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Fire in ice: two millennia of boreal forest 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 spanning millennia, and are thus useful to examine the role of fire in the carbon cycle and climate system. Here we use the specific biomarker levoglucosan together with black carbon and ammonium concentrations from the North Greenland Eemian (NEEM) ice cores (77.49° N, 51.2° W; 2480 m a.s.l) over the past 2000 years to infer changes in boreal fire activity. Increases in boreal fire activity over the periods 1000–1300 CE and decreases during 700–900 CE coincide with high-latitude NH temperature changes. Levoglucosan concentrations in the NEEM ice cores peak between 1500 and 1700 CE, and most levoglucosan spikes coincide with the most extensive central and northern Asian droughts of the past millennium. Many of these multi-annual droughts are caused by Asian 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 Greenland Ice Cap. 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, 2013). Biomass burning plays an important role in the carbon cycle, as it emits up to 50 % as much CO_2 as fossil fuel combustion (Bowman et al., 2009). Fire products such as black carbon (BC) have a radiative absorption forcing up to 55 % of 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. Biomass burning may have a short-term (10 years) net negative forcing on global climate, where long-lived greenhouse gasses may then cancel the cooling after decades (100 years) (Jacobson, 2004). The 5th IPCC report suggests a net radiative forcing of $+0.00 \pm 0.20\,\mathrm{W\,m^{-2}}$ (IPCC, 2013) due to biomass burning; however, human activities may have also changed the net radiative forcing of pre-industrial fires (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 decreases fuel flammability (Pechony and Shindell, 2010), but biomass burning is highest at intermediate moisture levels (Daniau et al., 2012); a balance must therefore be struck between climatic conditions that are wet enough to allow biomass to grow but 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 have also had an influence on biomass burning trends (Marlon et al., 2008) through deforestation and slash-and-burn agricultural practices (FAO, 2007) – practices which may have affected fire regimes during the entire past millennia (Bowman et al., 2009; Kaplan et al., 2011; Ruddiman, 2003). 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 has begun to influence the global fire regime more than natural causes (Marlon et al., 2008; Pechony and Shindell, 2010).

1.1 Boreal forest fires

The circumpolar boreal zone contains about 30 % of world's forests, with half of the world's forest carbon and about 30 % of world's terrestrial carbon. Any change to the boreal forest carbon balance may therefore considerably affect the global atmospheric CO_2 (Conard et al., 2002). On average, boreal and temperate vegetation fires represent 4 % of global biomass burning and up to 12 % during extreme burning years, when corresponding BC emissions account for about 20 % of the worldwide BC emissions from all fires (Lavoue et al., 2000). Thus, boreal forest fires are an important source of air pollutants throughout the Arctic, including BC (Quinn et al., 2008; Lavoue et al., 2000). Due to the light-absorbing properties of BC and the associated decrease in surface snow albedo, boreal forest fire BC has a significant radiative impact on the Arctic atmosphere (Quinn et al., 2008; Flanner et al., 2007; Hansen and Nazarenko, 2004).

North American and Eurasian boreal fire regimes differ due to distinctive fuel, weather and fire ecology (Wooster and Zhang, 2004; de Groot et al., 2013). Mean fire intensity and fuel consumption is higher in North America than in Russia, even though the spatial extent of forests is greater in Russia (Wooster and Zhang, 2004; de Groot et al., 2013). Different fire regimes suggest a dominance of crown fires in North America and surface fires in Russia (Wooster and Zhang, 2004; de Groot et al., 2013), thus suggesting the possibility of different chemical pathways in pyro-product production and emissions. Accordingly, Russian fires may burn less fuel than North American fires (de Groot et al., 2013) and thus emit less pyro-products into the atmosphere per unit of area burned (Wooster and Zhang, 2004).

Boreal forest fires in Siberia may have a larger impact on total global fire emissions than those in North America due to the larger Siberian burn area (Stohl, 2006; Stohl et al., 2006). Russian boreal forests, the majority of which are located in Siberia, accounted for 14–20 % of average annual global carbon emissions from forest fires in 1998 (Conard et al., 2002). However, little is known about the emission, transport and deposition of BC from Siberian forest fires to the Arctic. The characterization of Siberian fire plumes is uncertain, due to currently undefined biomass fraction consumed, combustion efficiency, plume injection heights and emission ratios (Paris et al., 2009). Models are sensitive to the conditions and types of fires, such as the injection height of fire emissions (Turquety et al., 2007), and the lack of data about Siberian fires introduces large potential errors in estimating the impact of boreal wildfires on the atmospheric composition (Conard et al., 2002).

Summer atmospheric conditions favour enhanced wet and dry BC deposition in the Arctic, while dry winters allow for longer aerosol lifetimes due to lower wet deposition (Stohl, 2006). Generoso et al. (2007) determined that the 2003 Russian fires contributed to approximately 40–56 % of the total BC mass deposited north of 75° N. In fact, model results indicate that boreal forest fire sources, especially Siberian fires, are the dominant summertime source of BC reaching the Arctic, exceeding all contributions from anthropogenic sources (Stohl, 2006). Northern Eurasian and Siberian fire emissions such as BC can reach the Arctic more easily than emissions from North American regions at similar latitudes (Stohl, 2006; Stohl et al., 2006; Lavoue et al., 2000).

1.2 Biomass burning proxies

There are many potential indicators or proxies of past fire activity, such as altered products of plant combustion (e.g. charcoal, BC), partially combusted biological material (e.g. fire scars in tree rings), or chemical markers directly produced and volatilized during vegetation combustion (e.g. resin acids, polycyclic aromatic hydrocarbons) (Conedera et

al., 2009). Some proxies are specific as they are produced solely from biomass burning, such as charcoal in sediment cores, but most others have multiple potential sources other than wildfires (e.g. coal burning or biogenic emissions). Ice cores from polar regions are widely used to reconstruct detailed climate records over the past hundreds of thousands of years, and are thus powerful tools in palaeoclimate research (Legrand and Mayewski, 1997).

Examining forest fires in ice cores started with the pioneering work of Legrand et al. (1992), which suggested that the concentration of ammonium and light carboxylic acids, such as formic and oxalic acids, significantly increase above background levels in Greenland ice layers during forest fire events. This was confirmed by Dibb et al. (1996) and Jaffrezo et al. (1998), who reported a sudden increase of the atmospheric levels of these species at Summit (central Greenland) in August 1994 and June 1993, when biomass burning plumes were transported to the site from the northern Canada. Finally, Kehrwald et al. (2012) showed that the Summit snow layer corresponding to the August 1994 event exhibits oxalate and levoglucosan concentration peaks.

The subsequent work of Legrand and DeAngelis (1996) demonstrates that, on average, high-latitude biomass burning contributes between 20 and 30 % to the formate, oxalate and ammonium deposited in central Greenland over the last 200 years. However, for a typical biomass burning input the authors report strong enhancements with respect to background values for ammonium (up to a factor of 20) and formate (up to a factor of 100), where the ammonium to formate molar ratio during the event is close to unity (Legrand and DeAngelis, 1996). In addition to strong increases of ammonium, formate and oxalate concentrations, Savarino and Legrand (1998) found an increase of nitrate coincident with some of the fire events recorded in Greenland ice, indicating that nitrate is not a good proxy of forest fire inputs to Greenland ice. Finally, potassium is sometimes used to identify the contribution of biomass burning in atmospheric air masses (Andreae, 1983; Gao et al., 2003). Of the available ionic compounds, K^+ cannot be considered a suitable indicator for forest fire plumes reaching Greenland due to marine and terrestrial sources, even if corrected to remove these contributions (e.g. non-sea-salt and non-crustal K^+) (Legrand and DeAngelis, 1996; Savarino and Legrand, 1998).

NH_4^+, NO_3^- and organic acids in Greenland ice were often successfully 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; Savarino and Legrand, 1998). Many high fire years are identifiable by enhanced NH4+ concentrations in continental environmental archives (Eichler et al., 2011), yet the use of ammonium is less straightforward than in Greenland due to the presence of large biogenic sources in non-polar regions. However, statistical attempts to recognize forest fire signals from multiple ice core data sets used empirical orthogonal function (EOF) analysis or principal component analysis (PCA), which identify chemical associations between pyro-ion records and differentiate between sources and transport characteristics (Yalcin et al., 2006; Eichler et al., 2011).

The isotopic ratio of atmospheric gases, such as methane (CH_4) in Antarctic (Ferretti et al., 2005) and Arctic (Sapart et al., 2012) ice cores, has also been used as a fire proxy, but such ratios are also not solely products of biomass burning (Fischer et al., 2008). The isotopic composition of methane (δ^{13}C of CH_4) in ice cores differentiates biogenic from pyrogenic sources (Sapart et al., 2012). Methane is stable in the atmosphere for longer than the atmospheric exchange time and is therefore globally distributed. Hence δ^{13}C of CH_4 variations are impacted by sources distributed all over the world (Blunier et al., 2007).

BC is emitted during incomplete combustion of fossil and biofuels in fires ignited by both natural and human sources (Preston and Schmidt, 2006; McConnell et al., 2007). BC is not a single chemical compound, however, and lacks well-defined characteristics (Goldberg, 1985; Masiello, 2004). However, comparison of BC with vanillic acid, a lignin phenol tracer of conifer combustion (Simoneit, 2002), in an ice core from west central Greenland allowed McConnell (2007) to separate out the contributions of biomass burning and fossil fuels to the BC flux reaching Greenland. Other specific tracers for biomass burning, including levoglucosan (vegetation fires) and *p*-hydroxybenzoic and dehydroabietic acids (conifer fires), have been analysed in high- and mid-latitude ice cores from the Kamchatka Peninsula, northeastern Asia (Kawamura et al., 2012) and from the Tibetan Plateau (Yao et al., 2013).

Monosaccharide anhydrides are one of the few fire proxies that have specific sources (Gambaro et al., 2008; Simoneit, 2002). Levoglucosan (1,6-anhydro-β-D-glucopyranose) is a monosaccharide anhydride released during biomass burning when cellulose combustion occurs at temperatures > 300 °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 preserved in the 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 as chemical processing causes short lifetimes (of the order of days to weeks) (Hennigan et al., 2010; Fraser and Lakshmanan, 2000; Ramanathan and Carmichael, 2008; Fuhrer and Legrand, 1997) in addition to efficient deposition 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 means that levoglucosan remains a strong potential tracer for fire activity even across remote distances

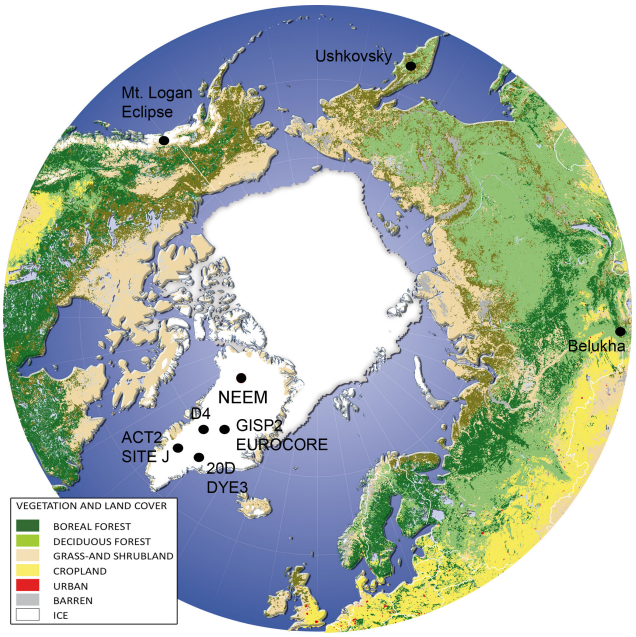

Figure 1. NEEM camp position and representation of boreal vegetation and land cover between 50 and 90° N. Modified from the European Commission Global Land Cover 2000 database and based on the work of cartographer Hugo Alhenius UNEP/GRIP-Arendal (Alhenius, 2003).

(Fraser and Lakshmanan, 2000; Holmes and Petrucci, 2006, 2007; Kehrwald et al., 2012).

Charcoal deposited both in non-polar ice and sediment cores can also help reconstruct past fire activity. The Global Charcoal Database (GCD) compiles individual charcoal records from terrestrial and marine sediment cores (Supplement Fig. S6) 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 (Supplement Fig. S6). As levoglucosan and BC can be transported hundreds to thousands of kilometres (Mochida et al., 2010), they complement GCD reconstructions that record combustion within tens of kilometres of the sampling sites, and provide fire activity data for regions where charcoal records do not exist.

1.3 Aim of the study

Here, we use a multi-proxy approach to reconstructing biomass burning in the northern high latitudes 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., 2013). We compare our results with Northern Hemisphere fire records, climate conditions obtained from historical records and palaeoarchives 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 °C; accumulation 0.22 m ice equivalent per year). The deep NEEM ice core (Fig. 1) was drilled from 2008 to 2012 and reached a depth of 2540 m. Ice core analyses were partly performed in the field and partly in the laboratories of the participating nations. Our levoglucosan data set consists of 273 samples from 4.95 to 602.25 m depth (1999 CE–1036 BCE), where each sample is a 1.10 m inner core section collected after being melted within 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 (see Supplement) using liquid chromatography/negative ion electrospray ionization – tandem mass spectrometry (HPLC/(-)ESI-MS/MS) at picogram per millilitre 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 $^{13}C_6$-labelled 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 the 2009 season (Kaufmann et al., 2008). All the major ions are available at millimetre resolution, and we calculated 1.1 m mean values in order to directly compare with levoglucosan.

The upper 1419 m of deep NEEM core $\delta^{18}O$, volcanic fingerprints, electrical conductivity measurement,

dielectrical profiling, and impurity records can be matched to the NGRIP (North Greenland Ice Core Project) GICC05 extended timescale (Supplement Fig. S7) (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 (Supplement Fig. S7) (Herron and Langway Jr, 1980).

Continuous flow analysis allows for sub-seasonal reconstructions of BC in the NEEM-2001-S1 core (Sigl et al., 2013) 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 ~ 1.10 m long samples with cross sections of ~ 0.033 m by 0.033 m. These longitudinal samples were analysed in early autumn 2011 using a well-established continuous ice core analytical system (McConnell, 2002; Banta et al., 2008), 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 with dating uncertainty of < 1 year. (Sigl et al., 2013). 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., 2013), as for the deep NEEM core dating (Supplement).

The BC and levoglucosan measurements were sampled at different resolutions from two parallel ice cores, where BC samples and levoglucosan samples represent similar, but not equal, average age of the ice. BC is also sampled at a higher resolution (sub-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 in dating, we chose not to calculate BC averages from the 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, instead leaving them as annual BC concentrations.

2.2 Statistics

One of the first challenges to understanding that levoglucosan reflects 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, and can provide clues as to 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 Holocene contribution of sea salts to total Greenland Ca^{2+} ice concentrations is estimated to be of the order of 10 % (Fischer et al., 2007). In order to avoid including possible errors due to data transformation by using the seawater Ca^{2+}/Na^{+} ratio, we use Ca^{2+} as a dust tracer without correcting for sea salt contribution.

2.3 Short- to long-term fire variation

For short-term analysis, we consider *megafires* as levoglucosan peaks with a Z score ≥ 1 [Z score $= (x_i - \bar{x})/\sigma$)], corresponding to a concentration $\geq 245\,\mathrm{pg\,mL^{-1}}$ (Table 1, Supplement Fig. S5). Multi-decadal fire activity was extracted from the levoglucosan and BC concentration profiles by analysing smoothed data after removing spikes above a fixed threshold (Supplement). 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 archives hundreds of sedimentary records of fire (Power et al., 2010). Regional and global synthesis allows for the examination of broad-scale patterns in palaeofire activity (Marlon et al., 2013). In order to compare levoglucosan data with decadal to centennial trends in other palaeoclimate records, we first applied the same standardized statistical procedures as were used to analyse the Global Charcoal Database (Marlon et al., 2008; Power et al., 2008). We modified this method for the NEEM data as it is from a single site (unlike the GCD) and correspondence to any outlying values resulting in artifacts into LOWESS (locally weighted scatterplot smoothing). A common approach to isolate the influence of spikes 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. In summary, to highlight long-term fire changes, we followed the following procedure (details are included in the Supplement):

- removing spikes above a fixed threshold,
- Z-score transformation,
- LOWESS modelling.

Table 1. Major levoglucosan peaks (with levoglucosan concentrations above the threshold 3rd $Q + 1.5$ IR) and their correspondence to 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." means that 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 data set (1036 BCE–1999 CE).
[2] Mean and standard deviation calculated for the common period (87–1992 CE) of the BC, levoglucosan and ammonium data set in order to compare data sets covering different temporal periods.
[3] Levoglucosan has been analysed in a four-bag sample (1593–1603 CE); ammonium value is referred to a two-bag sample (1598–1603 CE).

To analyse the cross-correlation between different time series which are irregularly spaced, we used cross-correlation analyses techniques based on Gaussian kernel methods (Bjornstad and Falck, 2001) which are suitable for palaeodata (Rehfeld et al., 2011). To further investigate cross-correlation, we performed a local analysis (local cross-correlation function), where we applied the abovementioned method to temporal windows.

3 Results

Pearson and Spearman correlations between 1.1 m averaged data (Supplement) demonstrate that, of the possible biomass burning-related products measured in the deep NEEM core, levoglucosan concentrations only significantly correlate with ammonium (p value $<10^{-6}$, with $\alpha = 0.05$). No correlation is noted between levoglucosan and crustal marker Ca^{2+} or dust (p values > 0.45, with $\alpha = 0.05$). Analysis of preliminary measurements of other crustal markers (i.e. Ti and Ba) in the deep NEEM ice core also shows a lack of correlation with levoglucosan (Gabrieli, J., 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 preindustrial periods, BC concentrations generally do not correlate with crustal particulate tracers but do with NH_4^+ and NO_3^- (J. R. McConnell, personal communication, 2014). Cross-correlation analyses between levoglucosan time series and high-latitude (north of 55° N) Northern Hemisphere charcoal synthesis, NEEM black carbon, Northern Hemisphere land temperature and NEEM pyrogenic methane (Fig. 3) are presented in the Supplement (Figs. S8–S15);

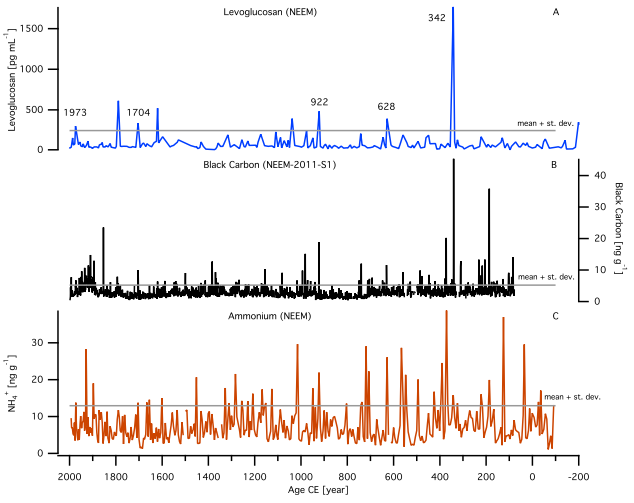

Figure 2. (a) Levoglucosan concentration profile measured in the deep NEEM core including the dates of megafires coincident between levoglucosan, black carbon and ammonium; **(b)** black carbon concentration profile measured in the NEEM-2011-S1 ice core; **(c)** ammonium concentration profile in the deep NEEM ice core.

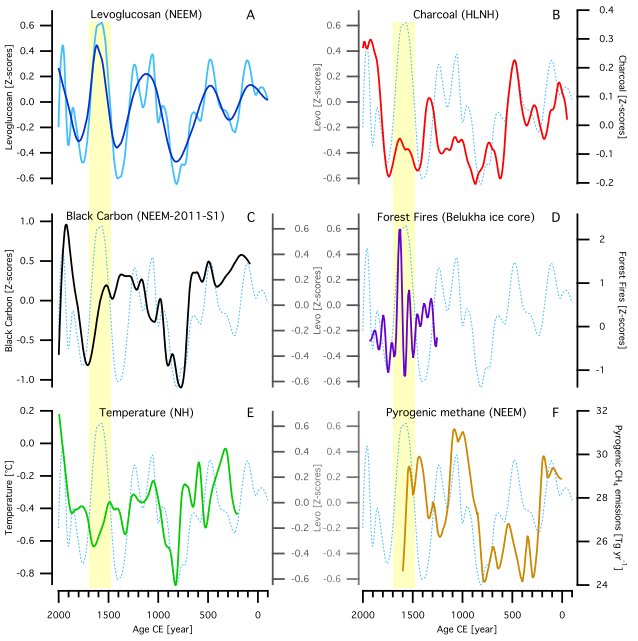

Figure 3. (a) LOWESS with SPAN parameter (f) 0.1 (light blue) and 0.2 (dark blue) of levoglucosan Z scores without peaks above the threshold 3rd_Q + 1.5 × IR; **(b)** high-latitude (above 55° N) Northern Hemisphere (HLNH) Z scores of charcoal influx (200-year LOWESS) as reported in Marlon et al. (2008) (red) and smoothed levoglucosan with $f = 0.1$ as in **(a)** (dashed line); **(c)** LOWESS 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); **(d)** Siberian forest fire reconstruction (Eichler et al., 2011) (purple) and smoothed levoglucosan (dashed line); **(e)** LOWESS with $f = 0.1$ of Northern Hemisphere land temperature (Mann et al., 2008) and smoothed levoglucosan (dashed line); **(f)** pyrogenic CH_4 emissions inferred from the deep NEEM core (Sapart et al., 2012) (light brown) and smoothed levoglucosan (dashed line). The yellow vertical bar indicates the period of strongest fire activity.

these analyses demonstrate that only the correlation with temperature tends to be significant over regional scales. On local scales, high (positive or negative) correlation is present in almost all measurements, depending on the time period. For example, the period before 500 BP highly positively correlates with temperature and highly negatively correlates with methane.

Atmospheric transport plays a fundamental role in recording fire signatures in polar ice cores. Levoglucosan spikes require that very intense fires occur in the source area and generate levoglucosan-rich plumes that are lifted rapidly. Transport conditions have to be favourable for transport of plumes from the location of fire events to the Arctic. Meteorological conditions regulate loss and dispersion by wet/dry deposition/scavenging of the plumes. If transport and/or meteorological conditions are unfavourable or if precipitation removes the pyro-products before the ice core site, fires will not be detected, or may be detected elsewhere on the Greenland Ice Sheet. In other words, levoglucosan spikes may be produced by mega-events occurring at remote distances or by modest events closer to the NEEM camp. Here we present the megafires recorded at the NEEM position.

Megafires (levoglucosan peaks with Z scores > 1) are observed at 1973, 1789, 1704, 1620, 1039, 976, 922, 628, 342 CE and at 199, 274, 330, 392 and 540 BCE (Fig. 2 and 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 and 1300 CE and 1500 and 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 and 1700–1800 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 and 1600 CE and 1600 and 1800 CE the BC profile shows a relative maximum and minimum, respectively, while 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 Major ions transport and deposition on the Greenland Ice Sheet

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 (Ruth et al., 2003; Fischer et al., 2007). The lack of correlation between crustal markers and levoglucosan suggests that levoglucosan enhancements are not only caused by increased wind strength or transport efficiency, but also reflect changes in source activity (forest fires). This evidence differs from the conclusion of Kawamura et al. (2012), who 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 biogenic emissions and temperature changes (Fuhrer et al., 1993; Fuhrer et al., 1996; Legrand et al., 1992; Fuhrer and Legrand, 1997). Basing NEEM fire reconstructions only on ammonium concentrations that are not corrected for the natural variability could potentially overestimate major fire events; however, comparing various fire proxies from the same core supplements biomass burning interpretations. The literature uses PCA for pyro-reconstructions (Yalcin et al., 2006; Eichler et al., 2011). Incorporating as many fire proxies as possible within the same ice core allows for 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 PCA 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", by localized fire events or by efficient transport of distant fires rather than by long-term increases in fire activity (Fig. 2 and S5 in the Supplement). In addition, the magnitude of spikes has to be high enough to not be lost/smoothed when analysing multi-year 1.1 m ice samples. As the mean accumulation rate in north central Greenland has been relatively constant over the past 3000 years (Andersen et al., 2006), we present levoglucosan and BC concentrations rather than fluxes.

Many levoglucosan spikes correspond to 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 data sets, 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).

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 whether relatively local metal production may have caused these levoglucosan peaks.

Other ice core records provide comparisons of fire events that allow for 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), even if this event is not recorded in ice core from other locations in Greenland (e.g. Savarino and Legrand, 1998). Contemporaneous fire signatures to 1787–1791 CE peak are reported in Greenland (GISP2, 20D) 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). Mega-events 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).

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. The highest ammonium peak in a Summit, Greenland, ice core spanning 1908–1989 CE occurred in 1908 CE and is synchronous with increases in formate, oxalate, glycolate and BC within the same core (Legrand et al., 1995). Legrand et al. (1995) ascribe this event to North American forest fires rather than the Tunguska event. This conclusion is supported by a vanillic acid peak, a source-specific tracer for conifers (Simoneit, 2002), in the D4 (central Greenland) ice core during this same time period (McConnell et al., 2007) and by a synchronous, extremely limited growth of regional trees (drought proxy) in northwestern Ontario (St. George et al., 2009). Levoglucosan and ammonium spikes in the NEEM (northwestern Greenland) ice core do not record this strong fire event. This evidence suggests that BC, NH_4^+ and NO_3^- peaks in ice cores may not be caused by forest fires during 1908 CE. Ammonium may have been produced by a Haber-like process (Melott et al., 2010) where high pressure and temperature conditions generated by the Tunguska comet entering the atmosphere created reactions in the atmospheric nitrogen and hydrogen from the comet ice, thereby accounting for both the ammonium and nitrate enhancements in some Greenland ice cores (Melott et al., 2010). In addition, 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). All three pyro-markers record a strong fire signature in the NEEM ice core in 1973 AD. This fire event is synchronous (within the age model error) and probably related to an anomalous heat wave and severe droughts in 1972 CE in Russia (Dronin and Bellinger, 2005; Golubev and Dronin, 2004; *The Palm Beach Post*, 9 August 1972; *The New York Times*, 9 August 1972, New York; Battisti and Naylor, 2009).

4.1.3 Decadal to centennial fire activity variability

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 and 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 those that centred around ∼ 1600 CE, have higher concentrations (Fig. 2a). Reasons for the general divergence of BC and levoglucosan profiles may be due to the fact that black carbon has an average atmospheric lifetime of a few days to 1 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 to the levoglucosan record. Levoglucosan yield is variable and strongly depends on combustion temperature, biomass composition and flame conditions (Gao et al., 2003; Oros and Simoneit, 2001a, b; Weimer et al., 2008). Thus, levoglucosan cannot be used to characterize and quantify BC in environmental samples (Kuo et al., 2008). In addition, different temporal resolution for BC and levoglucosan records may result in differences after removing spikes and applying LOWESS. 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.

The NEEM levoglucosan centennial-scale maximum (1500–1700 CE, Fig. 3a), the highest of the past two millennia, coincides with data from the Belukha ice core, Siberia (Eichler et al., 2011), that documents a period of increased fire activity between 1550 and 1700 CE, within which 1600–1680 CE recorded the highest forest fire activity from the last 750 years (Fig. 3d). High fire activity between 1500 and 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).

The high-latitude Northern Hemisphere (HLNH) > 55° N GCD composite curve registers a global long-term decline in fire activity from 0 to 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 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° N GCD (Fig. 3). The HLNH > 55° N GCD and levoglucosan data differ near 1600 CE when the HLNH > 55° N 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 probably under-represented 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.

Sapart et al. (2012) argue that changes in the δ^{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. High δ^{13}C values between 800 and 1200 CE, and to a lesser extent from 100 until 200 CE, corresponds to increased levoglucosan and BC concentrations. The pronounced levoglucosan peak centred on ∼ 1600 CE only partially overlaps with the pyrogenic methane record, where δ^{13}C peaks in the 1400s to 1500s. Despite these differences, the NEEM δ^{13}C of CH_4 isotopic record contains some similar features to the levoglucosan data (Fig. 3f), even though the two proxies have different sources and geographical distributions.

4.2 Source of NEEM fire products

4.2.1 Fuel loads and circulation patterns

Stable oxygen isotope (δ^{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., 2003; Bory et al., 2002; Lupker et al., 2010). The above 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.

Earth's boreal forests cover 9–12 million km^2 (Fig. 1), extending across Scandinavia, Russia and North America, representing approximately 10 % of Earth's 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).

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 and 6 % at 700 hPa) or from Europe and western Asia (only 6 % at 500 hPa). The pressure of 500 hPa corresponds to ∼ 5–6 km a.s.l and is typical of mid-tropospheric circulation, while 700 hPa is closer to the altitude of the Summit camp. 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. McConnell et al. (2007) demonstrate that variations in BC matched changes in the specific coniferous fire marker vanillic acid measured in the Greenland D4 ice core before 1850 CE yet do not correlate over the industrial period of 1850–1950 CE. This lack of correlation may result from the majority of BC arriving to Greenland between 1850 and 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 of 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.

4.2.2 Levoglucosan sources

Back-trajectory and model analyses suggest that North American and Siberian forests are the dominant sources of biomass burning aerosols, for which North America is likely the most important contributor due to the proximity to the NEEM location. 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. However, the back-trajectory analyses do demonstrate that Asia, although it is a lesser source, is a possible aerosol source.

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. Correspondences between regional droughts and increased fire activity are present in multiple northern and central Asian proxies (Table 2). Extreme droughts in northern central Asia (Thompson et al., 2000; Shen et al., 2007) correspond in timing with NEEM levoglucosan spikes (Table 2), therefore supporting that Asia may be an important fire source of burning signatures inferred in Arctic ice fields during strong fire events caused during severe dry years.

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).

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

4.3.1 Temperature and droughts

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 and 1250 CE (Mann et al., 2009), whereas Northern Hemisphere (and especially Russian) temperatures were elevated between 1000 and 1300 CE (Crowley and Lowery, 2000; Hiller et al., 2001) or 1000 and 1100 CE (Moberg et al., 2005). Such temperature anomalies correlate with increased fire activity in palaeofire 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 HLNH $> 55^\circ$ N GCD between 1000 and 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 and 1700 CE (Mann et al., 2009), 1580–1720 CE (Christiansen and Ljungqvist, 2012) and 1350–1850 CE (Wanner et al., 2008). Low fire activity is observed in Eurocore (Summit, Greenland) (Savarino and Legrand, 1998) from 1600 to 850 CE and in the global NH curve (based on GCD syntheses) centred around 1750 CE (Marlon et al., 2008). The levoglucosan profile suggests a correspondence between fire activity observed in the deep NEEM ice core and HLNH terrestrial temperature anomaly (Mann et al., 2008) during the 700–900 CE and 1700–1800 CE cooling, and during the 1000–1300 CE warming. Fire activity inferred from the deep NEEM ice core is consistent with the MWP and LIA climatic conditions, except for the strong difference between temperature and fire activity noted throughout the levoglucosan maximum centred around 1600 CE (Fig. 3e).

Ice core and 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 punctual events (1593–1603 CE and 1617–1622 CE) (Fig. 2, Table 1 and 2). The Siberian Belukha ice core identifies a period of exceptionally high forest-fire activity between 1600 and 1680 CE, following a drought period during 1540–1600 CE, which is coincident with the decadal to centennial highest 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 suggested to have contribuited 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 five 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. A network of tree-ring chronologies and historical documents (Grove, 1998; Cook et al., 2010) report extensive drought conditions in China and Mongolia throughout these periods (Table 2), while the Dasuopu ice core (Thompson et al., 2000) demonstrates that the 1790–1796 CE drought was the most intense arid period of the last millennium. A high-resolution determination of continental dust in the Dye-3 Greenland ice core Sr, Nd and Hf isotopic composition from 1786 to 1793 CE established that the majority of the samples have an Asian origin (Lupker et al., 2010). The highest levoglucosan peaks correspond to epic Asian droughts, as summarized in Table 2 – these droughts being among the most severe in the last millennium.

However, 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

Table 2. 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 coast	historical climatic records	Wang et al. (2001)	abrupt dry/wet climate change
1160–1290	Northern China (reconstruction based on different palaeo-climate archives)	flood/drought index	Yang et al. (2007) and refs. 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 palaeo-climate archives)	flood/drought index	Yang et al. (2007) and refs. therein	high dust fall frequency
1330s	Dasuopu ice core, Himalaya (India and Asia as source regions)	δ^{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**	**mega-event**[2]
1578–1582	Mongolia	tree ring	Davi et al. (2010)	2nd and 1st in order of severity since 1520
1600–1644				
1590s-1600s	Central High Asia	tree ring	Fang et al. (2010)	
1590s	Dasuopu ice core, Himalaya (India and Asia as source regions)	δ^{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 five centuries
1618–1635	Taihu drainage basin China, eastern coast	historical climatic records	Wang et al. (2001)	abrupt change to dry period
1702–1706	**NEEM ice core**	**levoglucosan**	**this work**	**mega-event**[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**	**mega-event**[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)	δ^{18}O, dust, Cl^-	Thompson et al. (2000)	monsoon failures "exceptionally large from a perspective of the last 1000 yr"
1789–1793	India	archival evidence of drought	Grove (1998)	
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 palaeo-climate archives)	flood/drought index	Yang et al. (2007) and refs. 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 years
1630s-1640s	China and Mongolia	tree ring	Li et al. (2009)	
1640s	Dasuopu ice core, Himalaya (India and Asia as source regions)	δ^{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, calculated 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.

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). Tree rings from Canada record low growth in 1595 CE and 1790s (St. George et al., 2009). A strong fire event is recorded in 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 and BC also peak ∼ 1789. 1790s was the driest decade in southwestern Canada (Sauchyn and Skinner, 2001; Sauchyn and Beaudoin, 1998; Wolfe et al., 2001) and in southern Alberta (Case and Macdonald, 1995), with a "prolonged absence of wet years around 1700" on the eastern side of the Canadian prairies (St. George et al., 2009).

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 kilogram of burnt fuel) and combustion characteristics (smoldering versus 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). 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.

4.3.2 Human impact on boreal fire activity

Climate (droughts and temperatures) 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). Marlon et al. (2008) argue 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. In addition, Marlon et al. (2008) and Prentice (2010) assert that the expansion of agriculture and fire suppression since the early twentieth century resulted in decreased global fire activity. Pyrogenic methane isotopic data (Ferretti et al., 2005; Mischler et al., 2009) show relatively high values in the first millennium CE, but do not support the prominent maximum of an anthropogenic peak at ∼ 1900 CE in the HLNH > 55° N GCD curve (Marlon et al., 2008) and inferred from CO isotopic measurements (Wang et al., 2010). Emissions modelling 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 data set (Supplement Fig. S5). 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, as inferred by Marlon et al. (2008) and Wang et al. (2010). In addition, levoglucosan data do not show the evident anthropogenic peak started from 1750 CE as inferred from the GCD synthesis, as the NEEM levoglucosan concentrations are low until the beginning of the 20th century, followed by a modest concentration increase.

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. Population growth and major anthropogenic land clearing in boreal 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, while Canadian ice cores also show peak biomass burning from 1920 to 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 have decreased global fire activity since the early 20th 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 Conclusions

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 palaeoclimate studies suggest North American/Canadian fires are the main sources of pyrogenic aerosols transported to the site, in particular during the pre-industrial period. However, Siberian forests may be an essential aspect of boreal fire reconstructions that have not yet been appropriately evaluated.

Temperature is an important controlling factor of boreal fire activity on centennial timescales. On multi-decadal or shorter timescales, however, boreal fires may be mainly influenced by droughts. Our results demonstrate the greatest amount of decadal-scale fire activity during the mid-1600s. We suggest that the 1500–1700 CE maximum in fire activity is due to increased boreal forest fires, caused by extensive dry conditions in the Asian region. NEEM levoglucosan concentrations suggest the major drought centred on 1640 CE may have been more extensive than inferred from local palaeoclimate reconstructions, and that this event dominates boreal fire activity over the past 2000 years. Fires are concurrent with known extensive droughts and monsoon failures, and levoglucosan concentrations during these droughts are greater 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. However, climate change and anthropogenic activity may increase future boreal fire activity, and have the potential to exceed the fire activity of the mid-1600s.

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