

Interactive comment on “Fire in ice: two millennia of Northern Hemisphere fire history from the Greenland NEEM ice core” by P. Zennaro et al.

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Interactive comment on “Fire in ice: two millennia of Northern Hemisphere fire history from the Greenland NEEM ice core”.

by P. Zennaro et al.

Thank you for the opportunity to publish my work in Climate of the Past. We appreciate the detailed comments from the referees. We followed most of the reviewer comments as suggested, unless otherwise noted. For the changes where we differ from the suggestions, we include explanations of our reasoning. Following the reviewers’ suggestions allowed us to improve the data interpretation and the quality and readability of the manuscript. Here we present our answers to each point outlined by the three referees,

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and we attach a new version of the work “Fire in ice: Two millennia of boreal forest fire history from the Greenland NEEM ice core”. Piero Zennaro

Anonymous Referee #1 Received and published: 17 March 2014

Zennaro and colleagues have analyzed multiple fire proxies for the past 2000 years using a Greenland ice core. They report the variability and compare this with what is in the literature and with climate data. This multi-proxy approach is an important step forward and the findings are very suitable for CP. It looks the manuscript was submitted under time pressure but it is still well written. My main comments are on the structure of the paper where I think the authors have to rethink it to avoid the discussion becoming overly long. For the rest I have no major concerns.

1) 811-25 "Biomass burning emits up to 50% as much CO₂ as fossil fuel combustion (Bowman et al., 2009) thereby affecting the climate system". Fires emit CO₂ that has been sequestered and are thus not a net contributor to enhanced CO₂ levels as fossil fuel emissions are, consider rephrasing.

We rephrased the sentence by changing “climate system” to “carbon cycle”. Lines 3 - 4 "Biomass burning plays an important role in the carbon cycle as it emits up to 50% as much CO₂ as fossil fuel combustion (Bowman et al., 2009)."

2) 812-8: "still not well defined but are in the range of $+0.03 \pm 0.12 \text{ W m}^{-2}$ (IPCC, 2007)". What does AR5 say?

We updated the radiative forcing range for biomass burning according to the AR5. In the AR5 the net contribution slightly decreases, while the error range increases.

Lines 10 - 11 “Estimates of the radiative forcing of combined biomass burning aerosols are still not well defined but are in the range of $+0.00 \pm 0.20 \text{ W m}^{-2}$ (IPCC, 2013).”

3) 812-14: "Precipitation affects fuel flammability, where conditions must be wet enough to allow biomass to grow, and dry enough to allow combustion (Pyne, 2001)." consider rephrasing

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Thank you for your suggestion, in order to be more precise we changed the sentence as follows:

Lines 16 - 19 “Precipitation decreases fuel flammability (Pechony and Shindell, 2010) but biomass burning is highest at intermediate moisture levels (Daniau et al., 2012), a balance must therefore be struck between climatic conditions that are wet enough to allow biomass to grow, but dry enough to allow combustion (Pyne, 2001).”

4) 812-20: How do household fires affect global fire activity? By reducing coarse woody debris on the ground?

We changed the sentence, since household fires are not increasingly altering global fire activity.

Lines 21 - 23 “Human activities have also had an influence on biomass burning trends (Marlon et al., 2008) through deforestation and slash-and-burn agricultural practices (FAO, 2007); practices which may have affected fire regimes during the entire past millennia (Bowman et al., 2009; Kaplan et al., 2011; Ruddiman, 2003).” Bowman, D., Balch, J. K., Artaxo, P., Bond, W. J., Carlson, J. M., Cochrane, M. A., D’Antonio, C. M., DeFries, R. S., Doyle, J. C., Harrison, S. P., Johnston, F. H., Keeley, J. E., Krawchuk, M. A., Kull, C. A., Marston, J. B., Moritz, M. A., Prentice, I. C., Roos, C. I., Scott, A. C., Swetnam, T. W., van der Werf, G. R., and Pyne, S. J.: Fire in the Earth System, *Science*, 324, 481-484, 10.1126/science.1163886, 2009. Kaplan, J. O., Krumhardt, K. M., Ellis, E. C., Ruddiman, W. F., Lemmen, C., and Goldewijk, K. K.: Holocene carbon emissions as a result of anthropogenic land cover change, *Holocene*, 21, 775-791, 10.1177/0959683610386983, 2011. Ruddiman, W. F.: The anthropogenic greenhouse era began thousands of years ago, *Clim. Change*, 61, 261-293, 10.1023/B:CLIM.0000004577.17928.fa, 2003.

5) 812-23: Century -> century

We changed the capital letter, at line 25

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“The observable impact of humans on fire regimes differs by geographic region (McWethy et al., 2009; Marlon et al., 2012; Power et al., 2008) but over the course of the 20th century human activity has begun to influence the global fire regime more than natural causes (Marlon et al., 2008; Pechony and Shindell, 2010).”

6) 812-29: are have -> have

We deleted the word “are”, at line 82 - 84 “Some proxies are specific as they are produced solely from biomass burning, such as charcoal in sediment cores, but most others have multiple potential sources other than wildfires (e.g., coal burning, volcanic eruptions or biogenic emissions).”

7) 813-11: that occurs during -> is emitted from

Thank you for your suggestions. At lines 110 - 111 we reworded the sentence and changed “occurs during” with “is formed during”.

“BC is emitted during incomplete combustion of fossil and biofuels in both naturally and anthropogenically created fires (Preston and Schmidt, 2006; McConnell et al., 2007).”

8) In the introduction there is quite some space reserved for detailing issues with fire proxies other than levoglucosan. I understand those issues but it would be nicer if you mention them but at the same time explain what we have learned from them (instead of why they are not as good as levoglucosan) and what information is lacking that this paper can contribute

We have substantially revised the introduction following the suggestions of Reviewer 2. Please see our response to Reviewer 2 to obtain all details. Generally, we now present a more balanced view of all proxies where we discuss both their positive and negative attributes.

9) 813-29: due to their short lifetimes and efficient removal processes -> isn't the short lifetime the result of efficient removal processes (as with BC) or are they also chemically broken down?

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Short lifetimes and removal processes, such as wet-deposition, of these indicators result in a non homogeneous distribution around the globe (or at hemispheric scales). Of course the short lifetime is the result of efficient removal processes, but the short lifetime of levoglucosan and ammonium is also the result of their instability with respect to the oxidizing character of the atmosphere. For example nitrate (NO_3^-) can be produced from the oxidation of ammonium (NH_4^+), or levoglucosan may be oxidised by radicals, and particularly with hydroxyl radicals.

10) 814-7: [high latitudes/hemisphere]?

It is not still clear which are exactly the source regions of levoglucosan reaching Greenland. A finding of this paper is that forests in the Asian regions contributed to biomass burning plumes, in particular during dry periods. Boreal forests at high latitudes are likely the major sources of biomass burning. We removed the square brackets and the word “hemisphere”, as they may confuse the reader.

Lines 153 -154 “Here, we use a multi-proxy approach to reconstructing biomass burning in the northern high latitudes during the past 2000 years.”

Methods - I cannot comment on the methods used but the authors have an excellent reputation

11) 2.3: I think it would be better to explain the statistical tools you used than to explain why you used them. Or do both

We explained the statistical tools we used, and reasons for their use, in the Supplementary Information. However, we now briefly report the statistical tools we used in the main article in the following lines (lines 246 -250):

”In summary, to highlight long-term fire changes we followed the following procedure (details are included in the Supplementary Information): Removing spikes above the fixed threshold $3\text{rd_Q} + 1.5 \times \text{IR Z-score}$ transformation LOWESS (Locally Weighted Scatterplot Smoothing) model”

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12) 819-7: (CITE)?

I am sorry for this problem. The missing citations were added in the final version of the paper.

Lines 281 -283 “Increased wind speeds result in greater dust concentrations and larger particles in ice cores (Ruth et al., 2003; Fischer et al., 2007).” Fischer, H., Siggaard-Andersen, M. L., Ruth, U., Rothlisberger, R., and Wolff, E.: Glacial/interglacial changes in mineral dust and sea-salt records in polar ice cores: Sources, transport, and deposition, *Rev. Geophys.*, 45, RG1002, 10.1029/2005rg000192, 2007. Ruth, U., Wagenbach, D., Steffensen, J. P., and Bigler, M.: Continuous record of microparticle concentration and size distribution in the central Greenland NGRIP ice core during the last glacial period, *J. Geophys. Res.-Atmos.*, 108, 10.1029/2002jd002376, 2003.

13) 820-12: Supplement -> Supplement)

Thank you for noting the missing parentheses.

14) 824-23: This is really a shining example of the strength of these ice core records over more local-scale measurements. This is new information we are eager for, please highlight these kind of findings more in the text and conclusions.

We are happy to provide information for which the scientific community is eager. We modified the conclusion as you suggested, at lines 646 – 657.

“Our results demonstrate the greatest amount of decadal-scale fire activity during the mid-1600s. We conclude that the 1500 – 1700 CE maximum in fire activity is due to increased boreal forest fires, caused by extensive dry conditions in the Asian region. Fires are concurrent with known extensive droughts and monsoon failures, and levoglucosan concentrations are greater than during the last 150 years when anthropogenic land-clearing rates were the highest in history. This evidence suggests that climate variability has influenced boreal forest fires more than anthropogenic activity over the past millennia in the boreal regions that supply biomass burning related species to

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Greenland. However, climate change and anthropogenic activity may increase future boreal fire activity, and have the potential to exceed the fire activity during the mid-1600s. Warmer, drier summers and increased deadwood availability due to past fire suppression, as well as insect outbreaks (Kurz et al., 2008; Wolken et al., 2011), and increased tree mortality from drought (van Mantgem et al., 2009) may amplify current and future fires. Increasing forest mortality in a warming climate (Anderegg et al., 2013) results in greater fuel availability with the potential to intensify future boreal fire activity.”

15) 4.2: I think it is worth considering moving the description of the charcoal and CH4 isotope analyses to the introduction to tell the reader what is known. Then 4.2 can be substantially shortened to show how the new data compares with what is known

Thank you for the suggestion, we moved the following text from Chapter 4.2 to the Introduction, and we merged Chapter 4.2.1 and 4.2.2 into Chapter 4.2.

We moved the following from Chapter 4.2.1 to the Introduction, at lines 139 – 150.

“Charcoal deposited both in non-polar ice and sediment cores can also help reconstruct past fire activity. The Global Charcoal Database (GCD) compiles individual charcoal records from terrestrial and marine sediment cores (Fig. S6) into global or regional reconstructions that provide a benchmark against which other biomass burning records can be compared (Power et al., 2010; Marlon et al., 2008). The charcoal database covers all climatic zones and all major biomes, but data for grasslands and dry shrublands, where low woody biomass limits charcoal production, are limited due to a lack of suitable sampling sites (Marlon et al., 2008). These vegetation types emit large quantities of levoglucosan when burned (Iinuma et al., 2007; Gao et al., 2003) resulting in a possible offset between charcoal and levoglucosan data. Many charcoal sampling sites are located in the US and Europe, and a limited number of GCD sites currently exist in northern Asia (Fig. S6). As levoglucosan and BC can be transported hundreds to thousands of kilometers (Mochida et al., 2010), they complement GCD reconstructions that record combustion within tens of km of the sampling sites, and provide fire

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activity data for regions where charcoal records do not exist. ”

We moved the following sentences from Chapter 4.2.2 to the Introduction, lines 107 - 113.

“The isotopic ratio of atmospheric gases, such as methane (CH₄) and carbon monoxide (CO) in Antarctic (Ferretti et al., 2005; Wang et al., 2010) and Arctic (Sapart et al., 2012) ice cores has also been used as fire proxy but such ratios are also not solely products of biomass burning (Fischer et al., 2008). The isotopic composition of methane ($\delta^{13}\text{C}$ of CH₄) in ice cores differentiates biogenic from pyrogenic sources (Sapart et al., 2012). Methane is stable in the atmosphere for longer than the atmospheric exchange time and therefore is globally distributed. Hence $\delta^{13}\text{C}$ of CH₄ variations are impacted by sources distributed all over the world (Blunier et al., 2007).”

16) 825-21: This is redundant. As you see, I am trying to find ways to shorten the discussion. Right now it is 2/3 of the written text and I think the paper would be easier to read if you restructure it. More to the introduction (for the reader who wants to know what the current state of the science is) and less in the discussion (for the reader who wants to know how the current work agrees or disagrees with what is known)

We agree with your suggestion to move parts of the discussion into the introduction (as detailed in the previous point and in our reponse to Reviewer 2). Generally, we moved many informative parts regarding each proxy from the discussion to the introduction.

17) 827: think this is the third time that atmospheric transport times are discussed, please introduce once and then refer to it if needed I must say that the whole discussion is difficult to follow. There is so much text detailing recorded drought periods etc, and peaks are related to droughts, they are related to higher temperatures, etc. It might be better to highlight the main findings and leave out all the detailed information (year numbers etc) but refer to Table 2.

In section 4.1.1 we argue that levoglucosan variability is not caused by rapid changes

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in transport patterns by analysing correlations with crustal ions. We discussed global transport patterns in section 4.3, in order to assess the likely geographic sources of levoglucosan reaching Greenland. Even if these two sections contain information about “transport”, the topic of these sections differs. Thus we prefer keep these sections separated. However, we removed the sentence “The closest source regions for the Greenland dust are boreal Canada and boreal Russia, which are similar landscapes with similar availability of entrainable dust.” from Chapter 2.2 (Statistics) in order to reduce redundant text.

Regarding Chapter 4.3, we understand it may be long-winded. We prefer to run this risk rather than delete or move text, which may therefore reduce readability. However we divided Chapter 4.3 in two parts: 4.3.1 Fuel Loads and Circulation Patterns (Lines 427) and 4.3.2 Levoglucosan Sources (Lines 472). We hope this strategy helps the reader.

We have been impressed by the high temporal resolution links between the well-dated NEEM fires and drought reconstructions in Asia. We looked for alternative methods to refer to both levoglucosan and droughts without the use of years in the main text. Unfortunately, our attempts failed and we think this format remains the best choice for the reader.

18) 827-16: North America and Siberian forests -> North American and Siberian forests

We changed the sentence as suggested, at line 473 – 476. “Back-trajectory and model analyses suggest that North American and Siberian forests are the dominant sources of biomass burning aerosols. A first-order approximation of aerosol concentrations along a trajectory is function of time where aerosol concentrations decrease exponentially with transport time (Fischer et al., 2007; Hansson, 1994).”

â– Anonymous Referee #2 Received and published: 28 March 2014

Zennaro et al. report ice core records of ammonium, levoglucosan and BC from

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the northern Greenland site of NEEM covering the last 2000 yrs. The topic of the manuscript is highly relevant with the scopes of the CP journal: interactions between climate and occurrence of boreal forest fires over the past. However, as detailed below, as it stands, the manuscript has too many weak aspects (structure of the paper, incomplete presentation of the state of the art sometimes missing key previous works, paragraphs sometimes very poorly written, some statements weakly supported). I encourage the authors to take time to improve the manuscript with major revisions specially concerning the structure of the paper, a more accurate presentation of the state of the art, and more in depth comparison of available high-resolution chemical ice core records for Greenland (previous works at Summit and D4, as well as the high-resolution ammonium record at NEEM).

1) Title: The title is too vague: Please specify that your records concern boreal forest fires (not Northern Hemisphere) and indicate the used proxies (ammonium, levoglucosan, and BC).

We appreciate your suggestion and we present the new title:

“Fire in ice: Two millennia of boreal forest fire history from the Greenland NEEM ice core”

2) Introduction : The introduction should be refocused on (1) boreal fires, (2) a more accurate presentation of previous studies using chemical ice core records to reconstruct past fire activities.

We appreciate your suggestions to improve the introduction and the presentation of the “State of Art” regarding proxies for biomass burning. We added the new sub-chapter “1.1 Boreal Forest Fires” (line 28) into the Introduction, and we reworded part of introduction into the sub-chapter “1.2 Biomass Burning Proxies” (line 78).

“1.1 Boreal Forest Fires The circumpolar boreal zone contains about 30% of world’s forests with half of the world’s forest carbon and about 30% of world’s terrestrial carbon.

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Any change to the boreal forest carbon balance may therefore considerably affect the global atmospheric CO₂ (Conard et al., 2002). Annual area burned (extent) and the global importance of fires in the boreal zone have often been underestimated (Conard et al., 2002). However, it is estimated that between 50,000 and 200,000 km² of boreal forests burn every year, mainly in Northern Asia and North America (de Groot et al., 2013; Wooster and Zhang, 2004). On average boreal and temperate vegetation fires represent 4% of global biomass burning and up to 12% during extreme events, when about 20% of global BC may be produced (Lavoue et al., 2000). Thus, boreal forest fires are an important source of air pollutants throughout the Arctic, and a major font of BC (Quinn et al., 2008; Lavoue et al., 2000). Due to the light-absorbing properties of BC and the associated decrease in surface snow albedo, boreal forest fire BC has a significant radiative impact on the Arctic atmosphere (Quinn et al., 2008; Flanner et al., 2007; Hansen and Nazarenko, 2004).

North American and Eurasian boreal fire regimes differ due to distinctive fuel, weather, and fire ecology (Wooster and Zhang, 2004; de Groot et al., 2013). Mean fire intensity and fuel consumption is higher in North America than in Russia, even though the spatial extent of forests is greater in Russia (Wooster and Zhang, 2004; de Groot et al., 2013). Different fire regimes suggest a dominance of crown fires in North America and surface fires in Russia (Wooster and Zhang, 2004; de Groot et al., 2013). Accordingly, Russian fires may burn less fuel than North American fires (de Groot et al., 2013) and thus emit less pyro-products into the atmosphere per unit of area burned (Wooster and Zhang, 2004). Over the period 2001-2007, the average size of large fires in Central Siberia was estimated as approximately 13.1 km², with an average 18,900 km² burned each year at an average fire intensity of 4858 kW m⁻¹ (de Groot et al., 2013). In western Canada the annual average size of individual large fires is 59.3 km², with a total of 5,600 km² burned annually and an average fire intensity of 6047 kW m⁻¹ (de Groot et al., 2013). The carbon emission rate is 53% higher in the Canadian study area, due to higher pre-burn forest floor fuel loads and higher fuel consumption by crown fires. However, the Russian study area had much higher total carbon emissions due

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to the greater annual area burned (de Groot et al., 2013). During the major fire year of 2004, an additional 31,000 km² of Canadian boreal forests and approximately 27,000 km² of Alaskan forests burned (Stohl et al., 2006) resulting in strong Pan-Arctic enhancements of light absorbing aerosols (Stohl et al., 2006). This strong burning year in Canada and Alaska is coincident with a pronounced decrease in snow albedo at Summit, Greenland due to the deposition of dark particles on the snow surface. Boreal forest fires in Siberia may have a larger impact on total global fire emissions than those in North America due to the larger Siberian burn area (Stohl, 2006; Stohl et al., 2006). Siberian forest fires are a major extratropical source of carbon, in particular carbon monoxide and black carbon (Lavoue et al., 2000). Russian boreal forests, where the majority are located in Siberia, accounted for 14–20% of average annual global carbon emissions from forest fires in 1998 (Conard et al., 2002). However, little is known about the emission, transport and deposition of BC from Siberian forest fires to the Arctic. The characterization of Siberian fire plumes is uncertain, due to currently undefined biomass fraction consumed, combustion efficiency, plume injection heights and emission ratios (Paris et al., 2009). Models are sensitive to the conditions and types of fires, such as the injection height of fire emissions (Turquety et al., 2007), and the lack of data about Siberian fires introduces large potential errors in estimating the impact of boreal wildfires on the atmospheric composition (Conard et al., 2002).

Summer atmospheric conditions favour enhanced wet and dry BC deposition in the Arctic (Stohl, 2006). Generoso et al. (2007) determined that the 2003 Russian fires contributed to approximately 40–56% of the total BC mass deposited north of 75 °N. In fact, model results indicate that boreal forest fire sources, especially Siberian fires, are the dominate summer-time source of BC reaching the Arctic, exceeding all contributions from anthropogenic sources (Stohl, 2006). Siberian fires, and northern Eurasia sources in general, can reach the Arctic more easily than emissions from other regions of similar latitude (Stohl, 2006; Stohl et al., 2006).

1.2 Biomass Burning Proxies There are many potential indicators or proxies of past

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fire activity, such as altered products of plant combustion (e.g. charcoal, BC), partially combusted biological material (e.g. fire scars in tree rings), or chemical markers directly produced and volatilised during vegetation combustion (e.g. resin acids, polycyclic aromatic hydrocarbons) (Conedera et al., 2009). Some proxies are specific as they are produced solely from biomass burning, such as charcoal in sediment cores, but most others have multiple potential sources other than wildfires (e.g., coal burning, volcanic eruptions or biogenic emissions). Ice cores from polar regions are widely used to reconstruct detailed climate records over the past hundreds of thousands of years, and thus are powerful tools in paleoclimate research. Examining forest fires in ice cores started with the pioneering work of Legrand et al. (1992), which demonstrates that the concentration of ammonium and organic acids including light carboxylic acids, significantly increase above background levels in Greenland ice layers during forest fire events. Among possible light carboxylic acids, formate and oxalate are the most useful to trace fires in Greenland ice cores (Jaffrezo et al., 1998; Legrand et al., 1992; Dibb et al., 1996; Kehrwald et al., 2012). The subsequent work of Legrand and DeAngelis (1996) demonstrates that high latitude biomass burning episodes increase light weight carboxylic acid concentrations by 20 – 30 % in Greenland ice. In addition to strong increases of ammonium, formate and oxalate concentrations, Savarino and Legrand (1998) found slight increases of nitrate coincident with fire events recorded in Greenland ice. However, lightweight carboxylic acids and major ions (potassium (K⁺), ammonium (NH₄⁺) and nitrate (NO₃⁻)) are not specific for biomass burning since they can be emitted by multiple sources. K⁺ and NH₄⁺, for example, are also emitted from numerous other sources and thus must be corrected accordingly in order to provide source-specific information (Savarino and Legrand, 1998). Of the available ionic proxies, K⁺ is less useful due to marine and terrestrial sources, even if corrected to remove these contributions (e.g. non-sea-salt and non-crustal K⁺) (Legrand and DeAngelis, 1996; Savarino and Legrand, 1998) Statistical attempts to recognize forest fire signals from multiple ice core data sets use Empirical Orthogonal Function (EOF) analysis or Principal Component Analysis (PCA), which identify chemical associations between

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pyro-ion records and differentiate between sources and transport characteristics (Yalcin et al., 2006; Eichler et al., 2011). NH_4^+ , NO_3^- , and organic acids in Greenland ice are often associated with biomass burning events, where NO_3^- in particular tends to increase in samples with higher NH_4^+ concentrations (Fuhrer et al., 1996; Whitlow et al., 1994; Savarino and Legrand, 1998). These indicators are a result of complex chemical reactions that are not necessarily a direct reflection of variations in biomass burning, however, many high fire years are identifiable by enhanced NH_4^+ concentration (Eichler et al., 2011). The isotopic ratio of atmospheric gases, such as methane (CH_4) and carbon monoxide (CO) in Antarctic (Ferretti et al., 2005; Wang et al., 2010) and Arctic (Sapart et al., 2012) ice cores has also been used as fire proxy but such ratios are also not solely products of biomass burning (Fischer et al., 2008). The isotopic composition of methane ($\delta^{13}\text{C}$ of CH_4) in ice cores differentiates biogenic from pyrogenic sources (Sapart et al., 2012). Methane is stable in the atmosphere for longer than the atmospheric exchange time and therefore is globally distributed. Hence $\delta^{13}\text{C}$ of CH_4 variations are impacted by sources distributed all over the world (Blunier et al., 2007).

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

Monosaccharide anhydrides are one of the few fire proxies that have specific

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sources (Gambaro et al., 2008; Simoneit, 2002). Levoglucosan (1,6-anhydro- β -D-glucopyranose) is a monosaccharide anhydride released during biomass burning when cellulose combustion occurs at temperatures $>300^{\circ}\text{C}$ (Gambaro et al., 2008; Simoneit, 2002). As with BC and NH_4^+ , levoglucosan is injected into the atmosphere in convective smoke plumes, deposited on glacier surfaces through wet and dry deposition, and preserved in the snow and ice (Gambaro et al., 2008; Kehrwald et al., 2012; McConnell et al., 2007; Fuhrer and Legrand, 1997). Unlike greenhouse gases, levoglucosan, NH_4^+ and BC are not homogeneously distributed in the atmosphere as chemical processing causes short lifetimes (on the order of days to weeks) (Hennigan et al., 2010; Fraser and Lakshmanan, 2000; Ramanathan and Carmichael, 2008; Fuhrer and Legrand, 1997) in addition to efficient deposition processes from the atmosphere. Although levoglucosan is oxidized by OH radicals in the gas phase (Hennigan et al., 2010) and in atmospheric water droplets (Hoffmann et al., 2010), its high concentration in biomass emissions means that levoglucosan remains a strong potential tracer for fire activity even across remote distances (Fraser and Lakshmanan, 2000; Holmes and Petrucci, 2006, 2007; Kehrwald et al., 2012).

Charcoal deposited both in non-polar ice and sediment cores can also help reconstruct past fire activity. The Global Charcoal Database (GCD) compiles individual charcoal records from terrestrial and marine sediment cores (Fig. S6) into global or regional reconstructions that provide a benchmark against which other biomass burning records can be compared (Power et al., 2010; Marlon et al., 2008). The charcoal database covers all climatic zones and all major biomes, but data for grasslands and dry shrublands, where low woody biomass limits charcoal production, are limited due to a lack of suitable sampling sites (Marlon et al., 2008). These vegetation types emit large quantities of levoglucosan when burned (Iinuma et al., 2007; Gao et al., 2003) resulting in a possible offset between charcoal and levoglucosan data. Many charcoal sampling sites are located in the US and Europe, and a limited number of GCD sites currently exist in northern Asia (Fig. S6). As levoglucosan and BC can be transported hundreds to thousands of kilometers (Mochida et al., 2010), they complement GCD reconstruc-

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tions that record combustion within tens of km of the sampling sites, and provide fire activity data for regions where charcoal records do not exist.”

2.1) To completely reword the text from end of page 811 to end of page 812 (with much more focus on boreal fires), I encourage the authors to visit the following papers on the role of boreal forest fires including those addressed in the 90’s with the ABLE campaigns and more recently with the POLARCAT campaigns.

Stocks, B.J., The extent and impact of forest fires in northern circumpolar countries, in *Global Biomass Burning*, edited by J.S. Levine, pp. 197-202, MIT Press, Cambridge, Mass., 1991. Harriss, R.C., et al., The Arctic Boundary Layer Expedition fieldwork (ABLE) 3A, July- August 1998, *J. Geophys. Res.*, 97, 16,383-16,394, 1992. Harriss, R.C., S.C. Wofsy, J.M. Hoell Jr., R.J. Bendura, J.W. Drewry, R.J. McNeal, D. Pierce, V. Rabine, R.L. Snell, The Arctic Boundary Layer Expedition (ABLE-3B): July– August 1990, *J. Geophys. Res.*, 1635–1643, doi: 10.1029/93JD01788. Paris, J.-D., Stohl, A., Nédélec, P., Arshinov, M. Yu., Panchenko, M.V., Shmargunov, V. P., Law, K.S., Belan, B.D., and Ciais, P.: Wildfire smoke in the Siberian Arctic in summer: source characterization and plume evolution from airborne measurements, *Atmos. Chem. Phys.*, 9, 9315–9327, doi:10.5194/acp-9-9315-2009, 2009. Schmale, J., J. Schneider, G. Ancellet, B. Quennehen, A. Stohl, H. Sodemann, J.F. Burkhart, T. Hamburger, S.R. Arnold, A. Schwarzenboeck, S. Borrmann, and K.S. Law, Source identification and airborne chemical characterisation of aerosol pollution from long-range transport over Greenland during POLARCAT summer campaign 2008, *Atmos. Chem. Phys.*, 11, 10097–10123, doi:10.5194/acp-11-10097-2011, 2011

The text from end of page 812 to line 10 of page 813 is very confusing (see details below) and the quite complex question of which chemical species represent good forest fire proxies in ice is out of place in an introduction. Furthermore, I disagree with numerous statements since the relevance of a given chemical species to trace back biomass burning is also dependant of the region of concern. The sentence (line 24, page 812) “Some proxies are produced solely from biomass burning, like charcoal, but

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most others, such as lightweight carboxylic acids, lignin and resin pyrolysis products, and major ions (K^+ , NH_4^+ and NO_3^-) are have multiple potential sources other than wildfires (e.g. coal burning, volcanic eruptions or biogenic emissions) (Eichler et al., 2011; Yalcin et al., 2006)” is very confusing. First, please make clear that charcoals are used as proxies in lake sediments whereas ions are used for ice cores.

We specify in the new version of paper that charcoal cannot be used as a proxy in ice cores, at line 82 – 84.

“Some proxies are specific as they are produced solely from biomass burning, such as charcoal in sediment cores, but most others have multiple potential sources other than wildfires (e.g., coal burning, volcanic eruptions or biogenic emissions).”

2.2) Second, please specify that among light carboxylates mainly formate and oxalate (as first shown by Legrand et al. 1992) were useful to trace fires in Greenland ice cores.

This useful information has been specified in the text, at line 88 – 90. “Among possible light carboxylic acids, formate and oxalate are the most useful to trace fires in Greenland ice cores (Jaffrezo et al., 1998; Legrand et al., 1992; Dibb et al., 1996; Kehrwald et al., 2012).”

2.3) Finally, it is no clear in your sentence which chemical species among K^+ , NH_4^+ or NO_3^- is related to volcanic eruptions ?! Please comment.

“(…) major ions (K^+ , NH_4^+ and NO_3^-) have multiple potential “sources other than wildfires (e.g., coal burning, volcanic eruptions or biogenic emissions)”. The aim of this sentence is to introduce major ions in the discussion, and advise the reader about their unspecificity as biomass burning markers. Coal burning, volcanic eruptions and biogenic emissions are few cited examples of sources for these species. However, not all these ions are emitted by all these sources.

We removed potential misinterpretation and modified the text as follows:

Lines 94 - 100. “However, lightweight carboxylic acids and major ions (potassium (K^+),

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ammonium (NH_4^+) and nitrate (NO_3^-) are not specific for biomass burning since they can be emitted by multiple sources. K^+ and NH_4^+ , for example, are also emitted from numerous other sources and thus must be corrected accordingly in order to provide source-specific information (Savarino and Legrand, 1998). Of the available ionic proxies, K^+ is less useful due to marine and terrestrial sources, even if corrected to remove these contributions (e.g. non-sea-salt and non-crustal K^+) (Legrand and DeAngelis, 1996; Savarino and Legrand, 1998).”

2.4) Again the sentence (lines 2-5, page 813) “Potassium (K^+) and ammonium (NH_4^+), for example, have traditionally been used in ice core studies as tracers of past fire events, but these ions are also emitted from numerous other sources (Eichler et al., 2011; Yalcin et al., 2006) and thus require to be corrected accordingly in order to provide source-specific information.” is very confusing and missed key references on previous Greenland ice core studies. In fact, the story of tracing back forest fires in ice cores started with Legrand et al. (1992) followed by Legrand and De Angelis (1996) showing that the concentration of ammonium, formate, and oxalate are very significantly enhanced above background level in Greenland ice layers corresponding to forest fire events. In contrast, it was shown that potassium is less useful due to marine and terrestrial source (Legrand and de Angelis, 1996). Later on, Savarino and Legrand (1998) found that some (but not all) of fire events recorded in Greenland ice exhibit a slight increase of nitrate (in addition to the strong ones of ammonium, formate, and oxalate). The work from Eichler et al. (2011) related to the Belukha glacier located in the continental Siberian Altai is very interesting to discuss later in your manuscript the role of Siberian fires but not here in the introduction to discuss which chemical tracers are relevant. Indeed, in contrast to Greenland, the relative contribution of continental biospheric emissions with respect to fire events may be larger at a Siberian glacier site compared to the case of remote Greenland sites. That weakens the powerfulness of ammonium and formate to trace back biomass burning plumes there.

We followed your suggestion and we reworded part of introduction into the now sub-

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chapter “1.1 Boreal Forest Fires” (Line 28).

2.5) Concerning your statement (line 6-10, page 813) that “NH₄⁺, nitrate (NO₃⁻), H₂O₂, and organic acids in Arctic ice are often associated with biomass burning events, where NO₃ in particular tends to increase in samples with higher NH₄ concentrations (Fuhrer et al., 1996; Whitlow et al., 1994), however these indicators are also a result of complex chemical reactions that are not necessarily a direct reflection of variations in biomass burning.”: The wording “Arctic” is confusing: please change to “Greenland”. I don’t think that H₂O₂ levels were extensively discussed in terms of biomass burning ? For nitrate, please also refer to Savarino and Legrand (1998).

We changed the word “Arctic” to “Greenland”, removed “H₂O₂” from the sentence, and referred to Savarino and Legrand (1998) when nitrate is discussed. Thank you for your precise suggestions

2.6) Lines 18-21, page 813: While levoglucosan is mainly produced by combustion via the breakdown of cellulose its emission factor can widely vary, depending on flame conditions (see Gao et al., 2003), please comment or advice the reader. Note also that for boreal fires the smoldering phase is far more important than flaming one (important for savanna fires) and would favor high levoglucosan emission factor, correct ?

Gao, S., D. A. Hegg, P. V. Hobbs, T. W. Kirchstetter, B. I. Magi, and M. Sadilek, Water-soluble organic components in aerosols associated with savanna fires in southern Africa: Identification, evolution, and distribution, *J. Geophys. Res.*, 108(D13), 8491, doi:10.1029/2002JD002324, 2003.

A mention of flaming conditions, as a reason for variable levoglucosan emission factor, has been added at line 393 – 395.

“Levoglucosan yield is variable and strongly depends on combustion temperature, biomass composition and flame conditions (Gao et al., 2003; Oros and Simoneit, 2001b, a; Weimer et al., 2008). Thus, levoglucosan can not be used to characterize

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and quantify BC in environmental samples (Kuo et al., 2008).”

2.7) Instead to launch in the introduction a discussion on the relevance of various chemical species to trace back fire events in ice, I suggest to totally reword this paragraph (from end of page 812 to line 10 of page 813) by reporting first on previous Greenland ice cores, namely :

Legrand et al. 1992 : Pioneering study on the fingerprint from forest fires in Greenland ice layers: ammonium, formate and oxalate. Legrand and De Angelis (1996), covered time period: 1762-1982 AD (seasonally- resolved), ammonium, formate, and oxalate (you missed to refer to this previous work published in the JGR). Savarino and Legrand (1998), covered time period: 1200-1982 AD (seasonally- resolved), ammonium, formate, and oxalate. MacConnell et al., 2007, 1788-2002 AD (seasonally-resolved), BC and vanillic acid. Then introduce : Eichler, 1250-2001, and emphasize that due to the influence of anthropogenic emissions in the last 60 years, the reconstruction based on fire proxies K⁺ and NO³⁻ was confined to the pre-industrial period AD 1250-1940, whereas charcoal data provided a fire history for the last 750 years. Kawamura et al., 2012, levoglucosan, dehydroabietic, vanillic and p-hydroxybenzoic acids, 1693-1997 AD, one-year resolution back to 1970, discontinuous below. Legrand M., and M. De Angelis, Light carboxylic acids in Greenland ice: A record of past forest fires and vegetation emissions from the boreal zone, J. Geophys. Res., 101, 4129-4145, 1996.

We followed your suggestion, and the history of reconstructing biomass burning event is reported in the new sub-chapter “1.2 Biomass Burning Proxies” (line 78).

3) General comments on data discussion My main concern is about the use of the NEEM data and first the lack of comparison with previous data: Given the previous works done by Legrand and De Angelis (1996), Savarino and Legrand (1998), both using high resolution ammonium records (together with those of formate) and since (as stated in lines 10-13 of page 815) the ammonium high resolution record is also available at NEEM, it is logic (and needed) to start data discussion by comparing them

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together. As discussed below, in the case of Greenland, there is no reason to discard ammonium to trace back forest fire event. So please show up with a comparison of high-resolution ammonium records from Summit and NEEM (also for the two last centuries with vanillic acid at D4 in Greenland). I believe that the multi-annual averaged NH₄ record (i.e., as reported in the manuscript) is less appropriate to reveal and describe fire events. Also please compare BC at NEEM and BC at D4. From that and well before starting the discussion with other proxies, identify common features and eventual discrepancies between the different high-resolution ammonium Greenland records. Here, depending of the result of the comparison, possibly interesting would be to compare air mass back-trajectories arriving in summer at Summit and NEEM (located further North) sites.

This paper focuses on the reconstruction of past fire events in the NEEM ice core. We are sure comparing different profiles from different environmental archives is a useful exercise, but perhaps this goes beyond of the purpose of this paper. In addition, many datasets exist for Greenland ammonium, and each dataset has different temporal resolution, or it is transformed accordingly for biomass burning events detections. Different sources, accumulation and deposition on different Greenland locations make this comparison a challenge that is not the goal of the paper. For this reason we compared levoglucosan and ammonium from the NEEM ice core, and we confirmed both the use of levoglucosan and ammonium to infer past fires, although ammonium should not be used alone to this purpose. However, we plan to discuss geographical variability of levoglucosan and ammonium across Greenland in ongoing works.

3.1) Section 2.2 entitled “statistics” (page 816): I don’t understand this section introducing the use of calcium in the discussion since calcium exhibit a maximum in spring in Greenland ice layers whereas forest fire events mainly occur in summer (Legrand and Mayewski, 1997).

We present major ions averaged for multi-year samples, the same sampling scheme of samples for levoglucosan analyses. In these multi-year samples, interannual variability

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could be ruled out from the discussion, and we discussed only “long term” correlation between levoglucosan and major ions.

3.2) Therefore I don't expect any correlation between calcium and levoglucosan and I am not at all surprised by your statement (line 5-7, page 818): “Preliminary results of ICP-MS analyses on the deep NEEM ice core support the lack of correlation between other crustal markers (i.e. Ti and Ba) and levoglucosan (J. Gabrieli, personal communication, 2014)”. So Please reconsider the usefulness of this section. Legrand, M., and P. Mayewski, Glaciochemistry of polar ice cores: A review, *Reviews of Geophysics*, 35, 219-243, 1997.

If levoglucosan correlates with a crustal marker, readers may think that levoglucosan variability may be dependent on both biomass burning plumes and transport efficiency such as wind strength. We think the “absolute” lack of correlation between levoglucosan and crustal markers is an important result to convince the reader about the lack of transport contribution or further data transformation (i.e. normalization for crustal tracer). We therefore prefer keep this result in this section.

3.3) Section 3: Line 7-12 page 818: You state “A similar correlation analysis of BC and other elements and chemical species (data not shown) measured in the NEEM-2011-S1 core indicates that during pre-industrial periods BC concentrations generally do not correlate with crustal particulate tracers but do with NH₄ and NO₃”. First, this statement would be at a better place in section 4.1. (When you are discussing biomass burning tracers at NEEM with other ions). Anyway, this sentence needs to be illustrated by at least one figure showing examples of BC peak accompanied ammonium and nitrate peaks and some statistics.

This statement refers to results from NEEM-2011-S1 core. Section 4.1 may be a good position to place this result. However we prefer keeping results from NEEM-2011-S1 core in chapter 3, where we present all results. Analyses and data from NEEM-2011-S1 core are still unpublished (Joe McConnell, Personal communication). As the NEEM-

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2011-S1 results further confirm our results from deep NEEM core, we prefer to present this evidence. However we think the discussion of the NEEM-2001-S1 results are not fundamental for our discussion, and we prefer keeping major ions analyses from the NEEM-2011-S1 core unpublished without presenting data in figures.

3.4) Section 4.1: lines 4-14 page 819: The sentence "Increased wind speeds result in greater dust concentrations and larger particles in ice cores (CITE)" remains a mystery for me ????

We include the correct citation in the new version of the manuscript (please, see point 12 of Referee #1). We apologize for the inconvenience.

3.5) Also the next sentence "Stronger winds may be expected to also increase levoglucosan concentrations as the increased wind strength could transport more biomass burning products in air plumes." has to be reworded (as it stands, this sentence sound strange for an atmospheric chemist).

We reworded the sentence in order to more clearly present our results.

Lines 283 – 284. "Stronger winds may be expected to also increase levoglucosan concentrations, as the increased wind strength could transport more efficiently air plumes and thus more biomass burning products."

3.6) Line 15-26 page 819: See my comment above: Checking high-resolution ammonium records it is easy to identify forest fire input in Greenland ice (mainly in the form of ammonium formate as discussed in details by Legrand and De Angelis, 1996). Also your suggestion about ammonium peaks linked to volcanic eruptions "During volcanic eruptions, when vast amount of sulfate is emitted to the atmosphere, NH_4^+ will irreversibly form NH_4HSO_4 and $(\text{NH}_4)_2\text{SO}_4$ which is then deposited on the ice. Therefore, peak concentrations in NH_4^+ measured during volcanic eruptions are most likely not directly linked to biomass burning" is totally incorrect. A convincing example showing that in no way volcanic eruptions disturbed ammonium deposition in Greenland ice

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is the case of the large Laki eruption (1783). Even during this large tropospheric eruption, no special ammonium spike was recorded in Summit ice layers (see Legrand and De Angelis, 1996).

We removed this sentence. Thank you for noting this weak point of the paper.

3.7) In addition to my previous comment on the lack of comparison with available high-resolution records, line 14-24 page 821 and lines 12-16 page 822 missed several previous works: line 14-24 page 821: You wrote "Other ice core records provide comparisons of fire events that allow an examination of the spatial extent of past short-lived fire activity. A prominent 1617–1622 CE levoglucosan peak coincides with high fire activity inferred from oxalic acid analyses recorded in the Greenland Site J ice core (Kawamura et al., 2001)" Please report here the work from Savarino and Legrand (1998) at Summit that does not report any ammonium, formate and oxalate perturbation at that time. Please comment.

We provided the requested reference regarding the absence of this fire event recorded at Summit.

Lines 336 – 338. "A prominent 1617 - 1622 CE levoglucosan peak coincides with high fire activity inferred from oxalic acid analyses recorded in the Greenland Site J ice core (Kawamura et al., 2001), even if this event is not recorded in ice core from other locations in Greenland (e.g. Savarino and Legrand, 1998)."

3.8) lines 12-16 page 822: You wrote "The maximum BC concentration (16 ng g⁻¹) in the entire Greenland D4 ice core occurred in 1908CE (McConnell et al., 2007). The second- highest BC peak (15 ng g⁻¹) over the past millennium in the NEEM-2011-S1 is dated to 1909.5CE (Fig. 2b). This timing is relatively coincident with the Tunguska Event, a bolide impact in western Siberia occurring in June 1908. An ammonium spike in 1910 CE in the GISP2 ice core (Taylor et al., 1996). Did you suggest that your NEEM BC spike is related to the Tunguska event ? Please make it clearer and report the previous discussion made by Legrand et al. (1995) on this specific event: In their

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1908-1989 ice core records from Summit (Greenland) Legrand et al. (1995) reported the highest ammonium peak corresponding to the 1908 year. They stated “Although some observations of burned trees have been made in Siberian area impacted by the Tunguska fall in 1908 (Kulik, 1927), it is difficult to conclude that the 1908 ammonium event revealed by our Summit ammonium profile is related to this phenomenon.” Reversely both Legrand et al (1995) and Taylor et al. (1996) emphasized that no forest fire event was detected in Summit ice layers corresponding to the 1915 year. Legrand et al. (1985) stated “check the impact of the particularly dry 1915 year in Siberia during which some 14 million of hectares were burned (Stocks, 1991), no significant increase of ammonium” Finally, note that the levoglucosan and p-hydroxybenzoic acids were disturbed in the 1915 ice layers in Kamchatka (see figure 4 from Kawamura et al., 2012). Please comment.

Legrand M., M. De Angelis, H. Cachier, and A. Gaudichet, Boreal biomass burning over the last 80 years recorded in a Summit-Greenland ice core, In NATO ASI Ser. "Ice cores studies of Global biogeochemical cycles", R. Delmas ed., 347-360, 1995. Kulik, L.A., Akademiia Nauk, SSSR, Co. R (Doklady), 23:399, 1927. Stocks, B.J., The extent and impact of forest fires in northern circumpolar countries, in Global Biomass Burning, edited by J.S. Levine, pp. 197-202, MIT

Thank you for the useful suggestions. We modified the text as follows.

Line 361 – 374. “The highest ammonium peak in a Summit, Greenland ice core spanning 1908-1989 CE occurs in 1908 CE and is synchronous with increases in formate, oxalate, glycolate and BC within the same core. Legrand et al. (1995) ascribe this event to North American forest fires rather than the Tunguska event. However, ammonium may be produced by a Haber-like process (Melott et al., 2010). High pressure and temperature conditions generated by the Tunguska comet entering the atmosphere in principle could have allowed atmospheric nitrogen and hydrogen from the comet ice to react, thereby accounting for both the ammonium and nitrate enhancements in some Greenland ice cores (Melott et al., 2010). The absence of levoglucosan and

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ammonium spikes in the NEEM (northern Greenland) ice core or vanillic acid peaks, a source-specific tracer for conifers (Simoneit, 2002), in the D4 (central Greenland) ice core during this same time period, supports the hypothesis that BC, NH₄⁺ and NO₃⁺ peaks may not be caused by forest fires during 1908 CE. In addition, comparisons of BC and toxic heavy metals in the ACT2 core from southern Greenland (McConnell and Edwards, 2008), as well as BC and non-sea-salt sulfur in the D4 ice core from central Greenland (McConnell et al., 2007), indicate that >80% of the BC during this period is associated with industrial emissions (primarily coal burning).”

3.9) Section 4.3. I found your arguments on the respective contribution of Siberian versus North American boreal fires rather weak. Line 25-30 page 826 and below: You first report “Arctic snow samples across the Greenland ice sheet and associated back-trajectories from a single year highlight the importance of Russian boreal forests as a biomass burning source and suggest that 55 % of modern BC reaching the Greenland ice sheet originates from Russian biomass burning, while 40 % is from North America, and only 5 % is from anthropogenic emissions (Hegg et al., 2009). A 44-year record of 10 day isobaric back-trajectories reaching Summit, Greenland (Kahl et al., 1997), indicates that more summer (fire season) trajectories originate over North America (46 % at 500 hPa and 85 % at 700 hPa) than from Eastern Asia (20 % at 500 hPa) or from Europe and Western Asia (only 6 %).” A few lines below you state “North America is likely the most important contributor due to the proximity to the NEEM location and the shorter routes travelled by aerosols reaching Greenland with respect to Siberian–Eurasian forests, but previous studies also demonstrate that air masses originating in Asia reach Greenland after a few days.” These different statements are rather confusing. First, the study from Kahl et al. (1997) is based on a good statistic (44 years with seasonal variation) compared to the Hegg’s study dedicated to a single year. For the reader, please make clearer that Kahl et al. (1998) report 10 day back-trajectories at 700 hPa (i.e; the elevation of Summit) as well as 500 hPa (i.e. 5-6 km asl) that reflect midtropospheric circulation.

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We added at line 460 -462 this useful information. “The pressure of 500 hPa corresponds to $\sim 5 - 6$ km asl and is typical of mid-stratospheric circulation, while 700 hPa is closer to the altitude of the Summit camp.” We agree that the Kahl study is more robust but we include Hegg as an example of how in individual years air masses originating from Russia can dominate the biomass burning signal recorded in Greenland ice.

3.10) When reporting the percentage “North America (46 % at 500 hPa and 85 % at 700 hPa) than from Eastern Asia (20 % at 500 hPa)” you miss to report the contribution from Eastern Asia (only 6% at 700 hPa).

We added the missing percentages, as suggested, at line 460 – 463. “A 44-year record of 10 day isobaric back-trajectories reaching Summit, Greenland (Kahl et al., 1997), indicates that more summer (fire season) trajectories originate over North America (46 % at 500 hPa and 85 % at 700 hPa) than from Eastern Asia (20 % at 500 hPa and 6% at 700 hPa) or from Europe and Western Asia (only 6 % at 500 hPa).”

3.11) I feel that the following sentences (line 20-25 page 828) overstated the contribution of Siberian fires to the Greenland ice sheet (it might be a coincidence !!!): “The Ushkovsky (Kawamura et al., 2012) and Belukha (Eichler et al., 2011) ice cores both contain up to multi-decadal periods of increased fire activity that are similar to peaks in the NEEM ice core suggesting that these sites may receive a contribution from Siberian fire activity (Fig. 3). Our work determines that Siberia is an important source of burning signatures inferred in Arctic ice fields during extreme fire events.” I would suggest more careful conclusion with respect to these apparent common trends.

Biomass burning plumes have been observed traveling around the world, and model results support the possibility that fire products originating in Asia may reach Greenland. We offered many examples of these studies in the text. Our conclusions are not only based on the observation of coincident fire peaks around the northern hemisphere, but also on observations of synchronous megafires and megadroughts in Asia. This evi-

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dence reduces the possibility of a coincidence. Even if few published measurements suggest the link between Asian droughts – Asian fires – biomass burning signature in Greenland, this result is not surprising (although interesting) since expected by models.

4.1) Minor comments: Line 12, page 818: The 1973 levoglucosan event is quite large: do we have direct observation available (e.g., satellite based), which would allow investigating the location of this particular recent fire?

Satellite observations of North American during this time period do not demonstrate large fires during this time period. Few satellite observations of Russian biomass burning are available for 1973. The newspaper articles which we cite are the only published information that we were able to obtain regarding the large Russian wildfires in 1973.

4.2) Line 25 page 814: I suggest to convert 1.1m of NEEM snow accumulation into years versus depth, so the reader has a direct information on the temporal resolution of the levoglucosan record.

NEEM ice samples for levoglucosan analyses are a 1.10 m core section, and have been collected by using a CFA analyses at the NEEM camp, then they were sent at Ca' Foscari University of Venice for levoglucosan analyses. In other words, we did not choose the sampling scheme. Due to ice compression each sample covers different temporal intervals, as specified at line 184 - 185 "Due to the ice compression and differences in Greenland snow accumulation, each 1.10 m sample covers 1 to 5 year-long temporal periods.' We do convert all samples into years, and all data are presented using the NEEM timescale. All discussions and figures discuss levoglucosan concentrations or flux relative to their age throughout the paper.

We now include the depth-age scale of the samples used in this study as a figure (Fig. S7) in the Supplementary Information. This depth-age scale is from the already published NEEM chronology (Rasmussen et al., 2013), but it also include the model dating for the upper 20 m of the NEEM ice core.

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"Figure S7 Age model representation. Blue points indicate the age-depth scale, as published in Rasmussen et al. (2013), black points are from the Herron-Langway firn densification model fit to the NEEM-NGRIP tie points for the 20-80 m long firn column."

4.3) Line 10 page 831: It is here difficult to follow the discussion that jumps from North America to monsoon, this is confusing.

The paragraph before the paragraph beginning at the original Line 10 page 831 also discusses possibilities of aerosols with an Asian source reaching Greenland. We mention North America in the sentence "A high-resolution determination of continental dust in the Dye-3 Greenland ice core Sr, Nd and Hf isotopic composition from 1786–1793 CE determine that the majority of the samples have an Asian origin (Lupker et al., 2010)". However, the point of this sentence is that the dust has an Asian origin. We mention North America (perhaps implied by Greenland?) simply because Greenland is the deposition location.

4.4) Line 13 page 813: The Wang et al. (2012) paper does not infer biomass burning history from firn record (it prescribes biomass burning emission to further evaluate changes in fossil fuel emission over the last 60 years).

We corrected the wrong citation to Wang's papers. We changed reference to (Wang et al., 2012) instead of (Wang et al., 2010).

Wang, Z., Chappellaz, J., Park, K., and Mak, J. E.: Large Variations in Southern Hemisphere Biomass Burning During the Last 650 Years, *Science*, 330, 1663-1666, 10.1126/science.1197257, 2010.

Wang, Z., Chappellaz, J., Martinerie, P., Park, K., Petrenko, V., Witrant, E., Emmons, L. K., Blunier, T., Brenninkmeijer, C. A. M., and Mak, J. E.: The isotopic record of Northern Hemisphere atmospheric carbon monoxide since 1950: implications for the CO budget, *Atmos. Chem. Phys.*, 12, 4365-4377, 10.5194/acp-12-4365-2012, 2012.

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Anonymous Referee #3 Received and published: 31 March 2014

This paper reports variation in concentrations of levoglucosan and ammonium measured in the NEEM ice core, and black carbon from the NEEM-2011-S1 core for the past two millennia. The authors used these reconstructions to infer fire activity from northern hemisphere high latitudes. In the light of previous studies (fire proxies transport, potential sources of fire proxies to the site, climate change and human activities) the authors raised the conclusion that fire activity recorded in the NEEM ice core reflects boreal forests fire from North America and Siberia. They suggest that this signal is mainly driven by temperature for the last two millennia except during periods of extreme Asian droughts that would trigger Siberian fires. The paper should be publishable after minor/major revisions suggested below to clarify the manuscript.

Detailed comments:

1) Line 5, page 811: “broad-scale” remains unclear (same line 4, page 814)

We removed “broad-scale” from the following sentences:

Abstract. “Biomass burning is a major source of greenhouse gases and influences regional to global climate. Pre-industrial fire-history records from black carbon, charcoal and other proxies provide baseline estimates of biomass burning at local to global scales, but there remains a need for fire proxies that span millennia in order to understand the role of fire in the carbon cycle and climate system.”

Lines 134 – 137. “Although levoglucosan is oxidized by OH radicals in the gas phase (Hennigan et al., 2010) and in atmospheric water droplets (Hoffmann et al., 2010), its high concentration in biomass emissions implies that levoglucosan is a strong potential tracer for fire activity even across remote distances (Fraser and Lakshmanan, 2000; Holmes and Petrucci, 2006, 2007; Kehrwald et al., 2012).”

2) Line 9, page 811: “over the past 2000 years” Add: to infer changes in boreal fire activity

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We add “to infer changes in boreal fire activity” as suggested by the Referee.

Abstract. “We use the specific biomarker levoglucosan, and multi-source black carbon and ammonium concentrations to reconstruct fire activity from the North Greenland Eemian (NEEM) ice cores (77.49° N; 51.2° W, 2480 masl) over the past 2000 years to infer changes in boreal fire activity.”

3) Line 15, page 811: “monsoon failures” Add Asian

We add “Asian” in the Abstract, as suggested. “Many of these multi-annual droughts are caused by Asian monsoon failures, thus suggesting a connection between low and high latitude climate processes.”

4) Line 28, page 812: remove “are” before “have multiple potential sources

We removed the verb “are”, as indicated at point 6 of Referee #1

5) Line 8, page 813: Add dot after “Whitlow et al., 1994). However (. . .)

We agree that the addition of dot increases the readability. However we modify the text as follows.

Lines 103 – 107. “NH₄⁺, NO₃⁻, and organic acids in Greenland ice are often associated with biomass burning events, where NO₃⁻ in particular tends to increase in samples with higher NH₄⁺ concentrations (Fuhrer et al., 1996; Whitlow et al., 1994; Savarino and Legrand, 1998). These indicators are a result of complex chemical reactions that are not necessarily a direct reflection of variations in biomass burning, however, many high fire years are identifiable by enhanced NH₄⁺ concentration (Eichler et al., 2011).”

6) Line 7, page 814: remove square brackets

We removed the brackets, as discussed at point 10 of Referee #1

7) Line 11, page 814: remove “to reconstruct fire activity for the past 2000 years”

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The text “to reconstruct fire activity for the past 2000 years” has been removed.

Lines 154 – 156. “Specifically, we report levoglucosan and ammonium concentrations in the upper part of the deep North Greenland Eemian (NEEM) ice core as well as BC measured in the 410 m NEEM-2011-S1 core (Fig. 1) which was collected adjacent to the deep core in 2011 (Sigl et al., 2013).”

8) Line 24, page 816: change title for “Sources of NEEM products”?

We would prefer keeping the original title “Statistics”, since this sub-chapter is a part of Chapter 2 “Methods”. Here we do not discuss the geographical source of NEEM pyro-products but only present the statistical methods and tools use to discuss levoglucosan variability. However, to avoid confusion and digressing, we removed the sentence from the “Statistics” chapter “The closest source regions for the Greenland dust are boreal Canada and boreal Russia, which are similar landscapes with similar availability of entrainable dust.”, and we add the sentence at line 226 - 228 “Then, we studied links between levoglucosan, ammonium and major ions by performing Pearson and Spearman correlations (Supplementary Information).” We discussed this problem at point 17 of Referee #1.

9) Line 9 page 817: Change “for punctual analysis” by “for short term analysis”

We changed “for punctual analysis” by “for short-term analysis”, as this nomenclature is reported in the title.

Line 231 – 232. “For short-term analysis, we consider megafires as levoglucosan peaks with a Z-score ≥ 1 [$Z\text{-score} = (x_i - \bar{x})/\sigma$], corresponding to a concentration ≥ 245 pg mL⁻¹ (Table 1, Figure S5).”

10) Line 4, page 818: give the p-values for correlations

As suggested we added the p-values and α parameters, at line 253 – 256.

“Pearson and Spearman correlations (Supplementary Information) demonstrate that,

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of the possible biomass burning-related products measured in the deep NEEM core, levoglucosan concentrations only slightly correlate with ammonium (p -value $< 10^{-6}$, with $\alpha = 0.05$). No correlation is noted between levoglucosan and crustal marker Ca_2 or dust (p -values > 0.45 , with $\alpha = 0.05$).

11) Line 12, page 818: add dates described in the text and Table 1 to figure 2.

We prefer to not add dates in Fig. 2, and instead to report only megafires inferred by all the three markers. NEEM fire reconstruction is based on a multi-proxy approach, and it is not exclusively based on levoglucosan measurements. If we add dates to Fig. 2, we should add dates for all the three markers. Figures would become unreadable, in particular regarding black carbon which was measured at very high temporal resolution and thus is able to record hundreds of fires. This would be even more complicated by adding strong-events from Table 1 to mega-events reported in Fig. 2.

12) Line 13-14, page 818: “(Fig. 2)”. Add Fig S5 and Table 1. Remove “other major peaks are reported in Table 1”.

We added Table 1 to “(Fig. 2)” and removed “other major peaks are reported in Table 1”, as suggested by the Referee.

13) Line 23, page 818: what about the low at 1700-1800 CE?

The period of low fire activity between 1700 and 1800 CE, coincident with the Little Ice Age, is discussed in the Chapter 4.4.1 (lines ...). For this reason we added these dates as you suggest.

Lines 269 – 272. “Strong fluctuations in the temporal profile dominate the record from decadal to centennial perspectives. Relative maxima in the levoglucosan profile are evident between 1000 - 1300 CE and 1500 - 1700 CE, and with a lesser extent around 500 CE and 100 CE, while the lowest fire activity is evident around 700 - 900 CE and with a lesser extent around 1300 - 1500 and 1700 - 1800 CE (Fig. 3A).”

14) Line 26, page 818: change “when” by “while” and remove “absolute” in “absolute

maximum in BC concentration” as you discussed about smoothed curve

We modified the text as suggested.

Lines 273 – 275. “Between 1000 - 1600 CE and 1600 - 1800 CE the BC profile show a relative maximum and minimum, respectively, while the absolute maximum in BC concentration peaks around 1900 CE.”

15) Line 3, page 819: change title to something like “major ions transport and deposition”

We made the suggested modifications, as this new title is more appropriate for a Chapter in the “Discussion and Interpretation” section.

Line 279. “4.1.1 Major ions transport and deposition on the Greenland ice-sheet”

16) Line 7, page 819: CITE ???

Please, see point 12, Referee #1

17) Line 25-28, page 820: please add a short sentence at the beginning to say that you discuss the 1973 CE fire peak in the NEEM core. Otherwise confusing with the 342 CE paragraph.

We appreciate your suggestion and restructured part of text at lines 318 – 324, as follows.

“The 1973 CE fire event is synchronous (within the age model error) and probably related to an anomalous heat wave and severe droughts in 1972 CE in Russia (Dronin and Bellinger, 2005; Golubev and Dronin, 2004). Thousands of Russian firemen, soldiers and farmers were mobilized to suppress a fire raging for several weeks in August 1972 in a bog-land east of Moscow (The Palm Beach Post, Aug. 09 1972; The New York Times, Aug. 09 1972, New York), and during the same week more than 1000 km² of forest burned in Central Alaska (The Milwaukee Journal, Aug. 09 1972).”

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18) Line 26, page 821: “The NEEM levoglucosan” centennial scale “maximum” 19)
Line 27, page 821: “from the Belukha ice core” located in Siberia

We modified the text as suggested by the Referee #3, at lines 346 – 349.

“The NEEM levoglucosan centennial scale maximum (1500 - 1700 CE, Fig. 3A), the highest of the past two millennia, coincides with data from the Belukha ice core, Siberia (Eichler et al., 2011), that documents a period of increased fire activity between 1550 - 1700 CE where 1600 - 1680 CE records the highest forest fire activity over the last 750 years (Fig. 3D).”

20) Line 20-30, page 822: add the other ice cores discussed in the text to figure 1 (and other archives in general).

We added all the environmental archives discussed in the text to Fig. 1.

21) Line 15, page 823: “can not be used to quantitatively characterize BC in environmental samples”. This sentence remains unclear.

We modify lines 393 - 395 to clarify text, as suggested by Referee #3

“Levoglucosan yield is variable and strongly depends on combustion temperature, biomass composition and flame conditions (Gao et al., 2003; Oros and Simoneit, 2001b, a; Weimer et al., 2008). Thus, levoglucosan can not be used to characterize and quantify BC in environmental samples (Kuo et al., 2008).”

22) Line 25, page 823: add “s” to read “compiles”

We corrected the error by adding “s” at lines 139 – 142.

“Charcoal deposited both in non-polar ice and sediment cores can also help reconstruct past fire activity. The Global Charcoal Database (GCD) compiles individual charcoal records from terrestrial and marine sediment cores (Fig. S6) into global or regional reconstructions that provide a benchmark against which other biomass burning records can be compared (Power et al., 2010; Marlon et al., 2008).”

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23) Line 4, page 824: (Linuma et al.

I checked and confirmed that the correct name is Dr. Yoshiteru linuma.

24) Line 15, page 824 : $\hat{A}n \hat{I}A$ but this trend is not is not $\hat{A}z \hat{I}\tilde{G}$ remove $\hat{A}n \hat{I}A$ is not $\hat{A}z \hat{I}\tilde{G}$

We corrected the typing error and we removed “is not” from the sentence at line 403 – 405. “A decreasing amount of ammonium originating from biomass burning is also deposited in the GRIP ice core (Fuhrer et al., 1996) but this trend is not clearly supported by levoglucosan data during the past two millennia (Fig. 3B).”

25) Line 24, page 824: “which are” probably “underrepresented

We added “probably” at lines 410 - 412. “We infer that the 1500 - 1700 CE maximum in fire activity is due to increased Eurasian boreal forest fires (Figure 3), which are probably underrepresented in the GCD, reflecting the bias in the geographical distribution of GCD sites.”

However Marlon et al. (2008) reports that “Although there are geographic gaps, there is good coverage of climatic zones and of all the major biomes except grassland/dry shrubland”, and this geographical gap is clear in the figures showing the charcoal records distribution (i.e. Marlon et al., 2013).

Marlon, J. R., Bartlein, P. J., Carcaillet, C., Gavin, D. G., Harrison, S. P., Higuera, P. E., Joos, F., Power, M. J., and Prentice, I. C.: Climate and human influences on global biomass burning over the past two millennia, *Nat. Geosci.*, 1, 697-702, 10.1038/ngeo313, 2008. Marlon, J. R., Bartlein, P. J., Danialu, A. L., Harrison, S. P., Maezumi, S. Y., Power, M. J., Tinner, W., and Vanniere, B.: Global biomass burning: a synthesis and review of Holocene paleofire records and their controls, *Quat. Sci. Rev.*, 65, 5-25, 10.1016/j.quascirev.2012.11.029, 2013.

26) Line 17, page 825: “the pronounced peak” of levoglucosan “centered on. . .”

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We modified lines 422 - 424 since your suggestion improves the text.

“The pronounced levoglucosan peak centered on ~ 1600 CE only partially overlaps with the pyrogenic methane record, where $\delta^{13}\text{C}$ peaks in the 1400s to 1500s.”

27) Line 9-14, page 827: sentence too long and unclear

We agree with your advice, and we divided the long sentence at line 465 – 470.

“Back trajectory modeling demonstrates the lack of correlation between BC and the specific coniferous fire marker vanillic acid measured in the Greenland D4 ice core. This lack may result from the majority of BC arriving to Greenland between 1850 to 1950 CE having a North American anthropogenic source, and from 1951 CE to the present having an Asian industrial source, separate from any fire activity (McConnell et al., 2007). Therefore, any analysis of Greenland BC from 1850 CE onward should not be considered a purely biomass burning indicator, and the sources for BC and fire emissions may substantially differ.”

28) Line 24, page 828: change “determines” by “suggest”

We changed the word “suggest” at line 503 - 504. “Our work suggests that Siberia is an important source of burning signatures inferred in Arctic ice fields during extreme fire events.”

29) Line 3, page 829: remove title “natural climate interactions” 30) Line 13, page 829: change 4.4.2 to 4.4.1

We removed the title, and the sub-chapter division 4.4.1, “Natural climate interactions”. Then we reorganized the following chapter numbers.

31) Line 23, page 829: “and in the GCD between 1000-1400 CE”, representing which composite curve?

We specified the GCD composite curve ($\text{HLNH} > 55^\circ$) that was compared to the levoglucosan trend, at lines 520 – 524.

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“Such temperature anomalies correlate with increased fire activity in paleofire reconstructions inferred from various environmental archives including the Eclipse Icefield in the Yukon, Canada (1240 - 1410 CE) (Yalcin et al., 2006), Eurocore (1200 - 1350 CE) (Savarino and Legrand, 1998) and GISP2 ice cores (1200 - 1600 CE) (Taylor et al., 1996), and in the HLNH > 55° GCD between 1000 - 1400 CE (Marlon et al., 2008).”

32) Line 3, page 830: “in the NH GCD” change to read “in the global NH curve (based on GCD syntheses)”

We modified the sentence as your suggestion improves the readability of the paper

Lines 528 – 530. “Low fire activity is observed in Eurocore (Summit, Greenland) (Savarino and Legrand, 1998) from 1600 - 1850 CE and in the global NH curve (based on GCD syntheses) centered around 1750 CE (Marlon et al., 2008).”

33) Line 12, page 830: change 4.4.3 to 4.4.2

Please, see points 29 and 30 of Referee #3

34) Line 10, page 831: add “in Asia” after “major droughts”

We added “in Asia” at lines 560 – 561. “Major droughts in Asia are associated with summer monsoon failures, and their multi-annual nature suggests that the droughts may have continued to occur through the winter (Table 2).”

35) Line 25, page 831: Change “our results show” to “we suggest that”

We modified lines 572 - 574 as suggested by Referee #3 “We suggest that Siberian fire activity is closely related to precipitation changes, where extreme biomass burning peaks are synchronous with precipitation anomalies, independently supporting GISS GCM model results (Pechony and Shindell, 2010).”

36) Line 19, page 832: change 4.4.4 to read 4.4.3

Please, see points 29 and 30 of Referee #3

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37) Line 24, page 832: change “the GCD synthesis” to read “Marlon et al. (2008) argues (. . .)” and remove reference at the end of the sentence.

We modified lines 595 - 597 as suggested.

“Marlon et al. (2008) argue that population growth and land-cover conversion rates, along with increases in global temperatures, are the main factors for the sharp increase in global biomass burning from 1750 CE to the late 19th century.”

38) Line 27, page 832: same comment, “However, Marlon et al. (2008) assert”

We modified lines 600 - 603 as suggested.

“However, Marlon et al. (2008) and Prentice (2010) assert that the expansion of agriculture and fire suppression since the early twentieth century resulted in decreased global fire activity, where current biomass burning rates may be lower than over the past 2000 years.”

39) Line 7, page 833: modify “observable in the GCD” to read “in the global NH curve (Marlon et al)”

We modified the text as suggested.

Lines 601 – 606. “Pyrogenic methane isotopic data (Ferretti et al., 2005; Mischler et al., 2009), which show relatively high values in the first millennium CE, do not support the prominent maximum of an anthropogenic peak at ~ 1900 CE in the HLNH > 55° GCD curve (Marlon et al., 2008) and inferred from CO isotopic measurements (Wang et al., 2010), and instead demonstrate that pyrogenic methane sources are still increasing with higher rates than during the last 2 millennia.”

40) Line 17, page 833: anthropogenic peak at 1900 CE or 1750 CE? Be consistent

In the chapter 4.4.3 we discuss both the onset of anthropogenic influences and the recent increasing trend in biomass burning, which is not supported by all the pyromarkers. In order to avoid a misunderstanding and to be consistent, we specified that

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at line 605 we are discussing the timing of the fire activity maximum (~ 1900 CE) and not the onset of the anthropogenic peak. We modified the text at line 601 – 606 as follows:

“Pyrogenic methane isotopic data (Ferretti et al., 2005; Mischler et al., 2009), which show relatively high values in the first millennium CE, do not support the prominent maximum of an anthropogenic peak at ~ 1900 CE in the HLNH $> 55^\circ$ GCD curve (Marlon et al., 2008) and inferred from CO isotopic measurements (Wang et al., 2010), and instead demonstrate that pyrogenic methane sources are still increasing with higher rates than during the last 2 millennia.”

41) Line 1-4, page 835: sentence too long and unclear

Please, see answer to Referee #1 point 14.

42) Line 21, page 835: remove lines 21 to 24.

We removed the retyped text from the acknowledgements section

43) Table 2a: add a comment of the meaning of tree-ring proxy in the table

We modified Table description, highlighting the importance of tree-ring analysis in inferring paleodroughts.

Line 1152. “Table 2. Asian droughts recorded in various proxies with strong fire events (a) and with centennial fire activity in the 16th and 17th century inferred from levoglucosan analysis (b). Generally drought is recorded in tree-rings as a sustained narrowness of growth rings.”

44) Figure 1: add locations of other records discussed in the text (such as the Belukha ice core)

We think adding positions of environmental archives discussed in the text strongly helps the reader realise the locations of the ice cores and have added these locations to Fig. 1.

45) Figure 2a: add all dates for significant peaks in levoglucosan; change scale in AD (year) by CE/BCE to be consistent with the main text.

We changed the scale in Fig. 2 to be consistent with the text. We did not add all dates of significant levoglucosan peaks since this paper is not a levoglucosan-based paper, but is a multi-approach reconstruction of past fire activity. If we add all dates for significant peaks, we should add all dates for significant peaks of other markers as well. But this would make the figure very confusing. Thus, we prefer indicate in figure only the fire events inferred by all the three markers. Other mega and strong levoglucosan events are indicated in Table 1.

46) Table S1 and S2: add the meaning of numbers in bold (significant correlation?)

We specify in the Table S1 and S2 captions the meaning of numbers in red and in bold, at lines 98 - 100 and 106 - 108 of Supplementary Information. Thank you for your suggestion.

“Red numbers emphasize correlations between levoglucosan and crustal markers Ca^{++} and dust. Numbers in bold emphasize correlations between levoglucosan and NH_4^+ .”

47) Figure S5: change 1974 to read 1973 as in Table 1

We changed “1974” to read “1973” in Fig. S5.

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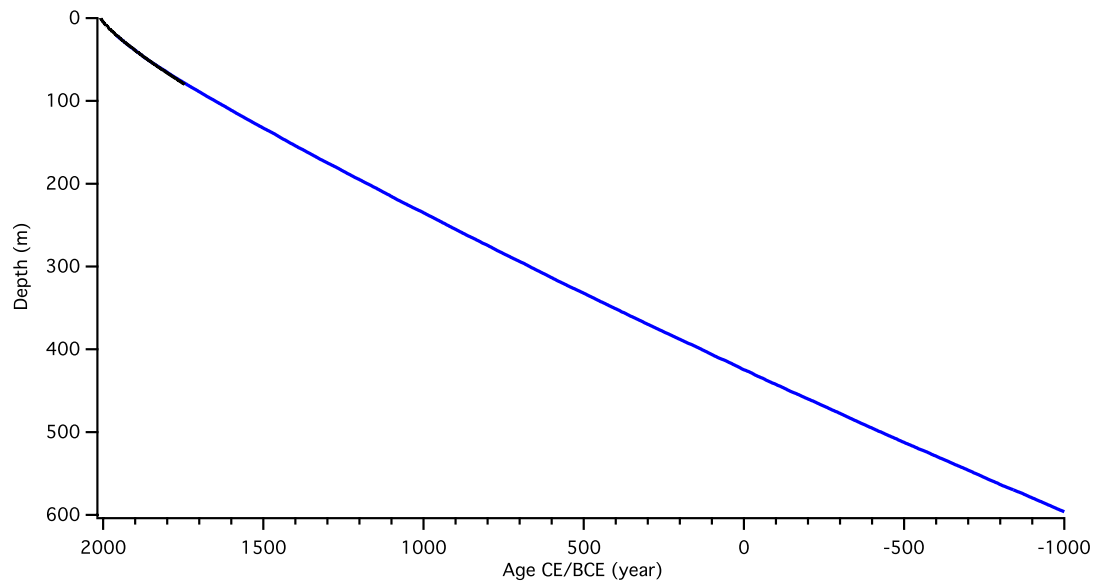


Fig. 1. Figure S7. Age model representation.

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