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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.

Anonymous Referee #2

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

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. 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 most others, such as lightweight carboxylic acids, lignin and resin pyrolysis products, and major ions (K+, NH4+ and NO3-) 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. 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. Finally, it is no clear in your sentence which chemical species among K+, NH4+ or NO3- is related to volcanic eruptions ?! Please comment.

Again the sentence (lines 2-5, page 813) "Potassium (K+) and ammonium (NH4+), 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.

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).

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.

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-hydroxybenzois 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.

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 together. As discussed below, in the case of Greenland, there is no raison 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.

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). 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.

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.

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 ????

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).

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 $(NH_4)_2SO_4$ 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 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).

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.

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 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 fore 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-hydroxybenzois 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

Press,Cambridge,Mass., 1991.

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 âĹij 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 hP (i.e; the elevation of Summit) as well as 500 hPa (i.e. 5-6 km asl) that reflect midtropospheric circulation. 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).

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.

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

End of the review

Interactive comment on Clim. Past Discuss., 10, 809, 2014.