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Sea ice in the northern North Atlantic through the Holocene: evidence from ice cores and marine sediment records

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Abstract

Sea ice plays a pivotal role in Earth’s climate and its past reconstruction is crucial to investigate the connections and feedbacks with the other components of the climate system. Among the available archives that store information of past sea ice are marine and ice cores. Recent studies on the IP25 biomarker extracted from marine sediments has shown great skill to infer past changes of Arctic sea ice. In ice matrixes, sodium, bromine and iodine have shown potential to store the fingerprint of sea ice presence. The development of an unambiguous sea ice proxy from ice cores, however, has proven to be a challenging task especially in the Arctic realm.

In this work we analyze the sodium, bromine and iodine records in the RECAP ice core, coastal eastern Greenland, to investigate the sea ice variability in the northern North Atlantic Ocean through the last 11,000 years of the current interglacial, i.e. the Holocene. We compare the RECAP records with marine sea ice proxy records available from the northern North Atlantic.
We suggest that RECAP sodium concentrations are associated with variability of sea ice extent, while the bromine-to-sodium ratios and iodine are associated respectively with seasonal sea ice and bioproductivity from open ocean and fresh sea ice surfaces.

According to our interpretation, we find that sea ice was at its lowest extent and seasonal in nature during the early Holocene in all regions of the North Atlantic. Increasing sea ice signals are seen ca. 8-9 ka b2k, in line with long-term Holocene cooling. The increasing sea ice trend appears uninterrupted in the Fram Strait and North Iceland while reaching a maximum ca. 5 ka b2k in the East Greenland region. Sea ice modifications during the last 5,000 years display great variability in East Greenland with intermediate conditions between the early and mid Holocene, possibly associated with local fjord dynamics. The last sea ice maximum was reached across all regions 1,000 years b2k.

1. Introduction

Sea ice reconstructions provide a long-term perspective for understanding the reductions of Arctic sea ice observed in the most recent decades (e.g. Comiso et al., 2008; Stroeve et al., 2007, 2012; Screen and Simmonds, 2010), as well as understanding the interactive role of sea ice in the coupled climate system. This is especially important for the northern North Atlantic region including the Nordic Seas, a complex region where warm, saline Atlantic water flows northward into the Arctic basin, and where sea ice and cold, fresher Arctic water is exported toward the Atlantic Ocean via the East Greenland Current (EGC), the largest sea ice and freshwater pathway in the Earth system (Hopkins, 1991). Sea ice variability in the northern North Atlantic is both an indicator and active agent of climate change, on regional and global scales. On the one hand, Atlantic Water in the northward-flowing boundary current can cause rapid warming and sea ice reductions in the Arctic margins near Svalbard (Lind et al., 2018); on the other hand, changes in sea ice in the EGC and Nordic Seas can lead to abrupt cold excursions (Miles et al., 2020).

Classic sea ice proxies from marine sediment cores, e.g. ice rafted debris (IRD), provide indirect evidence for sea ice occurrence (Stein et al., 2012). Recent advances have been made in the development of new proxies to reconstruct sea ice in the northern North Atlantic, notably the sea ice proxy biomarker IP$_{25}$ (Belt et al., 2007; Müller et al., 2011). Despite this progress, extracting a sea ice signature is often challenging and different proxies do not always agree (Stein et al., 2012; Belt and Müller, 2013; Belt, 2018, 2019). Moreover, marine sediment records often have low temporal resolution and substantial inherent dating uncertainties. Developing high-resolution, accurately-dated sea ice proxies is therefore a high priority goal of the Arctic paleoclimate community interested in reconstructing the past variability of sea ice, and understanding its role in the coupled climate system.

A complementary source of insight into sea ice variability with higher resolution and smaller dating uncertainties is offered by polar ice cores. Thus far, the development of unambiguous sea ice proxies from ice core records has proven to be a complex task. Sodium is associated with sea ice variability, especially in Antarctica (Wolff et al., 2006), whereas in the Arctic it is more often associated with modifications in the atmospheric forcing driving sea salt inputs from the open ocean (Rhodes et al., 2018). More recently, the presence of bromine and iodine
in ice cores have been suggested as indicators of sea ice. Particularly, the departure of bromine with respect to its expected abundance in sea water, known as “bromine enrichment” (Br\text{enr}), have shown potential as marker of past sea ice conditions in the aerosol source region, especially as an indicator of the newly formed sea ice areas, i.e., first-year (seasonal) ice. Br\text{enr} in Greenland ice cores is believed to reflect major changes in sea ice associated with glacial–interglacial transitions (Spolaor et al., 2016a; Maffezzoli et al., 2019; Vallelonga et al., 2021), as well as across Dansgaard–Oeschger climate transitions (Sadatzki et al., 2020). However, this potential has yet to be fully demonstrated. Open questions regarding chemical processes still remain, and the skill at detecting lower magnitude fluctuations across shorter timescales have not been extensively investigated. Ice core iodine has been associated with emissions from both open water and sea ice algae (Corella et al., 2019); incorporating information from this halogen element could therefore provide additional second-order evidence for past sea ice presence.

In contrast to the glacial millennial-scale climate variability and glacial–interglacial transitions, the Holocene climate is considered to be relatively stable and exhibits lower amplitude variability. The general development includes an early Holocene Climatic Optimum (HCO) with reduced sea ice, and a gradual transition to a mid Holocene deterioration (Neoglacial cooling), following the orbitally controlled insolation changes (e.g. Müller et al., 2012; Stranne et al., 2014). However, time-transgressive regional differences are apparent in several proxy records from marine sediments (Moros et al., 2006). Moreover, on suborbital time scales, temporal variability including abrupt changes within the Holocene is apparent in many records from the Nordic Seas, reflecting aspects of climate system dynamics – from internal interactions (Miles et al., 2020) and/or external forcing, e.g., volcanism (Miller et al., 2012). The question is whether and how such sea ice variability is recorded in the sea ice proxy records from Greenland ice cores.

Here, we investigate Holocene sea ice conditions in the northern North Atlantic based on the sodium, bromine enrichment and iodine records extracted from the RECAP (REnland ice CAP) ice core, located in coastal East Greenland, in comparison with marine sediment-based proxy records from the northern North Atlantic (Fig. 1). Because of its location, the Renland record should be sensitive to ocean processes and sea ice dynamics in its source area east of Greenland (Cuevas et al., 2018; Maffezzoli et al., 2019), a wide portion of the northern North Atlantic Ocean that includes multiple sediment records.

Both sea ice distribution and climate in and around the North Atlantic is strongly influenced by oceanography. As the warm salty Atlantic Water flows northward in the eastern Norwegian Sea, heat is gradually lost to the atmosphere. In the Fram Strait, the Atlantic Water submerges, and a part of it enters the Arctic Ocean; however, most recirculates in the Fram Strait and modified Atlantic Water flows southwards, subsurface, in the western Nordic Seas as the Return Atlantic Current (Mauritzen, 1996). Fresh, cold polar surface water and sea ice flows southwards from the Arctic Ocean via the EGC. In addition to these main current pathways, a fraction of Atlantic Water crosses the Greenland Scotland Ridge west of Iceland, through the Irminger Current, bending eastwards into the North Iceland Irminger Current (Fig. 1).
Through the Holocene, the EGC flow speed is suggested to be strongly controlled by the Atlantic inflow to the Nordic Seas, associated with how the Return Atlantic Current feeds into the EGC (McCave and Andrews, 2019). Hence, when setting the northern North Atlantic Holocene sea ice history in a climatic context, conditions of inflow and outflow regions of the Nordic Seas are both important.

Although marine aerosols reaching the Renland site are expected to be sourced from the northern North Atlantic, vast portions of sea ice found in the North Atlantic Ocean are nowadays created in the Arctic Ocean and eventually carried out from the Arctic basin through the Fram Strait via the Transpolar Drift (Supplementary Animation). As a result, although the sea ice chemical signature in the RECAP core is created in the North Atlantic, a sea ice reconstruction from the RECAP records is expected to represent a combination of sea ice formed in situ and Arctic Ocean sea ice export. Throughout the review we will adopt a ‘North Atlantic sea ice’ terminology but we note that such sea ice may also incorporate an Arctic fingerprint.

Here, we aim to:

1. Present and discuss the present knowledge on the use of sea ice proxies from ice and marine cores.
2. Present the Holocene Na, Br\textsubscript{env} and I records from the RECAP ice core (Renland ice cap, coastal East Greenland) and compare them with sea ice proxies derived from marine sediment cores, in order to assess the suitability of the RECAP sea ice proxies for reconstructing Holocene sea ice variability in the northern North Atlantic.
3. Produce a comprehensive synthesis of Holocene sea ice proxy reconstructions derived from marine cores focused on the northern North Atlantic, ensuring these are based upon comparable age models and sea ice proxy types.
4. Discuss the Holocene sea ice history, as seen from ice and marine sediment cores, relative to the Holocene climate development in the northern North Atlantic.

2. Sea ice proxies

2.1 Sea ice proxies from ice cores

Over the last two decades a substantial effort has been devoted to the development and the understanding of sea ice proxies from ice core records. We briefly introduce here, with a historical perspective and a primary focus on Greenland and the Arctic, the basic concepts behind the use of sodium, bromine and iodine in ice cores and discuss to which extent their quantification can provide information of past sea ice presence in the ice core source region. This section does not provide a full and exhaustive description of such markers, but rather presents the key aspects behind their interpretation in ice records. For a more in-depth analysis of sodium as a sea ice marker, see the review of Abram et al. (2013). Bromine and iodine are discussed as sea ice proxies in Vallelonga et al. (2021) and with a focus on atmospheric processes in Simpson et al. (2007) and Saiz-Lopez et al. (2012). The records that
are presented in this section serve the purpose of describing the progression in their knowledge in ice records and are not exhaustive. Specific records will be presented in Section 4.

2.1.1 Sea salt sodium

Sea spray aerosols are generated from bubble bursting over the ocean surfaces. A fraction of the sea salt oceanic input is eventually deposited onto polar ice caps and ice sheets. Among the markers of sea salt aerosols that have been routinely measured in ice core records, sodium is the preferred one, primarily due to the fact that, unlike chlorine, it does not undergo major chemical transformations after being emitted in the atmosphere (Legrand and Mayewski, 1997). Besides the marine origin, representing the majority of its signal, sodium has a minor contribution from mineral dust. The sodium crustal signature can be calculated using another element of crustal origin and its ratio with sodium within the dust source (Legrand and Delmas 1988; Wolff et al., 2010). At polar latitudes, this contribution equals a few percent during warm interglacial periods but may increase to up to ca. 30% during cold glacial periods (e.g. Maffezzoli et al., 2019).

Sodium records in ice cores have been associated with changes in atmospheric circulation and wind circulation patterns. In the GISP2 ice core, from Summit (Greenland), the sodium Holocene record has been associated with the modifications of the north polar vortex and consequent meridional circulation, responsible for bringing sea salt species to Summit (O’Brien et al. 1995). Building on this association, Dawson et al. (2003) suggest the GISP2 sodium time series is influenced by the North Atlantic seesaw and illustrated by its association with temporal changes in Iceland low pressure during winter (Meeker and Mayewski, 2002). This GISP2 record is the only complete Holocene sodium record available at present.

The association between the Summit sodium concentrations and past atmospheric modifications and North Atlantic ‘storminess’ changes across the Holocene was later challenged by a fundamental finding: the source of sodium was not only the ocean, but also sea ice. This advancement of knowledge was made possible, in particular, by studies in coastal Antarctica. By noting that sodium seasonality showed winter-spring maxima and that there was a sulphate fractionation (Wagenbach et al., 1998), it was suggested that sea ice could be a possible contributor, although no consistent link between sea ice extent and sea salt loadings was evident from the records (Weller et al., 2011). Originally, the sources and the emission mechanisms were not clear, and frost flowers on sea ice were proposed as the main saline formations responsible for such sea ice sodium inputs (Rankin et al., 2002). More recently, model studies (Yang et al., 2008; Huang and Jaeglè, 2017; Huang et al., 2020), and field campaigns (Frey, et al., 2020) have shown that the saline snow layer on fresh sea ice surfaces, known as blowing snow, is the main substrate for sea salt aerosols inputs at polar latitudes, with possible marginal contributions from frost flowers.

While the importance of sea ice as a source for ice core sodium was recognized, the question as to whether the main source of sodium was sea ice or the ocean was soon raised. If in
coastal Antarctica, the seasonal sodium peak during winter led to the conclusion that sea ice may represent the major source of sea-salt aerosol (Rankin and Wolff, 2003), despite winds being also stronger in that season, in Greenland it is generally more difficult to distinguish the relative contribution of open water and sea ice inputs to the final measured sodium concentrations. Differences between Arctic and Antarctic sea ice also pertain to the geographical setting and sea ice age distribution. Antarctic ice expands radially from the continent and is mostly seasonal (Nghiem et al., 2016), while Arctic sea ice can persist for more than one season (multiyear sea ice, MYSI) and grows in a more complex environment constrained by continental land. As a result, the interpretation of Arctic ice core sodium records has proven more challenging and site-dependent (Rhodes et al., 2018). Recent efforts have thus been directed at quantifying the relative importance of open water and sea ice as per the observed sodium signals and to understand which sites would be more suitable for Arctic sea ice reconstructions from sodium records in Greenland (Rhodes et al., 2018). According to model investigations, at NEEM, sea ice-related processes in the Baffin Bay and Canadian Arctic account nowadays for up to 50% of the sea salt aerosol inputs during wintertime, the other 50% being attributed to open water sea spray emissions from the North Atlantic Ocean, believed to be the source of marine aerosols (Fischer et al., 2007; Supp. Fig. 2 in Schüpbach et al., 2018). The sea ice related contribution is, however, expected to be greater during the glacial period. To infer past changes of the atmospheric sodium loading using the ice concentrations in the NEEM core, it was attempted to calculate the sodium air concentrations extending back 130,000 years to the Eemian period (Schüpbach et al., 2018). This step is necessary when the snow accumulation varies greatly across the investigated period, making neither the ice concentration nor the air-to-snow depositional flux parameters linearly linked to the air concentration, which is ultimately the variable of climatic significance. The calculation of the air concentration requires the knowledge of parameters like the dry deposition velocity, the snow (or rain, or both) scavenging factor and the atmospheric residence time, as well as a deposition model. Based on the Fischer et al. (2015) model, Schüpbach et al. (2018) estimated the sodium air concentration at NEEM, and found an increase in atmospheric sea salt aerosol during colder periods by only a factor of about 1.5. They attributed such cold period increases to a net expansion of sea ice surfaces, despite the concurrent growing multi-year sea ice that would reduce sodium emissions. Due to poor knowledge of the model parameters consequently affecting the reconstructed sodium air concentrations, however, no further robust conclusions could be drawn. 

The importance of sea ice as a source of sea salt aerosol has been increasingly recognized since the pioneering interpretations of Greenland sodium records based solely on circulation changes. Further studies, however, are still needed to more accurately assess the relative importance of sea ice and open water as sources for this marker in Arctic ice cores, as well as the importance of past changes in atmospheric transport processes.

### 2.1.2 Bromine and bromine enrichment factor

Similar to sodium, the main sources of bromine are sea salt aerosol emissions from open waters as well as sea ice, despite the emission mechanisms from the latter being different.
During springtime, a series of heterogeneous reactions involving saline and halide-rich substrates present over or in fresh sea ice lead to an exponential increase of gas-phase bromine, which can in turn be redeposited, triggering further bromine release (Abbatt et al., 2012). This cycle of catalytic reactions, which require sunlight, are known as 'bromine explosions' and were discovered in association with Ozone Depletion Events during the Arctic sunrise (Barrie et al., 1988). The regional link between bromine explosions and fresh sea ice was later evidenced by satellite-based measurements of gas phase BrO (e.g. Schönhardt et al., 2012).

With the aim of isolating the sea ice signal from that of open waters in ice core records, the bromine enrichment adimensional factor (hereafter Br\textsubscript{enr}) was introduced (Spolaor et al., 2014). In any ice sample, it is calculated by taking the ratio of bromine and sodium concentrations, then normalized by the same ratio in seawater (0.0062, Millero et al., 2008), assumed constant in time and space (Eq. 1):

\[
\text{Br}_{\text{enr}} = \frac{(\text{Br}/\text{Na})_{\text{ice}}}{(\text{Br}/\text{Na})_{\text{seawater}}}, \quad \text{with} \quad (\text{Br}/\text{Na})_{\text{seawater}} = 0.0062
\]  

(Eq. 1)

Equivalently, the non-sea-salt bromine concentrations (nssBr) can be calculated as the difference between the bromine content and the expected content from purely marine (non-sea ice) emissions (Eq. 2):

\[
\text{nssBr} = \text{Br} - \text{Na} \times (\text{Br}/\text{Na})_{\text{seawater}}
\]  

(Eq. 2)

While Br\textsubscript{enr} is adimensional, nssBr is a concentration and therefore more closely dependent on accumulation changes.

Building on these premises, bromine and especially Br\textsubscript{enr} in both Arctic and Antarctic ice records have been investigated as potential markers of seasonal sea ice (hereafter also referred to as first-year sea ice, FYSI). Annual studies both in the Arctic (Spolaor et al., 2014) and Antarctica (Maffezzoli et al., 2017) show that, as expected, the higher Br\textsubscript{enr} snow strata are deposited during the spring and summer seasons, the period of maximum combination of seasonal sea ice extent and solar radiation. Over decadal and centennial timescales, attempts have been made to link Br\textsubscript{enr} records with the few existing sea ice historical observations and satellite records in the Arctic (Spolaor et al., 2013; Spolaor et al., 2016b; Maselli et al., 2017) and Antarctic (Vallelonga et al., 2017) domains. Significant but generally weak correlations were found. Over glacial-interglacial time scales, Br\textsubscript{enr} values have been associated with modifications in the type of sea ice present in the source regions, with greater Br\textsubscript{enr} values believed to indicate more extensive FYSI, and lower values believed to indicate either aged and thick multi year sea ice (Spolaor et al., 2016a) or open water conditions (Maffezzoli et al., 2019). This dualism provides a challenge when considering a source region with mixed sea ice and open water conditions, as is the case of the North Atlantic, the source region for aerosols to the Renland site. Today, the Renland source area is dominated by open waters. The sea ice surfaces consist of both locally grown seasonal sea ice and multi year ice exported southwards from the Arctic Ocean along the East Greenland coasts (Supplementary...
Besides the challenge represented by the suggested dual nature of Br$_{en}$, a number of basic fundamental questions regarding the use of bromine as a sea ice marker are still being investigated. These can be grouped into three categories: activation, transport and post-deposition effects. On this topic, we refer the readers to Maffezzoli et al. (2019, main text and Appendix), for a more in depth discussion.

### 2.1.3 Iodine

Due to low concentrations in polar snow providing experimental challenges, iodine records from ice cores have only recently started to become more widely available. Oceanic micro and macro algae are the main source of atmospheric iodine species, in the form of both inorganic and organic compounds (Saiz-Lopez et al., 2012; Prados-Roman et al., 2015). A secondary source of iodine is represented by sea ice, especially the thin ice, which allows the permeation of light and consequently life of diatoms living within or below the sea ice surface (Saiz-Lopez et al., 2015). Biologically produced iodine compounds from sea ice algae can therefore be transported through the porous sea ice matrix and emitted into the overlying polar atmosphere (Saiz-Lopez et al., 2015). High iodine air concentrations have been detected from satellites above the high porosity Antarctic sea ice in the Weddell Sea (Atkinson et al., 2012), while lower iodine levels have been found over Arctic sea ice (Schönhardt et al., 2008; 2012). This polar asymmetry, referred to as the 'polar iodine paradox' (Saiz-Lopez and Blaszczak-Boxe, 2016), is suggested to be due to Arctic sea ice often being thicker than its Antarctic counterpart, preventing enough light permeation to sustain high biologic activity beneath its surfaces. Unexpectedly, early ice core seasonal investigations have shown higher iodine concentrations in the winter strata (Spolaor et al., 2014), opposite to the timing of the algal bloom, evidenced from maximum IO concentrations reported during spring and summer seasons from satellite sensors (Schönhardt et al., 2008). Laboratory and field measurements have confirmed that iodine is photochemically activated (Kim et al., 2016; Gálvez et al., 2016; Spolaor et al., 2019) from the snowpack after deposition during the spring-summer period, leaving higher concentrations in winter ice core strata (Raso et al., 2017). However, if assuming constant loss from the snowpack over the investigated period, iodine time series from ice cores could still provide a record of open ocean and sea ice biological activity in the source region. On this assumption, the Holocene iodine record from the Renland ice core in eastern Greenland has been associated with marine oceanic productivity in the northern North Atlantic, secondarily modulated by sea ice changes (Corella et al., 2019).

### 2.2 Sea ice proxies from marine sediment cores

Over the last decades, substantial progress has been made in the development and the understanding of sea ice proxies from marine sediment records. We briefly introduce here the two specific sea ice proxies that we compare the RECAP ice core records with: IP$_{25}$ and %quartz.

Using biomarkers such as IP$_{25}$, a mono-unsaturated highly branched isoprenoid (HBI) alkene...
seasonally produced by diatoms living in the sea ice, Brown et al. (2014) provides a rather new but well documented method for reconstructing past locations of the Arctic sea ice margin (e.g. Belt et al., 2007; 2015; Belt, 2019; Stein et al., 2017). The approach based on biomarkers represents the state-of-the-art and offers the most direct sea ice proxy available for this area. The presence of IP$_{25}$ in Arctic marine sediment reflects the presence of seasonal sea ice over the site at the time of sediment deposition (Kolling et al., 2020). Similar to the B$_{ren}$ ice core counterpart, the absence of IP$_{25}$ in the sediment can reflect either a permanent sea ice cover or open water conditions at the site (Müller et al., 2009; Stein et al., 2012; Belt and Müller, 2013; Belt, 2018). By simultaneously analyzing IP$_{25}$ and a phytoplankton biomarker, the latter indicating open water conditions (e.g. brassicasterol, dinosterol, HBI-III), the PIP$_{25}$ index can be calculated, providing a semi-quantitative sea ice estimate (Müller et al., 2011; Fahl and Stein, 2012; Belt et al., 2015; Berben et al., 2017; Stein et al., 2017; Belt, 2019).

Lithic indicators of sea ice such as ice-rafted debris and the presence of specific minerals can be used to infer sea ice. Quartz is considered an allochthonous mineral in the vicinity of the Iceland shelf (Moros et al., 2006). An increased relative quartz content is therefore considered indicative of increased transport of ‘drift ice’, a mix of first year, multiyear and icebergs, to this area. The concept of using %Quartz as an indicator of sea ice for the Iceland shelf area has been validated against historical sea ice data (Moros et al., 2006).

There are also other, indirect proxy indicators of sea ice, including biogenic tracers of sea ice such as diatoms and dinoflagellate cysts, as well as biogenic indicators of sea ice-free conditions, such as coccoliths and foraminifera (e.g., de Vernal et al., 2013). However, in this paper we restrict our comparison to marine sediment core proxy records based on IP$_{25}$ (PIP$_{25}$) and quartz.

3 Approach and methods

The RECAP ice core from the Renland ice cap in coastal East Greenland (71° 18’ 18” N; 26° 43’ 24” W; 2315 m above sea level, Fig. 1) was drilled in summer 2015. Here, we present the Holocene RECAP records of all the chemical markers that are considered to carry information on sea ice variability: sodium, bromine and iodine. The sodium and bromine records for the last glacial cycle have been presented by Maffezzoli et al. (2019), while Corella et al. (2019) have specifically considered the Holocene iodine record. For the details on the experimental methodology, we refer the readers to these two papers. Here, all the considered records consist of a total of 940 ice sample measurements at 55 cm resolution which comprise the 530 m depth range of the Holocene period. Due to the extreme thinning of the Renland ice cap, the time resolution varies greatly across the Holocene, from annual in the top section, decreasing to ca. 30 years at 5 ka b2k and to ca. 350 years at 11.3 ka b2k. The chronological framework is established by Simonsen et al. (2019). In 1988, another ice core was drilled at Renland, a few hundred meters away from the 2015 drilling site. Records from the 1988 core provide information from 10 to 120 kyr (Hansson, 1994). Hence, the 2015 RECAP core provides the only continuous Holocene record from east Greenland. The annual
accumulation rate at the coastal Renland site is much greater than that on the continental
Greenland plateau. Investigations of the 1988 and 2015 cores (Hansson, 1994; Corella et al.,
2019; Hughes et al., 2020) suggest an average Holocene annual accumulation rate of ca. 500
kg m⁻² yr⁻¹. We therefore suggest that wet deposition is the main deposition mechanism at this
site and hence the ice concentrations, and not the air-to-snow fluxes, should be considered the
variables linearly related to the atmospheric loading.

The source area for marine aerosols influencing the RECAP ice core was investigated by
Maffezzoli et al. (2019) by performing an atmospheric reanalysis for the recent period (2000-
2016). Among the whole batch of computed 72 hour back trajectories, the authors only
considered those that crossed the marine boundary layer (MBL, defined as the 900 hPa
isosurface) for at least 10 hours and computed the spatial distribution of such trajectory
endpoints. They concluded that 75% of the signal originates from the North Atlantic realm,
50-85°N (Fig. 1). Although such a method suffers from a known central-tendency bias
towards the core site, the ocean regions closer to Renland are indeed expected to be more
significant. Recent works on halogen records have shown that, due to its coastal location, the
RECAP site is sensitive to ocean-related processes from the northern North Atlantic Ocean
(Cuevas et al., 2018; Corella et al., 2019).

The RECAP core source area encompasses several regions where marine sediment records
have been extracted, and where information exists on past sea ice and ocean conditions: the
North Icelandic shelf, the East Greenland shelf, the Norwegian Sea and the Fram Strait (Fig.
1). To assess the skills and limits of bromine, sodium and iodine as sea ice markers, we
compare the RECAP records with the state-of-the-art knowledge about the Holocene sea ice
history as documented by records of established sea ice markers (IP₂₅ and %quartz) from
marine cores located within the RECAP source area (Fig. 1). Combined, the ice core and
marine core sea ice reconstructions provide an integrated overview of the Holocene sea ice
history for the Fram Strait area, the East Greenland margin and north of Iceland. This
approach is not exclusive, and other strategies should be also pursued, most notably the
development of transfer functions based on ice core chemical records and satellite-based sea
ice observations. Such approaches are, however, beyond the scope of this paper. The changes
in regional sea ice distribution inferred from the RECAP and the marine sediment records
will be also discussed in context of the Holocene climatic and oceanographic development of
the Nordic Seas realm.

To facilitate the comparison between the synthesised marine records and the RECAP records,
all age models of the marine sediment cores that were originally based on older versions of
the marine calibration dataset, have been updated using Marine13 (Reimer et al., 2013). We
are aware about the new Marine20 calibration dataset, however, this new calibration dataset
is not intended, or suitable, for calibration in polar regions (Heaton et al., 2020). Calib 7.1
(Stuiver and Reimer, 1993) was used to calibrate the radiocarbon dates to calendar ages,
using a ΔR value of 0±0 Table A.1). Core top ages were set to the year of coring. Ages are
calculated by linear interpolation between dated tie points. This approach accounts for
MD99-2269 (Stoner et al., 2007), MSM5/5-712-2 (Müller et al., 2012) and MSM5/5-723-2
(Müller et al., 2012). For MD99-2269, the age model follows Table 3 in Stoner et al. (2007), presenting a combined age model for MD99-2269 and MD99-2322, where the two cores are set at a common depth scale based on extensive radiocarbon dating of both cores and a further correlation of paleomagnetic secular variations. For site PS2641, we use the age model as published by Perner et al. (2015) and used by Kolling et al. (2017) but presented as b2k rather than before 1950 (BP). The PS2641 dataset published by Müller et al. (2012) has been updated to the Perner et al. (2015) chronology. Since Perner et al. (2015) and Kolling et al. (2017) only focus on the last 6000 years, the oldest dates from Müller et al. (2012) have been updated following the same approach as above. The age model for MD99-2272 is used as published by Xiao et al. (2017), solely based on identified tephra horizons. All ages of marine data are given as years before 2000 (b2k), as for the RECAP records.

Figure 1. Northern North Atlantic Ocean region map with all the investigated cores. The RECAP core is indicated with a star. The underlying distribution is the annual averaged 2000-2016 RECAP source region of marine aerosols, calculated from back trajectory crossings in the marine boundary layer (MBL), following the method described in Maffezzoli et al. (2019). The magenta solid and dashed lines indicate the 1981-2010 median sea ice extent in March and September respectively (data from NSIDC, Fetterer et al., 2017). The main warm and cold currents are indicated respectively with red and blue colors. ‘NAC’: North Atlantic Current; ‘NwAC’: Norwegian Atlantic Current; ‘WSC’: West Spitsbergen Current; ‘IC’: Irminger Current; ‘ECG’: East Greenland Current.
4. Results and discussion

The sodium concentrations measured in the RECAP core have been, on average, $17 \pm 4$ (1σ) ppb for the last 10 ka (Fig. 2). The lowest values, 10 ppb, are found during the Early Holocene, particularly before 8-9 ka b2k. Hereafter, sodium levels have increased almost uninterruptedly until a broad maximum at ca. 5.5 ka b2k. Following mid-Holocene maximum concentrations of 25 ppb, sodium concentrations decrease again to minimum values of 10 ppb at ca. 3.8 ka b2k. Generally low to below-average Na concentrations are observed from 3.8 to 2 ka b2k, after which they have risen to moderate to high values for the last 2,000 years, in association with increased variability. Compared to sodium, bromine concentrations show lower variability. As a result, the low frequency trend of Br\text{enr} (and nssBr) is mostly driven by and mimics the sodium signal, with high Br\text{enr} values during periods of low Na concentrations and low Br\text{enr} values during periods of high Na concentrations.

The iodine record has been discussed in Corella et al. (2019) and shows high concentration values (on average 0.04 ppb) during the Early Holocene up until ca. 6 ka b2k, followed by an evident decrease to generally low and more variable values reported from the remaining part of the Holocene (Fig. 2).

When presenting and discussing the datasets, we focus on four different time periods: approximately 10 to 5.5 ka b2k (Sect. 4.1); approximately 5.5 to 3.8 ka b2k (Sect. 4.2); 3.8 to 1 ka b2k (Sect. 4.3); and the last 1 ka b2k (Sect. 4.4). Such division is primarily but not solely based on the described changes seen in the RECAP sodium and Br\text{enr} records. The last 1,000 years are discussed in a dedicated section (Sect. 4.4) since several marine sediment studies are available for this period. For each time interval we will first present and discuss the new RECAP data, followed by a discussion of the results in context of marine sea ice proxy information and of Holocene climate and oceanography.

4.1 From approximately 10 to 5.5 ka b2k

The RECAP sodium concentration record displays some of the lowest values of the entire interglacial, ~10 ppb, from ca. 11 to ca. 8-9 ka b2k (Fig. 2c). Early Holocene bromine concentrations were ~0.4 ppb, slightly above Holocene average, and remained stable for over four millennia (Fig. 2b). As a result, centennial-averaged Br\text{enr} values during the early Holocene were some of the highest of the entire Holocene, ~6-8, and remained within 1σ until ca. 10 ka b2k, after which they start to decrease (Fig. 2d).

At present, Renland surface snow is affected by summer melting. The question as to whether summer melting affects the preservation of sodium and bromine should be addressed, especially during the early Holocene when air temperatures and insolation were at their maximum. Sodium is known to percolate towards deeper snow strata. The high annual layer thickness at Renland (55 cm of ice equivalent), however, would suggest that any remobilization would only affect the sodium variability on annual scales. Open questions still challenge the preservation of snowpack bromine and iodine, and specifically whether they are photolithically reactivated. Similar to sodium, we suggest that the high accumulation at the Renland site prevents effective loss from the photic zone of the snowpack at the timescales considered in this work. Seasonal or subseasonal records of halogens at Renland, however,
We suggest that the high RECAP Br\textsubscript{enr} values and non-sea-salt bromine concentrations around 10 ka reflect enhanced bromine explosions from seasonal sea ice surfaces in the North Atlantic, as compared to other periods of the Holocene (Fig. 2). Recent works have hypothesised that Br\textsubscript{enr} is linked to enhanced seasonal sea ice conditions (e.g. Spolaor et al., 2016a), while sodium concentrations have been associated with increased sea ice conditions (Schüpbach et al., 2018) and/or sea spray inputs from open waters (e.g. Mayewski et al., 1994), facilitated during periods of favorable, or stronger, atmospheric circulation patterns bringing sea spray to the ice core site (Sect. 2.1.1). The only other existing and complete sea salt Holocene record from Greenland is from the inland GISP2 ice core (Fig. 2c), retrieved at Summit. Unlike RECAP, which location in coastal East Greenland supports the North Atlantic Ocean as the main source region for sea salts, large-scale circulation to central Greenland is believed to be dominated by both westerly and meridional transport pathways (Kahl et al., 1997; Rhodes et al., 2017). As such, while GISP2 and RECAP sea salt records may share a common North Atlantic fingerprint, GISP2 sodium is also influenced by atmospheric transport from, and sea ice changes in, the Baffin Bay. Therefore, the sodium records at the two sites should be compared cautiously.

O'Brien et al. (1995) associated the GISP2 sodium variability with the strength of the Icelandic Low and expansion of the polar vortex, facilitating the atmospheric circulation, emission and transport of marine species from the North Atlantic to the Greenland plateau (Fig. 2c). The authors related the period of high sodium circulations ~11 ka b2k to atmospheric circulation patterns associated with the Younger Dryas, and the following decrease with an atmospheric reorganization that led to less favorable meridional circulation during the early Holocene. Note that O'Brien et al. (1995) did not consider the Baffin Bay as a particularly relevant source region for sodium deposition at Summit, and the importance of sea ice sea salt was largely unexplored at the time. The RECAP sodium concentrations, despite the greater accumulation at this site, are higher than at GISP2 (Fig. 2c, Supp. Table), suggesting that at Renland, not unexpectedly, the influence of the North Atlantic Ocean results in a greater atmospheric loading of sodium, facilitated by a closer source region and reduced loss during transport compared to Summit. We propose that the minimum RECAP sodium values during the early Holocene can be interpreted to represent a small sea ice extent, associated with mostly ice-free waters during summer in the North Atlantic, and with a generally weak atmospheric transport of marine aerosols, while maximum Br\textsubscript{enr} values would indicate that sea ice surfaces were preferentially seasonal. This situation, with a reduced and seasonal sea ice extent in the North Atlantic during the early Holocene is also suggested from the high iodine concentration values (Fig. 2e) that have been attributed to enhanced emission of biogenic iodine from the open ocean and seasonal sea ice phytoplankton colonies (Corella et al., 2019).

The comparison of both Na and Br\textsubscript{enr} series with modern values suggests that similar-to-present sea ice conditions existed ca. 8-9 ka b2k (Fig. 2c,d). From ca. 8-9 ka b2k the Br\textsubscript{enr} and Na RECAP series show decreasing and increasing values respectively. We suggest that, following the early Holocene minimum, sea ice gradually increased in the North Atlantic. We
also suggest a shift towards multiyear sea ice conditions as inferred from decreasing Br\textsubscript{enr} values. We note that to explain the increasing MYSI conditions as interpreted from the Br\textsubscript{enr} decrease and the sodium increase, the assumption that sodium is not only emitted from seasonal sea ice but rather a mixture of first and multiyear sea ice must be invoked. The sodium (Br\textsubscript{enr}) series display a broad maximum ~5 ka b2k, during the mid-Holocene. A sodium maximum is also measured at GISP2 ~500 years after the RECAP one. We point out once again, however, that the GISP2 source region includes both West and East Greenland, therefore such maxima may not be related to one another.

According to our interpretation of the RECAP sodium and Br\textsubscript{enr} records, we suggest that increasingly more favourable atmospheric circulation for transport of marine aerosols to the site and the increase of North Atlantic sea ice culminated in a maximum extent and multiyear sea ice conditions at ca. 5 ka b2k (Fig. 2). The interpretation of an advance of thicker/more extended perennial sea ice is consistent with an evident iodine concentration decrease from ca. 6 to 5.5 ka b2k (Fig. 2e, Corella et al. 2019). It is worth noting that while the Na and Br\textsubscript{enr} series show more gradual variations starting after ca. 10 ka b2k, the iodine drop at ca. 6 ka b2k is faster. This difference suggests a threshold in sea ice thickness, allowing light penetration through seasonal ice and iodine sea ice algae production up to 6 ka b2k, similar to modern Antarctic conditions (Saiz-Lopez and Blaszczak-Boxe, 2016).

Despite the higher accumulation, the RECAP Holocene average sodium concentrations, 17±4 ppb (Supp. Table), are similar to those found during the cold glacial stadials (17±6 ppb, Supp. Table). Based on general findings on Holocene-to-glacial accumulation ratios (about 4-5 times higher during the Holocene), a calculation of the sodium fluxes would yield higher Holocene values. Although a precise estimate of the atmospheric sodium loading would be needed to draw any robust conclusion, we suggest that this has been higher during the Holocene than during the last glacial period. The opposite is found at Summit (Mayewski et al., 1994, 1997) and NEEM (Schüpbach et al., 2018), with about a factor ~5-7 higher sodium ice concentrations during the glacial than during the Holocene on the Greenland plateau (Supp. Table). The latter resulted in about 1.5 times higher emissions of sea-salt aerosols during the glacial period, as estimated at NEEM from sodium air concentration calculations by Schüpbach et al. (2018). Based on these considerations, we suggest that, while glacial enhancement of the atmospheric circulation to the Greenland ice sheet from the North Atlantic ocean probably affected all sites similarly, the increase in sea spray emissions from the North Atlantic ocean during the Holocene yielded greater sodium depositions at Renland compared to the sites over the Greenland plateau. These considerations bring evidence that, as climate transitioned from the glacial into the Holocene, at Renland the effect of an increase in sea spray emissions from an ice-free ocean was greater than any potential sea salt emission increase from sea ice surfaces that occurred during the glacial, when the more saline seasonal sea ice surfaces were probably located further away from the Greenland coasts by multiyear sea ice.

We now examine the marine sediment records, which show general evidence for extensive open ocean and therefore minimal sea ice in the North Atlantic Ocean, except for the
Northeast Greenland shelf, during a period that we broadly define as starting from the early Holocene and continuing through to approximately 6 to 5 ka b2k (Fig. 3). At the eastern border of the Fram Strait, the lowest P_BIP25 values are found at 8 ka b2k (Fig. 3b). From 8 ka b2k, P_BIP25 slowly and consistently increases through to 5 ka b2k, indicating steady sea ice increases. Similarly, off eastern North Greenland, core PS93/025 biomarkers suggest reduced but variable sea ice cover until ca. 9.3 ka b2k, followed by increased seasonal conditions (Fig. 3c, Syring et al., 2020a; Zehnich et al., 2020) that continued until 5.5 ka b2k. On the other hand, extensive seasonal sea ice cover is inferred on the inner Northeast Greenland continental shelf from ca. 10 to ca. 7.5 ka b2k (core PS100/270), with prolonged seasonal sea ice conditions in the late Early Holocene (Fig. 3c, Syring et al., 2020b), after which predominately perennial sea ice cover was established. We note that the PS100/270 record is the only one where seasonal sea ice conditions are inferred and consistent with the high RECAP Br values during the Early Holocene (Fig. 3h). Further south near the East Greenland coast, a low-resolution P_BIP25 record from shelf site PS2641-4 shows consistently low values (Fig. 3d), likely indicating less sea ice, or even ice-free conditions (Müller et al., 2012), further supported by the low IRD counts in core PS1878 from the Greenland Sea (Fig. 3e, Telesinski et al., 2014A). On the North Icelandic shelf, cores MD99-2272 (Xiao et al., 2017) and MD99-2269 (Cabelo-Sanz et al., 2016) show minimal IP25 concentrations until ca. 5.5 ka b2k (Fig. 3f). The inferred absence of sea ice off the north coast of Iceland is supported by contemporaneous low quartz fluxes to site MD99-2269 (Fig. 3f, Moros et al., 2006). This striking image of an essentially ice-free northern North Atlantic, with possible sea ice only at the Fram Strait, is consistent with an IP25 record showing ice-free conditions at the Barents Sea gateway (Berben et al., 2014) and with a compilation of offshore and onshore sea ice proxy indicators from the Arctic Ocean basin and coast (Stranne et al., 2014), which suggest essentially ice-free conditions persisting from the early Holocene through to about 6 ka b2k, particularly in the Canadian Arctic Archipelago, Ellesmere Island and Northeast Greenland coast.

The early Holocene minimum sea ice extent coincides with peak insolation to the high northern latitudes (Fig. 3a) and the peak of HCO temperatures as reconstructed from Greenland and Arctic ice cores (Cuffey and Clow, 1997; Gkinis et al., 2014; Lecavalier et al., 2017). In the context of oceanographic changes, the advection of Atlantic Water into the Norwegian Sea reached a maximum between 11 and 9 ka b2k (Risebrobakken et al., 2011; Eldevik et al., 2014). This advection maximum was associated with warm subsurface temperatures, as seen by planktic foraminiferal assemblage data, and can be traced all along the Norwegian Sea and north into the Fram Strait (e.g. Consolaro et al., 2018; Risebrobakken et al., 2011; Werner et al., 2016, Fig. 4a and 4b). A contemporary maximum in abundance of thermophilic molluscs is seen around Svalbard, documenting significantly warmer temperatures than today (Mangerud and Svendsen, 2018, Fig. 4a). Depleted planktic oxygen isotopes document extensive freshwater influence (e.g. Consolaro et al., 2018; Risebrobakken et al., 2011), following the deglaciation of the Fennoscandian and Svalbard-Barents Sea ice sheets (Hughes et al., 2016). While the enhanced early Holocene advection of Atlantic Water can be seen in the Fram Strait, maximum temperatures in the subsurface central Greenland Sea first occurred at 8 ka (Telesinski et al., 2014, Fig 4c). At that time a limited sea ice cover
was already established, as inferred from a low content of IRD in the PS1878 core (Fig. 3e, Telesinski et al., 2014A).

Between ca. 9 and 6 ka b2k, the warmest early Holocene summer mixed layer and surface air temperatures are seen in the Norwegian Sea and the Fram Strait (e.g. Eldevik et al., 2014; Fig. 4b), reflecting the high early Holocene northern hemisphere summer insolation (e.g. Birks and Koc, 2002; Moros et al., 2004; Jansen et al., 2008; Risebrobakken et al., 2011). Within this time interval, both Norwegian and Svalbard glaciers were smaller than today (Nesje, 2009; Solomina et al., 2015). The advection of Atlantic water was, however, reduced relative to the prior interval (Risebrobakken et al., 2011).

Following the deglaciation, cold, fresh water prevailed at the southeast Greenland margin until ca. 9.5 ka b2k, suggested by combined planktic foraminiferal fauna and oxygen isotope records (Jennings et al., 2011). While the subsurface temperatures were highest from ca. 8 ka b2k (Jennings et al., 2011, Fig. 4c), the summer mixed layer temperatures were at a Holocene maximum between ca. 10.5 and 6 ka b2k (Andersen et al., 2004; Justwan and Koç, 2008). Greenland glaciers were smaller than today during the early Holocene (Solomina et al., 2015), and the highest Holocene Greenland temperatures are reconstructed at NGRIP (Gkinis et al., 2014) and GISP2 (Cuffey and Clow, 1997).

At the North Iceland shelf the summer mixed layer temperatures were at a Holocene maximum from ca. 10.5 to ca. 6 ka b2k, based on alkenones and diatoms (Fig. 4d, Kristjánsdóttir et al., 2017; Andersen et al., 2004; Jiang et al., 2015; Bendle and Rosell-Mele, 2007). The North Iceland shelf subsurface temperatures were also higher than present during the early Holocene, as seen by the Mg/Ca values from, and reduced relative abundance of, the polar foraminifera species *N. pachyderma* (Fig. 4d, Kristjánsdóttir et al., 2017). However, a high content of C$_{37:4}$ until 8 ka documents an enhanced influence of freshwater until 8 ka (Cabello-Sanz et al., 2016). From terrestrial sites around Iceland there is also evidence for overall warmer conditions during the early Holocene (Geirsdottir et al., 2019).

Hence, the Norwegian Sea, Fram Strait, Greenland Sea, Iceland Sea and surrounding land areas experienced overall warmer conditions during the early Holocene, in line with the above interpretation of a minimum sea ice extent in the RECAP source area prior to ca. 8-9 ka b2k, from when the amount of sea ice gradually increased.

### 4.2 From approximately 5.5 to 3.8 ka b2k

Following the 25 ppb maximum at ca. 5 ka b2k, the RECAP sodium concentrations decrease substantially to a minimum of around 10 ppb at 3.8 ka b2k (Fig. 2c). During this period bromine only slightly increases (Fig. 2b), implying that the increasing Br$_{enr}$ trend is mostly driven by the sodium signal (Fig. 2d). GISP2 sodium reaches minimum values ca. 4.5 ka b2k (Fig. 2c). Similar to the early Holocene, we associate the sodium decrease in both cores with less favourable atmospheric transport of marine aerosols from the North Atlantic Ocean to the Greenland ice sheet and possibly to a reduction in sea ice extent. The combined RECAP
Na and Br$_{enr}$ series would suggest similar sea conditions at ca. 3.8 ka b2k to those found during the early Holocene: reduced extent and preferentially seasonal. Following the sudden iodine drop to ~0.01 ppb at ca. 6 ka (Fig. 2e), associated with an advance of thicker perennial sea ice (Corella et al. 2019), the iodine values are similar until ca. 4 ka b2k and a small increase to ~0.015 ppb from ca. 4 to 3.5 ka b2k. This suggests that overall limited iodine production persisted from ca. 5.5 to 4 ka b2k, followed by slightly enhanced production, possibly related to increased seasonal sea ice conditions, from ca. 4 to 3.5 ka b2k, although the iodine levels reached at this time are half of those found during the early Holocene.

None of the marine sea ice records show a similar feature to the RECAP sodium marked decrease that culminated with the 3.8 ka b2k minimum. An enduring long-term sea ice growth is indicated by IP$_{25}$ from the Fram Strait (Fig. 3b, Müller et al., 2012; Müller and Stein, 2014; Werner et al., 2016), in parallel with a slightly IRD increase in the Greenland Sea site around 5.5-5 ka b2k (Fig. 3e, Telesinski et al., 2014). On the Northeast Greenland shelf, harsher sea ice conditions continue to characterize the middle to late Holocene (Fig. 3c), with seasonal to extended sea ice conditions inferred from medium to high PIP values (Syring et al., 2020b). On the eastern North Greenland continental shelf, the PS93/025 record suggests that stable seasonal to marginal sea ice conditions characterized the last 5 ka (Fig. 3c, Syring et al., 2020a; Zehnich et al., 2020). Further downstream at the North Iceland shelf, the first indications of sea ice expanse are evinced at two sites from increased IP$_{25}$ concentrations and concomitant increase in quartz (Fig. 3f, Cabedo-Sanz et al., 2016; Xiao et al., 2017; Moros et al., 2006). At the East Greenland margin the presence of harsh seasonal sea ice conditions with a nearly closed sea ice cover in winter and landfast ice is inferred during the mid Holocene from high P$_{8}$IP$_{25}$ values in the PS2641 core (Fig. 3d, Kolling et al., 2017), in line with our interpretation of a maximum extent of MYSI at ca. 5 ka b2k based on the combined sea ice proxies from RECAP. The similarity between RECAP and the east Greenland margin site, and how these to some extent contrast the other marine sea ice records, would suggest a local sea ice source of the aerosols.

Considering the regional climate development, gradual cooling took place over Norway and in the summer mixed layer of the Norwegian Sea after the warm early Holocene (e.g. Eldevik et al., 2014; Calvo et al., 2002; Fig. 4b), around Svalbard (Mangerud and Svendsen, 2018; Fig. 4a) and in the Fram Strait (Werner et al., 2016), documented by molluscs and dinoflagellates, respectively. In contrast, the northward advection of Atlantic water in the Norwegian Sea, associated with warmer subsurface temperatures, increased (Risebrobakken et al., 2011; Eldevik et al., 2014). However, both in the Fram Strait and in the central Greenland Sea, a subsurface cooling is seen ca. 5.5 to 4 ka b2k (Fig. 4a, 4c; Telesinski et al., 2014; Werner et al., 2016). The Fram Strait and Greenland Sea cooling corroborated initial sea ice growth as indicated by IP$_{25}$ (Müller et al., 2012).

As in the Norwegian Sea and in the Fram Strait, a gradual cooling of the surface water is seen at the southeastern Greenland shelf (Andersen et al., 2004), however, the subsurface stayed warm until ca. 4 ka (Fig. 4c; Jennings et al., 2011). At the North Iceland shelf, most surface proxies (diatoms, alkenones and dinoflagellate cysts) and bottom water indicators (Mg/Ca of
benthic foraminifera) show warm conditions (Kristjansdóttir et al., 2017; Andersen et al., 2004; Justwan and Koç, 2008; Solignac et al., 2006; Fig. 4d); however, this interval is still a continuing part of the long-term gradual cooling from the early Holocene (Kristjansdottir et al., 2017). From ca. 5.5 ka the first indication of renewed glacier growth in the interior highlands of Iceland is seen (Geirsdóttir et al., 2019), in line with the first indications of increased IP$_{25}$ concentrations at the North Iceland shelf (Cabedo-Sanz et al., 2016) (Fig. 3f). Hence, despite warm ocean conditions, the combined winter precipitation/temperature and summer melt must have allowed for initial establishment of glaciers and sea ice.

The marine sea ice and climate records suggest some inconsistencies between different regions within the RECAP source area. In the Fram Strait, cooling paralleled a gradual increase in IP$_{25}$. A similar relationship between temperature and sea ice is seen at the North Iceland shelf. Hence, gradually more ice is seen as climate cools. At the east Greenland shelf, such an initial increase in IP$_{25}$ is not seen (Fig 3d). However, based on multi-proxy evidence, the existence of seasonal sea ice and at times land fast sea ice is still argued (Kolling et al., 2017). The differences seen between the Fram Strait, East Greenland shelf and North Iceland shelf are linked to the different oceanographic influences at the sites (Kolling et al., 2017). In the Fram Strait, the occurrence of sea ice is strongly influenced by the temperature of the Atlantic Water, and similarly, the occurrence of sea ice at the North Iceland shelf will be influenced by the conditions of the Irminger current. Since the EGC is constantly cold, it is not surprising that the East Greenland shelf responds differently. The interpretation of land fast ice at parts of the East Greenland shelf, in the proximity of the Renland ice core, would therefore support the interpretation of the maximum extent of MYSI around 5 ka b2k. The abrupt sea ice reduction suggested by the RECAP sodium minimum at 3.8 ka b2k, however, is not recorded in any of the marine sediment records.

4.3 From approximately 3.8 to 1 ka b2k

If considering GISP2 sodium as indicative of atmospheric circulation from both West Greenland and the North Atlantic, stable and similar conditions to the early Holocene are found from 4 to 1 ka b2k, with low sodium concentrations suggesting weaker transport to the Greenland ice sheet throughout this period (Fig. 2c). Unlike GISP2, the last 4,000 years display great variability in all RECAP records, both at millennial and centennial timescales, thanks also to the combined effect of high accumulation rate at this site and limited ice compression towards the surface. RECAP sodium concentrations from 4 to 1 ka b2k have been intermediate between the early and the mid Holocene (Fig. 2c). If interpreting Br$_{enr}$ as the type of sea ice, this marker also suggests intermediate seasonality conditions with respect to the early and the mid Holocene periods (Fig. 2d). On multi centennial timescales, similar sea ice minima are found at 3.8 and 2.1 ka b2k, both showing similar values of Na and Br$_{enr}$. In between these two sea ice minima, the sodium record hints to a period of increased sea ice followed by a maximum ca. 3 ka b2k and a decline towards the 2.1 ka b2k minimum. During this period, Br$_{enr}$ suggests highly variable sea ice type conditions within time spans of a few centuries. Unlike sodium and Br$_{enr}$, iodine concentrations show less variability around a 0.015 ppb average (Fig. 2e) throughout the 4-1 ka b2k period.
From the 2.1 ka minimum to 1 ka b2k, RECAP sodium suggests an increase of sea ice that reached a maximum ca. 1,000 years ago with similar conditions as the mid-Holocene maximum. No increase appears in the GISP2 sodium record. As inferred from both the sodium increase and $Br_{enr}$ decrease, we relate such changes at RECAP to an increase in multiyear sea ice.

The Fram Strait and North Iceland shelf sediment records suggest a cooling and clear increase of sea ice from 4 ka b2k onwards (Fig. 3b,f). Such a sea ice increase is less evident in the East Greenland coast and central Greenland Sea (Fig. 3d,e). Where seasonal sea ice was already observed between 6 and 4 ka b2k, this sea ice signal becomes stronger and implies extensive seasonal sea ice. In some records, greater variability is observed, perhaps due to episodic drift or export excursions. In the Fram Strait, both sediment cores MSM5-5-723 and -712 show an increase in $PbIP_{25}$ through the period 4-1 ka b2k, with a peak in both records about 1.5 ka b2k (Fig. 3b). On the inner Northeast Greenland continental shelf, near perennial sea ice conditions endured (Fig. 3c, Syring et al., 2020b), while seasonal to marginal sea ice conditions are reported off eastern North Greenland (Syring et al., 2020a).

On the North Icelandic Shelf, increased concentrations of $IP_{25}$ and quartz are observed, indicating persistent seasonal sea ice presence and possible maxima at 2.5 and 1.3 ka b2k (Fig. 3f).

Along the East Greenland shelf, there are inconsistencies among the available sea ice proxies. The low resolution PS2641-4 core shows rather stable sea ice conditions through the period 4-1 ka b2k (Fig. 3d, Müller et al., 2012), while a higher resolved record from the same site shows more extended sea ice concentrations from ca 5.2 to 3.2 ka b2k, followed by a phase of reduced sea ice until ca. 1.3 ka b2k (Fig. 3d, Kolling et al., 2017).

Sea ice modifications along the East Greenland coast (Fig. 3d) appear different from the generally increasing sea ice development reported in the Fram Strait and North Iceland shelf records. Such discrepancies may be related to the occurrence of landfast ice in the local fjords (Kolling et al., 2017). The overall picture of developing sea ice conditions across the Nordic Seas is supported by similar indications across the Barents Sea (Berben et al., 2017) and Arctic Ocean. From 3 to 2 ka b2k, indications of more persistent sea ice conditions are also reported for the Canadian Archipelago and northeast Greenland coast (Polyak et al., 2010; Stranne et al., 2014).

From ca. 4 ka b2k, the subsurface water temperature of the Norwegian Sea increased (Risebrobakken et al., 2011), associated with a stronger northward heat advection (Eldevik et al., 2014). The summer mixed layer temperatures continued to decrease and reached the coldest Holocene values during the last ca. 3-2.5 ka. Surface air temperatures over Norway decreased continuously since the early Holocene and Norwegian glaciers started to grow again between ca. 5 and 3 ka b2k (Eldevik et al., 2014; Nesje, 2009). Glaciers at Spitsbergen also increased in size (Solomina et al., 2015). Ocean temperatures around Spitsbergen reached a Holocene minimum ca. 3.5 to 1 ka (Mangerud and Svendsen, 2018; Fig. 4a), in line with the cold subsurface temperatures recorded in the Fram Strait (Werner et al., 2016; Fig. 4b).
4a) and weaker warm recirculating Atlantic water advection on the inner Northeast Greenland continental shelf (Syring et al., 2020b) throughout the Late Holocene. Also, the Greenland Sea subsurface water masses reached their Holocene minimum temperatures and stayed cold until ca. 2-1.5 ka b2k (Telesinski et al., 2015).

At the southeast Greenland shelf, the coldest Holocene subsurface temperatures are seen from ca. 3 ka b2k (Jennings et al., 2011; Fig. 4c). The glaciers at Greenland stayed smaller compared to those of today (Solomina et al., 2015). Temperatures at Renland continued to gradually decrease (Vinther et al., 2009), while temperatures at NGRIP dropped at 4 ka b2k and stayed cold until ca 0.6 ka b2k (Gkinis et al., 2014). At the North Iceland shelf, temperatures reached a Holocene minimum between 4 and 3 ka b2k throughout the water column (Kristjansdottir et al., 2017; Fig. 4d), and Iceland glaciers grew further under overall cold conditions (Geirsdottir et al., 2019).

Hence, most of the Nordic Seas and surrounding areas experienced cold conditions between 4 and 1 ka b2k, also reflected by the increase in IP$_{25}$ both in the Fram Strait and at the North Iceland shelf. In contrast, it is argued for land fast ice at the east Greenland shelf until ca. 3.2 ka b2k, from when less harsh seasonal sea ice conditions established and lasted until ca. 1.3 ka b2k (Fig. 3d, Kolling et al., 2017). The east Greenland record (Fig. 3d) therefore appears to more closely support the RECAP Na and Br$_{enr}$ interpretations (Fig. 3g,h) of a more variable sea ice development during the late Holocene and a sea ice reduction associated with increasing FYSI conditions ca. 2.1 ka b2k. The available sea ice records overall suggest that, locally along the east Greenland margin the sea ice history has been different than in the Fram Strait and North of Iceland.

### 4.4 The last 1 ka b2k

The increasing RECAP Na trend that started ca. 2.1 ka b2k culminated in a maximum ca. 900 years ago (Fig. 2c). The following centuries are characterized by generally high and decreasing sodium concentrations, ~20 ppb, and by concomitant increasing Br$_{enr}$ values (Fig. 2d). The combined information would therefore indicate a sea ice maximum ca. 0.9 ka b2k, generally wide but slightly declining sea ice cover towards present, associated with increasingly FYSI conditions. A significant event of sodium decrease and concurrent Br$_{enr}$ increase is seen ca. 0.7 ka b2k, possibly related to the warm Medieval Climate Anomaly (MCA) period.

The RECAP iodine concentrations increase by 50% during the last 8 centuries, reaching mean values of 0.02 ppb (Fig. 2e). Such an increase has been associated with an increase of organic and inorganic iodine emissions from both open ocean and thinner sea ice (Corella et al., 2019), thus consistent with the above interpretation of a sea ice decline and increasing FYSI. The GISP2 record shows a significant increase in sodium concentrations ca. 610 years b2k and stable concentrations ever since (Fig. 2c). Such an increase was associated with a strengthened atmospheric circulation to the site during the Little Ice Age (O’Brien et al., 1995).
The marine sediment proxy records indicate generally increasing, but also highly variable, sea ice extent over the last millennium. The Eastern Fram Strait PBIP25 records show a strong increase and maximum Holocene values (Fig. 3b). The MSM5/5-723-3 exhibits a dip in the early part of the millennium – plausibly reflecting the MCA, followed by an increase during the LIA (Müller et al., 2012). The PBIP25 record from MSM5/5-712-2 has its Holocene peak value early in the millennium. A sharp decrease is seen ca. 0.5 ka, suggesting decreased sea ice influence. However, the PBIP25 and IRD records from the nearby site MSM5/5-723-2 (Fig. 3b) show that significant amounts of sea ice were still present in the Fram Strait (Müller et al., 2012; Werner et al., 2016). The PS93/025 record off eastern North Greenland (Fig. 3c) suggests stable seasonal sea ice conditions (Syring et al., 2020a). Evidence of extended sea ice is reported in the Barents Sea during the last millennium (Berben et al., 2014, 2017).

The Northeast Greenland shelf PBIP25 record from PS2641 shows signals of increasing sea ice over the last millennium (Fig. 3d). The low-resolution record has its highest recorded value ca. 0.7 ka b2k (Fig. 3d, Müller et al., 2012), while the high-resolution record exhibits a significant increase ca.0.8 ka b2k with high variability but generally high values in subsequent centuries (Fig. 3d, Kolling et al., 2017). Such an increase is also clearly evident in foraminifera from PS2641 indicative of sea ice (Perner et al., 2015, not shown).

The North Iceland shelf records from core MD99-2269 indicate the existence of a wide sea ice cover over the past millennium (Fig. 3f). The quartz percentage is generally higher during the last millennium (Moros et al., 2006; Fig. 3f) and the IP25 values reach their Holocene maximum (Cabeno-Sanz et al., 2016; Fig. 3f). The marked increase in IP25 from ca. 0.7 ka until the 20th century is consistent with an earlier IP25 study (Massé et al., 2008, not shown) from another core from the North Iceland shelf. Both IP25 records also record relatively low values in the early part of the last millennium, suggesting reduced sea ice at this time.

The Renland Na (Br\text{enr}) interpretations (Fig. 3g,h) are reasonably consistent with the marine records with generally higher (lower) values indicating generally extended sea ice and high variability possibly related to the centennial variability recorded in the sediment sea ice records. One notable example is the abrupt drop in Na concentrations and concomitant peak in Br\text{enr} values around 0.7 ka, suggesting a period of reduced and seasonal sea ice following the MCA; this century-scale excursion is similar to the “Great Sea Ice Anomaly” recently identified (Miles et al., 2020) in multiple marine sediment records from Fram Strait, northeast and southeast Greenland shelf and the North Iceland shelf, including the IRD records shown here in Figures 3b,d,e.

From an ocean perspective, over the last 1 ka, summer mixed layer temperatures of the eastern Nordic Seas stayed cold, and cooling took place in the subsurface. The Fram Strait also stayed cold until the last ca. 100 years when a strong temperature increase is seen (Werner et al., 2016; Spielhagen et al., 2011). The water around Spitsbergen has been as of today or warmer (Mangerud and Svendsen, 2018; Fig 4a). In the Greenland Sea, increased abundance of Turborotalita quinqueloba has been used to infer a warming (Telesinski et al., 2015). On the east and southeast Greenland shelf, climate has varied (Kolling et al., 2017;
Jennings et al., 2011), in association with the dark ages cold period, the MCA and LIA. Furthermore, the Greenland glaciers grew over the last 1 ka (Solomina et al., 2015), when the Renland temperature record shows the coldest Holocene conditions (Vinther et al., 2009). A warming is, however, seen at NGRIP over the last ca. 600 years (Gkinis et al., 2014). Near Iceland it remained cold and the shelf bottom water cooled over the last 1 ka, however, the summer mixed layer temperatures experienced a warming relative to the preceding cold interval (Kristjansdottir et al., 2017; Geirsdottir et al., 2019).

As noted above, the highest Holocene IP$_{25}$ and PIP$_{25}$ values are seen at the east Greenland shelf (Fig. 3d), in the Fram Strait (Fig. 3b) and north of Iceland (Fig. 3f) within the last 1 ka (Müller et al., 2012; Kolling et al., 2017; Cabedo-Sanz et al., 2016). The Na and Br$_{enr}$ records from Renland mostly show higher and lower values than today respectively (Fig. 3g,h), suggesting overall more extensive sea ice and MYSI conditions during the last millennium.
Figure 2. RECAP Holocene ice core records of $\delta^{18}$O, Br, nssBr, Na, Br$_{enr}$ and I.

a. RECAP $\delta^{18}$O (black) and daily mean 65 °N summer solstice insolation (red, Laskar et al., 2004). b. RECAP total bromine (red) and non-sea-salt bromine (black). c. RECAP sodium (black) and GISP2 sodium (blue, smoothed using a 100-yr gaussian kernel as in O’Brien et al., 1995)). The timing of the suggested maximum and minimum sea ice conditions mentioned in the text are indicated respectively with blue (5.5 and 0.9 ka b2k) and red arrows (3.8, 2.1 and 0.7 ka b2k). d. RECAP bromine enrichment. e. RECAP iodine (Corella et al., 2019).

The RECAP raw series in panels b,c,d,e are shown with light colors. The solid lines are low-pass gaussian kernel filters. FYSI = first year sea ice; MYSI= multi year sea ice; OW=open water.
Figure 3. Sea ice proxy records extracted from RECAP and from marine sediment cores in the North Atlantic Ocean. See Figure 1 for the core sites.

a. RECAP δ¹⁸O (black) and daily mean 65 °N summer solstice insolation (red, Laskar et al., 2004).

b. P₈IP₂₅ records from MSM5-5-712-2 (grey, left axis, combined from Müller et al., 2012 and Müller and Stein, 2014) and MSM5-5-723-2 (black, left axis, Werner et al., 2016). IRD record (150-200 µm range) from MSM5-5-723-2 (blue, right axis, Werner et al., 2016).

c. Records from PS93/025 (blue, Syring et al., 2020a) and PS100/270 (red, Syring et al., 2020b). Light and dark series correspond to P₈IP₂₅ and P₉IP₂₅ respectively.

d. P₈IP₂₅ from PS2641-4 (grey, Müller et al., 2012). P₈IP₂₅ from PS2641 (black, smoothed using a gaussian kernel, Kolling et al., 2017).

e. IRD from PS1878 (Telesinski et al., 2014A).

f. MD99-2269 IP₂₅ (grey, left axis, Cabedo-Sanz et al., 2016) and Quartz % (blue, right axis, Moros et al., 2006). MD99-2272 IP₂₅ (black, left axis, Xiao et al., 2017). Note that the MD99-2272 IP₂₅ concentrations are zero before ca. 7 ka b2k.

g. RECAP sodium (black) and GISP2 sodium (blue, smoothed using a 100-yr gaussian kernel as in O’Brien et al., 1995). The timing of the suggested maximum and minimum sea ice conditions mentioned in the text are indicated respectively with blue (5.5 and 0.9 ka b2k) and red arrows (3.8, 2.1 and 0.7 ka b2k).

h. RECAP bromine enrichment.

i. RECAP iodine (Corella et al., 2019).

The RECAP raw series in panels g,h,i are shown with light colors. The solid lines are low-
pass gaussian kernel filters. FYSI = first year sea ice; MYSI= multi year sea ice; OW=open water.

Figure 4. Climate proxy records from marine sediment cores in the North Atlantic Ocean. See Figure 1 for the core sites. The marine datasets presented are examples, not a full representation, of the datasets and studies discussed throughout the text.

a. MSM5-5-712-2 (yellow, left axis) and MSM5-5-723-2 (red, left axis) SST\textsubscript{100m} reconstructed from planktic foraminifera and the Husum and Hald (2012) transfer function (Werner et al., 2016 and references therein). West Svalbard August SST (black, right axis) estimated from the occurrence of molluscs (Mangerud and Svendsen, 2018).

b. MD99-2284 (black) and MD95-2011 (red) subsurface SST (SubT, ca. 100 m) based on planktic foraminifera and a Maximum Likelihood transfer function method (Risebrobakken et al, 2011). MD95-2011 SST based on alkenone (blue, Calvo et al., 2002).

c. PS1878 SST\textsubscript{100m} based on planktic foraminifera and the Husum and Hald (2012) transfer function (Telesinski et al., 2014A). MD99-2322 upper 200m-integrated temperature based on planktic foraminifera (red, Jennings et al., 2011).

d. MD99-2269 SST based on alkenone (black) and SubT based on Mg/Ca ratios in Neogloboquadrina pachyderma (sin) (red, Kristjánsdóttir et al., 2017).
e. RECAP sodium (black) and GISP2 sodium (blue, smoothed using a 100-yr gaussian kernel as in O’Brien et al., (1995)). The timing of the suggested maximum and minimum sea ice conditions mentioned in the text are indicated respectively with blue (5.5 and 0.9 ka b2k) and red arrows (3.8, 2.1 and 0.7 ka b2k).

f. RECAP bromine enrichment. g. RECAP iodine (Corella et al., 2019).

The RECAP raw series in panels e,f,g are shown with light colors. The solid lines are low-pass gaussian kernel filters. FYSI = first year sea ice; MYSI= multi year sea ice; OW=open water. SubT=sea subsurface temperature; SST$_{100m}$=sea subsurface temperature at 100 m water depth; SST=sea surface temperature.

5. Summary and conclusions

The reconstruction of past sea ice conditions using ice core records is still one of the most intriguing challenges in ice core science. Despite increasing efforts by the ice core community in recent years, uncertainties still exist. In this work we used the sodium, bromine and iodine records in the RECAP core to test their skills as proxies of sea ice in the northern North Atlantic Ocean region and compared them with sea ice and climate records from marine sediment cores. We suggest that RECAP sodium is preferentially sourced from sea ice and modulated by atmospheric transport to the Greenland ice sheet. We base the interpretation of Br$_{enr}$ as a marker of first-year sea ice on studies linking bromine explosions to atmospheric bromine species, although we acknowledge that a robust quantitative link has yet to be found. Iodine records are proven to be helpful for the interpretation of sodium and bromine records and we strongly recommend this marker to be measured in future ice core analysis campaigns. Overall we find some degree of agreement between ice core and marine sediment core records and summarize the following conclusions on the evolution of North Atlantic sea ice during the Holocene:

- In the early Holocene lowest extent of sea ice was found in the Fram Strait, the East Greenland shelf and north of Iceland, likely driven by maximum influx of warm Atlantic waters. Na and Br$_{enr}$ levels suggest that the sea ice cover was reduced and mostly seasonal in nature, possibly also formed in the Arctic Ocean basin and exported to lower latitudes.

- From ca. 8-9 ka b2k a gradual increase in sea ice is observed in the northern North Atlantic. A sea ice maximum is suggested from the RECAP records and at the east Greenland shelf ca. 5 ka b2k, contrasting the continued gradual increase seen in the Fram Strait and at the Icelandic shelf. This spatial difference could suggest a local east Greenland shelf source of marine aerosols to RECAP at this time. However, from 5 to 3.8 ka b2k the RECAP proxies suggest a rapid sea ice reduction that is not evidenced by any of the marine sediment records.

- In the late Holocene continuous cooling took place in parallel with an increased sea ice extent in the Fram Strait and over the North Icelandic shelf. At the East Greenland shelf great variability with generally intermediate sea ice conditions between early Holocene minimum and mid Holocene maximum is suggested from the RECAP records.
The maximum sea ice conditions are inferred during the last 1000 years, with evidence of sea ice reduction and more seasonal conditions following the warm Medieval Climate Anomaly ca. 0.7 ka b2k.

6. Acknowledgement
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7. Declaration of interests
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

8. Author contributions
PV and AS conceived the experiment. NM, RE, PV, HAK, CT, AS collected the ice core samples and ran the experimental analyses. NM, BR, MM wrote the paper with inputs from all authors.

9. Data Availability
All RECAP data will be made available on NOAA paleoclimate and PANGAEA online data archives.

10. References


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Appendix A

Table A.1. Radiocarbon dates, ash horizons and calibrated ages used to create the updated age models. We have updated the age models for MD99-2269 (Stoner et al., 2007), MSM5/—712—2 (Müller et al., 2012) and MSM5/—723—2 (Müller et al., 2012) using the Marine13 calibration dataset and Calib 7.1. Ages are based on linear interpolation between dated tie points. The core top age was set to the year of coring. For MD99—2269 the age model follows Tabel 3 in Stoner et al. (2007), presenting a combined age model for MD99—2269 and MD99—2322, where the two cores are set at a common depth scale(+) based on the extensive radiocarbon dating of both cores and a further correlation of paleomagnetic secular variations. For site PS2641 we use the age model as published by Perner et al., 2015(−). We have updated the age model for the PS2641 dataset published by Müller et al., 2012 to the age model from Perner et al., 2015. The oldest dates from Muller et al., 2012, not included by Perner et al, 2015 due to a shorter focus interval, have been updated following the same choices as for the other updated age models(−). ∆R=±0.± for all 13C AMS based chronologies. The age model for MD99—2272 is used as published in Xiao et al., 2017, solely based on identified tephras horizons. All age models for all records shown are presented as years before 2000 (b.k.).

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| KIA 45218 | MSM5/5-712-2 | 28 | NPS | 1985±25 | 0±0 | 1511-1584 | 1.00 | 1546 | Müller et al., 2012 |
| KIA 45219 | MSM5/5-712-2 | 41 | NPS | 2565±25 | 0±0 | 2199-2297 | 1.00 | 2241 | Müller et al., 2012 |
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