Clim. Past Discuss., https://doi.org/10.5194/cp-2017-158
Manuscript under review for journal Clim. Past
Discussion started: 21 December 2017

Climate of the Past Discussions

High latitude Southern Hemisphere fire history during the Mid-Late Holocene (750- 6000 yr BP)

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Abstract. High latitude Southern Hemisphere fire history was reconstructed by determining the specific biomarker levoglucosan in ice cores from the TALos Dome Ice CorE drilling project (TALDICE) during the Mid-Late Holocene (750-6000 yr BP). Potassium was also analyzed in order to provide a comparison with another fire proxy to create a more robust biomass burning record. The levoglucosan record is characterized by a long-term increase with higher rates starting at~4000 yr BP and higher peaks between 1500 and 2500 yr BP. Comparisons with charcoal syntheses help evaluate fire sources, showing a possible higher contribution from Patagonian fires rather than Australian biomass burning. We interpret the anomalous increase in levoglucosan centred at ~2000 yr BP as a combination of the atmospheric transport pathway and the interplay between climatic factors.

1 Introduction

Fire and climate reciprocally influence one another. Biomass burning affects the chemical composition of the atmosphere, the global carbon cycle and the radiative balance due to the emission of greenhouse gasses (carbon dioxide, carbon monoxide, methane and nitrous oxide) as well as aerosols (Andreae and Merlet, 2001; Akagi et al., 2011; Bowman et al., 2009; Keywood et al., 2013; Galanter et al., 2000; van der Werf et al., 2004; Harrison et al., 2010). Temperature, precipitation and atmospheric carbon dioxide (CO_2) control both fuel productivity and flammability where biomass growth, fire ignition and spread are favoured by the oscillation of wet and dry conditions (Westerling et al.,2006; Daniau et al., 2010; Marlon et al., 2013; Pyne, 2001; Rollins et al., 2002). Spatial variability in climate and the resulting fire-vegetation-climate interactions further complicate fire dynamics (Lynch et al., 2004). For example, Holocene fire records from the same region may not be synchronous over centennial to millennial timescales (Brunelle and Whitlock, 2003). Furthermore, regional

anthropogenic deforestation for the creation of open spaces for croplands and herding adds another variable to the fire-climate system. Anthropogenic impacts complicate the dynamics, as humans are able to both provoke and extinguish fires (Clare-Smith et al, 2016, Zohary et al., 2012; Tauger, 2013; Ruddiman, 2003; Marlon et al., 2008; Power et al., 2008, Chuvieco et al., 2008; Archibal et al., 2009). The early anthropogenic hypothesis proposed by Ruddiman (2003) is still debated in terms of the scale of the effect of early agriculture on the global climate system, but there is no doubt that land use changes affect climate at regional scales (Broeker and Stocker,2006; Joos et al., 2004; Singarayer et al., 2011; Mitchell et al., 2013, Kaplan et al., 2009 and 2011).

Charcoal is an accepted proxy for reconstructing past fire regimes from sedimentary archives. Distinguishing microscopic (< 100 µm) to macroscopic (>200 µm) charcoal allows reconstructing fire history at regional and local scales, respectively (Clark et al., 1996; Marlon et al., 2013; Carcaillet et al., 2002; Whitlock et al.,2007). The Global Charcoal Database (GCD) (Power et al. 2010, https://www.paleofire.org/) provides a useful dataset for research sedimentary records of fire (Blarquez et al., 2014; Marlon et al., 2008 and 2013; Power et al., 2008 and 2013; Daniau et al., 2012; Mooney et al., 2011; Vanniere et al., 2011) and the paleofire R package creates customized charcoal syntheses (https://github.com/paleofire/GCD, Blarquez et al., 2014). Although the recent Global Charcoal Database version 3 (GCDv3) compiles more than 700 charcoal records (Marlon et al. 2016), the sites included in GCD are not homogenously distributed, leading to geographical over- or underrepresentation. For example, Africa and Asia are poorly represented when compared to North America, Europe and Australia. The atmospheric transport of macrocharcoal is limited to a few kilometers (Carcaillet et al 2001) and therefore charcoal records cannot encompass some areas such as deserts or polar regions.

However, ice cores complement charcoal syntheses in that they can reconstruct fire histories at regional to semi-hemispheric scales (Legrand et al. 1992). Fire proxies in ice cores include a list of inorganic chemicals such as ammonium, nitrate and potassium (Legrand et al. 2016), black carbon (McConnel, 2007) and some organic compounds such as formate, oxalate, phenolic compounds and levoglucosan (Legrand et al., 2016; Zangrando et al., 2013 and 2016) that are biomass burning by-products. These proxies originate from biomass burning, but not all of them are exclusively emitted by combustion. Several organic compounds are strongly related to the type of fuel where, for example, dehydroabietic acid is produced by conifer combustion (Fine et al. 2002), but oxalate, phenolic compounds and potassium, for example, are not specific indicators of biomass burning. Emission factors and lifetimes differ between proxies and atmospheric transport between the source region and the drilling site may influence individual records (Rubino et al. 2016).

Levoglucosan (1,6- anhydro β-D-glucopyranose) is a specific marker of biomass burning, as it is only produced by cellulose combustion (Kuo et al., 2008). The atmospheric lifetime of levoglucosan is actively debated with estimates of a few days (Hoffman et al., 2010; Hennigan et al., 2010) to a few weeks (Bai et al., 2013; Slade and Knopf, 2013). Levoglucosan records extending back 15,000 years reconstruct high northern latitude fire history in Greenland (Zennaro et al., 2015).

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Mountain glacier levoglucosan records depict regional fire histories (Kawamura et al. 2012; Yao et al. 2013). Southern Hemisphere levoglucosan records still leave much to be explored. Antarctic levoglucosan records will likely differ from Arctic records due to the substantially smaller land masses surrounding near the ice sheet and the long distances required for atmospheric transport of biomass burning material. For example, the southernmost tip of Patagonia, the closest continental landmass to Antarctica, only extends to ~55°S.

Hemispheric fire history since the Last Glacial Maximum, derived from marine records (Daniau et al. 2012), demonstrates relatively high fire activity during the Holocene where biomass burning is more pronounced at high latitudes. The northern and southern hemisphere fire histories substantially differ from one another (Daniau et al. 2012) where the southern hemisphere fire history is characterized by a widespread spatial heterogeneity (Power et al., 2008). From the Mid-Late Holocene onward, sea levels approached near-modern levels, and most regional Southern Hemisphere glacial ice had melted. The increase in human population in Australasia and South America and the associated shifting vegetation types and/or the strengthening of the El-Niño Southern Oscillation (ENSO) activity explain the heterogeneity of fire patterns since 3000 y BP (Power et al 2008, Whitlock et al. 2007).

We aim to reconstruct Southern Hemisphere fire history by determining levoglucosan concentration in an ice core obtained from the TALos Dome Ice CorE drilling project (TALDICE) during the Mid-Late Holocene (750-6000 yr BP). Talos Dome (159°11'E, 72°49'S, 2315 m a.s.l., Fig.1) is located in the South Pacific/Ross Sea sector of the East Antarctic Plateau (www.taldice.org). The relatively high snow accumulation rates (80 kg m^{-2} yr^{-1}, average 2004-1259 AD; Stenni et al. 2002) enables accurate dating of the core (Buiron et al., 2011; Veres et al. 2013) and high-resolution climate analyses during the Holocene (Albani et al., 2012a; Delmonte et al., 2013). We also analyze potassium in order to provide a more complete biomass burning record through a comparison with another fire proxy. Charcoal syntheses provide information of biomass burning source regions and changing fire activity. These fire and climate records can provide a synthesized history of high latitude Southern Hemisphere biomass burning in a relatively stable climate regime that is subject to an increasing human influence.

2 Methods

2.1 Ice core samples and analysis

We analyzed 266 ice core samples from the TALDICE (TD) ice core, covering the depth interval between 90 and 403 m. Each sample represents the uppermost 15 cm of a 1 m ice core section. These sections were the ice remaining after other analyses performed by Italian TALDICE researchers. The samples are therefore non-continuous and equidistant in space but not in time. The ice core samples were stored at Ca' Foscari University of Venice at -20°C until analysis. Before the analysis, the ice samples were decontaminated by washing the outermost section three times using ultrapure water (ELGA

LabWater, Marlow, UK) in an ISO 5 clean room in order to remove possible drilling fluid residuals or impurities. During the washing the samples were held using PTFE tongs (Nalgene Corporation, Rochester, NY) previously immersed for two days in 5% and 1% HNO_3 solutions and rinsed with ultrapure water. The same procedure was used for decontaminating LDPE bottles and vials. Ice samples were melted at room temperature in 125 mL LDPE bottles and then transferred to 15 mL LDPE bottles and stored at -20°C in triple polyethylene bags. These precautions were necessary for minimizing any possible contamination of the samples.

Levoglucosan analysis was performed using a method specifically developed for the analysis of polar ice samples (Gambaro et al. 2008 and as modified in Zennaro et al., 2014 and 2015) based on liquid chromatography coupled with a triple quadrupole mass spectrometer (HPLC/(-)ESI-MS/MS). Briefly, we injected 300 µL of the sample in a HPLC system (Agilent 1100, Waldbronn, Germany).Chromatographic separation was obtained using a C-18 Synergy Hydro column (4.6 mm i.d., 50 mm length, 4 µm size particles; Phenemenex, Torrance, CA) and an autosampler was equipped with LOOP Multidraw 44 Upgrade Kit G1313 – 68711. The samples were eluted with methanol (ultragradient, H411, Romil Ltd. Cambridge, U.K) and ultrapure water (18.2 MΩ, TOC 1 ppb, PURELAB Pulse and PURELAB Flex, Elga). The mass spectrometer was a API 4000 (Applied Biosystem/MDS SCIEX, Toronto, Ontario, Canada) equipped with a ion spray source ((-)ESI) Turbo V that operated in negative polarity. Mass/charge (m/z) ratios used for the quantification were 161 and 113 for levoglucosan and 167 and 118 for ^{13}C-labelled levoglucosan. This method limits pre-analytical procedures that may affect the quantitative determination and the sample contamination and it allows analyzing levoglucosan at levels of a few pg mL^{-1}. We quantified the results using isotopic dilution ($^{13}C_6$-labelled levoglucosan as an internal standard; Cambridge Isotope Laboratoies Inc. (Andover, MA)) and instrumental response factors were determined prior, during and after each set of analyses in order to evaluate eventual deviation from the instrumental response. Procedural limit of quantification (LoQ) for levoglucosan was 4 pg mL^{-1}.

The analytical methods for the analysis of Na, Fe and K are previously reported (Vallelonga et al. 2013). Melted samples were acidified to pH 1 using sub-boiling distilled HNO_3 (Romil, Cambridge, UK) and analyzed at least 24 hours later by Inductively Coupled Plasma Sector Field Mass Spectrometry (ICP-SFMS; FinniganTM ELEMENT2, Thermo Fisher Scientific Inc., Bremen, Germany) coupled to an APEX Q desolvating introduction unit (Elemental Scientific, Omaha, NE, USA) in low (^{23}Na), medium (^{56}Fe) and high (^{39}K) resolution modes. The detection limits, defined as three times the standard deviation of the instrument blank, and analytical precisions were (0.06 ng mL^{-1}, 3%), (0.03 ng mL^{-1}, 16%) and (0.2 ng mL^{-1}, 3%) for Na, Fe and K, respectively.

2.2 Dating and data treatment

The ice cores samples ages were calculated using the AICC2012 age scale (Bazin et al. 2013; Veres et al. 2013). Levoglucosan and biomass-burning potassium (K_{bb}) concentrations were normalized into flux values by dividing by the

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accumulation rate for the corresponding ice section (Buiron et al. 2011). Charcoal syntheses were performed by using the paleofire R package associated with the Global Charcoal Database by extracting a 20-year bin time window (consistent with the time period covered by each sample analyzed for levoglucosan) from 6000 y BP to present. Detection and separation of anomalous intense peaks from the levoglucosan and K_{bb} series were based on Cook's distance (Cook, 1977), after generalized additive modeling (GAM) of the relationship between each series and age using the R package mgcv (Wood, 2006). A preliminary data analysis revealed that levoglucosan records were highly skewed, therefore they were log-transformed before any further analysis to reduce skewness. All other skewed variables were also log-transformed. Potential associations between levoglucosan and K_{bb}, as well as atmospheric transport indicators, fire sources and climate parameters, were analyzed the records using the slot correlation with Gaussian kernel, in order to take into account of the irregularly sampled data (Rehfeld et al., 2011). In order to investigate the statistical significance of the resulting correlation estimates (r), the 95% bias corrected and accelerated (BCa) confidence intervals (95% CI) were constructed based on 5000 bootstrap samples. Calculations were made in R (R Core Team, 2017) using the boot package (Canty and Ripley, 2017; Davison and Hinkley, 1997).

3 Results and Discussion

3.1 Fire tracers in the Talos Dome ice core

Levoglucosan is a source specific proxy for fire that is produced and emitted by biomass burning, with a maximum yield at ~250°C (Kuo et al., 2008). In the 266 TD ice core samples analyzed in this work, levoglucosan concentrations ranged from 4 (corresponding to the LoQ) to 1100 pg mL^{-1}. However, in 68 of the overall 266 samples the levoglucosan concentrations were below the LoQ. The highest obtained concentrations were lower, but of the same order of magnitude, than the major levoglucosan peaks detected in the NEEM (Greenland) ice core (185-1767 pg mL^{-1}) during the late Holocene (Zennaro et al., 2014) and in the Ushkovsky (Kamchatka Peninsula) ice cap (10-5000 pg mL^{-1}) during the last three centuries (Kawamura et al,. 2012). Levoglucosan concentrations from these remote regions are significantly lower than in environmental archives closer to more human-influenced areas such as in the Muztagh Ata and Tanggula ice cores where concentrations were 10-718 ng mL^{-1} and 10-93 ng mL^{-1}, respectively (Yao et al., 2013).

The complete Mid-Late Holocene Talos Dome levoglucosan record is shown in Fig. 2A. Levoglucosan spikes potentially indicate extreme fire events, more frequent fire activity during the short temporal interval, and/or more efficient atmospheric transport during the fire event. These anomalous intense peaks significantly affect the temporal trend of the record even when adopting different smoothing approaches (Zennaro et al., 2014). In order to better identify which individual peaks could influence the entire dataset, we used Cook's distance to indicate outliers (n=9) which resulted in peaks with values greater than ~ 16 μg m^{-2} yr^{-1} (labeled with red dots in Fig. 2A). We therefore separate these intense peaks from the longer-term fire history (Fig. 2A and Fig. 2A-1, respectively).

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The levoglucosan flux is lowest during the oldest part of the record (between 6000 and 4000 yr BP), while a significant long-term increase occurred after approximately 4000-3500 yr BP (Fig. 2A-1). Levoglucosan increases between ~2000 and ~2250 yr BP, and with a major increase between ~1250 to ~800 yr BP. The timing of the individual levoglucosan spikes (Table 1) roughly occur between 2200-1800 and 1200-900 yr BP, corresponding with the major long-term increases. Thus, these extreme peaks should be considered as part of a more widespread period when levoglucosan was generally higher in the Talos Dome Mid-Late Holocene sequence rather than isolated or punctual fire events.

Attributing these individual spikes only to megafire events, however, may oversimplify their source. OH• radicals, that may occur in the atmosphere in both the gas and aqueous phase, are able to oxidize levoglucosan, leading to degradation during transport (Hennigan et al., 2010; Hoffman et al., 2010). The extent of this degradation is currently unknown as recent studies also propose that levoglucosan can exist in the atmosphere up to 26 days (Bai et al. 2013). The atmospheric stability of levoglucosan and atmospheric transport dynamics both influence fire signals in polar ice (Zennaro et al., 2014; Legrand et al., 2016). Although large amounts of levoglucosan are emitted during biomass burning and are quantifiable even at remote distances from the source (Zennaro et al., 2014 and 2015, Kehrwald et al., 2012), the detected levoglucosan concentrations may be affected by a combination of transport and degradation, thus making the signal over or under-representative of the initial individual fire event. Conditions such as varying precipitation amounts, dry versus wet deposition, and the distance from the fire source can modulate the levoglucosan signal, thus resulting in reducing the effects of megafire events or amplifying modest fires. Conversely, the stability of levoglucosan once trapped in the ice may be less significant. Levoglucosan profiles in polar ice cores do not result from simple degradation curves, supporting the assumption of the stability of this proxy in ice cores (Zennaro et al., 2014 and 2015, Kehrwald et al., 2012).

Besides levoglucosan, several elements such as potassium have been proposed as tracers for biomass burning in ice cores (Cachier et al. 1991). The primary sources of potassium in aerosols are soil, sea-salt and biomass burning smoke. Some organic matter emits potassium when burned, which can help identify the contribution of biomass burning in atmospheric air masses. Unlike levoglucosan, potassium is not a source-specific proxy. Marine and terrestrial inputs affect the potassium contribution from fire plumes (Legrand and de Angelis 1996; Savarino and Legrand 1998). In aerosols, several authors propose the following formula to obtain biomass burning potassium (K_{bb}) by correcting for the terrestrial and marine contributions to the total potassium concentrations (Begum et al., 2006; Cahill et al., 1980):

$$[K_{bb}] = [K] - 0.6[Fe]\text{-}0.038[Na], \quad (1)$$

where [K], [Fe] and [Na]are the potassium, iron and sodium concentrations in the sample, respectively. Although Legrand and de Angelis (1996) proposed the use of calcium instead of iron for taking into account of the terrestrial contribution to potassium in Greenland firn cores ($[K^+]/[Ca^{2+}] = 0.04$), this approach may be incorrect at Talos Dome, where a significant

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contribution from local sources can affect the Ca^{2+} concentrations (see paragraph 3.2 for a more detailed discussion). Here, we chose to evaluate K_{bb} using Eq.(1).

We applied Cook's distance to the K_{bb} profile in the same manner as for the levoglucosan profile in order to determine anomalous peaks (n=9, red dots; Fig. 2B). The long-term K_{bb} profile significantly increased at ~4000 yr BP (Fig. 2B-1), with other high values at 2000-2500, 2750-3000 and 3500-3750 yr BP, followed by a further increase between 1250-750 yr BP. The K_{bb} profile only partially agrees with levoglucosan. For example, K_{bb} increases between ~3000 and ~4000 yr BP where this peak is not consistent with the levoglucosan record. These differences may be due to the non-specificity of potassium for biomass burning, where the multi-source aspect of potassium remains a critical issue when reconstructing fire histories using this proxy.

The outlying values of K_{bb} (Table 1) mainly occur after 2200 yr BP and are present in two distinct periods between 900-1200 yr BP (group I) and 1900-2200 yr BP (group III). These periods are similar to the time intervals with outlying values in the levoglucosan record. Levoglucosan and K_{bb} spikes in group I perfectly coincide at 1180 and 1193 yr BP, including their relative amounts after applying a minimax transformation. In group I, only sample TD108 does not have a direct match between K_{bb} and levoglucosan peaks. In the other samples included in group I, a direct comparison is not possible because the samples were not simultaneously analyzed for levoglucosan and K_{bb}. In contrast, between 1600-1900 yr BP (group II) there is no correspondence between the levoglucosan and K_{bb} peaks, as only levoglucosan was detected during this time period. Group III (between 1900-2200 yr BP) showed a perfect peak correspondence at 1920 yr BP, while in the short period between 2160-2200 yr BP levoglucosan and K_{bb} peaks were very close to one another, although not coincident, and are staggered by approximately 15-25 years. Considering that we analyzed the same ice samples, a possible uncertainty in chronology is not sufficient to explain this latter discrepancy. It is possible that potential differences in migration of K^+ ions and levoglucosan within the ice matrix and/or selective depositional/ post-depositional selective result in this offset between the two proxies, although migration of soluble inorganic ions appeared to be limited in several cases (Kreutz et al. 1998).

The individual levoglucosan and K_{bb} spikes occur during the same time windows, where these time intervals also correspond to high long-term values, suggesting periods of increased fire activity. The relationship between levoglucosan (log-transformed) and K_{bb} profiles was estimated using the slotting correlation approach. Data indicate a positive and significant correlation between the two variables, both including (r (95% CI) =0.264 (0.141, 0.536)) and excluding outliers (r (95% CI) =0.394 (0.280, 0.563)). This comparison between the specific biomarker levoglucosan and the multi-source K_{bb} suggests that K_{bb} can increase the information obtained from a single fire proxy, but that K_{bb} is often best used in conjunction with other biomass burning markers. In the following discussion, we focus on the levoglucosan record for reconstructing fire activity during the Mid-Late Holocene.

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3.2 Atmospheric transport

Unlike central East Antarctic Plateau locations (i.e. Vostok (Petit et al., 1999) and EPICA Dome C (EDC) (Lambert et al., 2008)), Talos Dome is significantly influenced by dust of local origin. Talos Dome contains dust grains larger than 5 µm (diameter) that are absent in the EDC core (Delmonte et al. 2010a). The finer dust fraction (1-5 µm) at Talos Dome is considered to be a mixture of local and remote sources, while this same fraction in EDC is due to long-range transport where the net dust amount is substantially less than at Talos Dome. However, a clear declining trend in dust flux since ~8000 y BP was evident in both ice core records (Albani et al., 2012a; Delmonte et al., 2005). The progressive opening of the Ross Sea embayment from the Early Holocene onward increased the relative contribution of regional dust (Hall et al., 2009; Stenni et al., 2011; Masson-Delmotte et al., 2011; Albani et al., 2012 a).

The composition of mineral dust provides useful indications of the original source and subsequent atmospheric transport. The isotopic composition of dust in central East Antarctica suggests that Southern South America was the primary dust source during glacial periods, with possible contributions from Australia (Delmonte et al., 2010b; Grousset et al., 1992; Basile et al., 1997; Delmonte et al., 2008; Gabrielli et al., 2010; Vallelonga et al., 2010). During the Holocene, the considerable contribution of dust from proximal ice-free areas of northern Victoria Land, Antarctica, located at similar altitude with respect to the drilling site, (Delmonte et al., 2010a) renders the isotopic analysis of Talos Dome dust unreliable. Nevertheless, the $^{87}Sr/^{86}Sr$ vs $^{143}Nd/^{144}Nd$ isotopic signature of mineral dust in other East Antarctica sites (Vostok and Dome C) prompted Revel-Rolland (2006) to consider a possible Eastern Australian provenience. Although the Patagonian (South American) source was reduced with respect to glacial periods, several authors proposed that the South American provenance also persisted during the Interglacials (Holocene and MIS-5), but where the source shifted to lower latitude regions (Delmonte et al., 2008; Gaiero et al., 2007). This hypothesis is also supported by the lithium enrichment factors from the EDC ice core that are similar to continental ice cores from Bolivia (Siggaard-Anderses et al., 2007; Correia et al., 2003). However, a relative weakening of the South American source during Interglacials cannot be excluded (Delmonte et al., 2007 and 2008; Gaiero et al., 2007) with a feasible combination of both Australian and South American contributions during the Holocene.

We cannot stress strongly enough that the source regions and atmospheric transport of mineral dust and organic materials in smoke plumes may not be the same. Comparisons between Talos Dome levoglucosan (log-transformed) and mineral dust <5 µm (log-transformed data from Albani et al. 2012) provide statistically significant negative correlation (r (95% CI) = -0.215 (-0.365, -0.103)). These results suggest that at least part of the levoglucosan record at Talos Dome may be affected by a different circulation pathway.

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3.3 Where from the Southern Hemisphere?

The levoglucosan profile likely reflects fire activity occurring in the nearby continents, as Antarctica contains essentially no biomass available for burning. Although charcoal is a local proxy for fire (Whitlock and Larsen 2002; Clark et al., 1996 and 1988), the syntheses provided by the Global Charcoal Database (Marlon et al. 2016; Blarquez et al. 2014) is a useful tool for reconstructing fire history across regions (Marlon et al. 2013 and 2016). We compiled charcoal syntheses from different potential source areas of the fire signal recorded in the Talos Dome ice core: (i) East Australia (including New Zealand) between 90° and 30°S (latitude) and between 160° and 180° E (longitude) (ii) Patagonia between 90° and 30°S (latitude) and between 60° and 80° W (longitude)and (iii) a synthesis of the Southern Hemisphere covering areas between 90° and 30° S (Fig.3). The number of charcoal records varies by region where the East Australia and Patagonia composite records were obtained by combining n=131 and 25 sites, respectively. In order to compare the levoglucosan record with the charcoal syntheses, we applied a Box-Cox transformation to the records (see Methods section; Power et al. 2008). As is evident from Fig.3, the levoglucosan increase observed at ~3500 yr BP coincides with the same increase in Patagonia and SH charcoal syntheses. Conversely, the levoglucosan increase centered at ~2000 yr BP, which also includes more frequent outliers is not supported by any charcoal records. The correlation analysis of Talos Dome levoglucosan (log-transformed; outliers excluded) and charcoal records produces a statistically significant positive correlation with Patagonia (r (95% CI) =0.333 (0.242, 0.438)) but not in East Australia (r(95% CI) =0.011 (-0.078, 0.095)). However, levoglucosan does positively correlate with the total SH charcoal synthesis (r (95% CI) =0.225 (0.138,0.317)).

These results may suggest a higher contribution from Patagonian fires rather than Australian biomass burning. However, it is also possible that this observation may be due to the higher regional variability in East Australian fire activity (Mooney et al., 2011). During the Mid-Holocene, sites in the far south of South-Eastern Australia showed higher fire activity due to a shift toward more moisture-stressed vegetation, while sites in the Southern Tablelands and east of the Great Dividing Range showed less fire activity with the region supporting relatively moisture-demanding vegetation (Pickett et al., 2004; Mooney et al., 2011). Thus, at a wider spatial scale, these contributions tend to cancel one another. As a result, although the South-Eastern Australia charcoal synthesis is characterized by higher values at about 6000 yr BP, during the Mid-Holocene, only small local increases at ~4000 y BP and ~2500 yr BP were observed. The Patagonia charcoal synthesis, instead, is characterized by biomass burning mainly driven by fuel availability rather than suitable climate conditions (Iglesias and Whitlock, 2014). Therefore, when dry conditions prevailed throughout Patagonia, the sparse steppe-dominated vegetation was the limiting factor for fire occurrence.

3.4 Fuel availability limitation or different atmospheric circulation pattern?

The iron content in Southern Chile marine sediments reconstructs rainfall changes due to latitudinal shifts of the Southern Westerlies during the Holocene (Lamy et al., 2001, see also Fig.4 in this paper). This reconstruction is consistent with

regional terrestrial paleoclimate data-sets (Villagran et al., 1990; Lamy et al., 2001) and indicates more arid conditions from 7700 to 4000 yr BP, followed by a general increase in rainfall from 4000 yr BP to the present day. This finding is consistent with the hypothesis that fire activity in Southern America is mainly driven by fuel availability, as previously discussed (Iglesias and Whitlock, 2014 and Iglesias et al. 2014). Interestingly, as shown in Fig.4, levoglucosan matches the iron record reported by Lamy et al. (2001). Bivariate analysis indicated a significant negative association between levoglucosan (log-transformed; outliers excluded) and iron records (log-transformed) (r (95% CI) =-0.183 (-0.277, -0.083)), suggesting that when arid conditions characterized southern South America, the levoglucosan signal was low and started to increase when the Southern Westerlies strengthened and shifted equatorward, bringing more humid conditions to the continent.

3.5 Megafires

The attribution of the levoglucosan spikes to megafire events is difficult to assess since atmospheric transport and stability may alter the signal (Section 3.1), resulting in an amplification of modest fires. Peaks in charcoal records can either be intense local fire events or can result from an increase in short-term transport. For example, the higher charcoal signals observed in Laguna Padre Laguna (Argentina) between ~1500 and ~2000 yr BP and at Laguna Zeta (Argentina) between ~2000 and ~2500 yr BP (Iglesias and Whitlock, 2014) are consistent with the spikes observed in the levoglucosan record, as well as intense fire episodes that were also recorded between ~2100 and ~2300 yr BP in the Wingecarribee Swamp (Southern Australia (de Montford, (2008); ID site=857 in GCD) and at 2160 yr BP in Eweburn Bog (New Zealand, (Ogden et al. 1998)); ID site=441 in GCD). A possible correspondence with levoglucosan and these individual charcoal spikes is only speculative. Conversely, we could interpret these anomalous spikes as a result of favourable transport, rather than assigning them to specific mega-fire events. Comparing trends in long-term fire activity, as identified by levoglucosan records and charcoal syntheses is more indicative of changes in biomass burning as opposed to comparing individual, likely localized, events.

3.6 Fire and major climatic parameters

Although the evaluation of a single levoglucosan spike is highly uncertain, the long-term change in fire activity may be linked to major climatic variables (Fig.4). As observed, the levoglucosan record is characterized by two main features: (a) a long-term increase (with higher rates starting at~4000 yr BP) and (b) higher peaks between 1500 and 2500 yr BP.

The long-term increase in levoglucosan is consistent with a general increase in fire activity in the Southern Hemisphere from 4000 yr BP to the present (Fig. 4). A significant positive correlation with solar radiation in December at 30°S (from Berger, 1978) (r (95% CI) =0.458(0.362, 0.562); see also Fig.5) is depicted by a long-term increase up to approximately 1000 yr BP. However, Southern Hemisphere temperature (r (95% CI) =-0.272 (-0.428, -0.147); see also Fig.5) substantially decreases between ~1500 and ~2500 yr BP, where this decrease is not evident in either the solar irradiance or greenhouse gas concentrations (Fig. 4). Carbon dioxide increases fairly steadily from the Mid-Late Holocene onward in the Taylor Dome ice core (Indermühle et al. 1999; Fig. 4), while the general levoglucosan trend increases beginning ~4000 yr BP, yet includes a

major dip in fire activity around ~2800 yr BP (r (95% CI) =0.455 (0.357 0.576)). This difference is to be expected, as CO_2 in ice cores represent a globally-mixed signal while levoglucosan in polar ice cores is a regional to semi-hemispheric signal (e.g. Zennaro et al., 2014 and 2015). Strong El Niño Southern Oscillation (ENSO) events and periods affect high latitude Southern Hemisphere climate by increasing mean Southern Hemisphere air temperature (Schneider and Steig, 2008), expanding sea ice east of the Antarctic Peninsula (Mayewski et al., 2017 and references therein), and delivering snowfall to East Antarctica (Roberts et al., 2015). The strengthening of ENSO beginning around ~5000 yr BP increased short-term fluctuations in Patagonian vegetation (Moy et al., 2002; Iglesias et al., 2014) and increased precipitation near northern Patagonia as evidenced by the iron record in marine sediments (Lamy et al., 2001; Lamy et al., 2004). The iron record and changes in ENSO strength (log-transformed) also correlate with the Talos Dome levoglucosan record (log-transformed) (r (95% CI) = 0.172 (0.092, 0.354)), although to a lesser extent than solar radiation and greenhouse gases. This correlation suggests that the impacts of ENSO on regional circulation and the amount of available burned vegetation in Patagonia, contributing to affect Southern Hemisphere fire activity, as recorded in the Talos Dome ice core.

Variable and generally wet conditions prevailed in South America after 4000 yr BP, resulting in a more available biomass and increased fire events. However, biomass burning increased even more when, at ~ 2500 yr BP, Patagonia became drier yet still contained an abundance of vegetation from the previous humid period (Iglesias et al. 2014). This 1000-year peak in levoglucosan occurs at the same time as a local decrease in temperature between 1500 and 2500 yr BP. As it is unlikely that decreased temperatures triggered an increase in fire activity, we propose three possible explanations: (i) aerosol feedbacks (ii) different atmospheric transport and (iii) interplay between climatic factors.

(i) Studies of long-term fire activity variations since the Last Glacial Maximum and through the Holocene demonstrate that temperature is the main driver of fire activity over a global scale, while the precipitation-evaporation (P-E) balance is the second-most important factor (Daniau et al 2012). The net effect of both temperature and P-E strongly depend on the initial conditions and fuel availability. Fire does not respond linearly to temperature, and an increase in temperature does not directly result in more fire under warm rather than cold conditions (Daniau et al. 2012). The relation between fire and P-E is unimodal, where an increase in P-E under dry/moist conditions leads to an increase/decrease in fire. This long-term observation contrasts with our record, where an increase in fire activity coincides with a net decrease in temperature. It must be noted, however, that our observations do not represent a global signature and the relatively cold period was limited to only approximately 1000 years in the framework of the current interglacial. Moreover, fires emit both large quantities of CO_2 and particulates. While CO_2 warms the atmosphere, many aerosols can decrease temperature by shielding the solar radiation that reaches the surface. As a net result, at least over local to regional scales, aerosol emissions lead to a cooling effect, as observed in the Kuwait oil fires, where a reduction in ground-level temperature resulted from the absorption of solar radiation by the smoke (Bakan et al. 1991). However, although aerosol emissions can impact the surface temperature, this hypothesis is very difficult to support for a time-period of ~1000 years and over such a large spatial scale. (ii) The

temperature decrease between 1500 and 2500 yr BP may coincide with less stable atmospheric circulation patterns. As previously discussed, a possible increase in transport efficiency to the Talos Dome site could at least partially explain the anomalous increase in levoglucosan signal between 1500 and 2500 yr BP. We cannot ignore that atmospheric transport may have played a role in our levoglucosan sequence recorded at the Talos Dome site. Additional insights may be achieved using more internal Antarctic cores where local transport effects are potentially minimized, such as the EPICA Dome C ice core. (iii) The cooling period coincides with a shift to drier conditions in South America characterized by initial conditions with available fuel, creating favourable conditions for fire.

While hypothesis (i) is more disputable, in our view a possible combination of (ii) and (iii) may more successfully support the anomalous increase in levoglucosan centred at ~2000 yr BP. It is also interesting to note that similar features occur in the NEEM (Greenland) levoglucosan record, with an increase in fire at ~4000 yr BP (a) and an unusual peak in biomass burning at ~2500 yr BP (b) (see Fig.1 in Zennaro et al. 2015). This coincidence may suggest a more global description of the fire history that should be further investigated in more detail.

3.7 Possible anthropogenic influence?

This Talos Dome levoglucosan record is limited by the available ice core samples and does not extend to the present, thereby not covering the time period of major regional population shifts, yet covering the time period of the advent of agriculture on a global scale. Several studies examine the anthropogenic influence on global to regional fire activity due to agriculture in the Mid-Late Holocene (e.g. Broeker and Stocker,2006; Joos et al., 2004; Singarayer et al., 2011; Mitchell et al., 2013, Kaplan et al., 2009 and 2011). Although the first agricultural practices in South America likely appeared ~10,000 yr BP, the development of agriculture in the SH was limited with respect to the NH (Martin and Sauerborn, 2013). The Hyde 3.2 database demonstrates that in the possible fire source regions of Patagonia and Southern Africa the demographic density was between 1-5 inhabitant per km^2 and even less than 1 inhabitants/km^2 in Australia until ~ 1000 yr BP (Goldewijk, 2016). Human populations in the region fluctuated wildly from 1000 yr BP to present due to European settlement and the associated decimation of indigenous populations. The human impact on fire activity is not directly proportional to population density, where small bands of humans can substantially change their environment, such as in the case of rapid burning of New Zealand forests (McWethy et al., 2010 and 2014). However, the Talos Dome ice core record from 6000 yr BP to ~750 yr BP currently does not provide clear evidence that the fire record may be strongly affected by anthropogenic activities during the Mid-Late Holocene, although we cannot exclude at least a partial influence.

4 Conclusion

We reconstructed the high latitude Southern Hemisphere fire history by using the specific biomarker levoglucosan and potassium in ice cores from the TALDICE during the Mid-Late Holocene (750-6000 yr BP). Potassium was analyzed in

order to provide a more complete biomass burning record through the comparison with another fire proxy. The comparison between levoglucosan and the multi-source potassium suggested that potassium can increase the information obtained from a single fire proxy, although it is often best used in conjunction with other biomass burning markers. The levoglucosan record is characterized by a long-term increase with higher rates starting at~4000 yr BP and higher peaks between 1500 and 2500 yr BP. The possible influence of atmospheric transport was considered by incorporating dust and charcoal synthesis. We demonstrated that, at least in part, the levoglucosan record at Talos Dome may be affected by a different circulation pathway. Moreover, comparisons with regional charcoal syntheses suggested a possible higher contribution from Patagonian fires rather than Australian biomass burning. The long-term trend of levoglucosan was considered in conjunction with other climatic parameters, such as carbon dioxide, solar radiation, ENSO activity, SH temperature synthesis, biomass availability, P-E balance, etc. In order to explain the levoglucosan long-term trend, especially focusing on the anomalous increase centred at ~ 2000 yr BP, we proposed three main explanations: (i) aerosol feedback (ii) different atmospheric transport and (iii) interplay between climatic factors. However, the combination of (ii) and (iii), rather than (i), was interpreted as a more conceivable explanation for this anomalous increase of fire activity, where a generalized cooling period in the Southern Hemisphere coincides with a shift to drier conditions in South America, characterized by initial conditions with available fuel, that, in turn, created favourable conditions for fire. Finally, although we cannot exclude a possible anthropogenic influence on the SH fire history, we did not find clear evidence for supporting this occurrence.

Acknowledgments

The research has received funding from the This work was financially supported by the Early Human Impact ERC Advanced Grant of the European Commission's VII Framework Program, grant number 267696, by Programma Nazionale di Ricerche in Antartide (PNRA) Grant: PEA 2013/B2.05, and by European Union Marie Curie IIF Fellowship (MIF1-CT-2006-039529, TDICOSO) within the VII Framework Program. The Talos Dome Ice Core Project (TALDICE), a joint European programme, is funded by national contributions from Italy, France, Germany, Switzerland and the United Kingdom. Primary logistical support was provided by PNRA at Talos Dome. The authors gratefully acknowledge the help of ELGA LabWater in providing the PURELAB Pulse and PURELAB Flex that produced the ultrapure water used in these experiments. The authors thank Barbara Delmonte for providing raw data of Talos Dome's dust.

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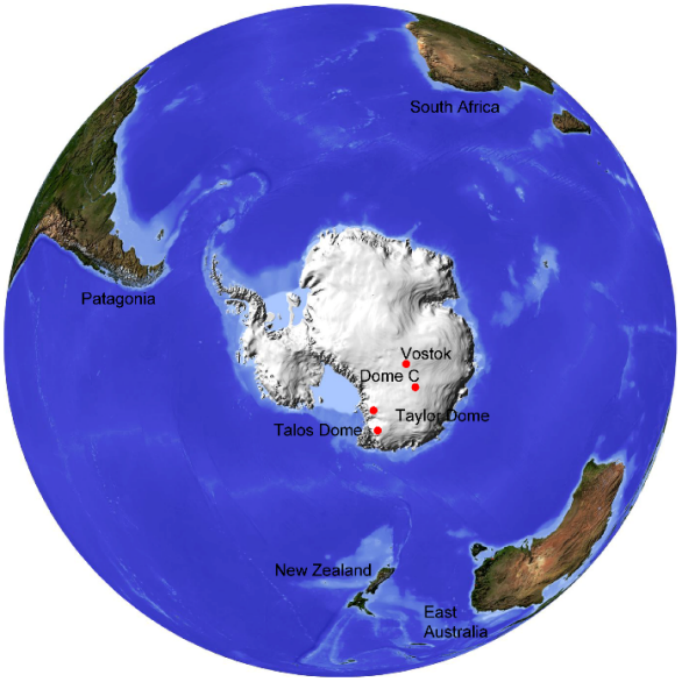

Figure 1: Map of the Antarctic region. Antarctic sites of Talos Dome, Vostok, Taylor Dome and Dome C, mentioned in this paper, are indicated.

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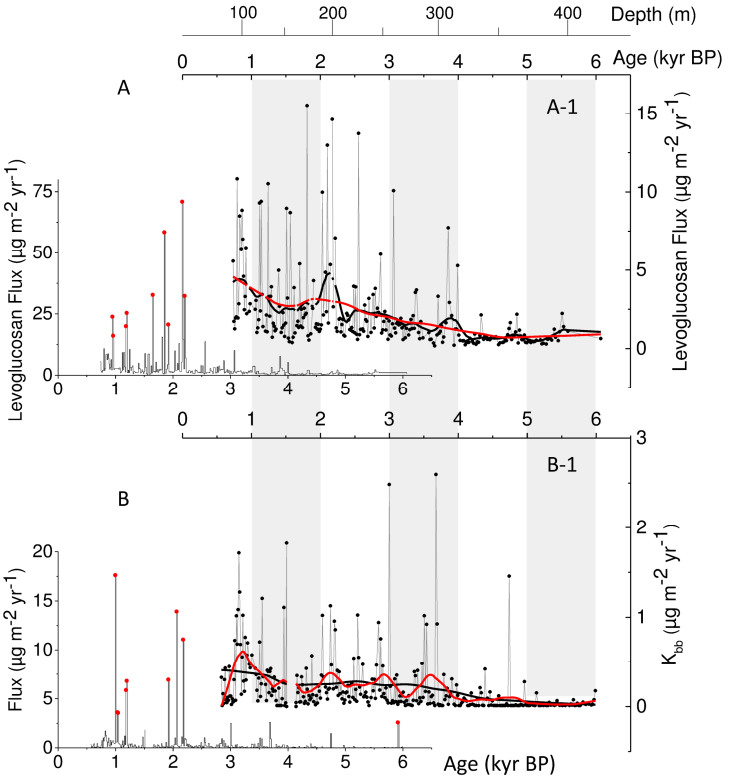

Figure 2: Complete levoglucosan (A) and K_{bb}(B) flux record obtained from TALDICE, where red dots represents anomalies following Cook's distance criterion. Levoglucosan (A-1) and K_{bb} (B-1) record without anomalies (LOWESS smoothing with SPAN parameter 0.15 (black line) and 0.3 (red line).

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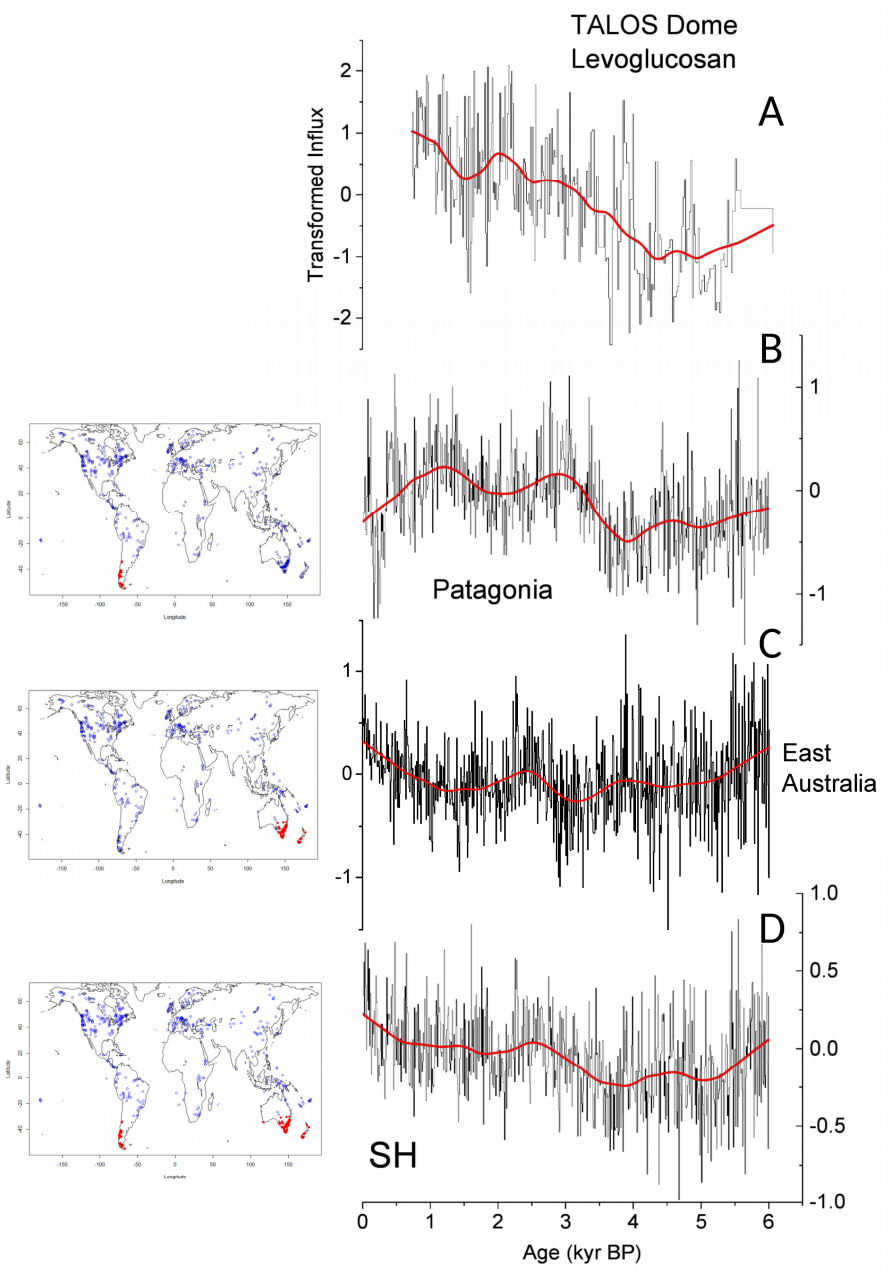

Figure 3: Box-Cox transformed levoglucosan influx and charcoal synthesis (bins= 20 years) from Patagonia, East Australia and Southern Hemisphere synthesis from GCD. LOWESS smoothing (in red) with SPAN parameters 0.2.

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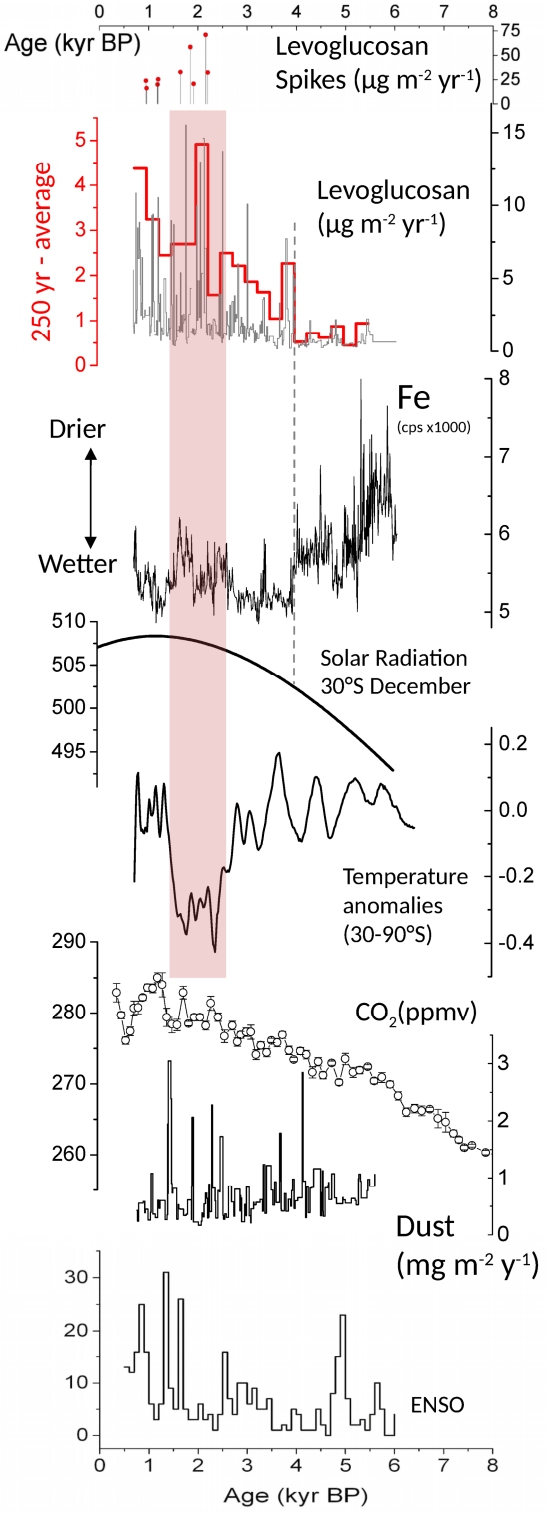

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Figure 4: Levoglucosan flux and 250 yr average window (red line), Fe content in marine sediments from SouthernChile (from Lamy et al. 2001); 30°S December solar radiation (from Berger and Loutre 1991), Temperature anomalies between 30° and 90° S (Marcott et al. 2013), CO_2 from Taylor Dome (Indermuhle et al. 1999), Dust from Talos Dome (Albani et al. 2002) and ENSO 100 yr frequency (Moy et al. 2002).

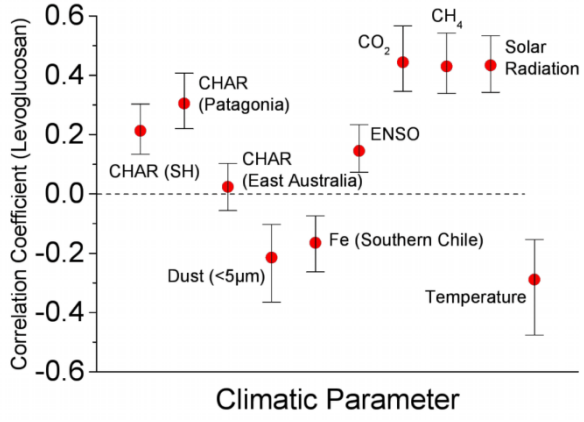

Figure 5: Correlation between levoglucosan recorded at Talos Dome (log transformed) and charcoal synthesis (CHAR) from Patagonia, East Australia and Southern Hemisphere, dust particles at Talos Dome (from Albani et al. 2012), Iron record from Southern Chile (Lamy et al. 2001), Carbon dioxide from Taylor Dome (Indermuhle et al. 1999), ENSO 100 yr frequency (Moy et al. 2002), 30°S December solar radiation (from Berger and Loutre 1991). (95% confidence interval (CI)).

Clim. Past Discuss., https://doi.org/10.5194/cp-2017-158
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Table 1: Levoglucosan and K_{bb} peak anomalies. Levoglucosan and K_{bb} values are minmax scaled. n.a. = not available ; n.s. = this values were not a anomalies.

Group	TD Sample	Age yr BP	Levoglucosan	K_{bb}	Match
I	108	945	0.34	**n.s.**	No
	109	957	0.23	**n.a**	
	112	994	**n.a**	1.00	
	115	1031	**n.a**	0.21	
	116	1043	**n.a**	0.20	
	127	1180	0.28	0.33	**Yes**
	128	1193	0.36	0.39	**Yes**
II	163	1647	0.46	**n.s.**	No
	178	1849	0.82	**n.s.**	No
III-a	183	1916	0.29	0.39	**Yes**
	193	2060	**n.a**	0.79	
III-b	200	2160	1.00	**n.s.**	No
	201	2174	**n.s.**	0.63	No
	203	2202	0.46	**n.s.**	No
	299	5911	**n.a**	0.15	