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Unexpected increase in elemental carbon values over the last 30 years observed in a Svalbard ice core

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Abstract

Black carbon (BC) is a light-absorbing particle that warms the atmosphere–Earth system. The climate effects of BC are amplified in the Arctic where its deposition on light surfaces decreases the albedo and causes earlier melt of snow and ice. Despite its suggested significant role in Arctic climate warming there is little information on BC concentrations and deposition in the past. Here we present results on BC (here operationally defined as elemental carbon (EC)) concentrations and deposition on a Svalbard glacier between 1700 and 2004. The inner part of a 125 m deep ice core from Holtedahlfonna glacier (79°8′ N, 13°16′ E, 1150 m a.s.l.) was melted, filtered through a quartz fibre filter and analysed for EC using a thermal optical method. The EC values started to increase after 1850 and peaked around 1910, similar to ice core records from Greenland. Strikingly, the EC values again increase rapidly between 1970 and 2004. This rise is not seen in Greenland ice cores and it seems to contradict atmospheric BC measurements indicating generally decreasing atmospheric BC concentrations since 1989 in the Arctic. Several hypotheses, such as changes in scavenging efficiencies, post-depositional processes and differences in the vertical distribution of BC in the atmosphere, are discussed for the differences between the Svalbard and Greenland ice core records, and the ice core and atmospheric measurements in Svalbard. In addition, the divergent BC trends between Greenland and Svalbard ice cores may be caused by differences in the analytical methods used, including the operational definitions of quantified particles, and detection efficiencies of different-sized BC particles. Regardless of the cause of the increasing EC values in the recent decades, the results have significant implications for the past radiative energy balance at the coring site.

1 Introduction

During the last century the Arctic has warmed twice as fast as the rest of the world which has been suggested to be explained by changes in albedo and related feed-

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backs in the Arctic covered with high reflectivity snow and ice (ACIA, 2005). Black carbon (BC) particles are produced by incomplete combustion of biomass as well as fossil and biofuels. Due to its color BC absorbs light and it is recognized as a strong warming agent in the atmosphere (e.g. Bond and Bergstrom, 2006; Ramanathan and Carmichael, 2008). The climate effects of BC are intensified in snow and ice where it lowers their reflectivity which leads to earlier spring melt (e.g. Warren and Wiscombe, 1980; Hansen and Nazarenko, 2004; Flanner et al., 2007; Bond et al., 2013). BC has been estimated to be the globally second most important climate warming agent after carbon dioxide, and in the Arctic even more important than greenhouse gases (Jacobson, 2001; Hansen and Nazarenko, 2004; Bond et al., 2013). Furthermore, the BC-albedo effect has been suggested to have caused 20 % of the warming and snow-ice cover loss in the Arctic during the last century (Koch et al., 2011). Yet, the effects of BC remain one of the largest sources of uncertainty in climate change analysis and models (e.g. Bond et al., 2013).

Due to its importance in climate perturbation BC distribution and concentrations have been increasingly studied around the world. The first continuous measurements on atmospheric BC concentrations in the Arctic started in the late 1980s (e.g. Hirdman et al., 2010a). The number and spatial coverage of BC measurements from arctic snow are increasing (e.g. Clarke and Noone, 1985; Forsström et al., 2009, 2013; Doherty et al., 2010, 2013; Aamaas et al., 2011), but comparing the individual results and establishing regional trends remain challenging because most snow measurements only represent a snap shot of BC concentrations in time and space. Despite the significance of BC in Arctic climate warming and geophysical processes, knowledge on its long-term concentrations and climate effects in the area beyond the observational data is very limited at present.

McConnell et al. (2007) and McConnell and Edwards (2008) were the first to report Arctic BC concentration and deposition trends, based on three ice cores from the Greenland Ice Sheet. McConnell et al. (2007) found a seven-fold increase in BC concentrations from 1850, peaking around 1910, followed by a decline to almost pre-

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industrial levels after 1950. This study was highly influential in introducing a new high resolution method to measure BC concentrations with a Single Particle Soot Photometer (SP2) in ice cores and was soon followed by other long-term studies on BC trends from Himalayan glaciers (e.g., Kaspari et al., 2011; Jenkins et al., 2013) and the Antarctic ice sheet (Bisiaux et al., 2012a, b), while European Alps glaciers had already been studied with other methods (e.g. Lavanchy et al., 1999; Jenk et al., 2006; Legrand et al., 2007; Thevenon et al., 2009). However, McConnell et al. (2007) recognized that the BC deposition at the high-elevation sites in Greenland could not be taken as representative of overall BC trends in lower elevation regions of the Arctic. The heterogeneity of the global distribution of BC is seen, for instance, in the different concentrations and trends between Himalayan, Arctic and European records, and even between individual records from these areas (e.g., Ming et al., 2008; Xu et al., 2009; Kaspari et al., 2011; Jenkins et al., 2013). To get a better overall view on its trends and effects, further investigations on BC concentrations and deposition are required from other locations in the Arctic.

Here we present results on BC (here operationally defined as elemental carbon, EC, due to the thermal-optical measurement technique used) concentrations and deposition from a Svalbard glacier, Holtedahlfonna, from 1700 to 2004. When discussing black carbon in general, we use the abbreviation BC, as is commonly used in the community. When discussing our specific measurements, or measurements made by others with similar thermal-optical methods, we use the abbreviation EC. For a recent review of BC terminology we refer to Petzold et al. (2013). We find an unexpected increase in EC values since the 1970s. Factors such as atmospheric BC concentrations, changes in BC scavenging efficiencies and post-depositional processes, are discussed in order to explain the observed trend. In addition to BC records from Svalbard being of particular interest in light of the limited data available from the Arctic, Svalbard glaciers are valuable as they are expected to have a different source attribution profile than Greenland (Hirdman et al., 2010b). Glaciers around the world are susceptible to melt and retreat caused by climate warming and possibly also BC deposition. As glaciers play an impor-

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tant part in climate feedbacks and the hydrological cycle, and as high-elevation glaciers at lower latitudes supply water to major human populations, it is crucial to learn more about the BC-glacier interactions.

2 Material and methods

2.1 Ice core recovery and sampling

Svalbard is located in the Arctic Ocean (Fig. 1) at the southern edge of the permanent sea ice. Despite its location at high latitudes, the archipelago has a relatively mild climate due to an intrusion of the North Atlantic current bordering western Svalbard and its location on the pathway of both Arctic and North Atlantic cyclones. About 60 % of Svalbard is covered by glaciers of which the majority has had a negative total volume change during the last 15–40 years (Nuth et al., 2010). Even the highest elevation glaciers on Svalbard, such as Lomonosovfonna (Fig. 1), can experience frequent surface melt in the summer (e.g. Beaudon et al., 2013).

Holtedahlfonna is the largest ice field in the western island of Spitsbergen in Svalbard, covering ca. 300 km^2 and situated 40 km northeast of the Ny Ålesund research station (Fig. 1). The ice core was drilled in April 2005 at 79°8′15′′ N, 13°16′20′′ E, 1150 m elevation, at a saddle point where the lateral ice flow velocity is expected to be minimal (Lefauconnier et al., 2001; Sjögren et al., 2007). The retrieved 125 m deep ice core did not reach the bedrock and thus the precise ice depth at the coring site is unknown, but radar measurements suggest it to be approximately 150 m (Sjögren et al., 2007; Beaudon et al., 2013). The ice core was retrieved in ca. 50–60 cm sections and immediately packed into plastic bags, subsequently stored frozen and transported to the cold room facilities at the Norwegian Polar Institute (NPI), Tromsø, Norway.

The ice core was cut and processed in a cold room (−22 °C) using a cleaned thin blade band saw. Each vertical ice-core section was split to subsamples assigned to tritium (van der Wel et al., 2011), oxygen isotope (Divine et al., 2011), organic contami-

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nant (Ruggirello et al., 2010), major ion (Beaudon et al., 2013) and EC measurements. EC measurements were performed on subsamples cut from the inner part of the core, i.e. the part best protected from contamination during drilling and handling of the ice. The samples were continuous except that a section representing the time period 1740–1755 was not available for EC analysis. After preliminary cutting each surface of the subsamples was scraped with a clean stainless steel knife under a laminar bench, after which the samples were placed in plastic bags and stored frozen. After subsampling there were 739 ice pieces of 5–20 cm vertical length and an average horizontal cross section of 2.8 by 3.5 cm, equal to around 10 cm^2 ($\pm 2\ cm^2$) surface area, available for EC analysis.

Dating of the ice core was performed using an age-depth scale based on the ice thinning model by Nye (1963) constrained by the depth of the 1963 radioactivity fallout layer at 28.5 m depth (van der Wel et al., 2011), and counting of annual $\delta^{18}O$ layers (Divine et al., 2011). In addition, a dating method based on statistical extraction of historically known volcanic eruptions (Moore et al., 2012) complemented the other dating methods. Volcanic signatures of five known eruptions, including the Laki eruption (1783) at 103.6 m depth, could be used as reference horizons in a refined Nye thinning model, and suggest that the core covers a period of 305 years with a mean accumulation rate of 0.38 m.w.e. yr^{-1} (Moore et al., 2012).

2.2 Filtering and EC analysis

The 739 ice samples were grouped for filtering so that ice samples representing the 18th century were filtered in ca. 10 year resolution, of the period 1800–1850 with a 5 year resolution, and samples from the industrial era with approximately a 2 year resolution. The ice was melted and immediately filtered through pre-burned (at 800 °C for 4 h) quartz fibre filters (Munktel) following the procedures described, for instance, in Forsström et al. (2009) and Svensson et al. (2013). This resulted in 88 EC filter samples. In order to check for possible contamination in the filtering system, blanks

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(nine) were prepared by placing a filter in the filtering system for a minute or by filtering distilled water through a filter (three).

The filters were analysed for EC using a thermal optical method (TO, Sunset Laboratory Inc., Forest Grove, USA; Birch and Cary, 1996) at Stockholm University. The analysis was performed with the latest recommended thermal sequence EUSAAR_2 (Cavalli et al., 2010) in order to separate EC and other (carbonate and organic) carbon from the filters. Carbonates were not eliminated from the filters with acid treatment before the analysis, since this has been shown to cause some errors in the analysis, e.g. induction of intense charring phenomena (Jankowski et al., 2008), and a decrease in BC particle size (Kaspari et al., 2011; Schwarz et al., 2013). Moreover, natural calcite evolves in the Helium-mode with the EUSAAR_2 protocol, and will be detected as organic carbon (Cavalli et al., 2010). Therefore we concluded that natural carbonates would not interfere with the EC measurements. All blanks showed EC concentrations from below detection limit to a magnitude lower than the detection limit of the analysis method (0.2 EC $\mu g\,cm^{-2}$).

2.3 Uncertainties

In the filter based method used in this study, a 1.5 cm^2 filter punch was analysed from the middle part of each filter (total area 11.34 cm^2). Additional punches from the same filters showed non-significant variation in EC loadings between different parts of the filters (n = 8; average relative standard deviation = 3 %; range of relative standard deviation = 1.1–4.9 %). However, some of the filters had a visible gradient of deposited matter with one edge being lighter than the other, possibly due to problems keeping the filtering system completely in level. In the cases where the EC was not uniformly deposited, based on ocular inspection and the deviation from the three punches, the EC concentration in the middle of the filter was close to the average of the whole filter whereas the light side contained lower and the darker side higher EC concentrations (n = 16, average relative standard deviation = 19.6 %; range of relative standard deviation = 9.1–38.3 %). These deviation values are in accordance to previously reported

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average standard deviations of 21 % in Svensson et al. (2013). As expected, the relative standard deviations were highest for filters with low EC loadings. The analysed filter punch was in all cases taken from the middle of the filter. For the samples from which several filter punched were analysed the reported concentrations are averages of the filter punches and the error bars are shown in Fig. 2.

A known error source of the filter approach relates to the efficiency of the used filter to capture EC particles in liquid samples. Previous studies have shown that significant amounts of EC particles in liquid samples may percolate through the filter resulting in undercatch (e.g. Forsström et al., 2013; Doherty et al., 2013; Torres et al., 2014). Most likely these penetrating particles are small in size as they are not even captured by additional filters in series (Torres et al., 2014). However, Schwarz et al. (2013) suggested that atmospheric processes during the formation of snow and repeated thaw–freeze cycles in deposited snow may lead to agglomeration of BC particles and a subsequent shift to bigger particles in snow samples compared to atmospheric samples. The Holtedahlfonna ice core samples have most likely experienced several thaw–freeze cycles as the glacier experiences annual summer melt (Beaudon et al., 2013). Therefore, the EC particles in the ice core samples may tend to be bigger and be filtered with higher efficiency. Possible additional losses of EC particles through the filter were avoided by refraining from the acid treatment to eliminate carbonates from the filters.

3 Results and discussion

3.1 Holtedahlfonna EC concentrations and comparison to other European data

EC concentrations in the Holtedahlfonna ice core have varied significantly between 1700 and 2004 (Fig. 2). Between 1700 and 1850, the concentrations are generally low, on average 23 $\mu g\,L^{-1}$ (Table 1), showing no specific trend. In samples corresponding to the period between about 1850 to 2004 the concentrations are generally higher than for the oldest part of the ice core, on average 39 $\mu g\,L^{-1}$. Two local minima in the 1920's

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and 1970 are show clearly in the EC record. Peaks in EC concentrations occur at about 1910, 1940, and 1998 at 80 $\mu g\,L^{-1}$, 58 $\mu g\,L^{-1}$ and 103 $\mu g\,L^{-1}$, respectively.

A clear feature of the Holtedahlfonna EC record is the steady increase in EC concentrations after 1970. This trend is surprising as it contradicts previous data from the Arctic. Atmospheric BC measurements from the Arctic only go back to 1989, but the stations at Alert (Ellesmere Island, Canada), Barrow (Alaska, USA) and Zeppelin (Ny Ålesund, Svalbard, Norway), all record a decrease of BC concentrations to the present. This trend is explained by a general decrease in BC emissions in northern latitudes since the 1990s, in particular associated with the collapse of the Soviet Union (Hirdman et al., 2010a; Sharma et al., 2013). The BC ice core records from Greenland show decreasing BC values to almost pre-industrial levels during the 20th century after a clear peak around 1910 (McConnell et al., 2007; McConnell and Edwards, 2008; McConnell, 2010).

The magnitude of the Holtedahlfonna EC concentrations seems to be in accordance with EC values measured with other filter-based thermal, thermal-optical or other similar methods in ice cores from the European Alps, presented in Table 1. It may seem surprising that EC concentrations on Svalbard, which is more remote from emission sources, are comparable or even higher than on the glaciers in the European Alps. However, this is likely partly explained by differences in the specific used methodologies in the studies. For instance, the EUSAAR_2 temperature protocol used in this study typically gives a factor of two higher EC content than the earlier commonly used NIOSH protocol, due to improved separation between the different types of carbon (Cavalli et al., 2010).

A clear similarity between the studies reported from the European Alps (Lavanchy et al., 1999; Jenk et al., 2006; Legrand et al., 2007; Thevenon et al., 2009) and the present study is the EC (or BC) trends indicating an increase in concentrations from the end of the 19th century to the present (Table 1), while the timing of peak values in the 20th century varies between the studies. Unfortunately, the record of Jenk et al. (2006) reaches only to the 1940s whereas Lavanchy et al. (1999) reaches to 1975 making

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the comparison to our results in the most recent trends impossible. The Thevenon et al. (2009) results reach 1980 and show increasing BC concentrations towards the end of the record whereas the results of Legrand et al. (2007) indicate a decrease from the 1960s until 1990. Interestingly, these latter BC results are from the same glacier (Colle Gnifetti, Switzerland) and the same elevation, but still show somewhat varying trends and non-synchronized peaks in the records.

BC and EC studies on Himalayan ice cores, as in Europe, have repeatedly shown different and contradicting trends when measured with different analytical methods even from the same glaciers (e.g. Ming et al., 2008; Xu et al., 2009; Kaspari et al., 2011; Jenkins et al., 2013). The SP2 method was originally designed to quantify BC mass concentrations and size distribution of atmospheric samples (e.g. Schwarz et al., 2006). Thereby, when applying the SP2 method to liquid samples, such as melted snow or ice, the liquid and particulates need to be nebulized into a dry aerosol phase. This added step of nebulization has shown to cause additional uncertainties in the BC concentration measurements as larger sized BC particles are not aerosolized as effectively as small ones (e.g. Schwarz et al., 2012, 2013). Consequently, typically only particles of smaller than ~ 500 nm core diameter have been quantified with the method from ice core samples (e.g. Kaspari et al., 2011; Bisiaux et al., 2012a, b; Jenkins et al., 2013). However, Schwarz et al. (2012, 2013) showed the presence of significantly larger BC particles in snow than are typically observed in the atmosphere. Therefore, Schwarz et al. (2012) recommend extending the size range of particles quantified with the SP2 method to at least 1.5 µm. At the same time the filter based methods may underestimate the occurrence of BC particles due to poor filtering efficiency at smaller particle sizes (e.g. Torres et al., 2014). Therefore, the European and Himalayan ice core results suggest that variations in observed EC and BC ice core trends can be expected also in the Arctic, especially when using different analytical methods.

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3.2 EC concentration vs. EC deposition

EC concentrations in snow are determined by numerous factors, such as BC concentration in the air, dry and wet deposition velocities, precipitation amounts, and post-depositional processes of wind drifting, sublimation and melt (Doherty et al., 2010, 2013). Ice core EC concentration trends may be particularly sensitive to changes in the snow and ice accumulation at the specific site through time. In addition, seasonal melt can lead to enhanced EC concentration at the surface of a glacier due to melt amplification (Doherty et al., 2013). These factors may lead to apparent fluctuations in recorded EC concentrations although EC deposition may have been constant over time.

Consequently, to further illuminate the processes leading to the recorded EC concentrations in Holtedahlfonna, we calculated EC deposition (EC $\mu g\,m^{-2}\,yr^{-1}$) by dividing the total amount of EC in a (filter) sample with the cross section of the ice sample and the amount of years covered in one filtered ice sample. We chose to calculate deposition rather than fluxes since flux calculation incorporate snow accumulation rates from the ice core and therefore add a source of uncertainty by introducing an additional variable. Deposition calculations eliminate post-depositional glaciological factors, such as snow accumulation rate and redistribution of EC in the snow pack, affecting the EC concentrations in the ice. The summer surface melt and subsequent redistribution of EC in snow packs will not affect deposition values as long as the redistribution is limited to the annual snow pack. From glaciers with summer melt Doherty et al. (2013), Ming et al. (2009) and Xu et al. (2012) have shown that BC is enriched at the top of the melt layer, and only about 10–20 % of the total BC is elusive and can percolate into deeper layers in the snow pack (Doherty et al., 2013). Percolation of BC is prevented by ice layers formed in the snow pack by refreezing of the previous year's melt layer (Doherty et al., 2013). As summer melt is occurring at Holtedahlfonna, ice layers are formed annually in the snow pack and firn (Beaudon et al., 2013), which suggests that

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the deposition values that we find in the ice core should not significantly be affected by EC redistribution.

The EC deposition trend is shown in Fig. 3b. Similar to the EC concentration trend it indicates lower deposition before the industrial era and high deposition in the beginning of the 20th century. The most evident difference is that the wide pronounced peak in the EC concentrations between 1920 and 1970 is not clear in the deposition. Consequently, it seems that the peak and variation in EC concentrations occurring 1920–1970 may be partly caused by changes in snow accumulation during this time period, rather than more EC being deposited on the glacier. Comparison of the EC concentrations with the measured snow accumulation of the ice core (Fig. 3c) indicates that snow accumulation at the ice core site was lowered during ca. 1930–1960 which may have led to higher concentrations measured in the ice core while EC deposition seems not to have increased in that period.

The EC deposition trend in Holtedahlfonna shows similarities to the BC trend recorded in the Greenland ice cores indicating a peak in both BC concentrations and deposition in the early 1900s and a decrease afterwards (McConnell et al., 2007, 2010; McConnell and Edwards, 2008). This BC peak is simultaneous in the Greenland and Svalbard ice cores although more pronounced in Greenland.

The rapid increase in the post-1970 EC concentrations in Holtedahlfonna is also apparent in the deposition (Fig. 3a and b). This indicates that more EC has been deposited at the site from 1970 to the present despite the measured decrease in atmospheric concentrations since 1989 in the Arctic (Hirdman et al., 2010a; Sharma et al., 2013). Consequently, the increase in EC concentrations cannot be simply explained by changes in the snow accumulation rate at the glacier. This example from Holtedahlfonna illuminates the importance of also studying the absolute deposition rather than focusing on the concentrations per se. The observed differences in the temporal evolution of EC deposition and concentration in the ice core show that additional processes to changes in atmospheric BC concentration are important for the final EC concentration in the ice core. Thereby, when comparing different ice cores to each other, both

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deposition and concentration of BC or EC are important to the analysis. In the following we will focus on explanations, which we find most plausible, for the increase in EC values in Holtedahlfonna during the last decades.

3.3 Black carbon emissions

The prevalent conception is that EC concentrations and deposition in the Arctic have decreased during the recent decades. In addition to atmospheric measurements (Hirdman et al., 2010a; Sharma et al., 2013) and emission inventories showing decreasing BC emissions in North America in the latter half of the 20th century (e.g., Novakov et al., 2003; Bond et al., 2007) this view is supported by ice core BC records from Greenland (McConnell et al., 2007; McConnell and Edwards, 2008). Model results also suggest that BC deposition in Svalbard was at a maximum around 1950 to 1960 and has decreased since then (Koch et al., 2011; Ruppel et al., 2013). At the same time the recorded EC deposition at Holtedahlfonna shows evident similarities with the historical global annual anthropogenic BC emission trend peaking in the early 20th century followed by a drastic rise since the 1970s (Fig. 4; Lamarque et al., 2010; Smith and Bond, 2014). The early 20th century global emission peak was mainly caused by European and American emissions whereas the increase since the 1970s is dominated by Asian emissions (Bond et al., 2007). However, northern Eurasia surface emissions have shown to be the dominant sources for atmospheric BC in the Arctic (e.g. Hirdman et al., 2010a, b and references therein), and therefore regional sources are expected to be mostly responsible for the recorded EC deposition trend in Holtedahlfonna.

Potential local emission sources on Svalbard are, among others, the coal mines in Longyearbyen, Barentsburg and Pyramiden, but these are not expected to have caused the observed trend in the Holtedahlfonna ice core. The nearest mine, located in Ny Ålesund, closed down in 1963 and therefore cannot account for the recent trend. In addition, Forsström et al. (2013) showed that local EC pollution on Svalbard snow is focused around the sources, and more specifically, snow around Ny Ålesund presents EC concentrations similar to arctic background levels (Aamaas et al., 2011). Apart

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from local terrestrial emissions, increased shipping in the Arctic has been proposed as a potential significant local BC source in the area (e.g., Eckhardt et al., 2013 and references therein). However, Eckhardt et al. (2013) showed that while cruise ships visiting Ny Ålesund in the summer months cause equivalent BC concentration peaks at the atmospheric Zeppelin measurement station (474 m a.s.l.) located less than 1 km away from the village, their influence on the annual mean concentration is minimal.

As mentioned, airmass transport to Greenland and Svalbard are rather different. The majority of atmospheric transport to Svalbard comes from Europe and northern Siberia (Stohl et al., 2006; Hirdman et al., 2010b) as opposed to North American emissions dominating Greenland ice core records (McConnell et al., 2007). In addition, the Greenland Ice Sheet and Svalbard glaciers receive divergent pollution deposition due to their different elevation. For instance, according to Hirdman et al. (2010b) the BC source regions for Summit on the Greenland Ice Sheet (3208 m a.s.l.) were specific for this high elevation site and did not reflect the low-level atmospheric transport of pollution from high latitude Eurasia that influences much of the rest of the Arctic which lies at lower elevations.

Of the Arctic monitoring stations recording atmospheric BC concentrations the Zeppelin station in Ny Ålesund has been shown to be particularly susceptible to BC emissions from Northern Russia, especially western Siberia (Eleftheriadis et al., 2009; Hirdman et al., 2010b; Stohl et al., 2013; Tunved et al., 2013). Stohl et al. (2013) pointed out that this is an intensive gas flaring region (circled in Fig. 1) and a previously underestimated major Arctic BC emission source. In fact, based on simulations run for 3 years with a Lagrangian particle dispersion model, Stohl et al. (2013) suggested that flaring in northern Russia may contribute to 42 % of the annual mean atmospheric BC concentrations near the surface in the Arctic.

Since flaring emissions from northern Russia originate at high latitudes their isentropic lifting in the polar dome is limited (Stohl, 2006; Stohl et al., 2013). The BC concentrations resulting from flaring are highest closer to the surface and decrease rapidly vertically in the atmosphere (Stohl et al., 2013). Furthermore, model results by Sand

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et al. (2013) suggest that a large fraction of BC emitted in the Arctic stays in the lowermost layers in the atmosphere and gets deposited at the surface. Therefore, despite their globally comparably small emission quantities, Arctic emissions may have a predominant impact on BC deposition at lower elevations of the Arctic (Sand et al., 2013). According to Stohl et al. (2013, Fig. 6 therein) emissions from flaring may contribute to 30–40 % of the simulated annual surface concentration of BC (ng m^{-3}) on Svalbard in comparison to 5–10 % on Greenland. It seems that the Greenland ice cores (e.g., at 2713 and 2410 m a.s.l.) are likely missing most of the flaring emissions both because of their different source areas as well as their elevation, whereas the Holtedahlfonna ice core is likely to capture more of these emissions. Interestingly, the large gas and oil fields in northern Russia, especially around the Gulf of Ob and west of it (circled area in Fig. 1), were discovered and established at the end of the 1960s to the 1970s and it can be expected that flaring commenced and increased soon after the extraction of oil and natural gas started there. Today, Russia is globally still the leading country in flaring which is mostly a consequence of missing infrastructure in the region to transport and utilize all extracted natural gas (Elvidge et al., 2009). According to satellite imaginary, flaring emissions from Russia have continued to increase from 1994 to 2005 (Elvidge et al., 2009). Therefore, it seems that the concurrent peak in the Greenland and Svalbard BC ice core records around 1910 could have been caused by the sites receiving same emissions from distant sources, most likely North America, since the BC peak in the Greenland ice cores was more pronounced than in Svalbard. The discrepancies between the sites since 1970 could partly be caused by Greenland not receiving major lower troposphere emissions that are recorded in Svalbard. However, it is unlikely that the post-1970 EC trend suggested by this Svalbard ice core was caused by Russian flaring emissions alone, as for instance, it does not seem to indicate clear or even short-term signs of lowered emissions associated with the collapse of the Soviet Union.

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3.4 Changes in scavenging efficiency?

While increasing post-1970 EC values in the Holtedahlfonna ice core may partly be explained by rising BC emissions from flaring (and possibly global BC emissions), these emission increases have not been captured by the atmospheric measurements at the Zeppelin station since 1998 (Eleftheriadis et al., 2009; Hirdman et al., 2010a) or any other atmospheric measurement station in the Arctic since 1989 (Hirdman et al., 2010a; Sharma et al., 2013). Although the overlapping time period of the ice core and atmospheric measurements is very short (three ice core data points), and the resolution of the ice core measurements too low for detailed temporal comparison with hourly atmospheric measurements, the recorded differences indicate further processes affecting the EC trends in the Holtedahlfonna ice core.

Variations in EC deposition that are not caused by changes in atmospheric concentrations may be caused by changes in the scavenging efficiency of BC. With constant or declining atmospheric EC concentrations, EC deposition may increase if BC scavenging efficiency increases. One possible pathway to influence the scavenging efficiency could be by changes in the temperature at which precipitation forms. In general, in-cloud scavenging of aerosols is less effective in ice clouds compared to liquid phase clouds. This is because at cold temperatures precipitation is formed via activation of especially efficient, but relatively few, ice nuclei that then grow by vapour deposition. At warmer temperatures, liquid precipitation is formed via collision and coalescence, which is a more efficient way to remove aerosols from clouds. Observations by Cozic et al. (2007) illustrate this effect clearly for BC particles. Below about −20 °C the scavenged fraction of BC is about 10 %. The fraction increases with temperature to about 60 % at temperatures just below the freezing point (Cozic et al., 2007). In the case of mixed phased clouds, precipitation can form through the Wegener–Bergeron–Findeisen process or through riming. In the former process, ice crystals grow by vapour deposition at the expense of the water droplets, which will have the same effect on the scavenging efficiency as cold cloud precipitation, i.e. low efficiency. Riming on the other

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hand yields much higher efficiency because ice crystals that are formed in the cloud will collide with the super-cooled droplets and the cloud condensation nuclei that formed the droplet will be scavenged in the process. As suggested by Cozic et al. (2007), the observed increase in scavenging efficiency with increasing temperature is consistent with a shift from non-rimed to rimed hydrometeors in mixed phased clouds. If there has been a shift over time in the temperature at which precipitation is formed above Holtedahlfonna, this could have resulted in a corresponding shift in the scavenging efficiency of BC.

Riming causes a preferential separation in snow chemistry as snowflakes scavenge nitrate (NO_3^-) more efficiently than sulphate (SO_4^{2-}) (Raynor and Hayes, 1983). Consequently, the nitrate to sulphate ratio is different in rimed as opposed to unrimed snow (Takahashi et al., 1996). This chemical signature was used by Hegg et al. (2011) to explain variation in BC concentration observed in fresh snow. The nitrate and sulphate records have been reported from Holtedahlfonna in Beaudon et al. (2013), but the hypothesis of riming controlled variation in BC scavenging efficiencies causing discrepancies between atmospheric and ice core EC trends cannot be verified with the available ion data from Holtedahlfonna, due to relocation of the nitrate and sulphate from the original snow layer during summer surface melt (e.g., Moore et al., 2005; Beaudon et al., 2013).

Whereas warmer temperatures could lead to an increased scavenging efficiency of BC in the Arctic, Browse et al. (2012) came to the conclusion that global warming may actually reduce the aerosol concentration and deposition rates in the Arctic. This is because enhanced BC scavenging efficiency en route from the source regions to the Arctic will reduce the amount of BC reaching the Arctic. While precipitation and temperature measurements are missing from the ice core drilling site, in general, observed annual precipitation and temperatures have increased in Svalbard in recent decades (Førland et al., 2011). Indirect observations of summer melt (see Sect. 3.5 below) are an indicator of strong changes at the Holtedahlfonna glacier over the last three to four decades.

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3.5 The linkage between summer melt and the EC concentration and deposition in Holtedahlfonna

In addition to the evident trend in EC values during the last decades, the ice core record suggests simultaneous changes in the summer melt of the glacier. Results from Beaudon et al. (2013) indicate that the Holtedahlfonna ice core has experienced summer melt throughout the last 300 years, but increasingly since the 1970s (Fig. 5a). The indicator of summer melt in ice cores is called melt index and in the ice core from Holtedahlfonna it is referred from log ($[Na^+]/[Mg^{2+}]$). The index is based on the fact that sodium (Na^+) and magnesium (Mg^{2+}) ions originate from same sources (sea salt), but percolate with different efficiencies in melting snow packs (Iizuka et al., 2002). The index has been defined as a good indicator of post-depositional processes of melt and percolation in Svalbard ice cores (Iizuka et al., 2002; Grinsted et al., 2006) with higher values reflecting more melt.

Beaudon et al. (2013) suggested the melt index to correspond to observed June–August air temperatures measured at the Svalbard airport in Longyearbyen (29 m a.s.l.) since 1911 (Fig. 5b). Summer temperatures in Svalbard started to increase in the 1960s which was accompanied by a distinct and steady increase in the ice core melt index, reaching unprecedented values during the 1980s. However, the increase in the melt index in 1970–1990 seems disproportionably strong when visually compared to the more variable and less pronounced temperature increase that does not exceed values of the early 1900s even by 2000 (Fig. 5b).

Though, when including the EC concentration trend in the assessment (Fig. 5c) it seems that there is some evident correspondence in the intensifying summer melt and the EC concentration in the ice core. We compare the melt index to EC concentrations since EC concentrations in snow are more relevant for the melt process than EC deposition. The turn of the melt index to a decrease in 1990 is not caused by a decrease in melting, but rather the melt index fails in the most recent part of the ice core where the summer melt has been much more extensive and thus the ions have been

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washed out from the annual snow pack (Beaudon et al., 2013) (though not penetrating into the snow of the previous year due to impermeable ice layers; c.f. Pfeffer and Humphrey, 1996). The importance of BC in accelerated glacier melt has been previously discussed, for instance, by Xu et al. (2009) for the Himalayas where glacier are retreating faster than what could be expected due to temperature increases alone. The current data supports the suggested process of increased BC (or EC) concentrations accelerating melt in glaciers.

At the same time the increased melt itself might actually cause the EC concentrations in the ice core to increase as melt is accompanied by water loss from the glacier surface to the atmosphere. During summer melt water can be lost from the annual snow pack by evaporation or sublimation as well as lateral water flow within the snow pack above the previous years' impermeable ice layer (Pfeffer and Humphrey, 1996). As EC tends to be hydrophobic, it is left behind within the snow pack during melt which leads to significantly higher EC concentrations at the snow surface (Conway et al., 1996; Doherty et al., 2010, 2013; Sterle et al., 2013). This melt amplification may have increased during the last 30 years in the ice core causing the increased EC concentrations. However, these summer time water losses are expected to have a rather small effect on the whole years' snow accumulation (Pfeffer and Humphrey, 1996). Thereby, increased melt is likely to have only a minor impact on the EC concentration trend. The fact that EC deposition, which is not affected by post-depositional processes, rises in tandem with the concentrations supports the inference that EC concentrations in the ice core are dominated by atmospheric processes, rather than post-depositional processes.

Although post-depositional processes do not seem to be overly significant for the ice core EC trends they may have a significant effect when comparing the results to snow measurements. For instance, at the Holtedahlfonna site EC concentrations of 1.4 $\mu g\,L^{-1}$, 9.4 $\mu g\,L^{-1}$ and 11.6 $\mu g\,L^{-1}$ were measured in 2007, 2008 and 2009, respectively (Forsström et al., 2013), which are roughly a magnitude lower than the ice core concentrations. These EC measurements have been made in springtime on surface snow which had most likely not significantly aged, i.e. experienced significant sublima-

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tion and/or melt. The ice core results on the other hand are the annual sum of all annual deposition as well as EC enrichment due to water losses and other aging processes throughout the year, and may therefore reach far higher concentrations than springtime snow measurements.

4 Summary and conclusions

Increasing EC concentrations and deposition since the 1970s have been recorded from a Svalbard ice core. This trend contradicts previous ice core records from Greenland and atmospheric measurements since 1989 which indicate decreasing BC concentrations in the Arctic during the last decades. However, the recorded trend agrees with estimated past global annual anthropogenic BC emissions and increased 20th century BC concentrations recorded in the European Alps glaciers.

The post-1970 EC trend in Holtedahlfonna is most likely explained by atmospheric processes. Of these, increased BC scavenging efficiency induced by rising temperatures is the most probable, as it is the only process that can simultaneously increase EC concentrations and deposition. This process may be responsible for the increasing EC deposition trend in the ice core record despite observed decreasing atmospheric concentrations in the Arctic.

Possible explanations for the differences in the recorded ice core BC trends from Greenland and Svalbard are partly different source areas, their different distances to sources, as well as the different elevations of the sites. Specifically, the post-1970 increase in Svalbard ice-core EC values may be affected by northern Russian flaring emissions which do not reach the Greenland ice core sites as effectively. The observed differences between the Greenland and Svalbard ice core records may also be caused by differences between the two analytical methods used. These differences may result from both the operational definition of what BC particle type is quantified by each method, and the size-dependent efficiency in the detection of particles. The SP2

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method may not incorporate largest BC particles and the filter based method may not include the smallest ones.

The results indicate that BC trends recorded in different ice cores may be quite different for various reasons. Results from a single spot glacier observation may not be easily extrapolated to a larger area. Therefore, more ice core and other sediment records from the Arctic are needed to better map the BC deposition in the region. Furthermore, comparison with atmospheric measurements and model estimations from the same area indicates that ice core measurements cannot a priori be converted to atmospheric concentrations as several factors independent of atmospheric concentrations can affect the concentrations and the deposition of BC to an ice core. In general, this may complicate model validation with help of ice core data.

The increasing EC values in Holtedahlfonna have major implications for the radiative transfer and forcing at the coring site. According to a calculated melt index from Holtedahlfonna, the glacier has experienced increasing summer melt from 1970. This trend is better explained by the increasing measured summer temperatures and the increasing EC concentration trend together, than by increasing temperatures alone. Whether our observed EC concentration and associated glacier summer melt trend is representative beyond the local scale remains to be determined. Consequently, further investigations of past BC values and its environmental impacts in the Arctic are necessary.

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Table 1. EC or BC concentrations measured with filter based thermal, thermal-optical or similar methods from the European Alps and Holtedahlfonna, Svalbard.

Reference	Glacier and location	Ice core time range	EC or BC concentrations (mean [± st. dev.] or range, in $\mu g\,L^{-1}$ in given time period)			Range in record (in $\mu g\,L^{-1}$)	Method used	Method reference
			Preindustrial	Industrial	Post 1940			
Lavanchy et al. (1999)	Colle Gnifetti, Switzerland	1755–1975	N/A	42 ± 22 (1890–1950)	72 ± 35 (1950–1975)	5–130	Thermal (2-step) including acid treatment	Cachier et al. (1989)
Jenk et al. (2006)	Fiescherhorn, Switzerland	1650–1940	15 (1650–1870)	27 (1870–1940)	N/A	8–60	Thermal (2-step)	Szidat et al. (2004)
Legrand et al. (2007)	Col du Dôme, France	1890–1990	N/A	1–5 (1890–1930)	7–16 (1940–1990)	1–16	Thermal-optical, including acid treatment	Pio et al. (2007)
Thevenon et al. (2009)	Colle Gnifetti, Switzerland	1000–1980	7 (1750–1850)	13 (1850–1950)	20 (1950–1980)	2–30	Elemental analysis after chemothermal oxidation of carbonates and organic carbon	Thevenon et al. (2009)
This paper	Holtedahlfonna, Svalbard	1700–2004	23 ± 9 (1700–1850)	36 ± 15 (1850–1950)	45 ± 19 (1950–2004)	9–103	Thermal optical, Sunset instrument, EUSAAR_2 protocol	Cavalli et al. (2010)

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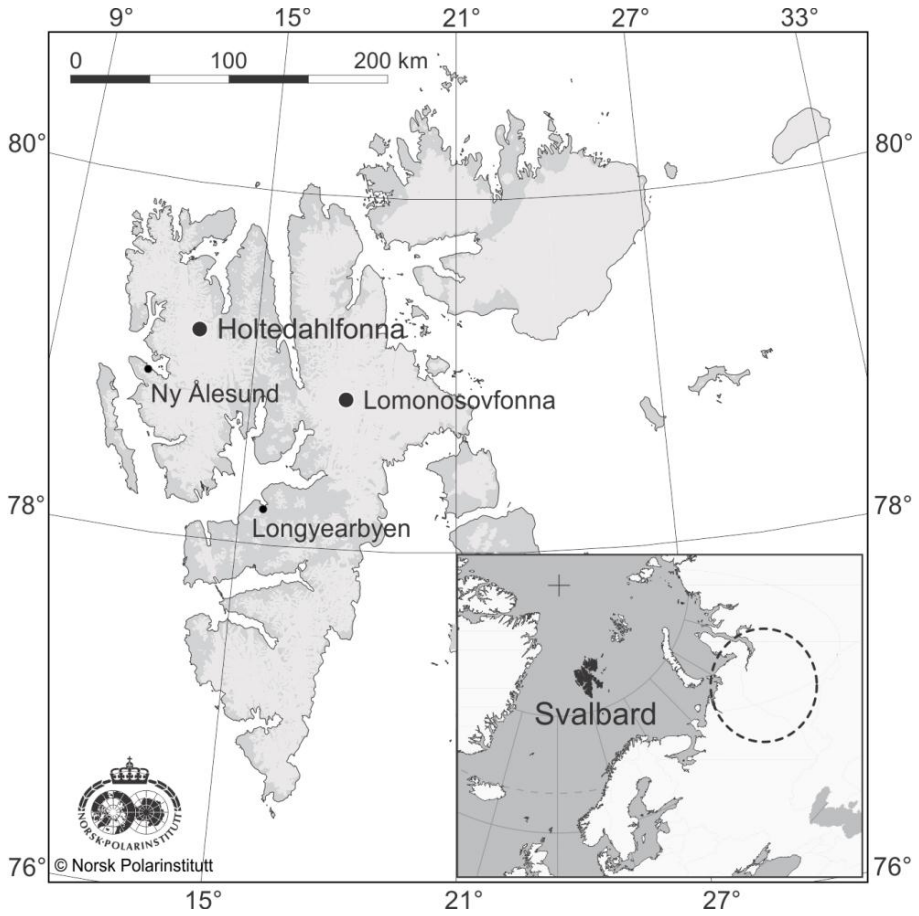

Figure 1. Map indicating the location of the Holtedahlfonna glacier on Svalbard and the geographical location of Svalbard. The circled area in the inset approximately indicates an area with substantial flaring activity in northern Russia.

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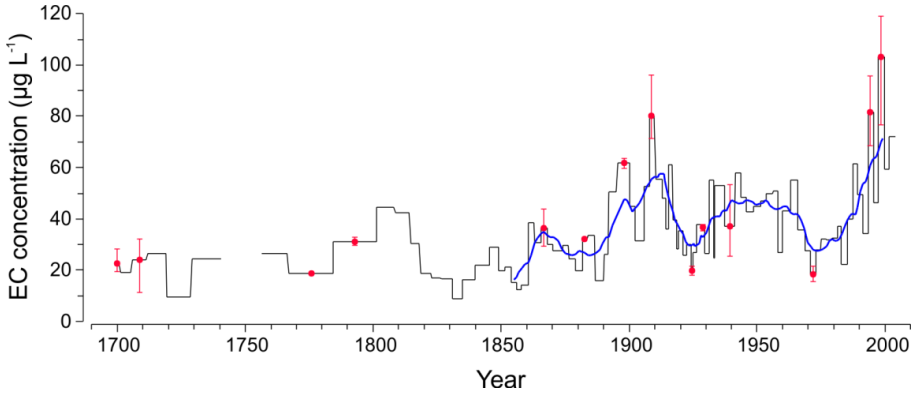

Figure 2. EC concentrations in the Holtedahlfonna ice core during the last 300 years. The black curve represents the concentrations at sample resolution and the blue line the running 10 year averages of samples made with approximately 2 year resolution. The red dots and error bars indicate average EC concentration and the absolute errors of samples from which multiple analyses were performed.

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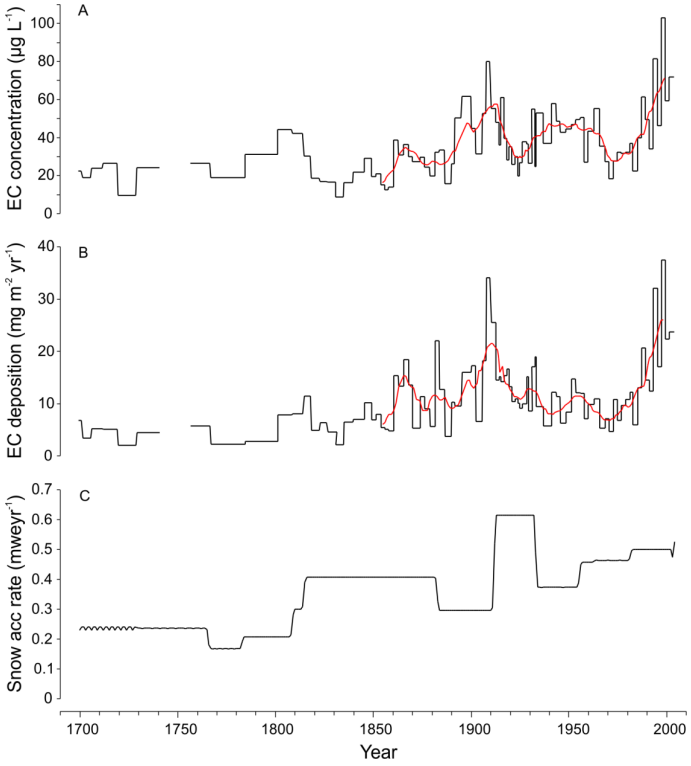

Figure 3. EC concentration and deposition compared to the snow accumulation rate in the Holtedahlfonna ice core. **(A)** EC concentration ($\mu g\,L^{-1}$) and **(B)** EC deposition ($mg\,m^{-2}\,yr^{-1}$) with 10 year running averages (red). **(C)** Snow accumulation rate ($m.w.e.\,yr^{-1}$) of Holtedahlfonna.

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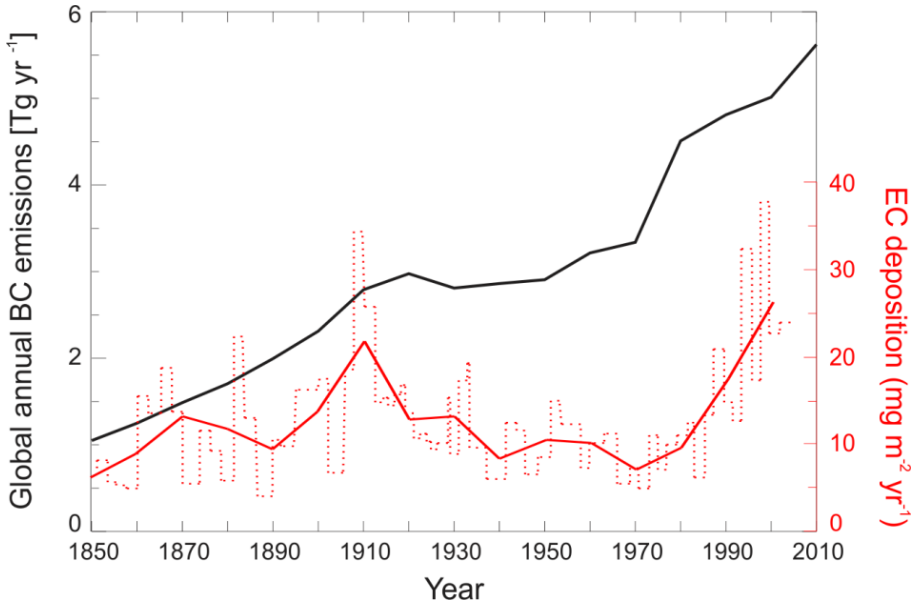

Figure 4. Global, annual anthropogenic BC emissions (emissions from Lamarque et al., 2010; redrawn after Ruppel et al., 2013) (black) compared to measured EC deposition in the Holtedahlfonna ice core (red) between 1850 and 2010. The EC deposition is shown in 10 year averages as well as in the sample resolution (dotted line).

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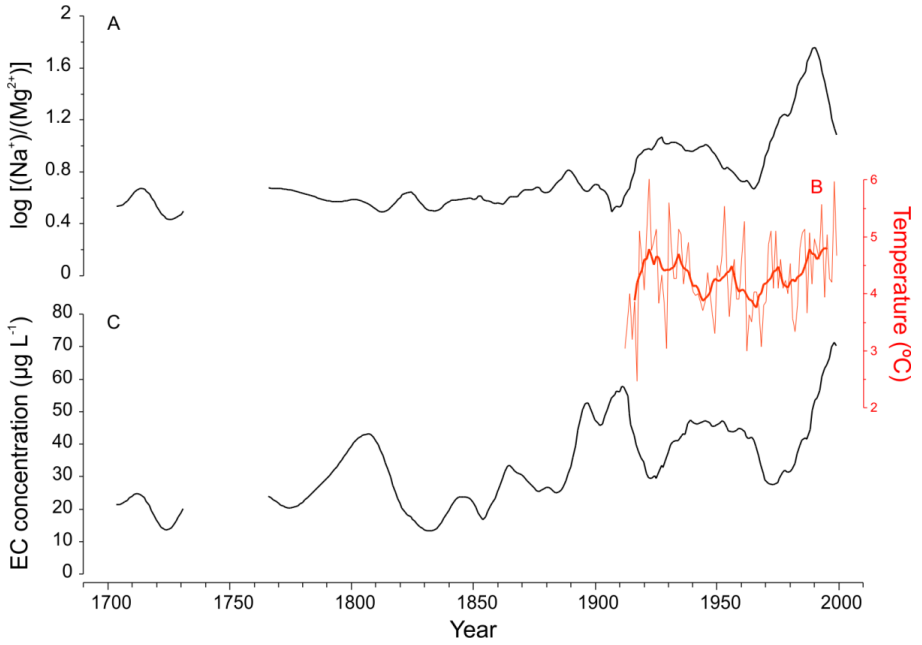

Figure 5. Holtedahlfonna melt index (log([Na^+]/[Mg^{2+}])) (Beaudon et al., 2013) compared to the measured June–August air temperatures at the Svalbard airport between 1911 and 2000 (source: http://climexp.knmi.nl/), and the EC concentration. The melt index **(A)** and EC concentration **(C)** are shown in 10 year running averages using linear interpolations in the resolution achieved by the ice samples. The temperatures **(B)** are shown in yearly summer resolution as well as 10 year running averages (thick red line).

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