800s (Fig. 3c).

Biomass burning was the major source of BC to central Greenland before 1850 CE, as revealed by the close correlation between BC and vanillic acid, an organic combustion marker released by coniferous trees (McConnell et al., 2007). After 1850 CE, the influence of industrial emissions, particularly coal, as a source of BC is clear through comparisons to non-sea-salt sulfur (McConnell et al., 2007) and industrial heavy metals such as lead, thallium, cadmium in the southern Greenland ACT2 core (McConnell and Edwards, 2008). The NEEM-2011-S1 BC record contains its highest concentrations from 1850 CE to the present, reflecting the input of these new sources.

The levoglucosan profile contains moderately high mean values during the 1900s, but other centennial-scale peaks, such as that centered ~ 1600 CE have higher concentrations (Fig. 2a). Black carbon has an average atmospheric lifetime of a few days to one week (Ramanathan and Carmichael, 2008; Bauer et al., 2013; McConnell et al., 2007). The diverse range of BC emission sources and the different atmospheric lifetime of levoglucosan may result in BC representing a separate emission source area than the levoglucosan record. Levoglucosan can not be used to quantitatively characterize BC in environmental samples, since levoglucosan yield is variable and strongly depends on biomass composition and combustion temperature (Kuo et al., 2008). In addition, different temporal resolution for BC and levoglucosan records may result in differences after removing spikes and applying LOWESS smoothing. Levoglucosan can only be produced by cellulose pyrolysis, thus the different pattern between the records post-1850 CE mainly reflects the differences between biomass burning and industrial activity.

4.2 Comparison with other pyro-reconstructions

4.2.1 Charcoal

The Global Charcoal Database compile individual charcoal records (Fig. S6 in the Supplement) into global or regional reconstructions that provide a benchmark against which other biomass burning records can be compared (Power et al., 2010; Marlon

et al., 2008). The charcoal database covers all climatic zones and all major biomes, but data for grasslands and dry shrublands, where low woody biomass limits charcoal production, are limited due to a lack of suitable sampling sites (Marlon et al., 2008). These vegetation types emit large quantities of levoglucosan when burned (Iinuma et al., 2007; Gao et al., 2003) resulting in a possible offset between charcoal and levoglucosan data. Many charcoal sampling sites are located in the US and Europe, and a limited number of GCD sites currently exist in northern Asia (Fig. S6 in the Supplement). As levoglucosan and BC can be transported hundreds to thousands of kilometers (Mochida et al., 2010), they complement GCD reconstructions that record combustion within tens of km of the sampling sites, and provide fire activity data for regions where charcoal records do not exist.

The high latitude Northern Hemisphere HLNH > 55° GCD composite curve registers a global long-term decline in fire activity from 0–1750 CE (Marlon et al., 2008). A decreasing amount of ammonium originating from biomass burning is also deposited in the GRIP ice core (Fuhrer et al., 1996) but this trend is not is not clearly supported by levoglucosan data during the past two millennia (Fig. 3b). The prominent levoglucosan minimum between 700 and 900 CE coincides with a minimum in the BC profile and HLNH > 55° GCD (Fig. 3). The HLNH > 55° GCD and levoglucosan data differ near 1600 CE when the HLNH > 55° GCD demonstrates a modest increase in fire activity, while the smoothed NEEM levoglucosan strongly peaks (Fig. 3b). This period of decadal-scale increased biomass burning is seen in both the levoglucosan and BC records in the NEEM ice cores, including individual levoglucosan peaks with Z-scores up to 2.8 (Table 1). We infer that the 1500–1700 CE maximum in fire activity is due to increased Eurasian boreal forest fires (Fig. 3), which are underrepresented in the GCD, reflecting the bias in the geographical distribution of GCD sites. Over 50 high latitude North American sites exist yet Siberia only has less than 10 lake cores > 60° N which span a distance of over 7000 km.

4.2.2 Isotopic composition of CH_4

The isotopic composition of methane ($\delta^{13}C$ of CH_4) in ice cores differentiates biogenic from pyrogenic sources of biomass burning. Methane is stable in the atmosphere for longer than the atmospheric exchange time and therefore is globally distributed. Hence $\delta^{13}C$ of CH_4 variations are impacted by sources distributed all over the world (Blunier et al., 2007). The natural diffusion of CH_4 in the firn column results in low-frequency CH_4 variations throughout ice core records (Spahni et al., 2003). Sapart et al. (2012) argue that changes in the $\delta^{13}C$ of CH_4 record cannot be explained without biomass burning variability. Methane isotopic variations during the past two millennia (Fig. 3f) correlate with anthropogenic activities, which have contributed to the methane budget before industrial times (Sapart et al., 2012). Methane emissions may be linked to early industrial activity including copper and lead smelting but levoglucosan is not released during coal combustion needed for early industrial activities. Despite the differences in sources and their geographical distribution, the NEEM $\delta^{13}C$ of CH_4 isotopic record contains similar features to the levoglucosan data (Fig. 3f). High $\delta^{13}C$ values between 800–1200 CE, and to a lesser extent from 100 BCE until 200 CE, corresponds with increased levoglucosan and BC concentrations. The pronounced peak centered on ~ 1600 CE only partially overlaps with the pyrogenic methane record, where $\delta^{13}C$ peaks in the 1400s to 1500s.

4.3 Source of NEEM fire products

Stable oxygen isotope ($\delta^{18}O$) data demonstrate that the circulation pattern of air masses reaching the Greenland ice sheet did not significantly change over the last 10 000 years (Vinther et al., 2006). We therefore assume that the atmospheric transport of biomass burning plumes should not have significantly changed during the past two millennia. The isotopic composition of dust in ice cores helps determine the geographic origin of particulate matter reaching Greenland. Holocene mineral dust in the NGRIP (North Greenland Ice core Project) ice core has an Asian origin with the Gobi

and Taklimakan deserts as the best source candidates (Bory et al., 2002, 2003; Lupker et al., 2010). These studies argue that North America and North Africa are not potential dust sources, as both regions have higher K/C ratios and higher smectite contents than dust extracted from NGRIP. Although mineral dust and biomass burning products are subject to different transportation styles and atmospheric interactions, these dust studies demonstrate the influence of Asian sources on material transported to the Greenland ice sheet. The Gobi and Taklimakan deserts have very little vegetation and so likely are not biomass burning sources, but these deserts border boreal forest regions that may be important biomass burning sources during major fire events, as discussed throughout this section (Table 2).

Earth's boreal forests cover 9–12 million km^2 (Fig. 1), extending across Scandinavia, Russia and North America (NA), representing approximately 10 % of Earth land cover and one third of the total global forested area (Zhang et al., 2003; Canadian Forest Canadian Forest Service, 2005). Boreal forest forms a green belt just below the Arctic Circle (55° to 70° N) interrupted only by the Pacific and Atlantic Oceans, and thus forms the major fuel source for fire emissions reaching the Greenland ice sheet. We consider both coniferous and deciduous forests at ≥ 55° N as boreal forests (Fig. 1). Russian forests alone represent ~ 25 % of global terrestrial biomass (Zhang et al., 2003). Two thirds of boreal forests are located within the 17 million km^2 of the Russian Federation (Zhang et al., 2003). Russian boreal forests have a high fire risk due to accumulated organic matter with slow decomposition rates and boreal regions have low precipitation during the summer fire season (Damoah et al., 2004). Canadian boreal forests comprise about one third of the NH circumpolar boreal forest (Canadian Forest Service, 2005).

Arctic snow samples across the Greenland ice sheet and associated back-trajectories from a single year highlight the importance of Russian boreal forests as a biomass burning source and suggest that 55 % of modern BC reaching the Greenland ice sheet originates from Russian biomass burning, while 40 % is from North America, and only ~ 5 % is from anthropogenic emissions (Hegg et al., 2009). A 44-year record

of 10 day isobaric back-trajectories reaching Summit, Greenland (Kahl et al., 1997), indicates that more summer (fire season) trajectories originate over North America (46 % at 500 hPa and 85 % at 700 hPa) than from Eastern Asia (20 % at 500 hPa) or from Europe and Western Asia (only 6 %). A model intercomparison demonstrates that North America and Europe each contribute 40 % of the total anthropogenic BC deposited on Greenland (Shindell et al., 2008). These models only investigate anthropogenic sources and explicitly state that biogenic Siberian emissions are not included in their estimates. Back trajectory modeling demonstrates that the lack of correlation between BC and the specific coniferous fire marker vanillic acid measured in the Greenland D4 ice core is likely a result of the majority of BC arriving to Greenland between 1850 to 1950 CE having a North American anthropogenic source, and from 1951 CE to the present having an Asian industrial source, separate from any fire activity (McConnell et al., 2007). Therefore, any analysis for Greenland BC from 1850 CE onward should not be considered a purely biomass burning indicator, and the sources for BC and fire emissions may substantially differ.

These back-trajectory and model analyses suggest that North America and Siberian forests are the dominant sources of biomass burning aerosols. A first-order approximation of aerosol concentrations along a trajectory is function of time where aerosol concentrations decrease exponentially with transport time (Fischer et al., 2007; Hansson, 1994). North America is likely the most important contributor due to the proximity to the NEEM location and the shorter routes travelled by aerosols reaching Greenland with respect to Siberian–Eurasian forests, but previous studies also demonstrate that air masses originating in Asia reach Greenland after a few days. Levoglucosan and BC atmospheric lifetimes are on the order of days to less than two weeks (Fraser and Lakshmanan, 2000; Hennigan et al., 2010), suggesting that these back-trajectory and model analyses include all possible levoglucosan and BC source regions. Asia has generally been ignored as a biomass burning aerosol source for Greenland due in part to the days required for air mass travel time compared to the aerosol atmospheric lifetimes. Atmospheric aerosol scavenging and seasonal circulation changes where more

air masses originating from Asia pass over Greenland during the winter months (Kahl et al., 1997) limit the quantity of boreal Asian biomass burning products incorporated into the Greenland ice sheet. However, the back trajectory analyses do demonstrate that although Asia is a lesser source, it is a possible aerosol source. Back-trajectories demonstrate that although many air masses originating from Siberia do have to travel eastward to reach Greenland, others have a north-west trajectory that substantially shortens the necessary travel distance (Kahl et al., 1997).

The correspondence of the major NEEM levoglucosan peaks with periods of extreme droughts in northern central Asia (Table 2) suggests that Asia may be an important fire source during major fire events while North America may be a more important source for background fire activity. Boreal wildfires can generate sufficient sensible heat during the combustion process to initiate deep convection and inject particles into the upper troposphere and lower stratosphere resulting in longer atmospheric lifetimes and more efficient transport of biomass burning aerosols (Damoah et al., 2004; Trentmann et al., 2006; Dentener et al., 2006; Hodzic et al., 2007). Regional droughts result in large amounts of available deadwood as fuel resulting in intense fires capable of generating deep convection (van Mantgem et al., 2009; Westerling et al., 2006; Trentmann et al., 2006; Hodzic et al., 2007). The correspondence between regional droughts and increased fire activity is present in multiple northern and central Asian proxies (Table 2). The Ushkovsky (Kawamura et al., 2012) and Belukha (Eichler et al., 2011) ice cores both contain up to multi-decadal periods of increased fire activity that are similar to peaks in the NEEM ice core suggesting that these sites may receive a contribution from Siberian fire activity (Fig. 3). Our work determines that Siberia is an important source of burning signatures inferred in Arctic ice fields during extreme fire events.

4.4 Climatic and anthropogenic influences on decadal to centennial changes in fire activity

4.4.1 Natural climate interactions

Fire frequency and intensity depend on regional climatic and environmental influences, where vegetation type, temperature and precipitation are the most important factors. Vegetation distribution impacts levoglucosan emission factors (the amount per kg of burnt fuel) and combustion characteristics (smoldering vs. flaming fires) (Simoneit et al., 1999; Engling et al., 2006). Large-scale variations in vegetation types likely do not cause the observed centennial oscillations, but may affect fire signatures over millennial timescales. Instead, elevated surface air temperatures and sustained drought affect fuel flammability and lead to increased global fire activity over seasonal to centennial timescales (Daniau et al., 2012).

4.4.2 Temperature

Temperature anomalies during the past 2000 years are not globally synchronous. The regional pattern of the Medieval Warm Period (MWP) results in differing times of maximum warming by region. Global temperature anomalies were highest between 950–1250 CE (Mann et al., 2009), whereas Northern Hemisphere (and especially Russian) temperatures were elevated between 1000–1300 CE (Crowley and Lowery, 2000; Hiller et al., 2001) or 1000–1100 CE (Moberg et al., 2005). Such temperature anomalies correlate with increased fire activity in paleofire reconstructions inferred from various environmental archives including the Eclipse Icefield in the Yukon, Canada (1240–1410 CE) (Yalcin et al., 2006), Eurocore (1200–1350 CE) (Savarino and Legrand, 1998) and GISP2 ice cores (1200–1600 CE) (Taylor et al., 1996), and in the GCD between 1000–1400 CE (Marlon et al., 2008).

The Little Ice Age (LIA) is characterized by relatively cold conditions between 1580 and 1880 CE (Pages 2k, 2013) but varies regionally between 1400–1700 CE (Mann

et al., 2009), 1580–1720 CE (Christiansen and Ljungqvist, 2012) or 1350–1850 CE (Wanner et al., 2008). Low fire activity is observed in Eurocore (Summit, Greenland) (Savarino and Legrand, 1998) from 1600–1850 CE and in the NH GCD centered around 1750 CE (Marlon et al., 2008). Comparisons of the levoglucosan profile with a high-latitude Northern Hemisphere terrestrial temperature anomaly record (Mann et al., 2008) suggests a correspondence between temperature and fire activity observed in the deep NEEM ice core, except during the levoglucosan maximum centered around 1600 CE (Fig. 3e). Fire activity inferred from the deep NEEM ice core is consistent with the MWP and LIA climatic conditions, with high values between the 11th and 13th century and with low decadal-scale levoglucosan concentrations in the 18th and 19th centuries, after removing the individual spikes attributed to mega-events.

4.4.3 Precipitation

Ice core, tree-ring proxy records and archival evidences document extensive north-central Asian droughts concurrent with the strongest centennial-scale levoglucosan concentrations during the 16th and 17th centuries including two strong events (1593–1603 CE and 1617–1622 CE) (Fig. 2, Tables 1 and 2). The Siberian Belukha ice core identifies a period of exceptionally high forest-fire activity between 1600–1680 CE, following a drought period during 1540–1600 CE, which is coincident with elevated NEEM levoglucosan concentrations (Eichler et al., 2011). This period coincides with the Late Ming Weak Monsoon Period (1580–1640 CE) (Zhang et al., 2008), which has been invoked as contributing to the decline of the Ming Dynasty. Two central and northern Asian droughts (1586–1589 CE and 1638–1641 CE) were the most extreme of the past 5 centuries (Shen et al., 2007), yet occurred during relatively cold periods (Li et al., 2009; Cook et al., 2010; Yang et al., 2002; Tan et al., 2003) and covered much of central Asia (Fang et al., 2010). The 1586–1589 CE drought resulted in the desiccation of Lake Taihu (the third largest freshwater lake in China) and the even more spatially extensive 1638–1641 CE drought event resulted in no outflow of the Yellow River (Shen et al., 2007). Levoglucosan and BC concentrations from the NEEM ice core contain outlying

peaks in 1702–1706 CE and 1787–1791 CE, during a period of low centennial-scale fire activity. Paleoclimate reconstructions report extensive drought conditions in China and Mongolia throughout these periods (Table 2). A high-resolution determination of continental dust in the Dye-3 Greenland ice core Sr, Nd and Hf isotopic composition from 1786–1793 CE determine that the majority of the samples have an Asian origin (Lupker et al., 2010). A network of tree-ring chronologies and historical documents (Grove, 1998; Cook et al., 2010) and the Dasuopu ice core (Thompson et al., 2000) demonstrate that the 1790–1796 CE drought was the most intense arid period of the last millennium.

Major droughts are associated with summer monsoon failures, and their multi-annual nature suggests that the droughts may have continued to occur through the winter (Table 2). The Siberian High is a seasonal high-pressure system from approximately 70° to 120° E and 45° to 60° N that is the dominant control on northern Asian winter and spring climate (Gong and Ho, 2002; Panagiotopoulos et al., 2005; Sorrel et al., 2007). A stronger winter Siberian High generally results in decreased precipitation and temperatures in the area covered by the high pressure system (Panagiotopoulos et al., 2005; Sorrel et al., 2007; Chen et al., 2010). An enhanced Siberian High during 1330–1750 CE is consistent with Aral Sea dust records demonstrating decreased temperatures and a regional drought including decreased Mediterranean precipitation arriving to western Central Asia (Huang et al., 2011; Sorrel et al., 2007) and with increased $nssK^+$ in the GISP2 ice core (Meeker and Mayewski, 2002). Summer Asian monsoon activity correlates with solar variability (Zhang et al., 2008). The northern and central Asian megadroughts coincide with periods of decreased sunspot activity (Weng, 2012). Low sunspot activity may increase the strength of the Siberian High (Weng, 2012) and therefore result in decreased precipitation. Our results show that Siberian fire activity is closely related to precipitation changes, where extreme biomass burning peaks are synchronous with precipitation anomalies, independently supporting GISS GCM model results (Pechony and Shindell, 2010).

North American droughts may have also affected fire activity archived in the NEEM ice cores. Elevated aridity and megadroughts during the MWP (900–1300 CE), coincident with levoglucosan peaks, affected large areas of North America and were more prolonged than any 20th century droughts (Cook et al., 2004; Stahle et al., 2000). Dry conditions during the 16th century were also inferred in the Canadian Prairie Provinces (St. George et al., 2009) and in western North America (Stahle et al., 2000). In particular, tree rings from southern Manitoba, Canada, record the lowest growth in 1595 CE (St. George et al., 2009), when a strong fire event is recorded by levoglucosan and ammonium in a 2.20 m NEEM sample dated 1593–1603 CE, and by a BC spike in the NEEM-2011-S1 in 1594.5 CE. Levoglucosan spikes in 1704 and 1789 CE are also synchronous with drought periods in the North America, inferred by tree-ring analysis. 1790s was the driest decade in Southwestern Canada (Sauchyn and Skinner, 2001; Sauchyn and Beaudoin, 1998; Wolfe et al., 2001), as in Southern Alberta (Case and Macdonald, 1995). However, St. George et al. (2009) indicate that tree growth was low in 1790s due to dry conditions across the Canadian prairies. The same authors observed a "prolonged absence of wet years around 1700" on the eastern side of the Canadian prairies, where this "notable" dry event lasted between 1688 and 1715 CE when tree growth was generally below the mean (St. George et al., 2009).

4.4.4 Human impact on boreal fire activity

Climate may be more important than anthropogenic activity in influencing high latitude boreal fires in areas responsible for levoglucosan emissions reaching the Greenland ice sheet even after 1750 CE (Marlon et al., 2008). Linkages between humans and fire regimes are intricate, where increasing population does not necessarily result in enhanced fire activity (Archibald et al., 2009; Prentice, 2010). The GCD synthesis argues that population growth and land-cover conversion rates, along with increases in global temperatures, are the main factors for the sharp increase in global biomass burning from 1750 CE to the late 19th century (Marlon et al., 2008). However, the GCD asserts that the expansion of agriculture and fire suppression since the early twentieth century

resulted in decreased global fire activity, where current biomass burning rates may be lower than over the past 2000 years (Marlon et al., 2008; Prentice, 2010). These results are supported by South Pole ice core carbon monoxide isotopic data that also record a downturn in global fire activity from the late 1800s to present (Wang et al., 2010). Pyrogenic methane isotopic data (Ferretti et al., 2005; Mischler et al., 2009), which show relative high values in the first millennium CE, do not support the prominent anthropogenic peak at ∼ 1900 CE observable in the GCD (Marlon et al., 2008) and inferred from CO isotopic measurements (Wang et al., 2010), and instead demonstrate that pyrogenic methane sources are still increasing with higher rates than the last 2 millennia. Emissions modeling does not agree with methane isotopes data, and suggests that higher-than-present levels of pre-industrial biomass burning are implausible (van der Werf et al., 2013). Levoglucosan concentrations are relatively high during the past century, but other higher multi-decadal maxima are present in our dataset (Fig. S5 in the Supplement). The number of post-1950 CE samples in our data set is limited, and so the levoglucosan analyses in the past decades are not sufficient to clearly assess the 20th century downturn in fire activity. Levoglucosan data do not show the evident anthropogenic peak started from 1750 CE as inferred from the GCD synthesis, as the NEEM levoglucosan profile contains low concentrations until the beginning of the 20th century, followed by a modest concentration increase. No significant increase in fire activity during the past 300 year is even observed on the Altai region, Southern Siberia (Eichler et al., 2011).

The differences between the GCD, $\delta^{13}C$ of CH_4, and levoglucosan in NEEM may be related to differing initiation times of extensive industrial and agricultural activity in boreal forest source regions. As previously discussed, Russian and Canadian boreal forests are likely the principal sources of levoglucosan reaching the Greenland ice sheet. Population growth and major anthropogenic land clearing in these regions only significantly increased during the past century (Kawamura et al., 2012). The former Soviet Union experienced the highest forest clearing rates and eastward cropland expansion into southern Siberia between 1940 and 1960 CE, while the high Canadian

land clearing rates occurred in the prairie provinces between 1900 and 1920 CE (Ramankutty and Foley, 1999). NEEM levoglucosan peaks during the same time interval as these land-clearing estimates. Canadian ice cores also show peak biomass burning from 1920–1940 CE (Yalcin et al., 2006) and 1930–1980 CE (Whitlow et al., 1994).

The dense GCD sampling in North America and Europe may affect the high latitude Northern Hemisphere charcoal synthesis. The conclusion that land-use and fire management practices decreased global fire activity since the early twenty century (Marlon et al., 2008) may be more appropriate for temperate North America and Europe than for boreal regions. NEEM levoglucosan concentrations may instead reflect the boreal source region land-clearance that was occurring even when anthropogenic land use and fire suppression were dominant in other parts of the Northern Hemisphere.

5 Conclusion

Levoglucosan, NH_4^+ and BC analyses in the NEEM ice cores provide a specific record of past biomass burning. Each biomass burning marker has a set of intrinsic strengths and limitations, and so a combination of fire proxies results in a more robust reconstruction. The NEEM records and connections with back trajectories and other paleoclimate studies suggest North American/Canadian fires are the main sources of pyrogenic aerosols transported to the site in particular during the preindustrial period. However, Siberian forests may be an essential aspect of boreal fire reconstructions that have not yet been appropriately evaluated. Temperature may be the controlling factor of boreal fire activity on centennial time scales. On multi-decadal or shorter timescales, however, boreal fires are mainly influenced by precipitation. NEEM levoglucosan, NH_4^+ and BC concentrations suggest the major drought centered on 1640 CE may have been more extensive than inferred from local paleoclimate reconstructions, and that this event dominates boreal fire activity over the past 2000 years.

Our results demonstrate the greatest amount of decadal-scale fire activity during the mid-1600s, concurrent with known extensive droughts and monsoon failures, rather than during the last 150 years when anthropogenic land-clearing rates were the highest in history. This evidence suggests that climate variability has influenced boreal forest fires more than anthropogenic activity over the past millennia in the boreal regions that supply biomass burning related species to Greenland. Recent climate change and anthropogenic activity may increase future boreal fire activity, and have the potential to exceed the fire activity during the mid-1600s. Warmer, drier summers and increased deadwood availability due to past fire suppression, as well as insect outbreaks (Kurz et al., 2008; Wolken et al., 2011), and increased tree mortality from drought (van Mantgem et al., 2009) may amplify current and future fires. Increasing forest mortality in a warming climate (Anderegg et al., 2013) results in greater fuel availability with the potential to intensify future boreal fire activity.

Supplementary material related to this article is available online at http://www.clim-past-discuss.net/10/809/2014/cpd-10-809-2014-supplement.pdf.

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References

Anderegg, W. R. L., Kane, J. M., and Anderegg, L. D. L.: Consequences of widespread tree Mortality triggered by drought and temperature stress, Nat. Clim. Change, 3, 30–36, 2013.

Andersen, K. K., Ditlevsen, P. D., Rasmussen, S. O., Clausen, H. B., Vinther, B. M., Johnsen, S. J., and Steffensen, J. P.: Retrieving a common accumulation record from Greenland ice cores for the past 1800 years, J. Geophys. Res.-Atmos., 111, D15106, doi:10.1029/2005jd006765, 2006.

Andreev, A. A., Pierau, R., Kalugin, I. A., Daryin, A. V., Smolyaninova, L. G., and Diekmann, B.: Environmental changes in the northern Altai during the last millennium documented in Lake Teletskoye pollen record, Quaternary Res., 67, 394–399, doi:10.1016/j.yqres.2006.11.004, 2007.

Archibald, S., Roy, D. P., van Wilgen, B. W., and Scholes, R. J.: What limits fire? An examination of drivers of burnt area in Southern Africa, Global Change Biol., 15, 613–630, doi:10.1111/j.1365-2486.2008.01754.x, 2009.

Banta, J. R., McConnell, J. R., Edwards, R., and Engelbrecht, J. P.: Delineation of carbonate dust, aluminous dust, and sea salt deposition in a Greenland glaciochemical array using positive matrix factorization, Geochem. Geophy. Geosy., 9, Q07013, doi:10.1029/2007gc001908, 2008.

Bauer, S. E., Bausch, A., Nazarenko, L., Tsigaridis, K., Xu, B. Q., Edwards, R., Bisiaux, M., and McConnell, J.: Historical and future black carbon deposition on the three ice caps: ice core

measurements and model simulations from 1850 to 2100, J. Geophys. Res.-Atmos., 118, 7948–7961, doi:10.1002/jgrd.50612, 2013.

Blunier, T., Spahni, R., Barnola, J.-M., Chappellaz, J., Loulergue, L., and Schwander, J.: Synchronization of ice core records via atmospheric gases, Clim. Past, 3, 325–330, doi:10.5194/cp-3-325-2007, 2007.

Bory, A. J. M., Biscaye, P. E., Svensson, A., and Grousset, F. E.: Seasonal variability in the origin of recent atmospheric mineral dust at NorthGRIP, Greenland, Earth Planet. Sc. Lett., 196, 123–134, doi:10.1016/s0012-821x(01)00609-4, 2002.

Bory, A. J. M., Biscaye, P. E., Piotrowski, A. M., and Steffensen, J. P.: Regional variability of ice core dust composition and provenance in Greenland, Geochem. Geophy. Geosy., 4, 1107, doi:10.1029/2003gc000627, 2003.

Bowman, D., Balch, J. K., Artaxo, P., Bond, W. J., Carlson, J. M., Cochrane, M. A., D'Antonio, C. M., DeFries, R. S., Doyle, J. C., Harrison, S. P., Johnston, F. H., Keeley, J. E., Krawchuk, M. A., Kull, C. A., Marston, J. B., Moritz, M. A., Prentice, I. C., Roos, C. I., Scott, A. C., Swetnam, T. W., van der Werf, G. R., and Pyne, S. J.: Fire in the Earth System, Science, 324, 481–484, doi:10.1126/science.1163886, 2009.

Brunelle, A., Whitlock, C., Bartlein, P., and Kipfmueller, K.: Holocene fire and vegetation along environmental gradients in the Northern Rocky Mountains, Quaternary Sci. Rev., 24, 2281–2300, doi:10.1016/j.quascirev.2005.11.010, 2005.

Canadian Forest Service: The State of Canada's Forests 2004–2005: The boreal forest, Ottawa, 2005.

Case, R. A. and Macdonald, G. M.: A dendroclimatic reconstruction of annual precipitation on the Western Canadian Prairies since AD 1505 from *pinus-flexilis james*, Quaternary Res., 44, 267–275, doi:10.1006/qres.1995.1071, 1995.

Chen, F., Yuan, Y. J., Wen, W. S., Yu, S. L., Fan, Z., Zhang, R. B., Zhang, T. W., and Shang, H. M.: Tree-ring-based reconstruction of precipitation in the Changling Mountains, China, since A.D.1691, Int. J. Biometeorol., 56, 765–774, doi:10.1007/s00484-011-0431-8, 2012.

Chen, H. F., Song, S. R., Lee, T. Q., Lowemark, L., Chi, Z. Q., Wang, Y., and Hong, E.: A multiproxy lake record from Inner Mongolia displays a late Holocene teleconnection between Central Asian and North Atlantic climates, Quatern. Int., 227, 170–182, doi:10.1016/j.quaint.2010.03.005, 2010.

Christiansen, B. and Ljungqvist, F. C.: The extra-tropical Northern Hemisphere temperature in the last two millennia: reconstructions of low-frequency variability, Clim. Past, 8, 765–786, doi:10.5194/cp-8-765-2012, 2012.

Chu, G., Sun, Q., Wang, X., and Sun, J.: Snow anomaly events from historical documents in eastern China during the past two millennia and implication for low-frequency variability of AO/NAO and PDO, Geophys. Res. Lett., 35, L14806, doi:10.1029/2008gl034475, 2008.

Conedera, M., Tinner, W., Neff, C., Meurer, M., Dickens, A. F., and Krebs, P.: Reconstructing past fire regimes: methods, applications, and relevance to fire management and conservation, Quaternary Sci. Rev., 28, 555–576, doi:10.1016/j.quascirev.2008.11.005, 2009.

Cook, E. R., Woodhouse, C. A., Eakin, C. M., Meko, D. M., and Stahle, D. W.: Long-term aridity changes in the western United States, Science, 306, 1015–1018, doi:10.1126/science.1102586, 2004.

Cook, E. R., Anchukaitis, K. J., Buckley, B. M., D'Arrigo, R. D., Jacoby, G. C., and Wright, W. E.: Asian monsoon failure and megadrought during the last millennium, Science, 328, 486–489, doi:10.1126/science.1185188, 2010.

Crowley, T. J. and Lowery, T. S.: How warm was the medieval warm period?, Ambio, 29, 51–54, doi:10.1639/0044-7447(2000)029<0051:hwwtmw>2.0.co;2, 2000.

Damoah, R., Spichtinger, N., Forster, C., James, P., Mattis, I., Wandinger, U., Beirle, S., Wagner, T., and Stohl, A.: Around the world in 17 days – hemispheric-scale transport of forest fire smoke from Russia in May 2003, Atmos. Chem. Phys., 4, 1311–1321, doi:10.5194/acp-4-1311-2004, 2004.

Daniau, A. L., Harrison, S. P., and Bartlein, P. J.: Fire regimes during the Last Glacial, Quaternary Sci. Rev., 29, 2918–2930, doi:10.1016/j.quascirev.2009.11.008, 2010.

Daniau, A. L., Bartlein, P. J., Harrison, S. P., Prentice, I. C., Brewer, S., Friedlingstein, P., Harrison-Prentice, T. I., Inoue, J., Izumi, K., Marlon, J. R., Mooney, S., Power, M. J., Stevenson, J., Tinner, W., Andrič, M., Atanassova, J., Behling, H., Black, M., Blarquez, O., Brown, K. J., Carcaillet, C., Colhoun, E. A., Colombaroli, D., Davis, B. A. S., D'Costa, D., Dodson, J., Dupont, L., Eshetu, Z., Gavin, D. G., Genries, A., Haberle, S., Hallett, D. J., Hope, G., Horn, S. P., Kassa, T. G., Katamura, F., Kennedy, L. M., Kershaw, P., Krivonogov, S., Long, C., Magri, D., Marinova, E., McKenzie, G. M., Moreno, P. I., Moss, P., Neumann, F. H., Norstroöm, E., Paitre, C., Rius, D., Roberts, N., Robinson, G. S., Sasaki, N., Scott, L., Takahara, H., Terwilliger, V., Thevenon, F., Turner, R., Valsecchi, V. G., Vannière, B., Walsh, M.,

Williams, N., and Zhang, Y.: Predictability of biomass burning in response to climate changes, Global Biogeochem. Cy., 26, GB4007, doi:10.1029/2011GB004249, 2012.
Davi, N., Jacoby, G., Fang, K., Li, J., D'Arrigo, R., Baatarbileg, N., and Robinson, D.: Reconstructing drought variability for Mongolia based on a large-scale tree ring network: 1520–1993, J. Geophys. Res.-Atmos., 115, D22103, doi:10.1029/2010jd013907, 2010.
Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys., 6, 4321–4344, doi:10.5194/acp-6-4321-2006, 2006.
Diamond, J. M.: Collapse: How Societies Choose to Fail Or Succeed, Viking, New York, 2005.
Dronin, N. M. and Bellinger, E. G.: Climate Dependence and Food Problems in Russia, 1900–1990: The Interaction of Climate and Agricultural Policy and Their Effect on Food Problems, Central European University Press, Budepest, Hungary, 2005.
Eichler, A., Tinner, W., Brusch, S., Olivier, S., Papina, T., and Schwikowski, M.: An ice-core based history of Siberian forest fires since AD 1250, Quaternary Sci. Rev., 30, 1027–1034, doi:10.1016/j.quascirev.2011.02.007, 2011.
Engling, G., Carrico, C. M., Kreldenweis, S. M., Collett, J. L., Day, D. E., Malm, W. C., Lincoln, E., Hao, W. M., Iinuma, Y., and Herrmann, H.: Determination of levoglucosan in biomass combustion aerosol by high-performance anion-exchange chromatography with pulsed amperometric detection, Atmos. Environ., 40, S299–S311, doi:10.1016/j.atmosenv.2005.12.069, 2006.
Fang, K. Y., Davi, N., Gou, X. H., Chen, F. H., Cook, E., Li, J. B., and D'Arrigo, R.: Spatial drought reconstructions for central High Asia based on tree rings, Clim. Dynam., 35, 941–951, doi:10.1007/s00382-009-0739-9, 2010.
Fang, K. Y., Gou, X., Chen, F., Frank, D., Liu, C., Li, J., and Kazmer, M.: Precipitation variability during the past 400 years in the Xiaolong Mountain (central China) inferred from tree rings, Clim. Dynam., 39, 1697–1707, doi:10.1007/s00382-012-1371-7, 2012.
FAO: Fire management: global assessment 2006, Food and Agriculture Organization of the United Nations, Rome, 2007.
Feng, S., Hu, Q., and Oglesby, R. J.: Influence of Atlantic sea surface temperatures on persistent drought in North America, Clim. Dynam., 37, 569–586, doi:10.1007/s00382-010-0835-x, 2011.

Ferretti, D. F., Miller, J. B., White, J. W. C., Etheridge, D. M., Lassey, K. R., Lowe, D. C., Meure, C. M. M., Dreier, M. F., Trudinger, C. M., van Ommen, T. D., and Langenfelds, R. L.: Unexpected changes to the global methane budget over the past 2000 years, Science, 309, 1714–1717, doi:10.1126/science.1115193, 2005.
Fischer, H., Siggaard-Andersen, M. L., Ruth, U., Rothlisberger, R., and Wolff, E.: Glacial/interglacial changes in mineral dust and sea-salt records in polar ice cores: sources, transport, and deposition, Rev. Geophys., 45, RG1002, doi:10.1029/2005rg000192, 2007.
Fraser, M. P. and Lakshmanan, K.: Using levoglucosan as a molecular marker for the long-range transport of biomass combustion aerosols, Environ. Sci. Technol., 34, 4560–4564, doi:10.1021/es991229l, 2000.
Fuhrer, K. and Legrand, M.: Continental biogenic species in the Greenland Ice Core Project ice core: tracing back the biomass history of the North American continent, J. Geophys. Res.-Oceans, 102, 26735–26745, 1997.
Fuhrer, K., Neftel, A., Anklin, M., and Maggi, V.: continuous measurements of hydrogenperoxide, formaldehyde, calcium and ammonium concentrations along the new grip ice core from summit, central greenland, Atmos. Environ. A-Gen., 27, 1873–1880, doi:10.1016/0960-1686(93)90292-7, 1993.
Fuhrer, K., Neftel, A., Anklin, M., Staffelbach, T., and Legrand, M.: High-resolution ammonium ice core record covering a complete glacial-interglacial cycle, J. Geophys. Res.-Atmos., 101, 4147–4164, 1996.
Gambaro, A., Zangrando, R., Gabrielli, P., Barbante, C., and Cescon, P.: Direct determination of levoglucosan at the picogram per milliliter level in Antarctic ice by high-performance liquid chromatography/electrospray ionization triple quadrupole mass spectrometry, Anal. Chem., 80, 1649–1655, doi:10.1021/ac701655x, 2008.
Gao, S., Hegg, D. A., Hobbs, P. V., Kirchstetter, T. W., Magi, B. I., and Sadilek, M.: Water-soluble organic components in aerosols associated with savanna fires in southern Africa: identification, evolution, and distribution, J. Geophys. Res.-Atmos., 108, 8491, doi:10.1029/2002jd002324, 2003.
Goldberg, E. D.: Black Carbon in the Environment: Properties and Distribution, J. Wiley, New York, 1985.

Golubev, G. and Dronin, N. M.: Geography of Droughts and Food Problems in Russia (1900–2000), Report of the International Project on Global Environmental Change and its Threat to Food and Water Security in Russia, Department of Geography, Moscow State University, Moscow, Russia, 2004.

Gong, D. Y. and Ho, C. H.: The Siberian High and climate change over middle to high latitude Asia, Theor. Appl. Climatol., 72, 1–9, doi:10.1007/s007040200008, 2002.

Grove, R. H.: Global impact of the 1789–93 El Nino, Nature, 393, 318–319, doi:10.1038/30636, 1998.

Hallett, D. J., Lepofsky, D. S., Mathewes, R. W., and Lertzman, K. P.: 11 000 years of fire history and climate in the mountain hemlock rain forests of southwestern British Columbia based on sedimentary charcoal, Can. J. Forest Res., 33, 292–312, doi:10.1139/x02-177, 2003.

Hansson, M. E.: The renland ice core – a Northern-Hemisphere record of aerosol composition over 120,000 years, Tellus B, 46, 390–418, doi:10.1034/j.1600-0889.1994.t01-4-00005.x, 1994.

Hegg, D. A., Warren, S. G., Grenfell, T. C., Doherty, S. J., Larson, T. V., and Clarke, A. D.: Source attribution of black carbon in Arctic snow, Environ. Sci. Technol., 43, 4016–4021, doi:10.1021/es803623f, 2009.

Hennigan, C. J., Sullivan, A. P., Collett, J. L., and Robinson, A. L.: Levoglucosan stability in biomass burning particles exposed to hydroxyl radicals, Geophys. Res. Lett., 37, L09806, doi:10.1029/2010gl043088, 2010.

Herron, M. M. and Langway Jr., C. C.: Firn densification: an empirical model, J. Glaciol., 25, 373–385, 1980.

Hiller, A., Boettger, T., and Kremenetski, C.: Mediaeval climatic warming recorded by radiocarbon dated alpine tree-line shift on the Kola Peninsula, Russia, Holocene, 11, 491–497, doi:10.1191/095968301678302931, 2001.

Hodzic, A., Madronich, S., Bohn, B., Massie, S., Menut, L., and Wiedinmyer, C.: Wildfire particulate matter in Europe during summer 2003: meso-scale modeling of smoke emissions, transport and radiative effects, Atmos. Chem. Phys., 7, 4043–4064, doi:10.5194/acp-7-4043-2007, 2007.

Hoffmann, D., Tilgner, A., Iinuma, Y., and Herrmann, H.: Atmospheric stability of levoglucosan: a detailed laboratory and modeling study, Environ. Sci. Technol., 44, 694–699, doi:10.1021/es902476f, 2010.

Holmes, B. J. and Petrucci, G. A.: Water-soluble oligomer formation from acid-catalyzed reactions of levoglucosan in proxies of atmospheric aqueous aerosols, Environ. Sci. Technol., 40, 4983–4989, doi:10.1021/es060646c, 2006.

Holmes, B. J. and Petrucci, G. A.: Oligomerization of levoglucosan by Fenton chemistry in proxies of biomass burning aerosols, J. Atmos. Chem., 58, 151–166, doi:10.1007/s10874-007-9084-8, 2007.

Huang, X. T., Oberhansli, H., von Suchodoletz, H., and Sorrel, P.: Dust deposition in the Aral Sea: implications for changes in atmospheric circulation in central Asia during the past 2000 years, Quaternary Sci. Rev., 30, 3661–3674, doi:10.1016/j.quascirev.2011.09.011, 2011.

Iinuma, Y., Bruggemann, E., Gnauk, T., Muller, K., Andreae, M. O., 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.

IPCC: Climate Change 2007: The Scientific Basis, Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge University Press, Cambridge, UK and New York, NY, USA, 2007.

Jacobson, M. Z.: The short-term cooling but long-term global warming due to biomass burning, J. Climate,17, 2909–2926, doi:10.1175/1520-0442(2004)017<2909:tscblg>2.0.co;2, 2004.

Kahl, J. D. W., Martinez, D. A., Kuhns, H., Davidson, C. I., Jaffrezo, J. L., and Harris, J. M.: Air mass trajectories to Summit, Greenland: a 44-year climatology and some episodic events, J. Geophys. Res.-Oceans, 102, 26861–26875, doi:10.1029/97jc00296, 1997.

Kaufmann, P. R., Federer, U., Hutterli, M. A., Bigler, M., Schupbach, S., Ruth, U., Schmitt, J., and Stocker, T. F.: An improved continuous flow analysis system for high-resolution field measurements on ice cores, Environ. Sci. Technol., 42, 8044–8050, doi:10.1021/es8007722, 2008.

Kawamura, K., Yokoyama, K., Fujii, Y., and Watanabe, O.: A Greenland ice core record of low molecular weight dicarboxylic acids, ketocarboxylic acids, and alpha-dicarbonyls: a trend from Little Ice Age to the present (1540 to 1989 AD), J. Geophys. Res.-Atmos., 106, 1331–1345, 2001.

Kawamura, K., Izawa, Y., Mochida, M., and Shiraiwa, T.: Ice core records of biomass burning tracers (levoglucosan and dehydroabietic, vanillic and *p*-hydroxybenzoic acids) and total organic carbon for past 300 years in the Kamchatka Peninsula, Northeast Asia, Geochim. Cosmochim. Acta, 99, 317–329, doi:10.1016/j.gca.2012.08.006, 2012.

Kehrwald, N., Zangrando, R., Gabrielli, P., Jaffrezo, J. L., Boutron, C., Barbante, C., and Gambaro, A.: Levoglucosan as a specific marker of fire events in Greenland snow, Tellus B, 64, 18196, doi:10.3402/tellusb.v64i0.18196, 2012.

Kuo, L. J., Herbert, B. E., and Louchouarn, P.: Can levoglucosan be used to characterize and quantify char/charcoal black carbon in environmental media?, Org. Geochem., 39, 1466–1478, doi:10.1016/j.orggeochem.2008.04.026, 2008.

Kurz, W. A., Stinson, G., Rampley, G. J., Dymond, C. C., and Neilson, E. T.: Risk of natural disturbances makes future contribution of Canada's forests to the global carbon cycle highly uncerain, P. Natl. Acad. Sci. USA, 105, 1551–1555, doi:10.1073/pnas.0708133105, 2008.

Legrand, M., Deangelis, M., Staffelbach, T., Neftel, A., and Stauffer, B.: Large perturbation of ammonium and organic-acids content in the Summit-Greenland ice core – fingerprint from forest-fires, Geophys. Res. Lett., 19, 473–475, 1992.

Li, J. B., Cook, E. R., Chen, F. H., Davi, N., D'Arrigo, R., Gou, X. H., Wright, W. E., Fang, K. Y., Jin, L. Y., Shi, J. F., and Yang, T.: Summer monsoon moisture variability over China and Mongolia during the past four centuries, Geophys. Res. Lett., 36, L22705, doi:10.1029/2009gl041162, 2009.

Liu, Y., Shi, J. F., Shishov, V., Vaganov, E., Yang, Y. K., Cai, Q. F., Sun, J. Y., Wang, L., and Djanseitov, I.: Reconstruction of May–July precipitation in the north Helan Mountain, Inner Mongolia since AD 1726 from tree-ring late-wood widths, Chinese Sci. Bull., 49, 405–409, doi:10.1007/BF02900325, 2004.

Liu, Y., Tian, H., Song, H. M., and Liang, J. M.: Tree ring precipitation reconstruction in the Chifeng-Weichang region, China, and East Asian summer monsoon variation since AD 1777, J. Geophys. Res.-Atmos., 115, D06103, doi:10.1029/2009jd012330, 2010.

Lupker, M., Aciego, S. M., Bourdon, B., Schwander, J., and Stocker, T. F.: Isotopic tracing (Sr, Nd, U and Hf) of continental and marine aerosols in an 18th century section of the Dye-3 ice core (Greenland), Earth Planet. Sc. Lett., 295, 277–286, doi:10.1016/j.epsl.2010.04.010, 2010.

Mann, M. E., Zhang, Z. H., Hughes, M. K., Bradley, R. S., Miller, S. K., Rutherford, S., and Ni, F. B.: Proxy-based reconstructions of hemispheric and global surface temperature variations over the past two millennia, P. Natl. Acad. Sci. USA, 105, 13252–13257, doi:10.1073/pnas.0805721105, 2008.

Mann, M. E., Zhang, Z. H., Rutherford, S., Bradley, R. S., Hughes, M. K., Shindell, D., Ammann, C., Faluvegi, G., and Ni, F. B.: Global signatures and dynamical origins of the Little Ice Age and Medieval climate anomaly, Science, 326, 1256–1260, doi:10.1126/science.1177303, 2009.

Marlon, J. R., Bartlein, P. J., Carcaillet, C., Gavin, D. G., Harrison, S. P., Higuera, P. E., Joos, F., Power, M. J., and Prentice, I. C.: Climate and human influences on global biomass burning over the past two millennia, Nat. Geosci., 1, 697–702, doi:10.1038/ngeo313, 2008.

Marlon, J. R., Bartlein, P. J., Gavin, D. G., Long, C. J., Anderson, R. S., Briles, C. E., Brown, K. J., Colombaroli, D., Hallett, D. J., Power, M. J., Scharf, E. A., and Walsh, M. K.: Long-term perspective on wildfires in the western USA, P. Natl. Acad. Sci. USA, 109, E535–E543, doi:10.1073/pnas.1112839109, 2012.

Marlon, J. R., Bartlein, P. J., Daniau, A. L., Harrison, S. P., Maezumi, S. Y., Power, M. J., Tinner, W., and Vanniere, B.: Global biomass burning: a synthesis and review of Holocene paleofire records and their controls, Quaternary Sci. Rev., 65, 5–25, doi:10.1016/j.quascirev.2012.11.029, 2013.

Masiello, C. A.: New directions in black carbon organic geochemistry, Mar. Chem., 92, 201–213, doi:10.1016/j.marchem.2004.06.043, 2004.

McConnell, J. R.: Continuous ice-core chemical analyses using inductively Coupled Plasma Mass Spectrometry, Environ. Sci. Technol., 36, 7–11, doi:10.1021/es011088z, 2002.

McConnell, J. R.: New Directions: historical black carbon and other ice core aerosol records in the Arctic for GCM evaluation, Atmos. Environ., 44, 2665–2666, doi:10.1016/j.atmosenv.2010.04.004, 2010.

McConnell, J. R. and Edwards, R.: Coal burning leaves toxic heavy metal legacy in the Arctic, P. Natl. Acad. Sci. USA, 105, 12140–12144, doi:10.1073/pnas.0803564105, 2008.

McConnell, J. R., Edwards, R., Kok, G. L., Flanner, M. G., Zender, C. S., Saltzman, E. S., Banta, J. R., Pasteris, D. R., Carter, M. M., and Kahl, J. D. W.: 20th-century industrial black carbon emissions altered arctic climate forcing, Science, 317, 1381–1384, doi:10.1126/science.1144856, 2007.

McWethy, D. B., Whitlock, C., Wilmshurst, J. M., McGlone, M. S., and Li, X.: Rapid deforestation of South Islands, New Zealands, by early Polynesian fires, Holocene, 19, 883–897, doi:10.1177/0959683609336563, 2009.

Meeker, L. D. and Mayewski, P. A.: A 1400-year high-resolution record of atmospheric circulation over the North Atlantic and Asia, Holocene, 12, 257–266, doi:10.1191/0959683602hl542ft, 2002.

Mischler, J. A., Sowers, T. A., Alley, R. B., Battle, M., McConnell, J. R., Mitchell, L., Popp, T., Sofen, E., and Spencer, M. K.: Carbon and hydrogen isotopic composition of methane over the last 1000 years, Global Biogeochem. Cy., 23, GB4024, doi:10.1029/2009gb003460, 2009.

Moberg, A., Sonechkin, D. M., Holmgren, K., Datsenko, N. M., and Karlen, W.: Highly variable Northern Hemisphere temperatures reconstructed from low- and high-resolution proxy data, Nature, 433, 613–617, doi:10.1038/nature03265, 2005.

Mochida, M., Kawamura, K., Fu, P. Q., and Takemura, T.: Seasonal variation of levoglucosan in aerosols over the western North Pacific and its assessment as a biomass-burning tracer, Atmos. Environ., 44, 3511–3518, doi:10.1016/j.atmosenv.2010.06.017, 2010.

Obrien, S. R., Mayewski, P. A., Meeker, L. D., Meese, D. A., Twickler, M. S., and Whitlow, S. I.: Complexity of holocene climate as reconstructed from a greenland ice core, Science, 270, 1962–1964, doi:10.1126/science.270.5244.1962, 1995.

Pages 2k: Continental-scale temperature variability during the past two millennia, Nat. Geosci., 6, 339–346, doi:10.1038/ngeo1797, 2013.

Panagiotopoulos, F., Shahgedanova, M., Hannachi, A., and Stephenson, D. B.: Observed trends and teleconnections of the Siberian high: a recently declining center of action, J. Climate, 18, 1411–1422, doi:10.1175/jcli3352.1, 2005.

Pechony, O. and Shindell, D. T.: Driving forces of global wildfires over the past millennium and the forthcoming century, P. Natl. Acad. Sci. USA, 107, 19167–19170, doi:10.1073/pnas.1003669107, 2010.

Pederson, N., Jacoby, G. C., D'Arrigo, R. D., Cook, E. R., Buckley, B. M., Dugarjav, C., and Mijiddorj, R.: Hydrometeorological reconstructions for northeastern Mongolia derived from tree rings: 1651–1995, J. Climate, 14, 872–881, doi:10.1175/1520-0442(2001)014<0872:hrfnmd>2.0.co;2, 2001.

Power, M. J., Marlon, J., Ortiz, N., Bartlein, P. J., Harrison, S. P., Mayle, F. E., Ballouche, A., Bradshaw, R. H. W., Carcaillet, C., Cordova, C., Mooney, S., Moreno, P. I., Prentice, I. C., Thonicke, K., Tinner, W., Whitlock, C., Zhang, Y., Zhao, Y., Ali, A. A., Anderson, R. S., Beer, R., Behling, H., Briles, C., Brown, K. J., Brunelle, A., Bush, M., Camill, P., Chu, G. Q., Clark, J., Colombaroli, D., Connor, S., Daniau, A. L., Daniels, M., Dodson, J., Doughty, E., Edwards, M. E., Finsinger, W., Foster, D., Frechette, J., Gaillard, M. J., Gavin, D. G., Gobet, E., Haberle, S., Hallett, D. J., Higuera, P., Hope, G., Horn, S., Inoue, J., Kaltenrieder, P., Kennedy, L., Kong, Z. C., Larsen, C., Long, C. J., Lynch, J., Lynch, E. A., McGlone, M., Meeks, S., Mensing, S., Meyer, G., Minckley, T., Mohr, J., Nelson, D. M., New, J., Newnham, R., Noti, R., Oswald, W., Pierce, J., Richard, P. J. H., Rowe, C., Goni, M. F. S., Shuman, B. N., Takahara, H., Toney, J., Turney, C., Urrego-Sanchez, D. H., Umbanhowar, C., Vandergoes, M., Vanniere, B., Vescovi, E., Walsh, M., Wang, X., Williams, N., Wilmshurst, J., and Zhang, J. H.: Changes in fire regimes since the Last Glacial Maximum: an assessment based on a global synthesis and analysis of charcoal data, Clim. Dynam., 30, 887–907, doi:10.1007/s00382-007-0334-x, 2008.

Power, M. J., Marlon, J. R., Bartlein, P. J., and Harrison, S. P.: Fire history and the Global Charcoal Database: a new tool for hypothesis testing and data exploration, Paleogeogr. Paleocl., 291, 52–59, doi:10.1016/j.palaeo.2009.09.014, 2010.

Prentice, I. C.: The burning issue, Science, 330, 1636–1637, doi:10.1126/science.1199809, 2010.

Preston, C. M. and Schmidt, M. W. I.: Black (pyrogenic) carbon: a synthesis of current knowledge and uncertainties with special consideration of boreal regions, Biogeosciences, 3, 397–420, doi:10.5194/bg-3-397-2006, 2006.

Pyne, S. J.: The fires this time, and next, Science, 294, 1005–1006, doi:10.1126/science.1064989, 2001.

Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon, Nat. Geosci., 1, 221–227, doi:10.1038/ngeo156, 2008.

Ramankutty, N. and Foley, J. A.: Estimating historical changes in global land cover: croplands from 1700 to 1992, Global Biogeochem. Cy., 13, 997–1027, doi:10.1029/1999gb900046, 1999.

Randerson, J. T., Liu, H., Flanner, M. G., Chambers, S. D., Jin, Y., Hess, P. G., Pfister, G., Mack, M. C., Treseder, K. K., Welp, L. R., Chapin, F. S., Harden, J. W., Goulden, M. L., Lyons, E., Neff, J. C., Schuur, E. A. G., and Zender, C. S.: The impact of boreal forest fire on climate warming, Science, 314, 1130–1132, doi:10.1126/science.1132075, 2006.

Rasmussen, S. O., Abbott, P. M., Blunier, T., Bourne, A. J., Brook, E., Buchardt, S. L., Buizert, C., Chappellaz, J., Clausen, H. B., Cook, E., Dahl-Jensen, D., Davies, S. M., Guillevic, M., Kipfstuhl, S., Laepple, T., Seierstad, I. K., Severinghaus, J. P., Steffensen, J. P., Stowasser, C., Svensson, A., Vallelonga, P., Vinther, B. M., Wilhelms, F., and Winstrup, M.: A first chronology for the North Greenland Eemian Ice Drilling (NEEM) ice core, Clim. Past, 9, 2713–2730, doi:10.5194/cp-9-2713-2013, 2013.

Running, S. W.: Is global warming causing more, larger wildfires?, Science, 313, 927–928, doi:10.1126/science.1130370, 2006.

Sapart, C. J., Monteil, G., Prokopiou, M., van de Wal, R. S. W., Kaplan, J. O., Sperlich, P., Krumhardt, K. M., van der Veen, C., Houweling, S., Krol, M. C., Blunier, T., Sowers, T., Martinerie, P., Witrant, E., Dahl-Jensen, D., and Rockmann, T.: Natural and anthropogenic variations in methane sources during the past two millennia, Nature, 490, 85–88, 2012.

Sauchyn, D. J. and Beaudoin, A. B.: Recent environmental change in the southwestern Canadian Plains, Can. Geogr.-Geogr. Can., 42, 337–353, doi:10.1111/j.1541-0064.1998.tb01350.x, 1998.

Sauchyn, D. J. and Skinner, W. R.: A proxy record of drought severity for the Southwestern Canadian Plains, Can. Water Resour. J., 26, 253–272, doi:10.4296/cwrj2602253, 2001.

Savarino, J. and Legrand, M.: High northern latitude forest fires and vegetation emissions over the last millennium inferred from the chemistry of a central Greenland ice core, J. Geophys. Res.-Atmos., 103, 8267–8279, 1998.

Shen, C., Wang, W.-C., Hao, Z., and Gong, W.: Exceptional drought events over eastern China during the last five centuries, Climatic Change, 85, 453–471, doi:10.1007/s10584-007-9283-y, 2007.

Shindell, D. T., Chin, M., Dentener, F., Doherty, R. M., Faluvegi, G., Fiore, A. M., Hess, P., Koch, D. M., MacKenzie, I. A., Sanderson, M. G., Schultz, M. G., Schulz, M., Stevenson, D. S., Teich, H., Textor, C., Wild, O., Bergmann, D. J., Bey, I., Bian, H., Cuvelier, C., Duncan, B. N., Folberth, G., Horowitz, L. W., Jonson, J., Kaminski, J. W., Marmer, E., Park, R., Pringle, K. J., Schroeder, S., Szopa, S., Takemura, T., Zeng, G., Keating, T. J., and Zuber, A.:

A multi-model assessment of pollution transport to the Arctic, Atmos. Chem. Phys., 8, 5353–5372, doi:10.5194/acp-8-5353-2008, 2008.

Sigl, M., McConnell, J. R., Layman, L., Maselli, O., McGwire, K., Pasteris, D., Dahl-Jensen, D., Steffensen, J. P., Vinther, B., Edwards, R., Mulvaney, R., and Kipfstuhl, S.: A new bipolar ice core record of volcanism from WAIS Divide and NEEM and implications for climate forcing of the last 2000 years, J. Geophys. Res.-Atmos., 118, 1151–1169, 2013.

Simoneit, B. R. T.: Biomass burning – a review of organic tracers for smoke from incomplete combustion, Appl. Geochem., 17, 129–162, 2002.

Simoneit, B. R. T., Schauer, J. J., Nolte, C. G., Oros, D. R., Elias, V. O., Fraser, M. P., Rogge, W. F., and Cass, G. R.: Levoglucosan, a tracer for cellulose in biomass burning and atmospheric particles, Atmos. Environ., 33, 173–182, 1999.

Sorrel, P., Oberhansli, H., Boroffka, N., Nourgaliev, D., Dulski, P., and Rohl, U.: Control of wind strength and frequency in the Aral Sea basin during the late Holocene, Quaternary Res., 67, 371–382, doi:10.1016/j.yqres.2006.12.003, 2007.

Spahni, R., Schwander, J., Fluckiger, J., Stauffer, B., Chappellaz, J., and Raynaud, D.: The attenuation of fast atmospheric CH_4 variations recorded in polar ice cores, Geophys. Res. Lett., 11, 1571, doi:10.1029/2003gl017093, 2003.

Stahle, D. W., Cook, E. R., Cleaveland, M. K., Therrell, M. D., Meko, D. M., Grissino-Mayer, H. D., Watson, E., and Luckman, B. H.: Tree-ring data document 16th century megadrought over North America, EOS Trans. Am. Geophys. Union, 81, 121–125, doi:10.1029/00eo00076, 2000.

St. George, S. S., Meko, D. M., Girardin, M. P., MacDonald, G. M., Nielsen, E., Pederson, G. T., Sauchyn, D. J., Tardif, J. C., and Watson, E.: The tree-ring record of drought on the Canadian Prairies, J. Climate, 22, 689–710, doi:10.1175/2008jcli2441.1, 2009.

Tan, M., Liu, T. S., Hou, J. Z., Qin, X. G., Zhang, H. C., and Li, T. Y.: Cyclic rapid warming on centennial-scale revealed by a 2650-year stalagmite record of warm season temperature, Geophys. Res. Lett., 2, 1617, doi:10.1029/2003gl017352, 2003.

Taylor, K. C., Mayewski, P. A., Twickler, M. S., and Whitlow, S. I.: Biomass burning recorded in the GISP2 ice core: a record from eastern Canada?, Holocene, 6, 1–6, 1996.

The Milwaukee Journal: Alaska, Russia battle blazes, West Palm Beach, Florida, Milwaukee, Wisconsin, 9 August 1972.

The New York Times: Smoke shrouds Moskow as peat-bog fire rages, New York, 9 August 1972.

The Palm Beach Post: Thousands fight Russia Peat fire, 9 August, 1972.
Thompson, L. G., Yao, T., Mosley-Thompson, E., Davis, M. E., Henderson, K. A., and Lin, P. N.: A high-resolution millennial record of the South Asian Monsoon from Himalayan ice cores, Science, 289, 1916–1919, doi:10.1126/science.289.5486.1916, 2000.
Trentmann, J., Luderer, G., Winterrath, T., Fromm, M. D., Servranckx, R., Textor, C., Herzog, M., Graf, H.-F., and Andreae, M. O.: Modeling of biomass smoke injection into the lower stratosphere by a large forest fire (Part I): reference simulation, Atmos. Chem. Phys., 6, 5247–5260, doi:10.5194/acp-6-5247-2006, 2006.
van der Werf, G. R., Randerson, J. T., Collatz, G. J., Giglio, L., Kasibhatla, P. S., Arellano, A. F., Olsen, S. C., and Kasischke, E. S.: Continental-scale partitioning of fire emissions during the 1997 to 2001 El Nino/La Nina period, Science, 303, 73–76, doi:10.1126/science.1090753, 2004.
van der Werf, G. R., Peters, W., van Leeuwen, T. T., and Giglio, L.: What could have caused pre-industrial biomass burning emissions to exceed current rates?, Clim. Past, 9, 289–306, doi:10.5194/cp-9-289-2013, 2013.
van Mantgem, P. J., Stephenson, N. L., Byrne, J. C., Daniels, L. D., Franklin, J. F., Fule, P. Z., Harmon, M. E., Larson, A. J., Smith, J. M., Taylor, A. H., and Veblen, T. T.: Widespread increase of tree mortality rates in the Western United States, Science, 323, 521–524, doi:10.1126/science.1165000, 2009.
Vinther, B. M., Clausen, H. B., Johnsen, S. J., Rasmussen, S. O., Andersen, K. K., Buchardt, S. L., Dahl-Jensen, D., Seierstad, I. K., Siggaard-Andersen, M. L., Steffensen, J. P., Svensson, A., Olsen, J., and Heinemeier, J.: A synchronized dating of three Greenland ice cores throughout the Holocene, J. Geophys. Res.-Atmos., 111, D13102, doi:10.1029/2005jd006921, 2006.
Wang, Z., Chappellaz, J., Park, K., and Mak, J. E.: Large variations in Southern Hemisphere biomass burning during the last 650 years, Science, 330, 1663–1666, doi:10.1126/science.1197257, 2010.
Wang, Z., Chappellaz, J., Martinerie, P., Park, K., Petrenko, V., Witrant, E., Emmons, L. K., Blunier, T., Brenninkmeijer, C. A. M., and Mak, J. E.: The isotopic record of Northern Hemisphere atmospheric carbon monoxide since 1950: implications for the CO budget, Atmos. Chem. Phys., 12, 4365–4377, doi:10.5194/acp-12-4365-2012, 2012.

Wang, Z.-H., Chen, Z.-Y., Kou, Y., and Chen, Y.: Dry/wet climate change since 960 A.D. in taihu drainage basin of China, Chinese Geogr. Sci., 11, 343–349, doi:10.1007/s11769-001-0050-0, 2001.
Wanner, H., Beer, J., Butikofer, J., Crowley, T. J., Cubasch, U., Fluckiger, J., Goosse, H., Grosjean, M., Joos, F., Kaplan, J. O., Kuttel, M., Muller, S. A., Prentice, I. C., Solomina, O., Stocker, T. F., Tarasov, P., Wagner, M., and Widmann, M.: Mid- to Late Holocene climate change: an overview, Quaternary Sci. Rev., 27, 1791–1828, doi:10.1016/j.quascirev.2008.06.013, 2008.
Ward, D. S., Kloster, S., Mahowald, N. M., Rogers, B. M., Randerson, J. T., and Hess, P. G.: The changing radiative forcing of fires: global model estimates for past, present and future, Atmos. Chem. Phys., 12, 10857–10886, doi:10.5194/acp-12-10857-2012, 2012.
Weng, H. Y.: Impacts of multi-scale solar activity on climate, Part I: Atmospheric circulation patterns and climate extremes, Adv. Atmos. Sci., 29, 867–886, doi:10.1007/s00376-012-1238-1, 2012.
Westerling, A. L., Hidalgo, H. G., Cayan, D. R., and Swetnam, T. W.: Warming and earlier spring increase western US forest wildfire activity, Science, 313, 940–943, doi:10.1126/science.1128834, 2006.
Whitlow, S., Mayewski, P., Dibb, J., Holdsworth, G., and Twickler, M.: An ice-core-based record of biomass burning in the arctic and sub-arctic, 1750–1980, Tellus B, 46, 234–242, 1994.
Wolfe, S. A., Huntley, D. J., David, P. P., Ollerhead, J., Sauchyn, D. J., and MacDonald, G. M.: Late 18th century drought-induced sand dune activity, Great Sand Hills, Saskatchewan, Can. J. Earth Sci., 38, 105–117, doi:10.1139/cjes-38-1-105, 2001.
Wolff, E. W., Barbante, C., Becagli, S., Bigler, M., Boutron, C. F., Castellano, E., de Angelis, M., Federer, U., Fischer, H., Fundel, F., Hansson, M., Hutterli, M., Jonsell, U., Karlin, T., Kaufmann, P., Lambert, F., Littot, G. C., Mulvaney, R., Rothlisberger, R., Ruth, U., Severi, M., Siggaard-Andersen, M. L., Sime, L. C., Steffensen, J. P., Stocker, T. F., Traversi, R., Twarloh, B., Udisti, R., Wagenbach, D., and Wegner, A.: Changes in environment over the last 800,000 years from chemical analysis of the EPICA Dome C ice core, Quaternary Sci. Rev., 29, 285–295, doi:10.1016/j.quascirev.2009.06.013, 2010a.
Wolff, E. W., Chappellaz, J., Blunier, T., Rasmussen, S. O., and Svensson, A.: Millennial-scale variability during the last glacial: the ice core record, Quaternary Sci. Rev., 29, 2828–2838, doi:10.1016/j.quascirev.2009.10.013, 2010b.

Wolken, J. M., Hollingsworth, T. N., Rupp, T. S., Chapin, F. S., Trainor, S. F., Barrett, T. M., Sullivan, P. F., McGuire, A. D., Euskirchen, E. S., Hennon, P. E., Beever, E. A., Conn, J. S., Crone, L. K., D'Amore, D. V., Fresco, N., Hanley, T. A., Kielland, K., Kruse, J. J., Patterson, T., Schuur, E. A. G., Verbyla, D. L., and Yarie, J.: Evidence and implications of recent and projected climate change in Alaska's forest ecosystems, Ecosphere, 2, 124, doi:10.1890/es11-00288.1, 2011.

Yalcin, K., Wake, C. R., Kreutz, K. J., and Whitlow, S. I.: A 1000-yr record of forest fire activity from Eclipse Icefield, Yukon, Canada, Holocene, 16, 200–209, doi:10.1191/0959683606hl920rp, 2006.

Yang, B., Braeuning, A., Johnson, K. R., and Shi, Y. F.: General characteristics of temperature variation in China during the last two millennia, Geophys. Res. Lett., 29, 1324, doi:10.1029/2001gl014485, 2002.

Yang, B., Brauning, A., Zhang, Z., Dong, Z., and Esper, J.: Dust storm frequency and its relation to climate changes in Northern China during the past 1000 years, Atmos. Environ., 41, 9288–9299, doi:10.1016/j.atmosenv.2007.09.025, 2007.

Zhang, P. Z., Cheng, H., Edwards, R. L., Chen, F. H., Wang, Y. J., Yang, X. L., Liu, J., Tan, M., Wang, X. F., Liu, J. H., An, C. L., Dai, Z. B., Zhou, J., Zhang, D. Z., Jia, J. H., Jin, L. Y., and Johnson, K. R.: A test of climate, sun, and culture relationships from an 1810-year Chinese cave record, Science, 322, 940–942, doi:10.1126/science.1163965, 2008.

Zhang, Y. H., Wooster, M. J., Tutubalina, O., and Perry, G. L. W.: Monthly burned area and forest fire carbon emission estimates for the Russian Federation from SPOT VGT, Remote Sens. Environ., 87, 1–15, doi:10.1016/s0034-4257(03)00141-x, 2003.

Table 1. Major levoglucosan peaks (with levoglucosan concentrations above the threshold 3rd $Q+1.5$ IR) and their correspondence with other biomass burning proxies. Samples in bold have levoglucosan concentrations above the mean plus one standard deviation. The NH_4^+ samples have the same ages as the levoglucosan samples. Blank spaces demonstrate the absence of high values (Z-scores > 1), "n.a." if no values were available.

Levoglucosan						Black Carbon			Ammonium	
Sample Age (yr CE)		Average age (yr CE)	[levoglucosan] ($pg\,mL^{-1}$)	Z-score[1]	Z-score[2]	Age (yr CE)	[BC] ($ng\,g^{-1}$)	Z-score[2]	[NH_4^+] ($ng\,g^{-1}$)	Z-score[2]
1975	**1972**	**1973**	**301**	**1.37**	**1.45**	1972.5	6.73	1.68	13.77	1.16
1791	**1787**	**1789**	**612**	**3.39**	**3.56**	1789.5	5.81	1.26		
1706	**1702**	**1704**	**336**	**1.59**	**1.69**	1703.5	10.02	3.15	13.75	1.16
						1702.5	5.85	1.28		
1622	**1617**	**1620**	**521**	**2.80**	**2.94**	1619.5	6.35	1.51		
1603[3]	1593[3]	1598[3]	170[3]	0.51[3]	0.55[3]	1594.5	5.29	1.03	14.98[3]	1.39[3]
1319	1313	1316	193	0.66	0.71					
1177	1171	1174	201	0.71	0.77	1170.5	5.80	1.26		
1112	1106	1109	229	0.90	0.96					
1041	**1036**	**1039**	**393**	**1.96**	**2.07**					
979	**973**	**976**	**245**	**1.00**	**1.06**	974.5	5.38	1.07		
924	**919**	**922**	**483**	**2.55**	**2.69**	922.5	18.85	7.11	21.93	2.71
745	739	742	208	0.76	0.81	739.5	12.02	4.05	13.01	1.02
631	**625**	**628**	**391**	**1.95**	**2.06**	630.5	11.58	3.85	26.07	3.50
562	556	559	192	0.65	0.70	559.5	9.35	2.85	15.07	1.41
451	446	449	188	0.63	0.68	446.5	8.47	2.46		
345	**339**	**342**	**1767**	**10.92**	**11.42**	345.5	6.26	1.46	15.83	1.55
						340.5	45.32	18.98		
225	219	222	170	0.51	0.56	224.5	8.23	2.35	16.02	1.59
						223.5	7.22	1.90		
						221.5	6.69	1.66		
−196	**−202**	**−199**	**349**	**1.68**	**1.78**	n.a.	n.a.	n.a.	26.44	3.57
−271	**−277**	**−274**	**401**	**2.02**	**2.13**	n.a.	n.a.	n.a.	22.08	2.74
−327	**−333**	**−330**	**382**	**1.89**	**2.00**	n.a.	n.a.	n.a.		
−389	**−396**	**−392**	**1445**	**8.82**	**9.23**	n.a.	n.a.	n.a.		
−524	−531	−527	192	0.65	0.70	n.a.	n.a.	n.a.	21.56	2.64
−537	**−543**	**−540**	**266**	**1.14**	**1.21**	n.a.	n.a.	n.a.	12.97	1.01
−584	−590	−587	185	0.61	0.66	n.a.	n.a.	n.a.		

[1] Mean and standard deviation calculated for the entire levoglucosan dataset (1036 BCE–1999 CE).
[2] Mean and standard deviation calculated for the common period (87–1992 CE) of the BC, levoglucosan and ammonium dataset in order to compare datasets covering different temporal periods.
[3] Levoglucosan has been analyzed in a 4-bag sample (1593–1603 CE); ammonium value is referred to a 2-bag sample (1598–1603 CE).

Table 2a. Asian droughts recorded in various proxies with strong fire events **(a)** and with centennial fire activity in the 16th and 17th century inferred from levoglucosan analysis **(b)**.

Age	Location	Proxy	Reference	Comment
1171–1177	**NEEM ice core**	**levoglucosan**	**this work**	**strong event**[1]
1170	Taihu Drainage Basin China, Eastern Cost	historical climatic records	Wang et al. (2001)	abrupt dry/wet climate change
1160–1290	Northern China (reconstruction based on different paleoclimate archives)	flood/drought index	Yang et al. (2007) and ref. therein	max frequency of dust storm
1313–1319	**NEEM ice core**	**levoglucosan**	**this work**	**strong event**[1]
1320–1370	Northern China (reconstruction based on different paleoclimate archives)	flood/drought index	Yang et al. (2007) and ref. therein	high dust fall frequency
1330s	Dasuopu ice core, Himalaya (India and Asia as source regions)	$\delta^{18}O$, dust, Cl^-	Thompson et al. (2000)	monsoon failure
1593–1603	**NEEM ice core**	**levoglucosan**	**this work**	**strong event**[1]
1617–1622	**NEEM ice core**	**levoglucosan**	**this work**	**megaevent**[2]
1578–1582 1600–1644	Mongolia	tree-ring	Davi et al. (2010)	2nd and 1st in order of severity since 1520
1590s-1600s	Central High Asia	tree-ring	Fang et al. (2010)	
1590s	Dasuopu ice core, Himalaya (India and Asia as source regions)	$\delta^{18}O$, dust, Cl^-	Thompson et al. (2000)	monsoon failure
exceptional events: 1586–1589; 1638–1641 decadal time scale: 1590s	Eastern China	drought/flood proxy data	Shen et al. (2007)	most severe events (exceptional) of past 5 centuries
1618–1635	Taihu Drainage Basin China, Eastern Cost	historical climatic records	Wang et al. (2001)	abrupt change to dry period
1702–1706	**NEEM ice core**	**levoglucosan**	**this work**	**megaevent**[2]
1694–1705	Changling Mountains, North-central China	tree-ring	Feng et al (2011)	coincident with Maunder Minimum[3]
1787–1791	**NEEM ice core**	**levoglucosan**	**this work**	**megaevent**[2]
1795–1823	Changling Mountains, north-central China	tree-ring	Chen et al. (2012)	coincides with Dalton Minimum[3]
1790–1799	North Helan Mountain, inner Mongolia	tree-ring	Liu et al. (2004)	
1791–1795	Northeastern Mongolia	tree-ring	Pederson et al. (2001)	
1779–1806	Chiefeng-Weichang region, China	tree-ring	Liu et al. (2010)	
1790–1796	Dasuopu ice core, Himalaya (India and Asia as source regions)	$\delta^{18}O$, dust, Cl^-	Thompson et al. (2000)	monsoon failures "exceptionally large from a perspective of the last 1000 yr"
1789–1793	India	archival evidences of drought	Grove (1998)	

Table 2b. Asian droughts recorded in various proxies with strong fire events **(a)** and with centennial fire activity in the 16th and 17th century inferred from levoglucosan analysis **(b)**.

Period	Location	Proxy	Reference	Comment
1500–1700 centennial scale: 16th, 17th century	**NEEM ice core** Eastern China	**levoglucosan** drought/flood proxy data	**this work** Shen et al. (2007)	**multi-decadal analysis**
1610–1710	Northern China (reconstruction based on different paleoclimate archives)	flood/drought index	Yang et al. (2007) and references therein	high dust fall frequency
1540–1600	Siberian Altai (Southern Siberia)	ice core pollen data and dust	Eichler et al. (2011)	extremely dry period followed by high fire activity
1414–1560	Lake Teletskoye, Republic of Altai (Russian Federation)	sediment core pollen data	Andreev et al. (2007)	
1450–1600	Eastern China	historical documents	Chu et al. (2008)	negative snow anomaly events
1400–1550	East Juyanhai Lake Inner Mongolia	sedimentology and geochemical parameters	Chen et al. (2010)	extremely arid conditions
1630s-1640s	Xiaolong Mountain, central China	tree-ring	Fang et al. (2012)	one of most severe droughts of the past 400 yr
1630s-1640s	China and Mongolia	tree-ring	Li et al. (2009)	
1640s	Dasuopu ice core, Himalaya (India and Asia as source regions)	$\delta^{18}O$, dust, Cl^-	Thompson et al. (2000)	monsoon failure
1640s-1650s	Central High Asia	tree-ring	Fang et al. (2010)	most severe megadrought

[1] This event represents levoglucosan concentration above the threshold 3rd Q + 1.5 IR. Where 3rd Q is the third quartile and IR is the interquartile range calculates as the difference between the third quartile and the first quartile. This peak was removed from the long-trend analysis.
[2] This event represents levoglucosan concentration above the mean plus one standard deviation.
[3] As noted by Chen (2012), the drying of the late 1600s to early 1700s, and the drying of the late 1700s to early 1800s, seen in the precipitation reconstructions, coincides with periods of low solar irradiance, the Dalton Minimum and the Maunder Minimum, respectively.
[4] Generally drought is recorded in tree-rings as a sustained narrowness of growth rings.

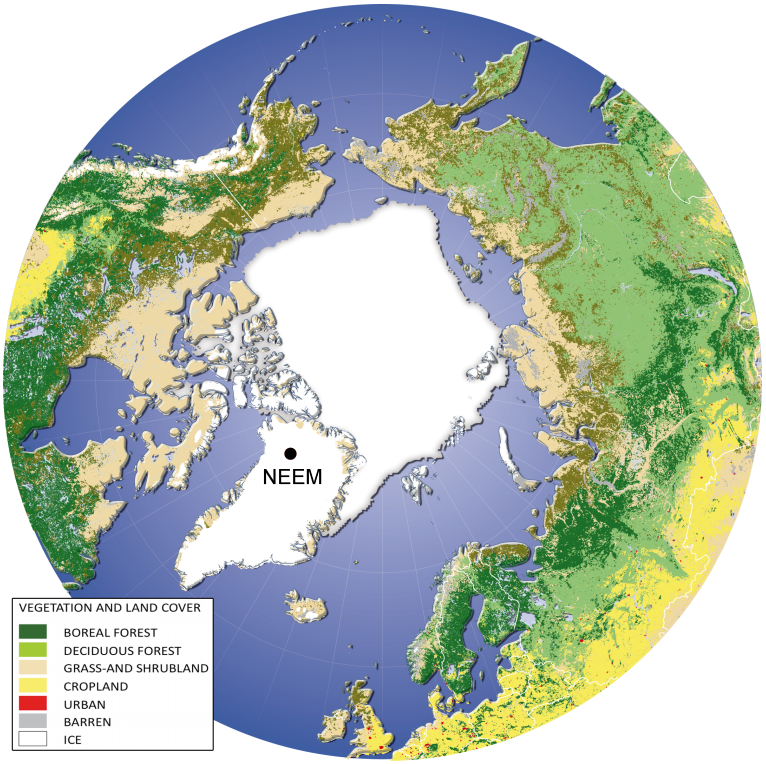

Fig. 1. NEEM camp position and representation of boreal vegetation and land cover between 50° and 90° N. Modified from the European Commission Global Land Cover 2000 database and based on the work of cartographer Hugo Alhenius UNEP/GRIP-Arendal (Alhenius, 2003).

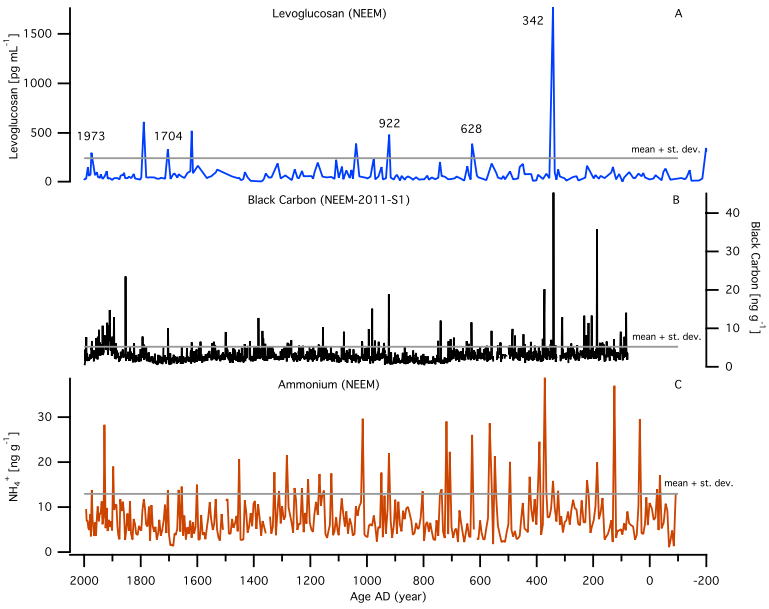

Fig. 2. (A) Levoglucosan concentration profile measured in the deep NEEM core including the dates of megafires coincident between levoglucosan, black carbon, and ammonium; **(B)** black carbon concentration profile measured in the NEEM-2011-S1 ice core; **(C)** ammonium concentration profile in the deep NEEM ice core.

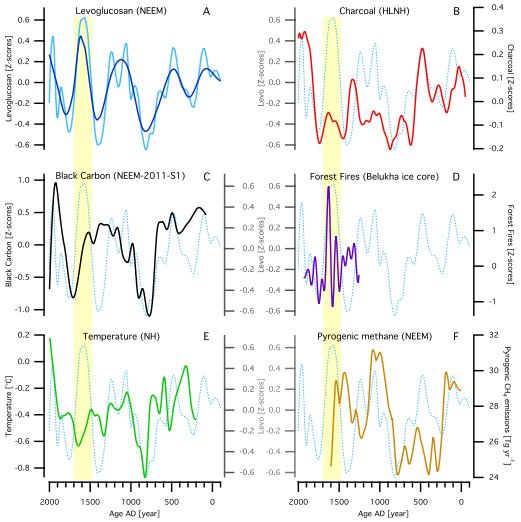

Fig. 3. LOWESS smoothing with SPAN parameter (f) 0.1 (light blue) and 0.2 (dark blue) of levoglucosan Z-scores without peaks above the threshold 3rd_Q + 1.5 × IR **(A)**; High Latitude (above 55° N) Northern Hemisphere (HLNH) Z-scores of charcoal influx (200 year LOWESS smoothing) as reported in Marlon et al. (2008) (red) and smoothed levoglucosan with f = 0.1 as in A (dashed line) **(B)**; LOWESS smoothing with f = 0.1 of Black Carbon Z-scores without peaks above the threshold 3rd_Q + 1.5 × IR (black) and smoothed levoglucosan (dashed line) **(C)**; Siberian forest fire reconstruction (Eichler et al., 2011) (purple) and smoothed levoglucosan (dashed line) **(D)**; LOWESS smoothing with f = 0.1 of Northern Hemisphere land temperature (Mann et al., 2008) and smoothed levoglucosan (dashed line) **(E)**; pyrogenic CH_4 emissions inferred from the deep NEEM core (Sapart et al., 2012) (light brown) and smoothed levoglucosan (dashed line) **(F)**. The yellow vertical bar indicates the period of strongest fire activity.