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Assessment for paleoclimatic utility of biomass burning tracers in SE-Dome ice core, Greenland

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ABSTRACT

We provide continuous records of biomass burning molecular tracers (levoglucosan and dehydroabietic acid) in a Greenland ice core collected from the Southeastern Dome (the SE-Dome ice core) over the past several decades to assess the paleoclimatic utility of these tracers in Greenland ice cores. An air mass backward-trajectory analysis indicates that eastern Canada is likely the primary source region of the biomass burning tracers. Comparisons of levoglucosan and dehydroabietic acid data in the SE-Dome ice core and area burned (vegetation fire) events in Canada suggests that the biomass burning tracers in the ice core document most of the pronounced biomass burning events in eastern Canada over the past several decades, confirming that analyses of biomass burning molecular tracers in Greenland ice cores are useful to reconstruct the frequency of significant biomass burning events in a local region. However, our study also highlights that the wind pattern when the biomass burning occurs is decisive for the registration of a biomass burning event in an ice core even though long-term changes in the wind regime associated with decadal-scale climate oscillations do not significantly influence the transport and deposition of biomass burning tracers on the Greenland ice sheet.

1. Introduction

Biomass burning (BB), which includes wildfires and other types of fires involving plant matter, is an important source of numerous greenhouse gases and aerosols, such as CO_2, CO, CH_4, black carbon (BC), alcohols, organic acids, and persistent organic pollutants (Andreae and Merlet, 2001; Crutzen et al., 1979; Lamarque et al., 2010; Mukai and Ambe, 1986). BB-derived gases and aerosols significantly influence the climate by changing the Earth's energy budget by scattering and absorbing the radiation or acting as cloud condensation nuclei (Aalto et al., 2001; Haywood and Ramaswamy, 1998; Jacobson, 2001). Aerosol particles emitted during burning may cause a short-term cooling of the global climate, while longer-lived greenhouse gases may cause warming after several decades (Jacobson, 2004). Therefore, the effects of BB products on the climate are complicated and their net radiative effects still contain large uncertainties (IPCC, 2013). Therefore, to better understand the substantial impact of BB on the climate, it is important to generate long-term reliable records that document the variability of BB events.

Paleoclimate archives containing annual layers (e.g., ice cores, tree rings, sediment cores, and coral reefs) record decadal-scale climatic oscillations in the past (Hodell et al., 1999; Jones and Mann, 2004). Because ice cores preserve key climate factors that influence the radiative balance in the atmosphere, such as greenhouse gases and atmospheric aerosols (Kawamura et al., 2012; Legrand et al., 2013; Legrand and Mayewski, 1997; Lüthi et al., 2008; Petit et al., 1999; Preunkert and Legrand, 2013; Zennaro et al., 2014), ice cores are often considered to be exceptional compared to other paleoclimate archives.

A number of tracers (e.g., inorganic: potassium and ammonium, formate; organic: levoglucosan, dehydroabietic acid, and vanillic acid; and the isotopic compositions of trace gases) have been proposed to reconstruct changes in the BB activity in the past (Bock et al., 2017; Legrand et al., 2016, 1992; Rubino et al., 2016; Simoneit et al., 1999) and have been applied to ice cores (Iizuka et al., 2018; Kawamura et al., 2012; Zennaro et al., 2014). Of these tracers, organic tracers are becoming increasingly common tools because they are produced solely by

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BB (Simoneit et al., 1999). Levoglucosan (LG) is the pyrolysis product of cellulose and hemicellulose, that are the major components of woody part of plant, at temperatures > 300 °C (Andreae and Merlet, 2001), has been proposed as a general tracer of BB in atmospheric aerosols, ice cores, and sediment cores (Elias et al., 2001; Fu et al., 2012; Hu et al., 2013; Kawamura et al., 2012; Schüpbach et al., 2015; Simoneit, 2002; Simoneit et al., 2000; Zennaro et al., 2015, 2014). Conversely, dehydroabietic acid (DA) has been proposed as a more specific tracer of the burning of conifer trees because DA is produced by the pyrolytic dehydration of abietic acid, which is a principal component of conifer resin (Simoneit, 2002; Simoneit et al., 1993). These BB tracers are injected into the atmosphere in convective smoke plumes and then are scavenged from the air column primarily by wet deposition and incorporated into firn and eventually glacial archives in the Arctic region (Gambaro et al., 2008). In recent years, these BB tracers have been applied to ice cores to reconstruct the variability of such aerosol loadings in the past (Grieman et al., 2017; Kawamura et al., 2012; Taylor et al., 1996; Zennaro et al., 2015, 2014). However, there are several factors that can potentially influence the records of BB molecular tracers in ice cores. Moreover, the emission of those tracers from the source and utility of BB tracers in ice cores has not been extensively evaluated.

To assess the paleoclimatic utility of these BB tracers in ice cores, it is vital to generate continuous records of BB aerosol tracers in ice cores with well-constrained chronologies and then compare the ice core tracer record with observationally based records of BB in the potential source region of the BB aerosol. In this study, we provide continuous records of BB aerosol tracers (LG and DA) in a well-dated Greenland ice core over the past 60 years to evaluate the reliability of the LG and DA data in ice cores as tracers of BB events.

2. Experimental section

2.1. Sample and age model

The ice core (90.45-m depth) was drilled in 2015 from the dome site in Southeast (SE) Greenland (67.18°N 36.37°W, 3170 m a.s.l.) (Fig. 1). The annual mean temperature at the SE-Dome site was −20.9 °C based on a 20-m-deep firn temperature measurement (Iizuka et al., 2016). From 1958, 2014, the SEIS2016 age scale, which is determined via the oxygen isotope matching method, was used. The SEIS2016 age scale was carefully evaluated with independent age markers, and its precision is within two months (Furukawa et al., 2017; Iizuka et al., 2018). The average accumulation rate of the SE-Dome ice core was 1.01 ± 0.22 m/yr, which is the highest rate of any ice core reported in Greenland. In addition, melt layers are relatively infrequent in the SE-Dome ice core. Therefore, the post depositional alteration of the deposited aerosols due to ice melt and low accumulation is thought to be minimal (Iizuka et al., 2018). This ice core is ideal to assess the paleoclimatic utility of aerosol tracers in ice cores by a precise comparison with observationally based records.

2.2. Organic tracer analyses

The SE-Dome ice core samples were cut into 50-cm-long pieces and were stored in a cold room (~20 °C) at the Institute of low Temperature Science, Hokkaido University, until analysis. The top 5 mm of the surfaces of the ice core sample were shaved off with a ceramic knife to remove possible contamination. Each sample was then melted in a pre-cleaned Pyrex beaker and treated with $HgCl_2$ to prevent microbial degradation of the organic compounds. Then, the samples were stored at 4 °C in pre-cleaned brown glass bottles until analysis.

The samples were prepared for analysis using a method described elsewhere (Fu et al., 2008; Kawamura et al., 2012). In brief, the melt water samples were transferred to a pear-shaped flask and concentrated to nearly complete dryness using a rotary evaporator under a vacuum. The total organic matter in the dried samples was extracted with a 2:1 v/v solution of CH_2Cl_2/CH_3OH using an ultrasonic bath. The extracts were passed through a glass column packed with quartz wool and further eluted with CH_2Cl_2 and CH_3OH to extract the organics potentially adsorbed on the particles. The eluents were then combined with the extracts, transferred to 1.5-mL glass vials, and dried under a pure nitrogen gas stream. Polar organic markers in the extracts were derivatized with 99% N,O-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) + 1% trimethylsilyl chloride for 3 h at 70 °C in a sealed glass vial (1.5 mL). The derivatives were then diluted by the addition of n-hexane containing C_{13} n-alkane as an internal standard prior to the determination via gas chromatography-mass spectrometry (GC-MS).

GC-MS analyses were performed on a Hewlett-Packard model 6890 GC coupled to a Hewlett-Packard model 5975 MSD using a capillary column (HP-5MS, 60 m × 0.32 mm I.D. × 0.32 μm film thickness) and a split/splitless injector. The GC oven temperature was programmed from 50 °C (2 min) to 120 °C at 30 °C/min, then to 300 °C at 5 °C/min, and maintained at 305 °C for 15.60 min. Helium was used as a carrier gas. LG and DA were identified by comparing the mass spectra with those of authentic standards (Fu et al., 2016; Kawamura et al., 2012). Recoveries for the standards or surrogates were better than 80%. The analytical errors in the triplicate analyses were within 15%. A laboratory blank was measured using Milli-Q water and showed no contamination for any target compounds.

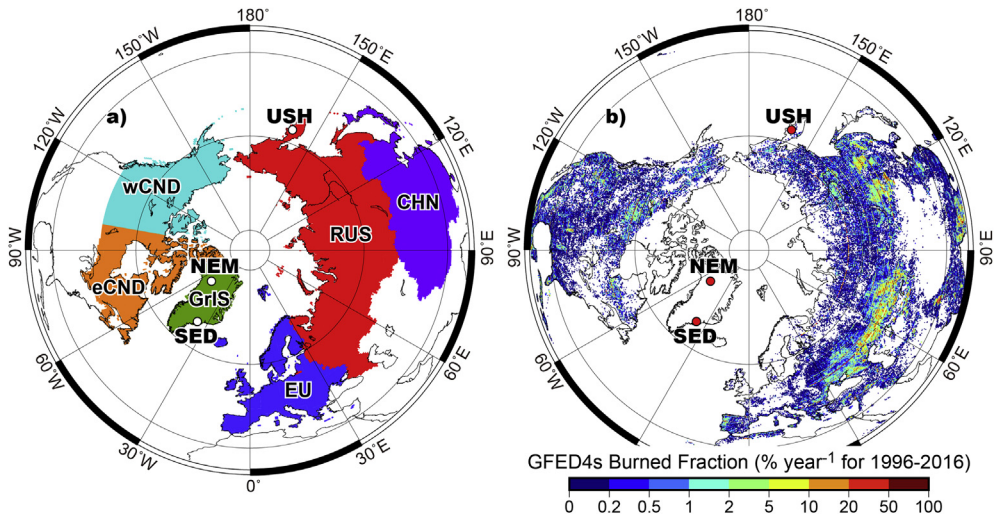

Fig. 1. Map showing (a) the location of the SE-Dome (SED) ice core in Greenland and six regions for calculating regional contribution (GrIS: Greenland, EU: Europe, RUS: Russia, CHN: China + Japan, wCND: western Canada, eCND: eastern Canada), and (b) the percentage of the burned fraction on the globe per year (averaged over 1997–2014) (http://www.globalfiredata.org/index.html), with the locations of the SE-Dome, NEEM (NEM), and Ushkovsky (USH) ice cores.

2.3. Backward-trajectory analysis

To investigate the source regions of the chemical species preserved in the ice core, air mass transport pathways were analyzed using HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) distributed by the National Oceanographic and Atmospheric Administration (Stein et al., 2015). Points at 10 m, 500 m, 1000 m, and 1500 m above ground level (a.g.l.), which correspond to 1560 m, 2050 m, 2550 m, and 3050 m a.s.l. in the model, were set as the starting points of the 10-day backward trajectories. The probability distribution of an air mass below 1500 m a.g.l. was calculated at a 1° resolution. We assumed wet deposition for the preserved aerosols and tracers (Iizuka et al., 2018). It has been reported that wet deposition produces about 90% of the black carbon, sulfate, and dust depositions in polar region (Breider et al., 2014). This suggests that wet deposition is also important process to eliminate water soluble organic aerosol such as LG and DA from the atmosphere. Therefore, the probability was weighted by the daily amount of precipitation when the air mass arrived at the core site. We used the daily precipitation in the reanalysis datasets of ERA-40 and ERA-Interim, both produced by ECMWF (European Centre for Medium-Range Weather Forecasts) (Dee et al., 2011; Uppala et al., 2005). To maintain consistency between the two precipitation products for the entire period (1958–2014), the daily precipitation of ERA-40 (p40) was calibrated with that of ERA-Interim (pi) via a linear regression obtained for the period of 1979–2001 (pi = 1.36p40, R^2 = 0.862, $p < 0.001$). Based on the probability distribution, we also calculated the regional contribution, for which land regions in the Northern Hemisphere was divided into 6 regions based on national boundary (Fig. 1a).

3. Results and discussion

3.1. Source region assignment of the BB tracers via backward-trajectory analyses

First, we attempted to constrain the potential source areas of the LG and DA deposited in the SE-Dome ice core based on the probability distribution of the integrated 7-day backward-trajectory analyses during the period from 1958 to 2014 (Fig. 2). According to the probability analyses, the source regions of these BB tracers are thought to be around southeastern and/or southern Greenland, the North Atlantic Ocean, and North America (more specifically eastern Canada); however, parts of Europe and Russia also have the potential to be important source regions. In addition to these regions, Iizuka et al. (2018) argues for a small possible contribution of inorganic aerosols from remote regions such as East Asia and India, where emissions of anthropogenic SO_x, NO_x, and NH_3 are enormous (Crippa et al., 2016). Given that lifetimes of LG and DA in the atmosphere are estimated to be less than 10 days (Fraser and Lakshmanan, 2000; Hennigan et al., 2010; Lai et al., 2015), organic BB tracers originating from remote areas are unlikely to reach Greenland. Greenland is also unlikely to be a primary source region because most of Greenland is covered with ice sheets and is inhospitable to vegetation. Iizuka et al. (2018) calculated the regional contributions of air mass origins to the SE-Dome and found that the contributions from North America are 2.5 times and 7 times larger than those from Europe and Russia, respectively (See pie diagram of Fig. 2). Therefore, it is thought that North America (Canada) is likely the primary source region of the BB organic tracers. Our estimate based on the backward air mass trajectory is consistent with previous estimates of BB tracer source regions in the Greenland ice sheet (Legrand et al., 1992; Whitlow et al., 1994; Zennaro et al., 2014).

3.2. Concentrations of the BB molecular tracers in SE-Dome ice cores

Historical changes in the concentrations of the two BB tracers (LG and DA) in the SE-Dome ice core are shown in Fig. 3a and b.

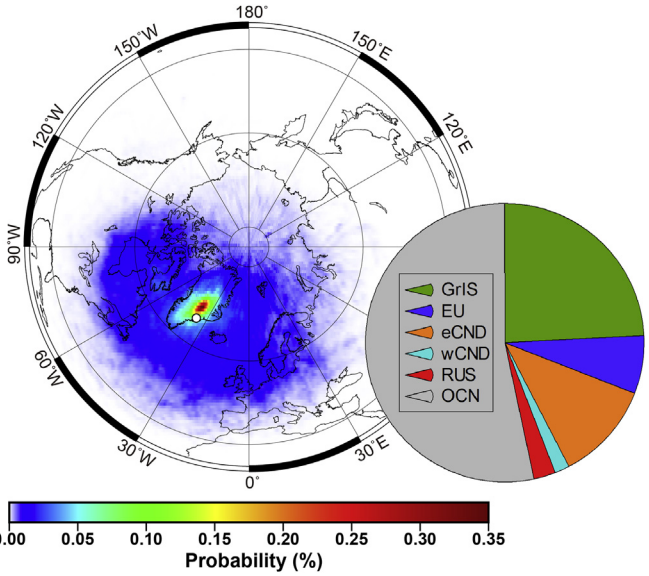

Fig. 2. Probability distribution of an air mass arriving at the SE-Dome site (10–1500 m a.g.l. as initial four points) from a 7-day 3D backward-trajectory analysis for the period of 1958–2014. A color scale indicating the probability percentage is shown at the bottom. Pie diagram shows the relative contribution of each region from where the airmass reached to SE-Dome ice core. Abbreviations of the regions are shown in Fig. 1a except for ocean (OCN). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Concentrations of LG in the SE-Dome ice core range from below the detection limit (0.005 ng/g-ice) to 217 ng/L with an average of 18.27 ng/L, showing no clear increasing or decreasing trends toward the present with the occurrence of sporadic peaks in a handful of years. Conversely, concentrations of DA in the SE-Dome ice core range from below the detection limit (0.003 ng/g-ice) to 45 ng/L with an average value of 5.20 ng/L. Relatively higher concentrations of DA were recognized in the period of 1959–1964. A gradual increasing trend is observed from 2009 to 2014 (Fig. 3b).

To examine regional differences in the concentrations of LG and DA in the ice core, we compared the average values of the BB molecular tracer concentrations in different ice cores. Table 1 represents the average concentrations of LG and DA in the SE-Dome, NEEM (Zennaro et al., 2014), and Ushkovsky (Kawamura et al., 2012) ice cores for the same time interval. The NEEM ice core site is located on the northern part of the Greenland ice sheet, while the Ushkovsky ice core was collected from the Ushkovsky mountain glacier on the Kamchatka peninsula in far eastern Russia (Fig. 1b). The average concentrations of LG and DA in the SE-Dome ice core are lower than those in the Ushkovsky and NEEM ice cores (Kawamura et al., 2012; Zennaro et al., 2014), with the highest concentration found in the Ushkovsky ice core. Such differences are likely attributable to differences in the sources, the snow accumulation rates, and the atmospheric transport of BB tracers to the respective glacier sites (Müller-Tautges et al., 2016). The average LG and DA concentrations at the Ushkovsky ice core site are approximately 5–30 times higher than those at the Greenland ice core sites (NEEM and SE-Dome). The higher concentration in the Ushkovsky ice core is unlikely to be explained by the difference in the enrichment effect due to the low accumulation rate, because the accumulation rate (0.87 m/yr) of the Ushkovsky ice core (Kawamura et al., 2012) is much higher than that of the NEEM ice core (0.22 m/yr). The higher concentrations of BB tracers in the Ushkovsky ice core likely reflect the greater magnitude of BB in the source regions and/or the site's proximity to the source regions. According to the air mass backward-trajectory analyses, the source of LG and DA in the Ushkovsky ice core is Siberia including Kamchatka (Kawamura et al., 2012), while eastern Canada is likely the

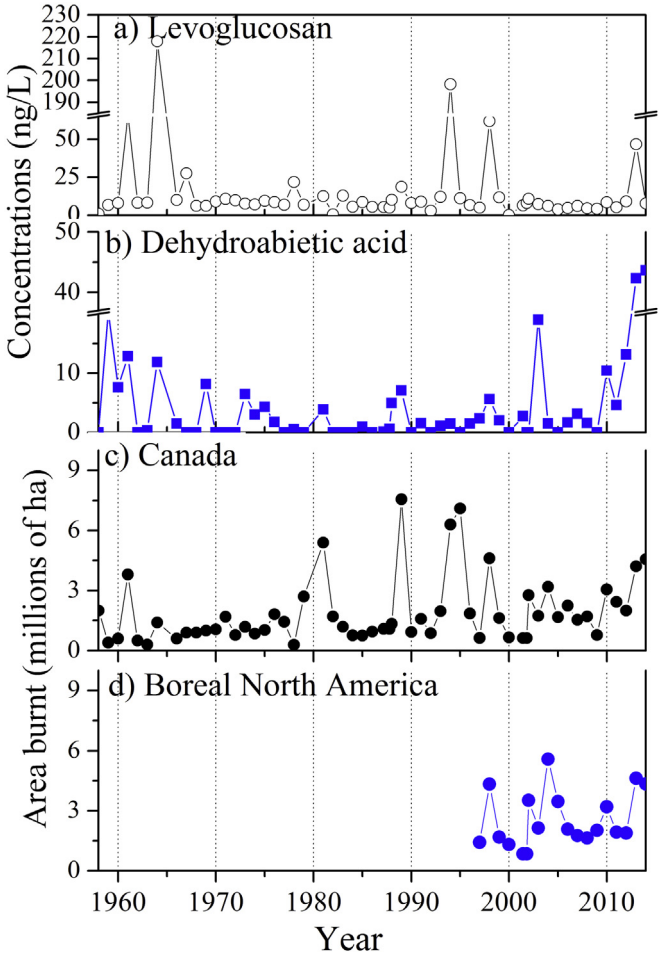

Fig. 3. Historical changes in the concentrations of (a) levoglucosan (LG) and (b) dehydroabietic acid (DA) in the SE-Dome ice core together with (c) the annual area burned in Canada (data adapted and compiled from the National Forestry Database (http://nfdp.ccfm.org/index_e.php), Macias Fauria and Johnson (2008) and Stocks et al. (2002)) and (d) the annual area burned due to vegetation fires in boreal North America (BONA) from 1997 to 2014 (http://www.globalfiredata.org/analysis.html). In GFED4, the continents have been divided into 14 basic regions to represents the BB data on a regional basis, where Canada is considered to be BONA.

Table 1
Mean concentrations of biomass burning tracers in ice cores.

Ice core	Period (AD)	LG (ng/L)	DA (ng/L)
SE-Dome (this study)	1958–2014	18.27 ± 38.85	5.17 ± 10.13
Ushkovsky (Kawamura et al., 2012)	1961–1997	542.6 ± 861.8	101.53 ± 99.77
NEEM (Zennaro et al., 2014)	1961–2001	98.26 ± 84.97	–

primary source of LG and DA in Greenland (Fig. 2). Siberia is one of the most intensive boreal forest BB regions (Giglio et al., 2013; Schultz et al., 2008; Scott et al., 2018). Emissions of BB aerosol tracers in Siberia are estimated to be several times higher than those in eastern Canada (Fig. 1b) (Schultz et al., 2008; Scott et al., 2018).

However, the relatively low concentrations in the SE-Dome ice core compared to the NEEM ice core are unlikely to be explained by differences in the magnitudes of BB between the source regions because the possible sources of LG in the two Greenland ice cores are nearly the same region. This difference is likely instead to be due to a difference in the snow accumulation rates between the two sites. In fact, the snow accumulation rate at the SE-Dome site (1.01 m/yr) is approximately 5 times higher than that at the NEEM ice core site (0.22 m/yr) (Zennaro et al., 2014). Therefore, the dilution effect is significant for the SE-Dome site compared to the NEEM site. Based on these lines of analysis, differences in the concentrations of BB tracers for the different ice core sites are likely explained by differences in the BB intensity between the potential source areas and the snow accumulation rates at the different sites.

3.3. Assessment of BB tracers in the SE-Dome ice core

Ice core records of BB aerosol tracers are potentially affected by several factors, including the emission of the tracers from the source regions (Zennaro et al., 2014), changes in the wind regime (Kawamura et al., 2012), and precipitation (wet deposition) (Zennaro et al., 2014). To examine the primary factors that determine the concentrations of the BB tracers in ice cores, we compared our BB tracer record to BB records in the source region and to instrumental records of decadal-scale climate oscillations. Because natural BB events increase during the warm season (spring to summer) in boreal forests (Johnson, 1992; Macias Fauria and Johnson, 2008; Seki et al., 2015), we compared our BB tracer records in the SE-Dome ice core to warm season climate oscillations that influence the regional wind regime around Greenland and Canada. We do not discuss the effect of precipitation because diurnal resolution BB data, that are required to assess the precipitation effect on aerosol deposition, are not available.

3.3.1. Comparison with the BB record

To evaluate the extent that LG and DA in the SE-Dome ice core record BB events in Canada in the past and to further constrain the source regions of LG and DA, we compared the BB tracer data to the observationally based records of areas burned for all types of vegetation fire (Fig. 3) in Canada. The area burned data were adapted and compiled from the National Forestry Database of Canada (http://nfdp.ccfm.org/index_e.php), Stocks et al. (2002) and Macias Fauria and Johnson (2008). In addition, area burned data of Canada (Fig. 3d) from the Global Fire Emission Database version 4.1s (GFED4.1s) were used for comparison. Area burned data from the National Forestry Database and Stocks et al. (2002) (Fig. 3c) covers the entire period (1958–2014) recorded by the SE-Dome ice core. Conversely, the area burned data in GFED4 (Fig. 3d) is limited to the period of 1997–2014. The area burned data from the National Forestry Database and Stocks et al. (2002) are not complete nor without some limitations. The data have been collected by different agencies. Therefore, the data completeness and quality may vary among agencies and years. Conversely, GFED4, which is used to reconstruct the area burned caused by BB, combined satellite information on fire activity and vegetation productivity to estimate the gridded monthly burned area and fire emissions; in addition, it used scalars to calculate higher temporal resolution emissions. Note that the area burned data used in this study account for burning events over all of Canada including the western region, which is not one of the potential source areas of the BB tracers in the Greenland ice core for most years, as inferred from the air mass backward trajectory (Fig. 2).

Our LG record shows large fluctuations over the past 60 years with sporadic high peaks in 1961, 1964, 1994, 1998, and 2013 with the largest peak in 1964 (Fig. 3a). Conversely, the DA record in the SE-Dome ice core shows marked peaks in 1961, 2003, 2010, 2013, and 2014. The burned area data in Canada (Fig. 3c and d) also show a number of pronounced peaks in 1961, 1981, 1989, 1994, 1995, 1998, 2004, 2010, 2013, and 2014. Six significant BB events in Canada during the past 60 years are found to correspond to the BB tracer peaks in the SE-Dome ice core. This correspondence suggests that the BB molecular tracers in the SE-Dome ice core document approximately 60% of the significant BB events in Canada over the last 60 years. This relatively high correspondence between the ice core record and the BB events in Canada suggests that analyses of BB molecular tracers in Greenland ice cores are useful to reconstruct the frequency of BB events in certain

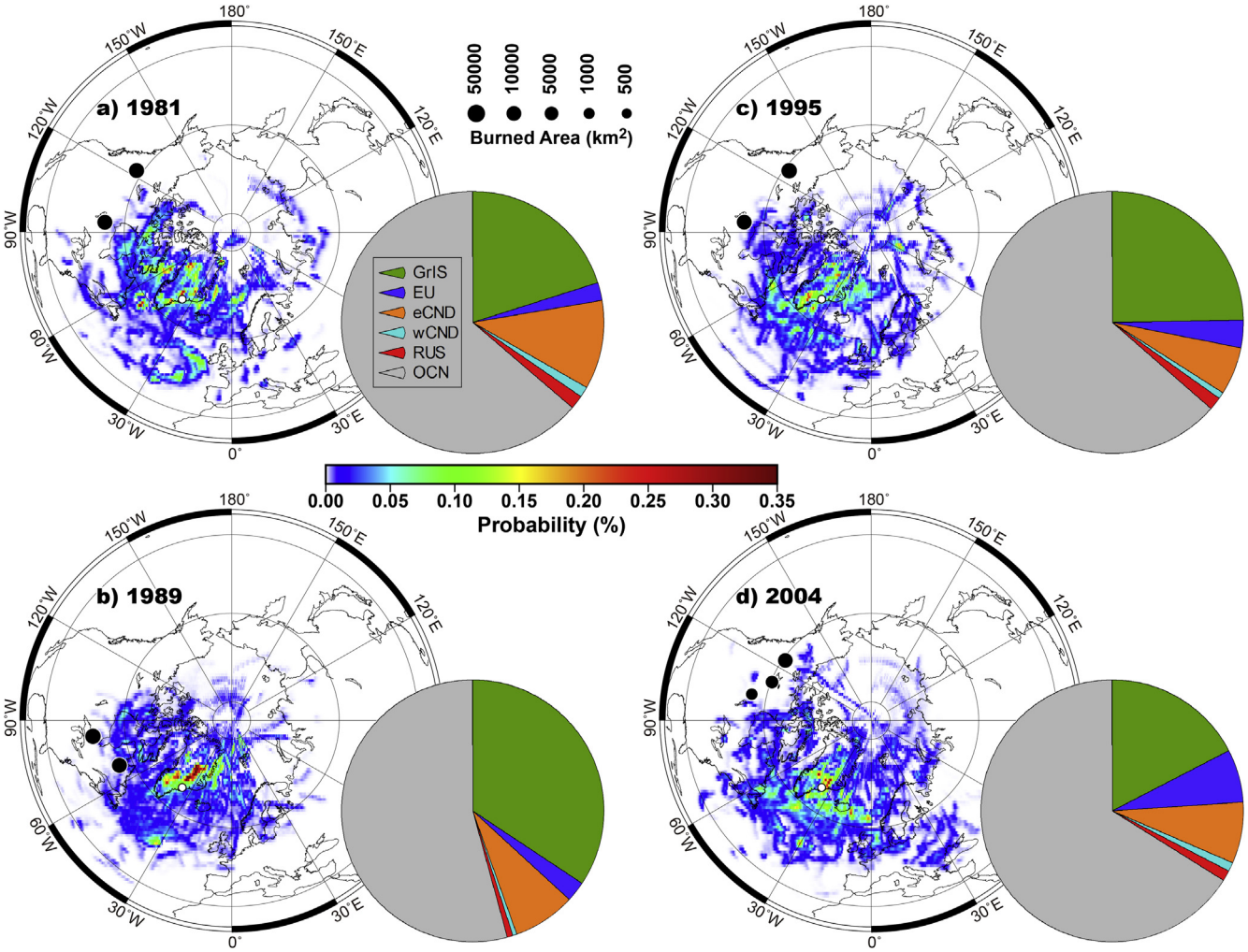

Fig. 4. Probability distributions of an air mass arriving at the SE-Dome site (10–1500 m a.g.l. as initial four points) from a 7-day 3D backward-trajectory analysis in the spring-summer seasons of (a) 1981, (b) 1989, (c) 1995 and (d) 2004, when BB events were prominent in Canada but the events were not recorded in the SE-Dome ice core LG and DA data. Pie diagrams show the relative contribution of each region from where the airmass reached to the SE-Dome ice core (abbreviations as of Fig. 1). Black dots represent the regions where significant BB occurred in those years and the size of the black dots indicates the magnitude of area burnt.

regions of Canada.

However, the four BB events in 1981, 1989, 1995, and 2004 were not registered in the SE-Dome ice core records of the BB molecular tracers (Fig. 3a and d). These mismatches suggest that LG and DA in the Greenland ice core do not record all the BB events in Canada. One possible explanation for the absence of BB signals in the ice core BB tracers is that these BB events took place in regions from which the emitted BB tracers could not reach the SE-Dome. Indeed, the prominent BB events in 1981, 1989, 1995, and 2004 were found to have primarily taken place in the western region of Canada (Stocks et al., 2002). To further examine the observed mismatch between the ice core record and the observational based BB events, we calculated the probability distributions of the 7-day backward trajectories for the spring-summer seasons (six months from March to August) in the selected years (Fig. 4). As shown in Fig. 4, the air masses indeed did not come from the significant BB areas in the mismatched years.

Conversely, in 1961, 1994, 1998, 2010, 2013, and 2014, the air masses came from regions (Fig. 5) where significant BB events occurred (http://nfdp.ccfm.org/fires/national_e.php). The 7-day air mass backward-trajectory analysis suggests that air masses originating in the eastern part of Canada (i.e., Newfoundland, Quebec, Ontario, and some parts of Manitoba) reached the SE-Dome ice core site during those years. Therefore, our result demonstrates that the registration of BB events in Greenland ice cores strongly depends on the origin of the air masses being transported to the ice core site.

Other discrepancies between the ice core and the BB events in Canada are observed in 1964 and 2003. In these years, significant peaks in LG and DA were recognized in the ice core while prominent BB events were absent in Canada (Fig. 3). However, a closer look reveals that, even though the total area burned peak for all of Canada is small in 2003, the burned area peaks in the eastern part of Canada (Ontario and Manitoba) were relatively high (http://nfdp.ccfm.org/fires/national_e.php). In addition, the probability distribution of the trajectory analysis shows that 29% of the air mass arriving at the SE-Dome site in the spring of 2003 came from Canada, which includes Quebec, Ontario, and Manitoba. Therefore, the significant DA peak in 2003 may reflect local BB events in Ontario and Manitoba.

As for 1964, the source region of the LG peak remains unclear because there were no significant area burned events in any region in Canada. One possible explanation for the mismatch is that the BB tracers originated from regions other than Canada such as the Greenland and Europe in 1964. The probability distribution of the 7-day air mass backward trajectory for 1964 (Fig. 6) shows that a significant portion of the air mass came from Greenland (11%) and Europe (11%) in spring to summer. However, most area of the Greenland is covered with ice and is inhospitable to life forms. As for Europe, the number of fires and area burned is reported to be prominent in East Germany in that year (Goldammer and Mutch, 2001) and fire weather index is also high in

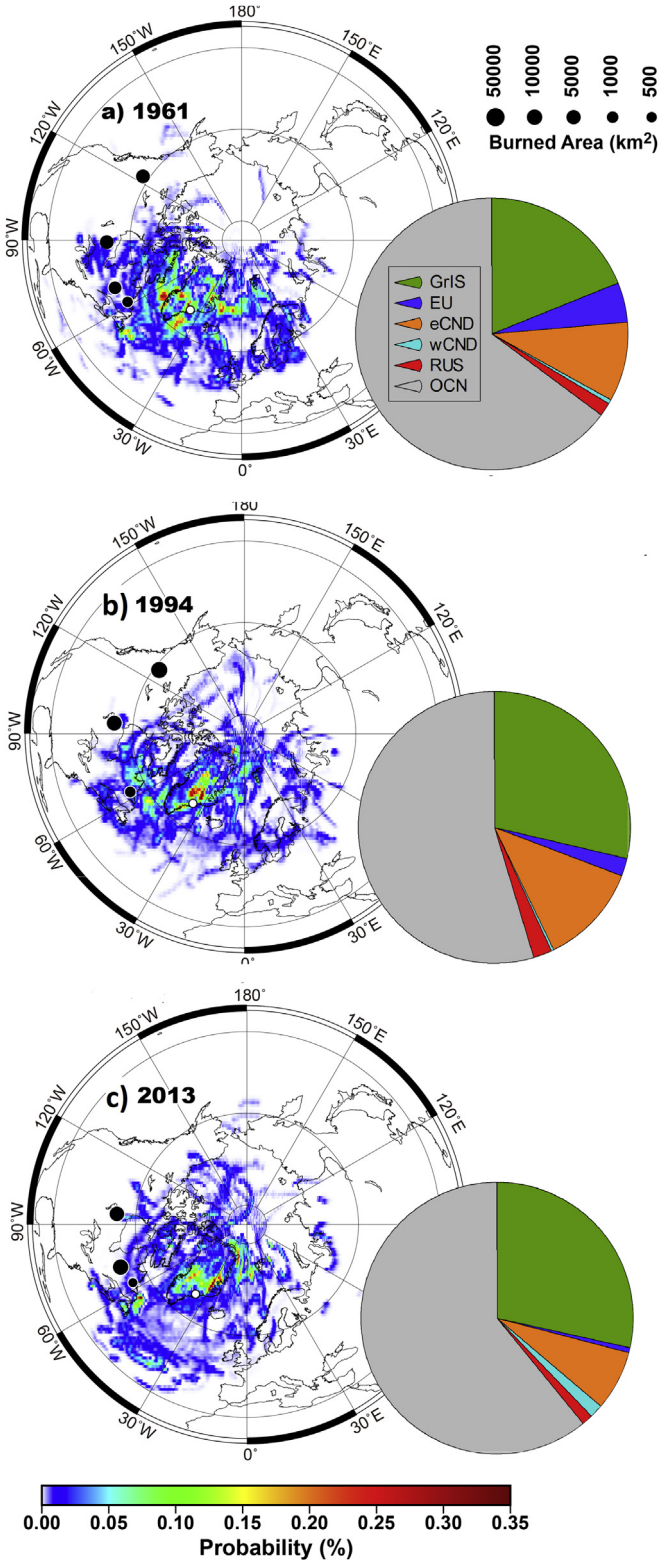

Fig. 5. Probability distributions of an air mass arriving at the SE-Dome site (10–1500 m a.g.l. as initial four point) from a 7-day 3D backward-trajectory analysis in the spring -summer season of (a) 1961, (b) 1994 and (c) 2013, when BB events were prominent in Canada and the LG and DA records in the SE-Dome ice core also show prominent peaks. Pie diagrams show the relative contribution of each region from where the airmass reached to the SE-Dome ice core (abbreviations as of Fig. 1). Black dots represent the regions where significant BB occurred in those years and the size of the black dots indicates the magnitude of area burnt.

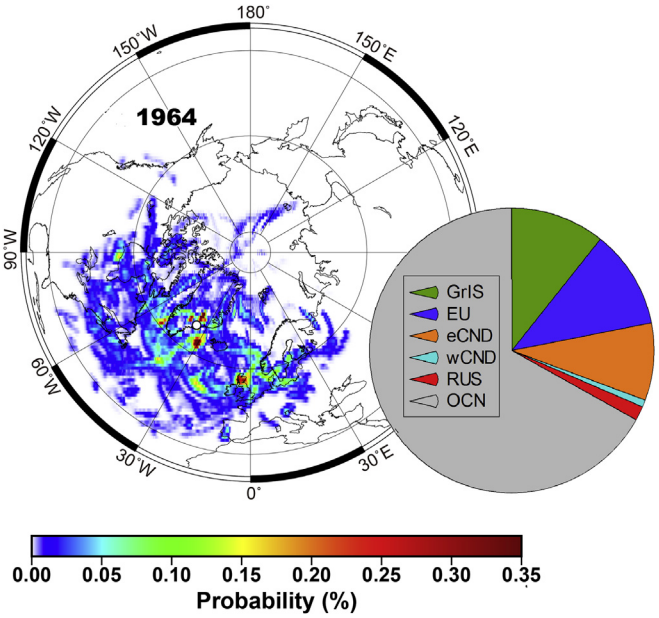

Fig. 6. Probability distributions of an air mass arriving at the SE-Dome site (10–1500 m a.g.l. as initial four point) from a 7-day 3D backward-trajectory analysis in the spring -summer season of 1964. Pie diagrams show the relative contribution of each region from where the airmass reached to the SE-Dome ice core (abbreviations as of Fig. 1).

Europe during 1960–1965 (Venäläinen et al., 2014). Hence, the LG peak in 1964 might reflect fire event in Europe.

Our data shows no correlation between LG and DA in the SE-Dome ice core, even though coniferous trees are more numerous than deciduous trees in Canada. Such a decoupling of LG and DA is also found in the Ushkovsky ice core (Kawamura et al., 2012). This indicates that the LG originated from other sources, such as deciduous trees, brown coal, and agricultural wood, rather than coniferous trees (Oros and Simoneit, 2000; Simoneit et al., 1999). An alternative explanation is the preferential degradation of DA during the transport process and after deposition onto ice sheet. The aromatic structure of DA has a higher sensitivity to photodegradation than LG (Elias et al., 2001; Simoneit, 2002). LG is also thought not to be significantly degraded in the early firnification process (Kehrwald et al., 2012). In addition, a lack of covariance between DA (tracers of conifer resin burning) and LG (cellulose burning tracers) may imply that conifer tree barks are more readily burned than woody parts during BB events (Eichler et al., 2011).

3.3.2. Effect of decadal-scale climatic oscillations on BB aerosol tracer transportation

Next, we examine the influence of changes in the decadal-scale atmospheric circulation on aerosol transport. Changes in the wind regime affect the atmospheric transport of aerosols. For example, sources of aerosols in East Asia are dramatically changed due to seasonal changes in the Asian monsoon circulation (Kawamura et al., 2003). Decadal-scale changes in the wind regime may also influence the transport of aerosols. The Arctic Oscillation (AO), which is a key feature of climate variability in the studied region, may influence the transport of aerosols to high latitude regions (Fu et al., 2016; Seki et al., 2015). The AO refers to an opposing pattern of pressure between the Arctic and the northern middle latitudes. When the AO index is positive, the surface pressure is low in the polar region, which helps the middle latitude jet stream blow strongly and consistently from west to east and induces warmer air to move northward from the mid-latitudes (Thompson and Wallace, 1998). Conversely, when the AO index is negative, the opposite situation occurs. Seki et al. (2015) postulated that a positive AO might intensify the long-range transport of BB aerosols from their source regions

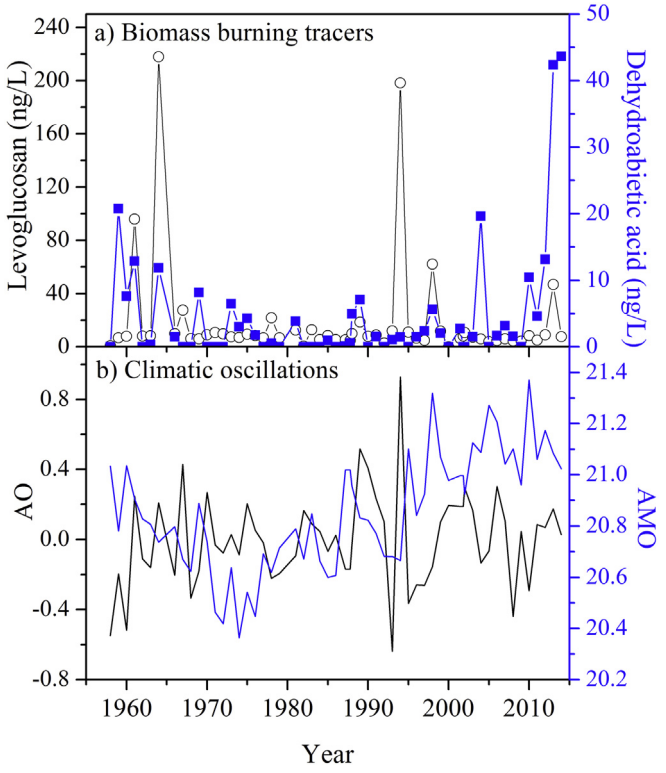

Fig. 7. (a) Historical trends in the concentrations of levoglucosan (LG) (black open circle) and dehydroabietic acid (DA) (blue solid square) in the SE-Dome ice core. (b) Instrumental records of the warm season Arctic Oscillation (AO) index (black line) (https://www.ncdc.noaa.gov/teleconnections/ao/) and the Atlantic Multi-decadal Oscillation (AMO) index (blue line) (http://www.esrl.noaa.gov/psd/data/timeseries/AMO/) during the period from 1958 to 2014. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

to ice core sites via the westerly jet. The Atlantic Multi-decadal Oscillation (AMO) is a variability in the North Atlantic sea surface temperatures with a periodicity of 60–80 years (Kerr, 2000) and might also influence the transport of aerosols around Greenland. The AMO coupled with the atmosphere may trigger the AO to vary on a multi-decadal time scale (Polyakova et al., 2006), control the position of the westerly jet, and potentially affect aerosol transport around Greenland.

We compared our BB tracer records of the SE-Dome ice core to the warm season AO and AMO indexes to examine the extent to which ice core records are linked to climate oscillations. The LG record is moderately correlated to the warm season AO ($R = 0.42$, $p < 0.001$), while DA exhibits no correlation (Fig. 7a and b). However, we found no correlation of either BB tracer records with the AMO. These results suggest that changes in the climatic oscillations do not significantly influence the transport of BB tracers that originate in eastern Canada. Hirdman et al. (2010) also argued that changes in the atmospheric circulation can only explain a small fraction of the long-term trends in the BC loading in the Arctic and that changes in the emissions from the source regions primarily dominate these trends.

4. Conclusions

We generated for the first time continuous records of BB molecular tracers over the last 60 years from a well-dated Greenland ice core (SE-Dome) and assessed the BB molecular tracers in the ice core. A comparison of our ice core tracer records with those of other ice cores indicates that concentrations of BB tracers in ice cores likely reflect differences in the BB magnitude in the source area and that the snow accumulation rate likely has an influence. LG and DA data in the SE-Dome ice core record most of the BB events in eastern Canada from 1958 to 2014, suggesting that ice core analyses of LG and DA in Greenland ice cores are a promising approach to reconstruct the BB frequency in eastern Canada, even though some BB events in Canada are not reflected in the ice core LG and DA records because these records also depend on the origins of the air masses reaching the ice core site. Decadal-scale climatic oscillations such as AO and AMO do not appear to significantly influence the transport of BB tracers. The apparent discrepancy between LG and DA suggests that along with boreal forest fires, other BB events, such as agricultural wood burning, deciduous plant burning, and coal combustion, are also important sources of the LG preserved in the SE-Dome ice core.

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