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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Historical black carbon deposition in the Canadian High Arctic: A 190-year long ice-core record from Devon Island" *by* Christian M. Zdanowicz et al.

Anonymous Referee #2

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Review Zdanowicz et al.: Historical black carbon deposition in the Canadian High Arctic: A 190-year long ice-core record from Devon Island

General Comments:

The authors present a reconstruction of black carbon and microparticle concentrations from an ice core from Devon Ice Cap in Canada covering the time period 1810-1990 AD. Where direct observations of atmospheric BC are scarce and limited to the most recent decades, ice-cores can – in principle – act as surrogates for direct observations and provide valuable information on the composition of the pre-industrial atmosphere and serve as benchmarks to assess the capabilities of models to realistically simu-

late the aerosol life-cycle and resulting forcing of past climate. In order to use such proxy reconstructions such glacio-chemical records need to realistically represent the atmospheric impurity content through time. In this respect, the current manuscript falls short in providing sufficient evidence for the reasons summarized below: The authors provide virtually no information that would allow to assessing their ability to achieve reproducible BC concentrations from ice cores or to repeat their experiments. They fail to report how they calibrated their measurements and omit to discuss any metrics (e.g., detection limit, stability, linearity, stability, repeatability, reproducibility) commonly considered necessary when introducing new instrumentation in analytical chemistry (see for example (Lim et al., 2014;Wendl et al., 2014;Mori et al., 2016;Bigler et al., 2011).

The same is true for the age-scales. The entire dating depends critically on the correct identification of the Laki signal in 1783 AD at this specific ice-core site and also for the other ice cores from Devon, but I can find in none of their cited papers a graph showing the full EC or SO4 record used to make this attribution. The same is true for the proxy signature of the 1963 AD nuclear bomb testing fallout. With no electrical or glacio-chemical signature of Laki provided in the manuscript and with having a huge sulfate spike recorded in 1847 AD which is not recorded in any other ice core from nearby Greenland, I consider it equally likely that the latter signal may as well be from Laki 1783, and your timescale off by over 60 years.

I consider it very unfortunate and not sufficiently well explained why you chose to limit your analyses to 1810-1990 AD and to only two new parameters. As stated above, Laki is crucial for the depth-age scale; the past 10 years would allow to have overlap with aerosol observations (e.g. from Alert); and analyzing additional aerosols in DV99.1 would allow you to 1) assess the effects of melting on your impurity records, 2) improve the relative dating among the different ice cores and timescales, and 3) to attribute specific sources to BC using for example NH4+ and SO42- as unique source tracers. The fact that the ice is fractured (>38m) or not consolidated (>4m) does not make measurements impossible, and half of an ice-core minus 2.5 x 2.5 cm consumed for your

CFA measurements should provide you with enough material for additional analyses.

Specific Comments:

Page 2: L. 25: How deep was the core, did you reach bedrock. Is this the same 170.6m long core as described by (Zheng et al., 2007) as D1999 core?

Page 3: L. 6: Surprising to see half of the core consumed for EC measurements and low resolution d18O analyses. What is the diameter of the cores? L. 9: What is the reference for the initial age estimate? L. 10-11: Zheng et al., (2007) report that the core quality was good for the entire core D1999? Is this the same core? If so, which statement is correct? Why don't you add a table with all meta data for ice cores and analyses you discuss in your manuscript? L.10: Why should unconsolidated snow not be useable for analyses? It is virtually impossible to contaminate with BC and analyses could have easily been performed with an SP2 on discrete samples. L12: When did the analysis take place? Over how many days, weeks, months? Did you observe sublimation on the ice surface after >15 years of storage? L13: What was class100? The cold room? The lab space? L12-14: I am missing references and I have never heard of an Advanced Ultra-clean Environmental facility. Is this the first time you are performing this kind of analyses in this lab? L14: At which melt rates did you melt the ice? How do you assure the flowrate is constant? It must be difficult with the frequent change of sold ice lenses (40-60% on average) and soft firn. If the flowrate is not constant, how do you correct for this? L14-21: Provide a chart with the analytical setup of all instruments. Provide information on calibration (standard material, linearity, stability) and reproducibility of the results. L.17-18: These citations are all for a lab in the USA L.20: How does the microparticle content connect to your scientific problem? What is the motivation? L. 24-25: Why did you not analyze these other aerosol species directly in DV99.1? There should be half of a core minus 2.5 x 2.5 cm of cross section left. This would allow you attribute with more confidence sources to biomass burning and coal burning. Comparisons to other ice cores drilled at different sites only allow you to compare some general trends. According to your age model you have but one

common age marker between these ice-core records (the alleged 1783 signal), strong spatial gradients in accumulation caused by wind erosion and/or melting.

Page 4: L. 4-12: Is any of this age models published? If so, please add the citation and the timescale name for the ice cores, respectively. It appears 2 of 3 citations at the end of this section are based on Agassiz ice cap. I do not find any figure showing the signatures attributed to Laki and Katmai in Kinnard et al. (2006). L. 4-12: Since your study is critically dependent on the correct identification of the two reference horizons 1783 and 1963, I expect to see all the data that was used for these attributions. It is very plausible that very large acidity in the Arctic were caused by the Laki eruption (Kekonen et al., 2005) but there may also other large acid layers recorded in the Arctic in e.g., 1765, 1815 (Wendl et al., 2015). Equally, the Arctic nuclear fallout signals are in general much broader (1954-1963, (Arienzo et al., 2016)) than described here. How sharp is your signal compared to these other ice cores? Maybe this could tell you something about potential redistribution of impurities caused by melting. L. 15: Provide these EC measurements for the DV99.1 ice core. How reliable is this peak, if it was recorded in the fractured ice-core sections >38m. Was it reproduced by sulfate analyses? L. 17: What accumulation rates to you get between year of drilling, 1963 and 1783 for each of the DV ice cores? Consider providing this information in supplement. L. 24-26: Please show these common signatures from DV99.1, DV98.1, DV98.3 L. 26: What do you mean with adopted? How much depth-age models exist? Where are they published? Are there any isochrones between these age-models and ice cores? L. 28: How much meters apart were DV98.3 and DV2000 drilled? Is it appropriate to use the same chronology for two different cores given that snow fall and snow conservation on summits are varying on very small spatial scales. I would only adopt a timescale for another ice core if this was supported by a number of isochrones. L. 31: I doubt the true effective resolution of the measurements is at a mm scale. You may be recording data at a rate equivalent to mm in depth, but you need to account for the uncertainty in the depth registration and dispersion of the signals through mixing (Bigler et al., 2011).

Page 5: L. 4-12: Add analytical uncertainties as well. L. 6: (e.g. wind scouring, melt induced relocation). According to Kinnard et al. (2006) average melt rates in D99 are 50% after 1850 AD, which appears to me a significant factor that could modify the impurity records one way or the other. L. 9-18: This approach assumes the layer thickness variation is the only source of uncertainty in estimating the age error. It assumes the Laki event is correctly attributed, and must take into account some prior knowledge of the snow accumulation rates and its variability. I don't see how you can estimate the variability "between reference layers of known age" without being able to count annual layers.

Page 6: L. 17-18: Microparticle concentrations do not follow the same trend than nssS and Pb but have a clear step-function. On which observation do you base your attribution of "anthropogenic pollution"? L. 22: Such source attributions would strongly benefit of having all parameters analyzed on the same core. L. 23: Black carbon concentrations

Page 7: L. 12: Necessary not only legitimate L. 14-30: I agree on this point.

Page 8: L. 6-17: Are the sections in the ice with the increased melt layer occurrence believed to be the periods experiencing more melting? Or are they accumulating the meltwater (plus impurities) from the ice sections above? In other words: how deep does percolation go? Is there surface runoff carrying impurities away? Given that DV is only 700 km away from NEEM and Humboldt ice cores and all ice cores agreeing on showing strong BC deposition in early 20th century I tend to believe the differences in BC deposition is not from differences in atmospheric burden, but from some aspect specific to the Devon ice cap. The low elevation and observed melt features appear to make a strong case that the impurities along the ice core may be subject to severe loss and/or redistribution in particular during warmer time periods (e.g. Arctic warming 1920-40s, (Yamanouchi, 2011)), which would smear and bias any atmospheric information. L. 18-28: Post-depositional coagulation moving BC sizes out of the detectable range also seems a very plausible explanation for reduced BC recovery during es-

pecially warm periods. Low reproducibility of replicate measurements when samples were subject to melting and freezing cycles is reported by several research groups performing BC analyses in ice and snow (apparent "loss" rates in the order of 50%); as a result performing BC analyses on samples that have been refrozen is strongly discouraged (see e.g. (Lim et al., 2014;Wendl et al., 2014)).

Page 9: L. 2-9: This may be true if you compared Devon Ice Cap with Central and Southern Greenland ice cores, but NEEM and Humboldt are just 700-800km away from Devon and are thought to have largely similar source regions for aerosols and precipitation (Zennaro et al., 2014).

Page 10: L. 2-26: How meaningful is such a comparison given the low degrees of freedom (resulting from decadal data), dating uncertainties and the inability to differentiate industrial from BB BC in the Devon ice core? Are these correlation stables if you varied binning, removed the common declining trend over most of the 20th century? L. 29-31: This is not surprising; as you outline below: K+, NH4+ and BC have multiple sources and probably also different chemical properties making them more or less susceptible to melt-induced relocation.

Page 11: L. 2: Could these low numbers for the most recent time period indicate some loss from melting caused by the rapid warming of the Arctic? To my knowledge and supported by Figs. 5 and 6 concentrations of rBC lower than during the pre-industrial baseline are not recorded for any other ice-core in the Arctic. L. 15: Given the potential limitations from inadequate nebulization, potential loss and redistribution of impurities, I strongly doubt that this value is a realistic approximation of the true atmospheric BC influx. L. 25: Note that you use the same abbreviation EC for both electrical conductivity and elemental carbon.

Page 12: L1-2: Does Humboldt show similar melt features than Devon Ice cap? The agreement between Humboldt and the other Greenland records (in both BC and nssS) seems very high. L9-12: None of the emission inventories or any ice core suggests

that mean BC emissions from 1960-1990 were below preindustrial (i.e. before 1850 AD) levels, as the Devon ice-core seems to imply.

Figures: Fig. 4: Extend the x-axis to include your only reference marker in 1783 AD. Units in panel b are missing and it is nssSO42-. I do not see any signature related to the largest VEI=6 eruptions of Katmai, Krakatao, Tambora and 1809, but a huge SO4 signal around 1847 AD. Do you have any explanation? How do you know this is not from Laki 1783 or Tambora 1815? How do you calculate K+BB? Fig.4 and Fig.6: DV98.3 nssSO4 (panel b, Fig 4) appears to be different from nssS (Fig. 6), the latter is peaking in the 1960-1980s, the first starts to peak only after 1980. Which one is correct? Fig. S3: Check the lower panel. There should be only one y-value for a given depth value

Technical Corrections: Page 2: L. 4: atmospheric chemistry climate models L. 14: add e.g. since the list is incomplete L22: ice core were previously drilled Page 3: L 29-30: Na+ (twice)

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