Clim. Past Discuss.,
https://doi.org/10.5194/cp-2018-46-AC2, 2018

Climate of the Past Discussions
Open Access
EGU

Interactive comment on "Burning-derived vanillic acid in an Arctic ice core from Tunu, Northeastern Greenland" *by* Mackenzie M. Grieman

Mackenzie M. Grieman

mgrieman@uci.edu

Received and published: 31 July 2018

We greatly appreciate the referee's comments. The manuscript has been edited as described below to take them into account.

1. "There should be some discussion of what in particular might be most informative/useful going forward in terms of new proxy comparisons, or modeling, etc."

"Moreover, the influence of different modes of climate variability on the VA record are nearly impossible to disentangle given that the decadal-scale variability in the VA record raises more questions than it addresses – it seems unclear still what exactly is producing the short-term variability in VA (in terms of fire), let alone what would cause the variations in fire themselves IF that is what the VA variations are reflecting."

Printer-friendly version

Discussion paper

The following has been added to the conclusion: “At this stage, ice core VA should be regarded as a qualitative tracer because it is not known to what extent the signals reflect paleofire emissions, paleofire frequency, or changes in air mass transport and deposition. Further work comparing VA in shallow ice cores to satellite measurements and modeling of fires during recent decades would improve our understanding of the origins of the VA signals in Greenland ice.”

2. “Furthermore, how might we better understand the source areas or relationships between VA and other fire proxies from ice (e.g. what benefits does VA provide in comparison with BC, ammonium, levoglucosan, etc.)? Where are the greatest sources of uncertainty? Clarifying the limitations of the existing study and adding some thoughts about ‘next steps’ would be helpful for those outside the ice core community in particular.”

The following description of other ice core biomass burning proxies has been added to the introduction: “A range of ice core proxies have been used to reconstruct biomass burning (Legrand et al., 2016; Rubino et al., 2015). Stable isotope ratios of ice core methane (δ13CH4) have been used to infer global biomass burning, using end member isotopic compositions of methane sources (Ferretti et al., 2005; Mischler et al., 2009). Elevated ammonium concurrent with elevated levels of other chemicals has been used to reconstruct regional biomass burning. The difficulty of using ice core ammonium is that it is derived from several sources (Rubino et al., 2015). Ice core black carbon has been used as a tracer for preindustrial burning. Differences between these records could be due to variability in combustion conditions and transport (Rubino et al., 2015). The incomplete combustion of biomass produces organic aerosols. A large percentage of the biomass burning-derived organic aerosol is composed of levoglucosan, which is emitted by all plant matter containing cellulose (Simoneit et al., 1999). However, levoglucosan has the potential for rapid degradation during atmospheric transport (Hennigan et al., 2010; Hoffmann et al., 2010; Slade et al., 2013).”

Added References:

Ferretti, D. F., Miller, J. B., White, J. W. C., Etheridge, D. M., Lassey, K. R., Lowe, D. C., Meure, C. M. M., Dreier, M. F., Trudinger, C. M., van Ommen, T. D., and Langenfelds, R. L.: Unexpected Changes to the Global Methane Budget over the Past 2000 Years, Science, 309, 1714–1717, doi:10.1126/science.1115193, 2005.

Hennigan, C. J., Sullivan, A. P., Collett, J. L., and Robinson, A. L.: Levoglucosan stability in biomass burning particles exposed to hydroxyl radicals, Geophysical Research Letters, 37, L09 806, doi:10.1029/2010GL043088, 2010.

Hoffmann, D., Tilgner, A., Iinuma, Y., and Herrmann, H.: Atmospheric stability of levoglucosan: a detailed laboratory and modeling study., Environmental Science and Technology, 44, 694–699, doi:10.1021/es902476f, 2010.

Sapart, C. J., Monteil, G., Prokopiou, M., van deWal, R. S.W., Kaplan, J. O., Sperlich, P., Krumhardt, K. M., van der Veen, C., Houweling, S., Krol, M. C., Blunier, T., Sowers, T., Martinerie, P., Witrant, E., Dahl-Jensen, D., and Rockmann, T.: Natural and anthropogenic variations in methane sources during the past two millennia, Nature, 490, 85–88, 10.1038/nature11461, 2012.

Slade, J. H. and Knopf, D. A.: Heterogeneous OH oxidation of biomass burning organic aerosol surrogate compounds: assessment of volatilization products and the role of OH concentration on the reactive uptake kinetics., Physical Chemistry Chemical Physics, 15, 5898–915, doi:10.1039/C3CP44695F, 2013.

3. “Section 3.3 – This section is describing methods but is located in the Results section – it seems more appropriate to methods. In addition, the 17% of the summer back trajectories reaching North America is quite low – where do all the trajectories that are not accounted for come from?”

The first paragraph of section 3.3 has been moved to the methods section (Section 2.3). They primarily originate from the ocean. A low percentage of the trajectories also transect Europe and Siberia.

4. “Section 3.6 also needs work. The methods referenced are entirely absent from the “Methods” section again and need to be moved and expanded there to understand what exactly was done (Pg. 7 line 19 is insufficient).”

This section describes a comparison to several climate proxy records presented in the publications referenced. The methods used to produce these records are described in the referenced publications and we think it is beyond the scope of this paper to describe them here.

5. “Furthermore, the features that are most robust in the VA record – high values during the RWP and MCA, and low values during the LALIA and LIA – are still not well understood. The RWP itself is not a well-known or widespread climate feature, so more information and background about that could be provided instead of discussion of possible climate modes that might control fire.”

The following has been added to the first paragraph of section 3.6 to define the lesser-known climate periods: “Climate proxies from the North Sea, the Qinghai–Tibet Plateau, southwest Greenland, Spain, Iceland, and other Northern Hemisphere locations have shown increased climatic variability around the period of the Roman Warm Period (Wang et al., 2012; Bianchi et al., 1999). Temperature records using tree ring chronologies from the Russian Altai and European Alps show the cooling episode defined as the Late Antique Little Ice Age. This period of cooling followed large volcanic eruptions (Buntgen et al., 2016). This period overlaps the Dark Ages Cold period, a period of colder climates spanning the Northern Hemisphere. The contributing factors for this period are under debate, but may involve ice-rafting events, North Atlantic Oscillation, and/or El Nino-Southern Oscillation (Helama et al., 2017). The characteristics of these climate periods depend on the location. For instance, proxy records of the Roman Warm Period indicate that the Mediterranean experienced a wet and humid climate episode (Wang et al., 2012).”

Reference added:

Bianchi, G. G. and McCave, 5 I. N.: Holocene periodicity in North Atlantic climate and deep-ocean flow south of Iceland, Nature, 397, 515, doi:10.1038/17362, 1999.

6. “I would rather see more investigation into understanding the potential relationships between the VA record and other fire proxies than a detailed investigation into the interannual climate controls on a very uncertain biomass burning reconstruction.”

The following has been added to section 3.4: “Levoglucosan and black carbon in the NEEM ice core from Northern Greenland are elevated from 200-600 CE and 100-700 CE, respectively (Fig. S8) (Zennaro et al., 2014). These periods overlap the period of elevated Tunu VA from 280-400 CE. They are still elevated 200-300 years after the Tunu VA record has declined. There is also an overlapping peak in the GISP2 ice core ammonium record from 320-330 CE (Chylek et al., 1995). NEEM levoglucosan, black carbon, and ammonium are also elevated from 1000-1200 CE, 1000-1600 CE, and 1200-1500 CE respectively, at about the same time as the peak in the Tunu VA record from 1080-1240 CE (Legrand et al., 2016; Zennaro et al., 2014). This peak is slightly earlier than a period of elevated ammonium, oxalate, and potassium in an ice core from the Eclipse ice field in western Canada from 1240-1410 CE (Yalcin et al., 2006). Periods of elevated burning in Greenland and Canadian ice core records after 1240 CE are not pronounced in the Tunu VA record. These periods include elevated NEEM levoglucosan from 1500-1700 CE, 2D and GISP2 ammonium from 1790-1810 CE and 1830-1910 CE, and periods of elevated burning in the Mt. Logan and Eclipse ice cores from western Canada in the 18th-20th centuries (Whitlow et al., 1994; Yalcin et al., 2006; Zennaro et al. 2014).”

Figure S8 showing a timeline of elevated periods of biomass burning in ice core records was also added to the supplement.

Added References:

Chylek, P., Johnson, B., Damiano, P. A., Taylor, K. C., and Clement, P.: Biomass burning record and black carbon in the GISP2 Ice Core, Geophysical Research Letters, 22,

89–92, doi:10.1029/94GL02841, 1995.

Whitlow, S., Mayewski, P., Dibb, J., Holdsworth, G., and Twickler, M.: An ice-core-based record of biomass burning in the Arctic and Subarctic, 1750–1980, Tellus B: Chemical and Physical Meteorology, 46, 234–342, doi:10.3402/tellusb.v46i3.15794, 1994.

Yalcin, K., Wake, C. P., Kreutz, K. J., and Whitlow, S. I.: A 1000-yr record of forest fire activity from Eclipse Icefield, Yukon, Canada, The Holocene, 16, 200–209, doi:10.1191/0959683606hl920rp, 2006.

7. “Pg 2. VA is described as resulting from the combustion of lignin. McConnell et al. (2007) indicated that conifers in particular are expected to be the primary source of VA – it seems worth noting this again, and also providing more information that might be useful based on the extensive previous research on VA that is cited. E.g. what is the expected atmospheric residence time of VA?”

The following has been added to paragraph 4 of the introduction: “These studies have shown that North American and European conifer and deciduous tree species produce vanillic acid (VA) (Simoneit, 2002, Oros and Simoneit et al. 2001a, b, Iinuma et al., 2007). VA is observed in atmospheric aerosols in the Arctic and Antarctic after long-distance transport (Zangrando et al., 2013, 2016}. The lifetime of aromatic acids in the gas phase is on the order of a day due to oxidation by the hydroxyl radical. However, modeling studies suggest that in aerosols these compounds may be shielded from oxidation, resulting in atmospheric lifetimes of several days (Donahue et al., 2013). Laboratory and field studies have also shown decreased volatility of low molecular weight organic acids in aerosol form as a result of interactions with sea salt and other cations. Such studies have not yet been carried out on aromatic acids (Häkkinen et al., 2014, Laskin et al., 2012).”

Added References:

Donahue, N., Chuang,W., Epstein, S., Kroll, J.,Worsnop, D., Robinson, A., Adams,

P., and Pandis, S.:Why do organic aerosols exist? Understanding aerosol lifetimes using the two-dimensional volatility basis set, Environmental Chemistry, 10, 151–157, doi:10.1071/EN13022, 2013.

Häkkinen, S. A. K., McNeill, V. F., and Riipinen, I.: Effect of Inorganic Salts on the Volatility of Organic Acids, Environmental Science & Technology, 48, 13 718–13 726, doi:10.1021/es5033103, 2014.

Laskin, A., Moffet, R. C., Gilles, M. K., Fast, J. D., Zaveri, R. A., Wang, B., Nigge, P., and Shutthanandan, J.: Tropospheric chemistry of internally mixed sea salt and organic particles: Surprising reactivity of NaCl with weak organic acids, Journal of Geophysical Research: Atmospheres, 117, D15 302, doi:10.1029/2012JD017743, 2012.

Zangrando, R., Barbaro, E., Zennaro, P., Rossi, S., Kehrwald, N. M., Gabrieli, J., Barbante, C., and Gambaro, A.: Molecular markers of biomass burning in Arctic aerosols, Environmental science & technology, 47, 8565–8574, doi:10.1021/es400125r, 2013.

Zangrando, R., Barbaro, E., Vecchiato, M., Kehrwald, N. M., Barbante, C., and Gambaro, A.: Levoglucosan and phenols in Antarctic marine, coastal and plateau aerosols, Science of The Total Environment, 544, 606 – 616, doi:10.1016/j.scitotenv.2015.11.166, 2016.

8. “Pg 4 Line 6: Were higher thresholds tried? Would thresholds in the 80s or 90s produce very different results? Why did you choose this range?”

The following has been added to paragraph 3 of section 3.4: “Increasing the threshold from the 80th-95th percentiles reduces the number of peaks, but does not significantly alter their timing.”

9. “Pg 4 Line 8: remove “Clearly” and explain what ‘these’ refers to.”

Edit complete and removed sentence: “These are robust features of the Tunu VA record.”

10. “Pg 4 Line 12: should say ‘fires”’

Edit Complete

11. “Pg 4 line 20: define the terms in the equation and make your words consistent with the equation. Pg 4 Lines 26-80 – the equation and its description are not rendering properly (esp. check the capitalization and subscripting; vd, rscav).”

Equation and equation term definitions edited as follows:

“Ftotal = Fdry + Fwet = Cair Vd + Cair Rscav PH2O,

where Ftotal is the total VA flux, Fdry is the VA dry deposition flux, Fwet is the VA wet deposition flux, where Cair is the concentration of VA in the atmosphere, Vd is a dry deposition velocity for the VA-containing aerosols, Rscav is the wet deposition scavenging ratio, and PH2O is the snow precipitation rate (Saltzman et al., 1997).”

12. “Nor is there discussion of the more recent variations and how they compare with historical data, which is a large omission that needs to be addressed, especially given the 2.5-year temporal resolution of the ice core record noted in Section 2.1.”

“Fig. 4 – Please discuss this figure and how it relates to the known fire history of NA. Fire history data are available from tree-ring (e.g. Giardin’s papers, the Canadian Large Fire Database) and historical studies and the data raise important concerns that warrant more discussion. Specifically, the high VA values in the 1960’s and 70’s are puzzling given that this pattern is directly opposite what historical records show (e.g. Mouillot and Field, 2005; Mouillot et al. 2006), where burning was very low in the middle part of the century. Many more recent NA fire records generally show that burning has increased most rapidly from low levels since the 1980’s (NIFC, Littell et al. 2009). Is it possible that VA could at times have been influenced by some intensive industrial or lumber/logging-related processes that might have been occurring during this time in NA? In any case, in light of these data it is good that the authors state that VA is best considered a qualitative proxy at this stage and requires further exploration – this

acknowledgement is appreciated."

The following has been added to section 3.1: "Burned area in the Boreal forest of North America was high in the early 1900s, declined into the middle of the twentieth century, and began to increase again beginning in the 1960s (Mouillot and Field, 2005). The short-term period of elevated Tunu VA from 1955-1985 does not mirror the trend in this burned area record. A record of large boreal wildfires in Canada, with burned areas exceeding 200 ha, shows periods of large wildfires around 1960, the late 1970s to the early 1980s, around 1990, and the mid-1990s (Stocks et al., 2003). The similarity between the Tunu VA record and large boreal fire record from Canada suggests that that twentieth century VA record may be showing large fires. These large fires represent 3.1% of the number of Canadian fires from 1959-1997, but 97% of the area burned (Stocks et al., 2003)."

Added References:

Mouillot, F. and Field, C. B.: Fire history and the global carbon budget: a 1_ 1 fire history reconstruction for the 20th century, Global Change Biology, 11, 398–420, doi:10.1111/j.1365-2486.2005.00920.x, 2005.

Stocks, B., Mason, J., Todd, J., Bosch, E., Wotton, B., Amiro, B., Flannigan, M., Hirsch, K., Logan, K., Martell, D., et al.: Large forest fires in Canada, 1959–1997, Journal of Geophysical Research: Atmospheres, 107, FFR–5, doi:10.1029/2001JD000484, 2002.

Interactive comment on Clim. Past Discuss., https://doi.org/10.5194/cp-2018-46, 2018.