

Response to P. Vallelonga (Referee #1)

General comments

The manuscript “Boreal fire records in Northern Hemisphere ice cores: A review” by Legrand and co-authors provides a thorough evaluation of the major proxies used to evaluate biomass burning activity from Greenland and alpine ice cores. There is an appropriate treatment of the chemical precursors to biomass burning proxies and the fire processes involved in their emission. The manuscript provides a good summary of the state-of-the-art for these techniques as well as an outlook on problems and opportunities for future research. The manuscript could benefit from some small improvements suggested below in the specific comments, but is already in a very good state. I will be happy to provide NEGIS data to develop the discussion regarding the 1908 event described below.

Specific comments

Continuous measurement of TOC. The Sievers 900 technique described in section 2 is novel and should be described in additional detail. Given the technique is susceptible to contamination by drill liquid, it would be good to show data from the dry-drilled TUNU core, for which ammonium data is also available. *We agree and in the revised version we added (section 2) “Recent improvements in sample handling and other techniques in the CFA system at DRI enable arguably the first reliable high-resolution measurements of total organic carbon (TOC). In this new method, a Sievers 900 TOC analyzer is coupled to an ice core melter. The constantly flowing sample stream (isolated from any interaction with laboratory air and after minimal contact with plastics and other sources of contamination in the flow lines) is analysed within a few minutes of initial melting. Total OC is determined in the Sievers 900 analyzer as the difference in total carbon and inorganic carbon. UV radiation and ammonium persulfate are used to oxidize organic compounds to CO₂, and the CO₂ is measured with a patented selective membrane-based conductometric detection method (details available in the Sievers 900 Series Total Organic Carbon Analyzers, Operation and Maintenance Manual, GE Analytical Instruments, 2011). We emphasize that for both DOC (Legrand et al., 2013) and TOC, measurements are most reliable in ice cores drilled without use of organic drilling fluids and in samples from below the pore close-off depth where potential contamination from circulating modern air through the core is eliminated.”*

Concerning the possible contamination when a drill fluid is used we wrote (section 3.2.4) : “As seen in Fig. 8, a very similar picture was obtained at DRI using CFA coupled with a TOC analyser. Although limited, these data highlight three important points. First, the similar OC background levels observed in the dry-drilled Summit-2015-Place ice core and in the wet-drilled North GRIP2 and NEEM cores (Fig. 8), suggests that the use of drill fluid has not significantly contaminated the North GRIP2 and NEEM ice.”

Data used for evaluation. The treatment of geographical distribution of ammonium signals in Greenland (discussed in section 4 and shown in figures 9, 14, 15) would benefit from the inclusion of ice core data from Northeast Greenland, such as the TUNU and NEGIS (Vallelonga et al., 2014, The Cryosphere, doi: 10.5194/tc-8-1275- 2014) sites. For example, the 1908 ammonium peak is visible in both the TUNU and NEGIS records, pointing to a widespread signal across Greenland, that is curiously absent at NEEM. The absence of such a signal at NEEM may be indicative of an atmospheric transport path that does not arrive at NEEM and is perhaps distinct from that which transports aerosols from North America. *This*

point was further discussed following email exchanges between Paul and Michel. Since the recommendation of the second reviewer was to minimize the discussion of the Tunguska and that, at the opposite of the case of Summit, NEEM and D4, for which continuous records covering the last 100 years or the last millennium are already published and presented in this paper (see Figures 9 and 14), we decided that we will mention at the end of the first paragraph of section 2 (Previously published and unpublished data) as follows: “Finally, recent and new chemical investigations, including ammonium (not shown), document biomass-burning fallout in ice cores extracted at the NEGIS (Vallelonga et al., 2014) and Tunu-2013 sites (Sigl et al., 2015). Ongoing studies conducted on northeast Greenland ice cores will complement results discussed here.”

Levoglucosan (section 3.2.5) The authors have rightly pointed out that levoglucosan records are not available in annual or sub annual resolution, which limits comparisons to other biomass burning proxies such as ammonium or BC. Given that levoglucosan measurements are relatively slow and laborious, it is unlikely that annually-resolved data will be produced in the foreseeable future. Consequently, it is important for a critical review such as this one to provide a critical evaluation of the available data. The authors should be more explicit regarding their observation that levoglucosan data produced to date is inconsistent with ammonium and BC records (sections 4.2 and 4.3) and this should be highlighted as an important concern to be resolved in future studies. *The discussion on Levoglucosan was updated (also considering comments from the second reviewers). First, at the end of section 3.2.5 (Levoglucosan) we wrote: “These laboratory studies suggest an atmospheric lifetime of levoglucosan against chemical degradation of approximately two days (Lai et al., 2014), or from two days to two weeks (Slade and Knopf, 1013). Given the lower range of estimated levoglucosan lifetime against chemical degradation we cannot rule out that chemical loss represent a significant loss for levoglucosan additional to the depositional loss that would apply to all biomass burning aerosol. “*

Second, at the end of paragraph 4.2, we added “Note that this conclusion conflicts with the hypothesis of a significant chemical degradation of levoglucosan suggested by laboratory studies (Sect. 3.2.5). More work is needed to elucidate why (1) levoglucosan might reflect Siberian fire activity in Greenland ice but not BC or ammonium, and (2) the finding that the levoglucosan record seems to mirror changes of fire activity at a larger spatial scales (Eurasia plus Canada) than ammonium records.”

Third, in the conclusion we added “Further work dedicated to high-resolution measurements of levoglucosan also would be welcome. Such measurements would enable improved understanding of the cause of the observed difference in past fire activity changes derived from levoglucosan and ammonium records. A large amount of ice would be needed to achieve high-resolution levoglucosan ice core profiles, but sampling Greenland snow in snow-pits would be a useful alternative. For instance, a 5 m depth pit at Summit would span the last 10 years during which numerous fire events have occurred in North America and Siberia that are well documented by satellite observations. “