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Record of North American boreal forest fires in northwest Greenland snow



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HIGHLIGHTS

- North American boreal fires in 2004 and 2005 are imprinted in Greenland snow.
- Peaks of Levoglucosan in Greenland snow reflect large boreal fires.
- Levoglucosan coincides with ammonium and oxalate in Greenland snow.
- Trajectory analysis and satellitebased CO support a North America source.

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G R A P H I C A L A B S T R A C T



ABSTRACT

We present boreal forest fire proxies in a northwest Greenland snowpit spanning a period of six years, from spring 2003 to summer 2009. Levoglucosan ($C_6H_{10}O_5$) is a specific organic molecular marker of biomass burning caused by boreal forest fires. In this study, levoglucosan was determined via liquid chromatography/negative ion electrospray ionization-tandem mass spectrometry, wherein isotopedilution and multiple reaction monitoring methods are employed. Ammonium (NH_4^+) and oxalate $(C_2O_4^{2-})$, traditional biomass burning proxies, were determined using two-channel ion chromatography. In the northwest Greenland snowpit, peaks in levoglucosan, ammonium, and oxalate were observed in snow layers corresponding to the summer-fall seasons of 2004 and 2005. Considered together, these spikes are a marker for large boreal forest fires. The levoglucosan deposited in the Greenland snow was strongly dependent on long-range atmospheric transportation. A 10-day backward air mass trajectory analysis supports that the major contributors were air masses from North America. In addition, satellitederived carbon monoxide (CO) and ammonia (NH₃) concentrations suggest that chemicals from North American boreal forest fires during the summer-fall of 2004 and 2005 were transported to Greenland. However, large boreal fires in Siberia in 2003 and 2008 were not recorded in the snowpit. The subannual resolution measurements of levoglucosan and ammonium can distinguish between the contributions of past boreal forest fires and soil emissions from anthropogenic activity to Greenland snow and ice.

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1. Introduction

Given the increasingly hot and dry climate, boreal forest fires in the Northern Hemisphere are becoming more frequent, larger, and intense. Climate change and boreal forest fires synergistically enhance widespread melt events of the Greenland ice sheet (Keegan et al., 2014). These fires can significantly impact the Earth system (atmosphere, biosphere, and climate) by altering atmospheric chemistry, permafrost structure, surface albedo, and carbon cycles (Bowman et al., 2009). An increase in boreal forest fires would likely result in positive feedback on global warming and could accelerate climate change (Oris et al., 2013).

Boreal forest fires release various chemicals into the atmosphere through smoke (Bhattarai et al., 2019). These emitted chemicals and particles are transported over long distances through the atmosphere and deposited on snow and ice in the polar regions, and can thus serve as biomass burning proxies in glacier snow and ice (You and Xu, 2018). Ammonium and potassium are commonly used as traditional biomass burning proxies in Greenland snow and ice cores (Legrand et al., 1992; Whitlow et al., 1994; Dibb et al., 1996; Savarino and Legrand, 1998). Additionally, light carboxylic acids, such as formate, acetate, glycolate, and oxalate, can be used to identify past boreal forest fires recorded in Greenland ice (Legrand and De Angelis, 1996). Black carbon released from incomplete combustion of biomass and fossil fuels has been used as a tracer for preindustrial biomass burning in Greenland ice cores (McConnell et al., 2007). Moreover, high concentrations of ammonium concurrent with black carbon is a useful marker for large boreal forest fires (Legrand et al., 1992). Recently, several organic compounds, such as vanillic acid in Arctic ice, have been used as proxies for past events of biomass burning (Grieman et al., 2017; Pokhrel et al., 2020). However, these compounds could also result from vegetation and soil processes, industrial processes, fossil fuel burning, and other anthropogenic sources (Rubino et al., 2016).

In contrast, levoglucosan (1,6-anhydro- β -D-glucopyranose; C₆H₁₀O₅) is a specific organic marker of biomass burning caused by boreal forest fires and it can only be generated from the thermal breakdown of cellulose when the burning temperature exceeds 300 °C (Simoneit et al., 1999). It can be transported away from the source region through long-range atmospheric circulation and, when formed at high temperatures in smoke plumes, can uplift to the upper troposphere or the lower stratosphere under strong thermal convection dynamics (Luderer et al., 2006). Levoglucosan remains stable for more than 10 days under most atmospheric conditions (Hennigan et al., 2010). Moreover, it can be easily scavenged by atmospheric precipitation and conserved for long periods of time with only negligible degradation in snow and ice. As a result, levoglucosan in snow and ice cores serves as a unique organic marker for past wildfire activities.

Recently, snow and ice records of boreal forest fires in the Northern Hemisphere have received significant attention (Legrand et al., 2016; Rubino et al., 2016; You and Xu, 2018). High-resolution measurements of fire proxies would improve the understanding of the causes of past changes in fire activity (Legrand et al., 2016). Biomass burning events in the Northern Hemisphere can be imprinted in Greenland ice and are characterized by co-occurring peaks in fire-specific organic aerosol compounds (Fischer et al., 2015). The imprint of these events is strongly influenced by atmospheric circulation at the time of the boreal forest fires. Greenhouse gases, chemical species, and atmospheric aerosols from extensive fires in North America and Siberia spread to Greenland via long-range atmospheric transport. However, not all biomass burning events in the Northern Hemisphere are recorded in Greenland snow and ice. Moreover, despite past studies on the snow records of boreal forest fires, studies of levoglucosan are still sparse and its significance remains poorly understood.

Therefore, in this paper, we present sub-annual variations of boreal forest fire proxy data acquired in a snowpit from northwest Greenland. The study focuses on the measurements of levoglucosan, oxalate, ammonium, and potassium in samples from the snowpit covering the period 2003 to 2009. In our previous studies, seasonal variations in snow chemistry and mineral dust were investigated in the same snowpit for the purpose of source region tracking (Kang et al., 2015, 2017). The present study confirms the source of the biomass burning influence on the Greenland ice sheet and improves the understanding of the causes of past fire activity recorded in Greenland snow and ice.

2. Materials and methods

2.1. Field sampling and sample handling

A continuous series of 70 snow samples was collected from a 3.2-m deep snowpit in northwest Greenland on June 26, 2009 (Kang et al., 2015). The site was located ~1.0 km southeast of the North Greenland Eemian Ice Drilling (NEEM) deep ice coring project site (77°26'N, 51°03'W, 2461 m a.s.l.). The snow samples were placed in pre-cleaned 1-L low-density polyethylene (LDPE) bottles. All samples were transported from the NEEM camp to the laboratory of the Korea Polar Research Institute (KOPRI) and kept frozen at -20 °C until analysis.

2.2. Sample preparation and levoglucosan analysis

The sample preparation and analytical methods for the direct determination of levoglucosan in polar snow followed the methods evaluated by Gambaro et al. (2008) with minor modifications (Gambaro et al., 2008). In brief, samples were melted at room temperature in a class 10 laminar airflow clean bench and then aliquoted into 1.5 mL screw neck glass vials (Macherey-Nagel GmbH & Co. KG, Germany) using pipettes with polyethylene tips (Eppendorf, Germany). Each individual sample consisted of 900 μ L of melted snow and 100 μ L of an internal standard of ${}^{13}C_6$ -levoglucosan. Response factors were determined using several levels of $^{2}C_{6}$ -levoglucosan and $^{13}C_{6}$ -levoglucosan calibration standards. Concentrations of calibration and internal standards are presented in Table S1. All calibration and internal standards were made by dilution with 100 μ g mL⁻¹ of ¹²C₆-levoglucosan (ULM-8000-1.2, Cambridge Isotope Laboratories Inc., USA) and 100 μ g mL⁻¹ of ¹³C₆levoglucosan (CLM-4748-1.2, Cambridge Isotope Laboratories Inc., USA).

Levoglucosan was analyzed at the Analytical Service Center of Diatech Korea Co., Ltd. A sample analysis was performed using liquid chromatography/negative electrospray ionization-tandem mass spectrometry (LC/(-)ESI-MS/MS). A Shimadzu Prominence UPLC system (Shimadzu Corporation, Japan) with a C18 Synergy Hydro-RP 80 Å column (2.0 mm i.d. \times 50 mm length, 4 μm particle size; Phenomenex, USA) and QTRAP 5500 triple quadrupole mass spectrometer (AB SCIEX, USA) was used to determine levoglucosan in the samples. For each analysis, 100 µL of the sample were injected into a C18 Synergy Hydro-RP column. The oven temperature was 40 °C. Isocratic elution was employed at 0.18 mL using a 15% methanol solution in water (v/v). Methanol (JT Baker, USA) and water (Merck Millipore, USA) were purchased at HPLC grade. Then, 13 mM of ammonium hydroxide (Fluka, USA) solution was added on-line after the chromatographic column using a syringe pump with a flow of 5 μ L min⁻¹. The retention time of ¹²C₆-levoglucosan was 1.56 min and the run lasted 4 min. Data were collected in negative ion mode by multiple reaction monitoring (MRM) with a

100-ms dwell time/transition. For quantification, the transitions of 161/101 m/z for ¹²C₆-levoglucosan and 167/105 m/z for ¹³C₆-labeled levoglucosan were used. A summary of the monitored transitions and the compound parameters are presented in Table S2. Data were collected and analyzed using the Analyst 1.5.2 Software (AB SCIEX, USA) and quantification and peak assignment was conducted using the MultiOuant 2.1 software (AB SCIEX, USA). The isotope dilution method was used for levoglucosan determination. Samples were spiked with 100 μ L of the ¹³C₆-labeled internal standard (500 pg mL⁻¹). The detection limits of levoglucosan, defined as three times the standard deviation of the procedure blanks, were 3 pg mL $^{-1}$. Repeatability was calculated as the relative standard deviation from five consecutive measurements and ranged from 20% for a levoglucosan concentration of 100 pg mL⁻¹ to 50% for a concentration of 20 pg mL⁻¹. Recovery in replicate analysis of samples spiked with 50 pg mL⁻¹ of a levoglucosan was 95% with a coefficient of variation (CV%) of less than 2%.

2.3. Ion analysis

Ammonium (NH₄⁺), potassium (K⁺), and oxalate ($C_2O_4^{2-}$) were analyzed using a two-channel ion chromatography (IC) system at KOPRI (Kang et al., 2015). The IC system simultaneously determines both anions and cations using a Dionex model ICS-2000 (Thermo Fisher Scientific Inc., USA) with an IonPac AS 15 column and KOH eluent (6–55 mM), and a Dionex model ICS-2100 (Thermo Fisher Scientific Inc., USA) with an IonPac CS 12A column and MSA eluent (20 mM), respectively. Detection limits, calculated as three times the standard deviation of blank measurements, were 0.14, 0.16, and 0.05 ng mL⁻¹ for ammonium, potassium, and oxalate, respectively.

2.4. Satellite-based ammonia and carbon monoxide concentrations and backward air trajectories

Mid-tropospheric ammonia and carbon monoxide data at 500 hPa from NASA's Atmospheric InfraRed Sounder (AIRS) on the Earth Observing System (EOS)/Aqua satellite were used to investigate the trajectories of fire-driven smoke (Warner et al., 2013, 2016). To assess the origin of the air masses arriving at the sampling site, backward air trajectories were computed using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) 4 model (https://ready.arl.noaa.gov). National Centers for Environmental Prediction/National Center for Atmospheric Research reanalysis model (NCEP/NCAR reanalysis model) data with a resolution of 2.5° were used as meteorological data in HYSPLIT. The 10-day backward air trajectories were calculated every 6 h for an altitude of 500 m above ground level during the period 2003 to 2009.

3. Results and discussion

3.1. Levoglucosan concentrations

The depth profiles of levoglucosan concentrations are presented in Fig. 1 and Fig. S1. Levoglucosan concentrations in the snowpit ranged from 75 to 3956 pg mL⁻¹ with a mean concentration of 315 pg mL⁻¹. The NEEM snowpit samples had been previously dated by combining the depth profiles of multiple parameters such as stable water isotopes (δ^{18} O and δ D) and ionic species (Na⁺, Ca²⁺, Cl⁻, SO₄²⁻, and methanesulfonic acid (MSA)) (Kang et al., 2015, 2017). Based on strong seasonal patterns of these parameters, the NEEM snowpit was shown to cover a 6-year period from spring 2003 to summer 2009 (Fig. 1). We found two significant levoglucosan peaks conserved in the 187–192 cm and 242–251 cm depth layers, corresponding to summer–fall 2004 and 2005, respectively (Fig. 1). The mean levoglucosan concentration of three continuous



Fig. 1. Vertical profiles of levoglucosan, ammonium, oxalate, and non-sea salt potassium. Red vertical lines represent winter layers and blue dashed lines represent summer layers. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

snow samples for the peak in 2004 was 1929 pg mL⁻¹, while that for the peak in 2005 was 874 pg mL⁻¹. The levoglucosan spikes were more than two times the mean concentrations. Except for these high levoglucosan spikes, we found levoglucosan peaks in spring-summer 2003, winter 2007, and spring 2008 layers, which concentrations were more than 300 pg mL⁻¹ with two continuous samples (Fig. S1). There was no gradually increasing or decreasing trend between 2003 and 2009, despite the short time period.

This study reports one of the highest levoglucosan concentrations in Greenland snow and ice compared with previous studies (Table 1). Measurements of levoglucosan concentrations in Greenland snow and ice are very limited (Kehrwald et al., 2012; Zennaro et al., 2014, 2015). The first study of levoglucosan concentrations was conducted on snowpit samples from the Summit, Greenland; concentrations ranged from 11 to 597 pg mL⁻¹ for the 2-year period between 1994 and 1995 (Kehrwald et al., 2012). Zennaro et al. (2014) reported levoglucosan concentrations in the NEEM ice core (4.95-602.25 m depth) that exhibit high variance with abrupt changes, ranging from 9 to 1767 pg mL $^{-1}$; the ice core covers the period from 1999 CE to 1036 BCE and contains levoglucosan with a mean concentration of 92 pg mL^{-1} (Zennaro et al., 2014). Zennaro et al. (2015) extended the levoglucosan record in the NEEM ice core back to 15 kyr BP (1493.25 m depth) (Zennaro et al., 2015).

Table 1

Levoglucosan concentrations (pg	mL^{-1}) in snow and ice from the	ne Northern Hemisphere.
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Site	Region	Sampling	Period	Altitude (m a.s.l.)	Method	Mean Concentration (pg mL^{-1})	Range (pg m L^{-1})	Ref.
NEEN	Greenland	Snowpit	2003-2009	2461	LC-MS/MS	315	75-3956	This study
Summit	Greenland	Snowpit	1994-1995	3270	LC-MS/MS	NA	11-597	Kehrwald et al. (2012)
NEEM	Greenland	Ice core	1036-1999	2461	LC-MS/MS	92	9-1767	Zennaro et al. (2014)
Ushkovsky	Kamchatka	Ice core	1693-1997	3903	GC-MS	750	BLD-18,620	Kawamura et al. (2012)
Aurora Peak	Alaska	Ice core	1665-2008	2825	GC-MS	543	BLD-20,800	Pokhrel et al. (2020)
Yulong (YL)	S TP	Snowpit	2013-2014	4720	LC-MS/MS	2410 ± 1470	NA	Li et al. (2018)
Gurenhekou (GRHK)	S TP	Snowpit	2013-2014	4520	LC-MS/MS	11,720 ± 15,610	NA	Li et al. (2018)
Donkemadi (DKMD)	C TP	Snowpit	2013-2014	5721	LC-MS/MS	290 ± 130	NA	Li et al. (2018)
Yuzhufeng (YZF)	C TP	Snowpit	2013-2014	5477	LC-MS/MS	250 ± 80	NA	Li et al. (2018)
Meikuang (MK)	C TP	Snowpit	2013-2014	5080	LC-MS/MS	330 ± 190	NA	Li et al. (2018)
Muztagh (MZTG)	N TP	Snowpit	2013-2014	5780	LC-MS/MS	750 ± 430	NA	Li et al. (2018)
Qiyi (QY)	N TP	Snowpit	2013-2014	4767	LC-MS/MS	500 ± 60	NA	Li et al. (2018)
Laohugou LHG	N TP	Snowpit	2013-2014	4780	LC-MS/MS	440 ± 320	NA	Li et al. (2018)
Tienshan (TS)	N TP	Snowpit	2013-2014	4130	LC-MS/MS	460 ± 70	NA	Li et al. (2018)
Yala (YL)	Himalaya	Surface snow	2012	5400	LC-MS/MS	26,660 ± 4770	21,940-32,900	You et al. (2016)
Dasuopu (DSP)	Himalaya	Surface snow	2012	5800-6700	LC-MS/MS	310 ± 480	BLD-1560	You et al. (2016)
Zuoqiupu (ZQP)	SE TP	Snowpit	NA	5150-5600	LC-MS/MS	1410 ± 1690	BLD-6070	You et al. (2016)
Demula (DML)	SE TP	Surface snow	2012	5400	LC-MS/MS	280 ± 470	BLD-1130	You et al. (2016)
Cuopugou (CPG)	E TP	Ice core	NA	5460	LC-MS/MS	910 ± 920	220-3950	You et al. (2016)
Qiyi (QY)	N TP	Surface snow	2012	5100	LC-MS/MS	2560 ± 530	2060-3120	You et al. (2016)
Muji (MJ)	NW TP	Snowpit	NA	5550	LC-MS/MS	1130 ± 890	1180-2120	You et al. (2016)
Kuokuosele (KKSL)	NW TP	Surface snow	2012	4850	LC-MS/MS	1970 ± 140	BLD-2780	You et al. (2016)
Muztagh Ata	W TP	Ice core	1900s-1990s	6350	LC-MS	3300	10,000-718,000	Yao et al. (2013)
Tanggula	C TP	Ice core	1990-2000	5645	LC-MS	3900	10,000-93,000	Yao et al. (2013)

*BLD: below detection limit; NA: Not available.

Snow and ice levoglucosan concentrations from the Northern Hemisphere are summarized in Table 1. As levoglucosan is ubiquitous in the atmosphere of the Northern Hemisphere, it can be deposited on the snow surface and detected in snow and ice (Simoneit et al., 1999; Simoneit, 2002; You and Xu, 2018). Levoglucosan concentrations from Greenland are much lower than those of other sites in the Northern Hemisphere due to the proximity of biomass burning sources. Kawamura et al. (2012) measured levoglucosan concentrations in the Ushkovsky ice core from the Kamchatka Peninsula; concentrations ranged from below the detection limit (BDL) to 18,620 pg g^{-1} with an average of 750 pg g ⁻¹. Pokhrel et al. (2020) recently reported levoglucosan concentrations in the Aurora Peak ice core from Alaska, ranging from BDL to 20,800 pg g⁻¹ for 1665–2008 CE with a mean concentration of 543 pg g^{-1} (Pokhrel et al., 2020). The Aurora Peak ice core conserved higher levoglucosan concentration than those of the Ushkovsky ice core from the Northern Hemisphere (Table 1), because Aurora Peak lies adjacent to the most likely source regions of biomass burning, such as Alaska. The highest levoglucosan concentrations in the Aurora Peak ice core were found for the 1660s-1830s.

Several studies measuring the levoglucosan concentrations in snow and ice have been conducted in the Tibetan Plateau (TP) (Yao et al., 2013; You et al., 2016; Li et al., 2018). Levoglucosan concentrations in the TP vary widely for different sampling sites (Table 1). For example, levoglucosan concentrations from the central TP showed similar concentration levels to Greenland snow and ice (Li et al., 2018). However, the highest levoglucosan concentration of the TP reached 718,000 pg mL^{-1} in the western TP ice core (Yao et al., 2013). The southern TP showed higher levoglucosan concentrations compared to the northern TP (You et al., 2016; Li et al., 2018). Interestingly, the levoglucosan concentrations in the Yala glacier were 86 times higher than those of the Dasuopu glacier, even though these two sites are located in very close proximity on Mt. Himalaya at over 5000 m a.s.l. (You et al., 2016). Intense biomass burning events are widespread in Asia and impact on the TP glaciers and it is still poorly understood in the TP (You et al., 2016). Thus, to understand the local sources of levoglucosan concentrations and long-range transportation of biomass burning over the TP, further studies are needed.

3.2. Ammonium, oxalate, and potassium concentrations

Depth profiles of ammonium (NH₄⁺), oxalate ($C_2O_4^{2-}$), and nonsea-salt potassium (nssK⁺) concentrations are shown in Fig. 1. Ammonium and potassium have been used as traditional biomass burning proxies in Greenland snow and ice (Dibb and Jaffrezo, 1997; Jaffrezo et al., 1998). To improve confidence in biomass burning event reconstruction, we combined these traditional biomass burning proxies with levoglucosan, a biomass burning specific proxy. The two high levoglucosan peaks co-existed with ammonium and oxalate peaks, but some differences were observed in those of potassium. We observed significant correlations between levoglucosan and ammonium (Pearson coefficient, r = 0.732, p < 0.01), and between levoglucosan and oxalate (Pearson coefficient, r = 0.464, p < 0.01); however insignificant correlations between levoglucosan and nssK⁺ (Pearson coefficient, r = 0.014).

High ammonium concentrations coinciding with high levoglucosan concentrations indicate the influence of biomass burning emissions. Ammonium concentrations ranged from 0.2 to 62.8 ng mL⁻¹ with a mean concentration of 10.5 ng mL⁻¹ in the northwest Greenland snowpit. It was reported that ammonium concentrations of more than 20 ng mL^{-1} reflect boreal forest fire inputs (Legrand and De Angelis, 1996). We detected coincident ammonium and levoglucosan peaks with more than two continuous points for layers corresponding to summer-fall 2004 and 2005, respectively. In addition, owing to the sub-annual resolution measurements of our study, we observed other excess ammonium peaks during the spring season both in 2004 and 2005. Ammonium concentrations in Greenland snow and ice have been linked to both soil and biomass burning emissions at sub-annual resolution (Fischer et al., 2015). The increase in ammonium concentration during the spring could reflect soil emissions caused by biological activities of ammonia (NH₃). In addition, some ammonia is converted to ammonium through the phenomenon of Arctic haze, as spring-time ammonia emissions rise owing to anthropogenic activities (i.e., farming). We observed ammonium peaks for every spring season, which can be related to soil emissions and anthropogenic activities. Overall, the concurrent peaks of ammonium and levoglucosan strongly suggest a boreal forest fire source (Gfeller et al., 2014).

Oxalate concentrations of the northwest Greenland snowpit ranged from 0.1 to 4.6 ng mL⁻¹ with an average of 1.5 ng mL⁻¹. The oxalate concentrations are similar to those from Summit, Greenland (Kehrwald et al., 2012). Along with concurrent peaks in levoglucosan, the elevated oxalate concentrations at Summit were interpreted to reflect biomass burning (Kehrwald et al., 2012). The presence of oxalate in the remote atmosphere is an evidence for biomass burning because oxalate can be originated from biomass burning sources (Legrand and De Angelis, 1996; Gillett et al., 2007). At the summit of the Greenland Ice sheet, air masses with high oxalate concentrations also contain increased atmospheric ammonium and potassium concentrations, suggesting the influence of biomass burning emissions (Jaffrezo et al., 1998).

We found no potassium peaks related with biomass burning proxies in the northwest Greenland snowpit. As potassium arise mainly from sea spray, we calculate non-sea-salt potassium (nssK⁺) by the equation: $nssK^+ = K^+ - 0.036 \times Na^+$, where K⁺ and Na⁺ are the total concentration of potassium and sodium and 0.036 is the K⁺/Na⁺ ratio in sea water (Bowen, 1979). In 2004 and 2005, the nssK⁺ peaks were not occurred in a snow layer coincident with levoglucosan peaks (Fig. 1). Several enhancements of nssK⁺ were related to ammonium events in summer 2004, spring 2006, and spring 2007. Moreover, the variations of nssK⁺ concentration are limited in the snowpit, it remains difficult to use as a boreal forest fires proxy (Legrand et al., 2016).

3.3. Northern Hemisphere boreal forest fires between 2003 and 2009

Boreal forests in North America and Siberia are important biomass burning sources owing to their relative proximity to Greenland (Zennaro et al., 2014). In the Northern Hemisphere, boreal forests play a crucial role in climate change because they contain over 30% of the global terrestrial carbon, even though the boreal zone covers less than 17% of Earth's land surface (Kasischke, 2000). Fires from boreal forests release substantial amounts of carbon, with 60 TgC year⁻¹ released in North America, and 124 TgC year⁻¹ released in Siberia (van der Werf et al., 2017).

The annual burned area is one of the most important parameters in the evaluation of biomass burning emissions. Table 2 shows estimates of annual burned areas in North America and Siberia between 2003 and 2009 (Alaska Interagency Coordination Center, 2010; Vivchar, 2011; Kukavskaya et al., 2012). These areas vary annually, resulting in large temporal and spatial variabilities in boreal forest fire activity. We examined the relationship between the sum of the annual levoglucosan concentrations and the estimated annual burned areas. The results show highly significant correlations between annual levoglucosan concentrations and annual burned areas in North America as a whole, and in Alaska and the Yukon Territory specifically (Pearson coefficient, r = 0.958, 0.881, and 0.924, respectively; p < 0.01); however, we observed insignificant correlations with Siberia.

North America experienced particularly high fire activity in 2004 and 2005. According to the U.S. National Interagency Fire Center (http://nifc.gov) and the Canadian Interagency Forest Fire Centre (www.ciff.ca), the 2004 Taylor Complex fire was a record-breaking wildfire, exceeding the previous record fire in 1957. The fire was concentrated on the Alaska/Canada border. In 2004, the total North American burned area was 4.9 Mha; Alaska accounted for ~2.7 Mha, the Yukon Territory accounted for 1.4 Mha, and north-central Canada accounted for 0.9 Mha (Turquety et al., 2007). The Taylor Complex fire began in late June and ended in September. In 2005, over 3.3 Mha burned across Alaska and the Yukon Territory. Furthermore, we found numerous large fires across central Alaska and the Yukon Territory in August 2004 and August 2005 on Moderate Resolution Imaging Spectroradiometer (MODIS) images from NASA's Terra satellite (Fig. S2).

In contrast, Siberia experienced low boreal forest fire activity in 2004 and 2005, but high wildfire activity in 2003 and 2008. The estimated annual burned area of Siberia was 4.1 Mha in 2003 and 2.6 Mha in 2008, but much lower in 2004 and 2005 with 0.3 Mha and 0.4 Mha, respectively (Vivchar, 2011). According to satellite data, the lowest fire activity was in 2004, while the boreal fire of 2003 was characterized by a large burned area detected by all fire products (Kukavskaya et al., 2012). In 2003, an absence of winter precipitation and dry weather conditions in the early spring and summer caused unusually large fires burning in the southeast regions of Siberia resulting in the high fire activity. Generally, the Siberian fire season begins earlier than that in North America, starting in late March and extending into October. From 1996 to 2000, the greatest fire activity in Siberia occurred in the spring season between April and May. However, we observed no significant levoglucosan peaks in the snow layers corresponding to the spring 2003 and spring 2008 seasons. Even though we detected an ammonium peak of more than 20 ng mL⁻¹ in snow layers corresponding to spring 2008, levoglucosan and oxalate peaks were not detected in the same layers. The spring ammonium peak can be attributed to soil emissions of anthropogenic activity during the spring season.

3.4. Source regions of boreal forest fires

The imprint of boreal forest fires on the Greenland ice sheet can be enhanced by forest fire activity, transport efficiency, and atmospheric deposition. At the NEEM, the mean annual snow accumulation rate is 22.5 g cm⁻² yr⁻¹; further, summer precipitation at the NEEM is more pronounced than at other locations on the Greenland ice sheet (Steen-Larsen et al., 2011). The record of boreal forest fires is strongly dependent on atmospheric circulation at the time of the fire, which must be favorable for long-range atmospheric transport from the source to the Greenland ice sheet (Fischer et al., 2015). In

Table 2	2
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Year	North America	Alaska	Yukon	Other Canada	Siberia	Sum of annual levoglucosan
2003	1.2	0.2	0.01	0.9	4.1	1742
2004	4.9	2.7	1.4	0.9	0.4	8352
2005	3.3	1.8	0.2	1.4	0.3	4687
2006	1.8	0.1	0.1	1.6	1.2	2285
2007	1.7	0.2	0.04	1.5	0.7	1505
2008	1.3	0.03	0.02	1.2	2.6	2477
2009	1.7	1.2	0.1	0.4	0.9	1037

particular, boreal forest fires in North America are more commonly transported towards Greenland owing to the tree species (Rogers et al., 2015). North American boreal forests support more high-intensity crown fires than Siberia, where lower-intensity surface fires are common (Rogers et al., 2015). Boreal crown fires consume 20–30 tones ha⁻¹ of fuel and produce towering convection columns that can reach the upper troposphere and lower stratosphere (Kasischke et al., 1999; Ansmann et al., 2018), which increases the likelihood of long-range atmospheric transport of smoke from North America to Greenland (Zennaro et al., 2014).

To identify the source of the boreal forest fires, 10-day air mass trajectories were run every 6 h in the backward mode for an altitude of 500 m above the ground level. Seasonal trajectories for 2004 and 2005 are presented in Fig. 2. Number of endpoints of backward air trajectories for 2004 and 2005 are presented in Fig. S3 and seasonal back trajectories in each of major transport patterns are presented in Fig S3. For the summers (June, July, and August) of 2004 and 2005, backward air mass trajectories show that the major contributors to Greenland's aerosols are air masses passing over North America and the Atlantic Ocean. Westerly transport patterns in North America are dominant. The trajectories are short and the atmospheric circulation weak (Kahl et al., 1997; Coumou et al., 2015). Minor contributors come from Siberia and East Asia. Specifically, the summer seasonal trajectories are consistent with the 25-year averaged values (Legrand et al., 2016). On average, backward air mass trajectories arriving at NEEM spend more time over eastern North America than over western North America. However, several air masses traveled over western North America (Alaska and Yukon) before reaching NEEM.

Air masses not from North America mainly traveled over the Atlantic Ocean and only rarely originated from Eurasia (Kahl et al., 1997). In fall (September, October, and November) of 2004 and 2005, the majority of air masses traveled over the major burned areas of western North America. Even though there was no significant change in air mass origin during the summers, longer air mass trajectories occurred when large fires occurred in western North America. The air masses traveling over western North America provide transport for levoglucosan from Alaska and the Yukon territory. Based on this multiple seasonal analysis of air masses reaching NEEM, we conclude that boreal regions of North

America were the main sources of the 2004 and 2005 fire signals in Greenland snow. In 2003 and 2008, the spring transport pattern was similar to that in summer, but the trajectories came south over the fire region of Siberia.

Backward air mass trajectory analyses are performed to identify the possible source regions of biomass burning events. Previous studies at Summit, Greenland suggested north-central Canada as a source region of a boreal forest fire event in August 1994 (Kehrwald et al., 2012), as confirmed by backward air mass trajectory analyses (Dibb et al., 1996). The 10-day backward trajectories arriving at Summit in June 1993 showed consistent transport from northcentral Canada (Jaffrezo et al., 1998).

Satellite observations of chemicals are useful for identifying mid-tropospheric transport (Fisher et al., 2010; Jacob et al., 2010). Biomass burning emissions include not only levoglucosan and ammonium, but also other short-lived constituents such as carbon monoxide (CO), nitrogen oxides (NO_x), and carbonaceous aerosols. Among them, CO is a good tracer gas of wildfires because its lifetime is a few weeks to two months and the spatial pattern of the CO concentration can be detected by satellites (Monks et al., 2012; Field et al., 2016). Pfister et al. (2005) estimated a total of 30 ± 5 Tg CO emitted from Alaskan wildfires during the summer of 2004 based on atmospheric inverse model analysis (Pfister et al., 2005).

Fig. 3 shows high levels of CO over North America drifting to Greenland. CO from the North American boreal forest fires spread from Alaska across northern North America in the summers of 2004 (Figs. 3) and 2005 (Fig. S5). High CO concentrations were observed on July 4, 2004 in Alaska (Fig. 3a). CO concentrations in Alaska then fell rapidly over two days (Fig. 3b) after the reduction of active fire detection, and northern North America exhibited high CO concentrations (Fig. 3b). Based on the spatial patterns of satellite-derived CO concentrations, the CO then moved further eastward and was transported to Greenland over a few days (Fig. 3c). The spatial patterns of the CO distribution are further evidence supporting that chemicals and particles from North American fires can be transported to Greenland, as shown for 2004.

In addition, satellite-based ammonia (NH₃) concentrations clearly show that Alaska had relatively high ammonia concentrations during the summers of 2004 (Fig. 4) and 2005 (Fig. S6), while Siberia exhibited negligible ammonia concentrations. This result



Fig. 2. Seasonal backward air mass trajectories arriving at the North Greenland Eemian Ice Drilling (NEEM) site (yellow star) for spring (MAM), summer (JJA), fall (SON), and winter (DJF) in (a) 2004 and (b) 2005. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)



Fig. 3. Daily CO concentrations (ppbv) at 500 hPa on (a) July 04, 2004, (b) July 06, 2004, and (c) July 08, 2004. Horizontal winds at 500 hPa (orange vector) are from National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis (Kalnay et al., 1996). Red dots denote active fires from Moderate Resolution Imaging Spectroradiometer (MODIS) observations. Stars show the location of the North Greenland Eemian Ice Drilling (NEEM) deep ice coring site. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

supports that high ammonium and levoglucosan concentrations in the 2004 and 2005 snow layers were driven by intense fire activity in North America rather than Siberia.

4. Conclusions

This paper presents a record of levoglucosan, ammonium, oxalate, and potassium concentrations from a northwest Greenland snowpit corresponding to the period between 2003 and 2009. The data provide proxies for boreal fire activity and confirm intense fire activities transported from North America (and Alaska and the Yukon Territory) in 2004 and 2005. Both 10-day backward air mass trajectory analysis and satellite-based CO concentrations support a North America source, with long-range atmospheric transport carrying smoke to Greenland. Levoglucosan concentrations in the northwest Greenland snowpit are dominated by the North American boreal forest fire sources, while Siberian biomass burning contributes little. Our results support the use of measurements of levoglucosan in snow and ice to elucidate past fire activity. North American boreal forest fires more significantly influence Greenland snow and ice than those of Siberia for the studied period.



Fig. 4. Monthly ammonia concentrations (ppbv) at 500 hPa on (a) June 2004, (b) July 2004, and (c) August 2004. Stars indicate the North Greenland Eemian Ice Drilling (NEEM) deep ice coring study site.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2021.130187.

Credit author statement

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