1	Sea-ice dynamics in an Arctic coastal polynya during the past
2	6500 years
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ABSTRACT

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The production of high-salinity brines during sea-ice freezing in circum-arctic coastal polynyas is thought to be part of northern deepwater formation by supplying additional dense waters to the Atlantic meridional overturning circulation system. In order to better predict the effect of possible future summer ice-free conditions in the Arctic Ocean on global climate, it is important to improve our understanding of how climate change has impacted sea-ice and brine formation, and thus eventually dense water formation during the past. Here, we show temporal coherence between sea-ice conditions in a key Arctic polynya (Storfjorden, Svalbard) and patterns of deep water convection in the neighboring Nordic Seas over the last 6500 years. A period of frequent sea-ice melting and freezing between 6.5 and 2.8 ka BP coincided with enhanced deep water renewal, while near-permanent sea-ice cover and low brine rejection after 2.8 ka BP, likely reduced the overflow of high salinity shelf waters, concomitant with a gradual slow-down of deep water convection in the Nordic Seas, which occurred along with a regional expansion in sea-ice and surface water freshening. The Storfjorden polynya sea-ice factory restarted at ~0.5 ka BP, coincident with renewed deep water penetration to the Arctic and climate amelioration over Svalbard. The identified synergy between Arctic polynya sea-ice conditions and deep water convection during the present interglacial is an indication of the potential consequences for ocean ventilation during states with permanent sea-ice cover or future Arctic ice-free conditions.

INTRODUCTION

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The sinking of dense waters on Arctic and Antarctic shelves through recurrent cooling and rejection of salt during sea-ice growth is a key contributor to global ocean circulation (Killworth 1983) with 10% of contemporary deep waters formed in the Arctic Ocean and the Barents Sea derived from these brine-enriched shelf waters (Quadfasel et al. 1988). High seaice production in Arctic coastal polynyas facilitates dense water production and ocean stratification, thus inhibiting the upward mixing of warm Atlantic water and sea-ice melt (Aagaard et al. 1981). Coastal polynyas are persistent and recurrent areas of open water that occur within locations of otherwise consolidated and thicker ice cover. Amongst these, the Storfjorden coastal polynya in southern Spitsbergen (Fig. 1) is known to be an important seaice factory (Haarpaintner et al. 2001) and a significant source of brine rejection (Quadfasel et al. 1988). Dense brine-enriched waters from Storfjorden cascade downslope before flowing north (Schauer 1995), where they descend to depths of more than 2000 m (Jungclaus et al. 1995) and account for up to 15% of the total dense water generated in the entire Arctic (Cavalieri and Martin 1994; Skogseth et al. 2004). However, this has likely changed in the past, either as a contributor to, or as a result of, climate change at high latitudes. Indeed, millennialscale reconstruction of past brine formation in the Storfjorden polynya based on the sedimentary distribution of calcareous and agglutinated benthic foraminifera has revealed a systematic pattern of high (low) intensities during cold (warm) climate periods over the last 15,000 years (Rasmussen and Thomsen 2014; Rasmussen and Thomsen 2015). In contrast, large annual variability in brine formation has also been observed during the most recent warm periods during the last century. Thus, reduced brine formation and, hence, strongly reduced export of dense water to the Arctic Ocean occurred during periods with exceptionally warm Atlantic water advection and reduced sea-ice coverage in the Barents Sea, while intense brine formation was re-established during periods of recurrent cooling (Arthun et al. 2011).

Accordingly, since the process of brine rejection is largely dependent on the seasonal formation of sea-ice, past reconstruction of sea-ice coverage coupled with environmental inferences from benthic foraminifera assemblages in the Storfjorden polynya (Rasmussen and Thomsen 2014; Rasmussen and Thomsen 2015) provides a more direct indication of past brine formation and thus, potentially, a new measure for evaluating the significance of Arctic coastal polynyas with respect to dense water formation on a glacial-interglacial timescale. This approach provides an alternative to the still disputed use of benthic foraminiferal stable isotope records as a measure of the influence of brine-enriched shelf waters on deep water production (Dokken and Jansen 1999; Mackensen and Schmiedl 2016; Rasmussen and Thomsen 2009). In this study, we combine downcore records of organic geochemical biomarkers of sea-ice variability (IP25) (Belt et al. 2007) and open-water phytoplankton (brassicasterol) with source-specific, sea-ice derived terrigenous sediments, supplemented by published agglutinated foraminifera (% of total benthic foraminifera) (Rasmussen and Thomsen 2014; Rasmussen and Thomsen 2015). We hereby present evidence that changes in sea-ice coverage and inferred brine formation in the Storfjorden polynya over the past 6500 years, coincide with past variability in deep water renewal in the Nordic Seas. As such, we highlight the importance of Arctic coastal polynyas as one significant driver of deep water renewal processes during the present interglacial.

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REGIONAL SETTING

Storfjorden, in southeastern Spitsbergen, is a ca. 200-km-long inlet, separated from the open ocean by a shallow sill (~120 m). Surface waters are seasonally stratified, with sea-ice and brine formation taking place each winter in the inner fjord (Schauer 1995). Strong northeasterly winds blow sea-ice away from the eastern shelf, producing a large latent heat polynya, where high sea-ice production and continuous freezing generates cold (<-1.9°C) and salty (34.8 to >35.8 psu) water (Haarpaintner et al., 2001; Skogseth et al., 2004), which sinks and fills the

central basin, eventually overflowing the sill. Depending on its salinity (34.3-35.3 psu) (Skogseth et al., 2004), the brine may continue downslope reaching 2000 m into the deep-intermediate water of the Greenland Sea (Jungclaus et al. 1995; Quadfasel et al. 1988). Surface currents in Storfjorden are controlled by southwestward flowing, ice-covered polar waters from the Arctic Ocean. The East Spitsbergen Current (ESC) balances the bottom currents that transport the dense water out of Storfjorden towards the deep ocean. Sediments deposited in Storfjorden are enriched in organic carbon (up to 2.4 wt.%) and largely dominated by terrigenous derived organic matter (Winkelmann and Knies 2005). Terrigenous sediments are largely supplied by local (fast) ice entrainment processes and episodic freezing/melting processes in the polynya. Alternatively, terrigenous sediments transported by polar surface waters (ESC) to Storfjorden are released during frequent melting episodes and deposited in Storfjorden (Winkelmann and Knies 2005).

MATERIAL AND METHODS

We studied inorganic elements and organic biomarkers in sediment surface samples (0-1 cm) taken with multicorer equipment from the western Barents Sea (Fig. 1, Tab. 1) and a gravity core JM10-10GC (77.41 °N, 20.10 °E, 123 m water depth, hereafter referred to as JM10), taken within the Storfjorden polynya where brines form today (Fig. 1). The surface samples were sliced onboard, frozen, and subsequently freeze-dried prior to analysis.

Inorganic geochemistry

All surface samples were analysed for major and trace elements by using a Philips PW 1480 WD XRF instrument equipped with an Rh X-ray tube. For XRF major elements about 2 g of finely-ground sample was pre-heated over a gas burner to remove any organic material before pre-ignition at $1000 \pm 50^{\circ}$ C for at least 1 hour. 4.200 ± 0.005 g Li₂B₄O₇ (Claisse, Quebec,

Canada) is mixed with 0.600 ± 0.005 g pre-ignited sample and fused to glass beads in Pt - 5% Au-crucible. The method for determination of trace element with XRF is based on pressed pellets. 1.2 ± 0.005 g Hoechst wax was mixed with 5.4 ± 0.005 g dried and fine-ground sample material in a Spex Mixer/Mill for at least 1 minute. The mixture was pressed to a pellet in a Herzog pelletizing press, with an applied force around 20 kN for 20 seconds. Methods accuracy for arsenic (As) and aluminium (Al) was tested with several certified reference materials (CRM), as shown in Table 2 and 3. Relative percent difference between the duplicate samples was within \pm 10%. Al-normalisation was applied for As data in core JM10 to avoid dilution due to variable sedimentation rates (19-104 cm/ka) in the record (Rasmussen and Thomsen 2014). A correlation coefficient $R^2 = 0.88$ between As/Al ratio and As concentrations (ppm) in core JM10 indicate no dilution effects on the As concentrations in the sediments. Concentration of leachable elements in the same sample set was measured by ICP-AES with the instrument PerkinElmer 4300 DV. Nitric acid extraction was used to estimate the amounts of As and Al present in the non-silicate fraction of the sediment in all surface sediments and core JM10. 1.000 ± 0.001 g of freeze-dried sediment was digested with 20 ml 7 M HNO₃ for 30 min at 120 ± 4°C in autoclave (CertoClav Sterilizer, CV-EL 18LGS), following the procedure described in the Norwegian Standard NS 4770 from 1994. After cooling the sample was filtered through Whatman grade 597 and further diluted. The analysed solution contains 10 ppm Rh as internal standard and about 10% HNO₃ (v/v). Method quantification limits, respectively 20 mg/kg Al and 2 mg/kg As, is based on 10 times the standard deviation for 10 replicates of method blanks. Relative percent difference between the duplicate samples was within \pm 10%. Certified reference material Mess-3 (marine sediment for trace elements and other constituents, NRC-CNRC Canada) was routinely analysed to test methods analytical performance. The correlation coefficients between XRF and ICP-AES based arsenic and

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aluminium concentrations of 73 surface samples is r^2 =0.95 and 0.75, respectively. Arsenic concentrations in the remaining text are based on the ICP-AES extraction method to allow comparison with published As concentration in floodplain and overbank deposits from Spitsbergen (Ottesen et al., 2010) (Fig. 1).

Biomarkers

The biomarkers IP₂₅ (Belt et al. 2007) and brassicasterol were quantified following addition of internal standards (9-octylheptadec-8-ene, 10 μL; 10 μg mL⁻¹; 5α-androstan-3β-ol, 10 μL; 10 μg mL⁻¹, respectively), extraction (DCM/Methanol; 3 x 3 mL, 2:1 v/v) and purification of extracts using silica column chromatography (IP₂₅: hexane, 6 mL; brassicasterol: 20:80 methylacetate/hexane, 6 mL). Further purification of the IP₂₅ containing fraction was achieved by Ag-ion chromatography (Supelco discovery Ag-Ion; 0.1 g) with saturated hydrocarbons (hexane; 1 mL) and unsaturated hydrocarbons (including IP₂₅: acetone; 2 mL) eluted as two single fractions. All partially purified fractions were analysed using gas chromatography - mass spectrometry (GC – MS) according to established methods (Belt et al. 2012). Brassicasterol was derivatized (BSTFA; 50 μL, 70°C, 1h) prior to analysis by GC – MS.

Chronology

The chronology of the upper 325 cm of JM10 is based on 7 AMS ¹⁴C radiocarbon dates obtained on bivalves and monospecific samples of the benthic foraminiferal species *N*. *labradorica* (Table 4) (see details in Rasmussen and Thomsen, 2015). All AMS ¹⁴C dates were calibrated to calendar ages by applying the Calib7.02 programs (Stuiver and Reimer 1993) and the Marine13 calibration curve (Reimer et al. 2013). The applied age model is consistent with the published model of Rasmussen and Thomsen (2015). The sedimentation rates vary between 19 and 104 cm/ka, with highest values (104 cm/ka) in the upper part of the

sediment cores (\sim 1.0 – 0.5 ka BP), and lowest values (19 cm/ka) between \sim 2.8 and \sim 1.0 ka BP. Sedimentation rates in the lowermost part of the record (2.8 - \sim 6.5 ka BP) vary between 49 and 76 cm/ka. The quality of the dated material was checked by measuring bivalves and *N. labradorica* in two different samples within the same depth interval (324-326 cm). The dates are identical within error (Table 4), excluding the possibility of re-deposition of the bivalves in this environmental setting. However, we caution the reader that the observed changes in sedimentation rates between 2.8 and 1.0 ka BP are based on dating results from bivalves only, due to the lack of sufficient planktic or benthic foraminifera in this interval.

RESULTS AND DISCUSSION

Proxies for sea-ice dynamics

In order to interpret our down-core record, we first provide the background to our combined proxy data by presenting measurements obtained from surface sediments that reflect the modern physico-geography of the region. Arsenic (As) concentration in near-shore unpolluted marine sediments is normally between 5 and 10 ppm (Wedepohl 1991). Sedimentary arsenic is principally associated with sesquioxide material (mostly hydrous iron oxides) as shown by a positive correlation between As and Fe ($r^2 = 0.65$). Arsenic concentration in our Barents Sea surface sediments varies between 2 and 105 ppm, with a clear geographical boundary along the Marginal Ice Zone (MIZ) (Fig. 2). South of the MIZ, the mean As concentration (7 ppm) resembles values in uncontaminated soils from northern Scandinavia (Reimann et al. 2009), while for sites north of the MIZ, a mean concentration of 27 ppm is significantly higher than the global average for coastal marine sediments (5-10 ppm; (Wedepohl 1991)). The enrichment in the northern sediments is, however, probably not related to diagenetic redox-cycling processes seen in other shelf environments (Sullivan and Aller 1996) since As anomalies are not correlated with other redox-sensitive elements such as Mn ($r^2 < 0.2$). Instead, it is more

likely that natural sources of As-rich deposits and dissolved As in the water column are the causes of the sedimentary enhancements. As-rich sediments are most likely transported by seaice and released along the MIZ (Hölemann et al. 1999), while dissolved As can be taken up by phytoplankton blooms in the MIZ and thus incorporated into the sedimentary cycle (Broecker and Peng 1982). Indeed, local As anomalies are reported from Paleogene sequences, SW Spitsbergen (Fig. 1) (Ottesen et al. 2010) and As concentrations as high as 225 ppm have been recorded in coal seams interbedded with marine and lacustrine siltstones and shales (Jensen 2000). Arsenic anomalies (>50 ppm) also occur in nearby floodplain sediments sourced from Carboniferous-Cretaceous organic-rich deposits along the coastline adjacent to Storfjorden (Fig. 1) (Ottesen et al. 2010). Coastal freezing processes along the shoreline or within coastal polynyas (Eicken et al. 1997) allow entrainment of As-enriched sediments in sea-ice with subsequent release during melt within the MIZ. Other As anomalies in sediments are reported from the Laptev Sea and Kara Sea shelves (Hölemann et al. 1999; Loring et al. 1998; Loring et al. 1995), where incorporation of As-enriched particles in newly formed sea-ice and transportation within the Transpolar Drift and East Spitsbergen Current may have caused the As-anomalies identified below the MIZ in the northwestern Barents Sea (Fig. 2). Hence, we use the As-anomalies in the sedimentary record as evidence for newly formed sea-ice that allowed incorporation of terrigenous (As-rich) particles in coastal areas, and subsequent seaice melting and release of As-rich ice-rafted sediments within the MIZ. To complement the As data, we also measured the distribution of the organic geochemical sea-ice proxy IP₂₅ in the same surface sediments. IP25 is a highly specific lipid biosynthesized by certain diatoms residing in the underside of seasonal Arctic sea-ice (Brown et al. 2014) and whose presence and abundance in sediments is strongly associated with overlying sea-ice cover (Belt and Müller 2013; Belt et al. 2007) including the Barents Sea (Belt et al. 2015; Navarro-Rodriguez et al. 2013). In general, higher or increasing sedimentary abundances of IP25 are positively

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associated with seasonal sea-ice occurrence (or change) as shown through various surface and downcore records from across the Arctic (Belt and Müller 2013). However, lower IP₂₅ abundances have been found in sediments from regions of much higher or near-permanent sea-ice cover including East Greenland (Alonso-Garcia et al. 2013) and the High Arctic (>80°N) (Vare et al. 2009; Xiao et al. 2015). In such settings, the abundances of phytoplankton biomarkers including brassicasterol, are also low; both observations being consistent with light-inhibited, and therefore low, biological productivity.

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Sea-ice dynamics in an Arctic coastal polynya

The accumulation of IP₂₅ in the MIZ sediments (Navarro-Rodriguez et al. 2013) closely resembles the spatial distribution of As (Fig. 2) consistent with recurrent freezing and melting of sea-ice in the region. Furthermore, the release of sea-ice debris is known to stimulate phytoplankton blooms during spring, resulting in high export production rates during peakbloom stages within the MIZ (Reigstad et al. 2011). Through particle scavenging, this provides an additional mechanism that leads to enhanced sedimentary As. Downcore analyses of these sea-ice (IP25, As) and phytoplankton (brassicasterol) proxies (Fig. 3) therefore provide a temporal measure of variable sea-ice coverage in the Storfjorden polynya and, by inference, changes in high-salinity brine rejection due to variable polynyal activity resulting from freezing/melting processes. The results are discussed for three different time intervals (6.5-2.8 ka, 2.8-0.5 ka, <0.5 ka BP), with the boundary at 2.8 ka based on the gradual decline of the IP₂₅ concentration between 3.0 and 2.5 ka and the abrupt increase in percentages of agglutinated forams at this time (Fig. 4). Notched box-whisker plots for the distributions of As/Al, IP25, and brassicasterol in these time intervals (Fig.3) confirm that, for all parameters, the median in the interval 2.8-0.5 ka is largely different from the median in the time intervals 6.5-2.8 ka and 0.5-0 ka on 5% level. However, on a 5% level, notched regions of As/Al distribution in intervals 2.8-0.5 ka and 0.5-0 ka do overlap (Fig. 3), implying that paleoenvironmental conditions for sedimentary As deposition during these time intervals were not significantly different compared to the interval 6.5-2.8 ka (see discussion below). Consistent with the surface sediment data, As/Al and IP25 co-vary in the 6500-year record (core JM10), with highest values between 2.8 and 6.5 ka, a decreasing trend towards 0.5 ka, and an increase towards the core-top (Fig. 4). The occurrence of IP25 at the core-top is consistent with modern observations of annual sea-ice formation in the polynya (Haarpaintner et al., 2001), while its presence throughout the record demonstrates persistent (but variable) seasonal seaice occurrence. Highest IP25 concentrations and As/Al ratios between 6.5 and 2.8 ka are accompanied by enhanced brassicasterol concentrations and lower relative abundances of agglutinated foraminifera (Fig. 4), implying a variable sea-ice margin and recurrent melting/freezing periods with associated phytoplankton blooms. These modern-like conditions, with seasonal sea-ice formation and increased polynyal activity, are in accordance with environmental inferences from calcareous and agglutinated foraminiferal assemblages in the fjord during this time interval (Rasmussen and Thomsen 2015). Our proxy data are also consistent with simulations of increased sea-ice production (+15%) and extent (+14%) in the circum-Arctic (Blaschek and Renssen, 2013), likely as a consequence of the flooding of the Arctic Siberian shelf (Bauch et al. 2001) and potentially positive ocean-sea ice-atmosphere feedbacks in the Barents Sea (Semenov et al. 2009), and further evidenced by reduced sea surface temperatures off western Svalbard around 5 ka (Werner et al. 2013) (Fig. 5). Elsewhere, a gradual southward expansion of the MIZ has been reconstructed for the Canadian Archipelago (Vare et al. 2009) and the Fram Strait (Müller et al. 2012). Werner et al. (2013) hypothesized that the occurrence of heavy winter sea-ice off the western Svalbard coast after 5.2 ka BP is due to established modern sea-ice production in the Arctic Ocean after the Holocene transgression. The distinct cooling trend in the Nordic Sea connected to the sea-ice

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expansion as a consequence of the flooding (Blaschek and Renssen 2013) and declining insolation (Laskar et al. 2004) (Fig. 5) provides the prerequisite for the advection and persistent presence of seasonal sea-ice in the Storfjorden polynya. A distinct change in sea-ice coverage in the Storfjorden polynya occurred after 2.8 ka BP. While a seasonally fluctuating MIZ similar to its present (winter) location prevailed along western Spitsbergen (Müller et al. 2012), reduced sea-ice and phytoplankton biomarkers, together with higher mean proportions of agglutinated foraminifera (Fig. 4), demonstrate a clear change in sea-ice conditions in the Storfjorden polynya between 2.8 and 0.5 ka, with low entrainment/freezing of terrestrial sediments, diminished surface water productivity and dense/packed sea-ice coverage. At the same time, on the western Svalbard/Barents Sea margin, decreasing values in planktic δ^{13} C records and a downward migration of the planktic foraminifera Neogloboquadrina pachyderma sin., also point to surface water freshening and saltier, warmer sub-surface waters (Fig. 5) (Sarnthein et al. 2003; Werner et al. 2011), thus preconditioning the setting for extensive sea-ice formation. The dominance of the calcareous benthic foraminifera species Elphidium excavatum in the Storfjorden sediments provides further evidence for more extensive seasonal ice cover (Rasmussen and Thomsen 2015). A permanent sea-ice cover in Storfjorden is also in agreement with observations from western coastal Svalbard, where enhanced formation of shore-fast sea-ice and/or dense sea-ice coverage has been suggested (Forwick and Vorren 2009). On the other hand, pulses of advected Atlanticwater along the Barents and Svalbard margin during this period (Sarnthein et al. 2003; Werner et al. 2014) did not influence the persistent sea-ice coverage in Storfjorden. However, confirmation of the latter requires a higher resolution IP25 record, as intervals with more variable sea-ice conditions inferred from highly fluctuating proportions of agglutinated foraminifera are not covered with the current IP25 dataset (Fig. 4). In the meantime, the highresolution As/Al record of constantly low values (<0.5) throughout this interval implies dense

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sea-ice coverage, suggesting that increased proportions of agglutinated foraminifera in some intervals may reflect variable preservational conditions under the dominant influence of Arctic waters rather than strong polynyal activity and thus brine formation. However, the latter needs to be explored further with additional records from the Storfjorden area and adjacent trough. Rapidly increasing phytoplankton production, and enhanced IP25 concentrations demonstrate that the sea-ice factory restarted abruptly ~0.5 ka BP, at which time, sediment entrainment/release processes also recovered, with higher As/Al ratios towards the core-top (Fig. 4). The establishment of a highly fluctuating sea-ice boundary would have eventually led to formation of a coastal polynya with seasonally variable sea-ice conditions. Enhanced IP25 and brassicasterol concentrations are largely consistent (except one interval centered around 0.3 ka) with increased proportions of agglutinated foraminifera (Fig. 6) supporting inferences by Rasmussen and Thomsen (2014) of an intensified, but variable polynyal activity. Thus, constantly high sea-ice production throughout the last ~500 years is likely the result of inferred mild summer temperatures on Spitsbergen including the Little Ice Age (Fig. 5) (D'Andrea et al. 2012). These modern-like conditions in Storfjorden, with variable sea-ice coverage over the last 500 years, contrast the more dense/packed sea-ice conditions in the preceding interval (~2.8 to 0.5 ka BP), but corroborate a recent biomarker-based sea-ice reconstruction for western Svalbard, which showed a gradual decline in spring sea-ice concentration over the past 400 years (Cabedo-Sanz and Belt 2016).

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Relationship between sea-ice, brines and deep water production

In modern times, it is well known that dynamic sea-ice production and brine rejection within the wind-driven polynyas in the circum-Arctic are important contributors for deep water convection in the Nordic Seas and Arctic Ocean (Aagaard et al. 1985; Schauer 1995; Skogseth et al. 2004). Further, Bauch et al. (2001a) suggested that for the Last Glacial Maximum,

enhanced sea-ice production and dense bottom water formation could be attributed to the formation of katabatic wind-driven polynyas in front of the western Svalbard-Barents Sea ice sheet. Similarly, based on calcareous and agglutinated foraminifera, Rasmussen and Thomsen (2014, 2015) showed that the strength of brine formation in the Storfjorden polynya over the last 15 ka BP was largely related to climatic conditions, with enhancements during cold periods (and vice versa). However, such studies were based on rather unselective proxies for sea-ice reconstruction (i.e. stable isotopes and assemblages of benthic and planktic foraminifera), which potentially limits their value in terms of confirming the significance of brine rejection on deep water formation in palaeo records.

In the present study, we demonstrate temporal coherence between our more direct proxy-based sea-ice reconstruction (and inferred brine intensity changes) and changes to deep water convection obtained from local and other regional records from the Nordic Seas (Fig. 5). Thus, the recurrent freezing/melting of sea-ice in the Storfjorden polynya and associated strong brine formation between 6.5 and 2.8 ka BP coincides with less radiogenic ε_{Nd} values (-9.4 – -10.6) from western Spitsbergen, as seen for present-day deep water penetration to the Arctic Ocean (Werner et al., 2014) (Fig. 5). During the same interval, high convection rates in most areas of the Nordic Seas is evident from high carbon isotope values in both planktic and benthic foraminifera (Bauch et al. 2001; Sarnthein et al. 2003), together with a period of maximum ventilation in the Greenland Sea (Fig. 5) (Telesiński et al. 2015; Telesiński et al. 2014) and AMOC strengthening (Hall et al. 2004). In contrast, more permanent sea-ice cover and probably subdued brine formation in Storfjorden polynya after 2.8 ka, is accompanied by a prominent shift to more radiogenic ε_{Nd} along the western Spitsbergen continental margin (Fig. 5) (Werner et al., 2014). At the same time, freshening of surface waters and intensification (thickening) of sea-ice in the Fram Strait has been deduced from carbon isotope data of planktic

foraminifera (Werner et al. 2013) (Fig. 5) and elevated IP₂₅ abundances (Müller et al., 2012), while increased sea-ice production in the Arctic and export through Fram Strait also coincides with a proposed reduction of deep convection in the Greenland Sea (Telesiński et al., 2014) (Fig. 5). Modelling results also suggest that negative anomalies in total solar irradiance ~2.7 ka may have been responsible for local shutdown of deep water formation in the Nordic Seas at this time (Renssen et al. 2006) which, when superimposed on decreasing insolation (Fig. 5), may have stimulated positive oceanic feedbacks such as enhanced stratification, expansion of sea-ice and less deep water formation leading to additional cooling and more sea-ice (e.g. Telesiński et al. 2014; 2015). Regardless of the ultimate trigger for the abrupt changes in sea-ice coverage in Storfjorden polynya at ~2.8 ka, the timing of such solar-forced cooling events demonstrates that the most severe climatic conditions in the Nordic Seas and circum-Arctic reduced the contribution of Arctic sea-ice factories (i.e. polynyas) to deep water production.

The enhancement of the sea-ice factory and phytoplankton production in Storfjorden at \sim 0.5 ka BP, when recurrent freezing/melting of sea-ice in the polynya coincides largely with the increased admixture of deep waters from the Nordic Sea (less radiogenic ϵ_{Nd}) (Fig. 5) and increased proportions of agglutinated foraminifera (Fig. 6), supports the notion of enhanced brine formation during stronger polynyal activity. The transition to more intense polynyal activity \sim 0.5 ka BP, coupled to higher sea-ice variability thereafter, also aligns with observations from western Svalbard, where spring sea-ice concentration has steadily declined over the past 400 years (Cabedo-Sanz and Belt 2016) and heat transport into the Arctic via the West Spitsbergen Current has increased (D'Andrea et al. 2012; Spielhagen et al. 2011).

IMPLICATIONS AND CONCLUSIONS

The Arctic Ocean halocline is maintained by the contribution of cold and brine-enriched deep waters (Aagaard et al. 1981; Cavalieri and Martin 1994), which are formed as a consequence of high sea-ice production in coastal polynyas over the continental shelves (Fig. 7) (Tamura and Ohshima 2011). Tamura and Ohshima (2011) showed that the current polar amplification of global warming will lead to negative trends in sea-ice production in most of the Arctic polynyas and with future projections of a summer ice-free Arctic Ocean (IPCC 2013), sea-ice factories in Arctic coastal polynyas may lose their significance entirely (Fig. 7). A likely cause for this trend could be delayed sea-ice freezing and increased Arctic air temperatures (Tamura and Ohshima, 2011). The last time a similar scenario occurred was during the Holocene Thermal Maximum when Arctic Ocean sea-ice cover was believed to be less than half of the minimum summer extent in 2007 (Funder et al. 2011). Indeed, Arthun et al. (2011) showed that during periods of maximum warming in the central Barents Sea, formation of brineenriched shelf waters and thus, export of deep waters to the Nordic Seas and Arctic Ocean, was strongly reduced. Whether this reduced export contributed to the slowdown in AMOC in the twentieth-century (Rahmstorf et al., 2015) remains speculative. However, from the present study we conclude that sea-ice production in Arctic coastal polynyas are highly sensitive to variable, externally forced climate or ocean feedback mechanisms. The correspondence between high (low) polynyal activity and variable sea-ice conditions in one important Arctic sea-ice factory and observations of stronger (weaker) deep water renewal processes in the Nordic Seas during the present interglacial highlights the potential consequences for ocean ventilation during states with permanent sea-ice cover or future Arctic ice-free conditions.

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FIGURE CAPTIONS

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Fig. 1 Study area and investigated marine sediment surface, floodplain and core samples.

Major oceanographic features and the maximum sea ice extent are indicated. Inset shows

outline of Storfjorden Polynya (grey shaded) superimposed on the geology of Svalbard.

Arsenic concentrations (ppm) in both floodplain sediments onshore and marine surface

sediments are shown.

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Fig. 2 Proxy data for modern sea-ice variability in the Barents Sea. Left: Arsenic

concentration (As in ppm) in Barents Sea surface samples. Right: Sea-ice biomarker IP25

concentration in Barents Sea surface samples (Navarro-Rodriguez et al. 2013). Storfjorden

Polynya (stippled polygon), studied core position JM10 (green square), maximum of marginal

ice zone (MIZ) (black line) and the Barents Sea polar front (stippled line) are indicated.

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Fig. 3 Notched box-whisker plots for all downcore measurements of JM10-10GC for the

parameters As/Al, IP₂₅, and brassicasterol in the time intervals 0-500 a BP, 500-2800 a BP,

and 2800-6500 a BP. White lines mark the estimated positions, and notched intervals the

95%-confidence limits for the medians of the distributions.

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Fig. 4 Proxy data for sea-ice variability in Storfjorden polynya (core JM10-10GC) over the past

6500 years BP. Bottom to top: IP₂₅ concentration (µg/gSed and µg/gTOC), As/Al ratio (*1000),

agglutinated foraminifera (% of total benthic assemblages), and brassicasterol concentration

(µg/gSed). Stippled lines indicate the mean values for each proxy in the three intervals

discussed in the main text. Note that IP₂₅ concentrations normalized to $\mu g/g$ Sediment and $\mu g/g$

TOC indicate no dilution effect on biomarker records due to variable sedimentation rates.

Fig. 5 Sea-ice reconstruction, brine formation and deep water penetration to the Arctic over the past 6500 years. Bottom to top, IP₂₅ concentration (µg/gSed) in Storfjorden, seawaterderived Nd isotope variations expressed as \(\epsilon \)Nd in the eastern Fram Strait (Werner et al., 2014), planktic foraminifera δ^{13} C from western Svalbard/Barents Sea (Sarnthein et al., 2003; Werner et al., 2013), benthic foraminiferal δ^{13} C from Greenland Sea (Telesiński et al., 2014), June/July/August (JJA) air temperatures over Svalbard (D'Andrea et al., 2012), sea surface temperatures (SST) off western Syalbard (Spielhagen et al. 2011, Werner et al. 2013, Werner et al. 2011), Greenland ice core data from DYE-3, GRIP and NGRIP on the GICC05 timescale (Vinther et al. 2006) and solar irradiance (Laskar et al. 2004). Fig. 6 Downcore variability of sea ice (IP₂₅), phytoplankton (brassicasterol), and agglutinated foraminifera indicators in Storfjorden polynya over the past ca. 500 years. Orange bars indicate correspondence of high sea ice variability, phytoplankton production and strong polynyal activity as inferred from higher proportions of agglutinated foraminifera (Rasmussen and Thomsen 2014). Blue bar shows no response. Fig. 7 Location of coastal polynyas in the Arctic with variable sea-ice dynamics. (A) Modern sea-ice distribution and strong deep convections (crosses) with vigorous sea-ice factories (orange polygons) for brine-enriched shelf water formation. Blue arrows: cold, ice-covered surface currents. Red arrows: warm, saline Atlantic-derived water masses. (B) Less sea-ice in the Arctic with shut down of sea-ice factories (open polygons) and slow-down of deep convection (minus). NWP: North Water polynya, NEWP: Northeast Water polynya, NB: Nordbukta, WBP: Whaler's Bay polynya, SP: Storfjorden Polynya, KSP: Kara Sea polynya, LSP: Laptev Sea polynya.

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Table 1: Inorganic geochemical data from Barents Sea surface sediments

Station	Latitude	Longitude	Water Depth	As (pp	om)	Al (p	pm)	Mn (ppm)	Fe (p	pm)
ID			(meter)	ICP-AES		ICP-AES	XRF	ICP-AES	XRF	ICP-AES	XRF
623	71.05	21.65	166	13	6	20200	62768	933	929	31300	33573
627	72.32	24.06	264	4	6	13100	54353	435	620	17500	24690
629	73.01	24.25	404	5 9	14 10	19700 22400	66949 68907	704 483	1084 620	25600 32300	38050 39449
631	73.67 74.34	24.47 24.69	451 373	22	26	13700	57264	471	465	29000	36931
635	75.00	24.94	182	12	16	17200	60016	204	232	25100	29866
639	75.57	27.90	263	19	23	22700	71236	277	310	33800	40148
643	76.49	29.91	291	7	9	22600	72294	190	232	29400	36581
645	75.86	29.46	296	25	23	18200	67372	163	232	33900	39309
647	75.20	29.01	343	14	28	19600	72241	277	465	29000	44345
649	74.54	28.58	394	22	27	20700	64885	543	697	32700	39239
651	74.64	26.08	317	18	29	20100	64461	745	929	32500	38959
653	73.97	25.81	441	9	11 15	22700 19800	73512 67319	251 539	310 697	32000 30300	40008 38190
655 657	73.31 72.64	25.54 25.27	412 268	6	10	11000	54035	261	387	16500	24341
659	71.98	25.06	256	2	5	9730	49325	245	387	13400	20144
661	71.37	22.76	408	3	5	19400	64779	421	620	25300	35322
663	71.61	25.99	291	1	3	6560	42392	187	387	9480	15808
665	72.17	28.41	289	3	6	12600	56629	193	310	16800	24830
667	72.84	28.76	305	11	13	13300	54618	336	387	23100	27278
669	73.50	29.15	414	8	11	19600	67478	375	465	28700	36022
671	74.15	29.55	366	13	21	22600	73194	330	387	35100	43925
673	74.67	32.49	165	20	19	11300 15700	50595	198	232	23900 31700	27488
675	75.33	33.07	209	18 14	58 21	22900	52395 78433	199 193	232 232	31700	28118
677 679	75.97 76.62	33.73 34.45	276 193	105	157	20100	65097	523	620	58400	38959 75261
681	76.43	37.17	249	13	15	11000	59645	102	155	18100	22802
690	71.02	30.96	283	8	8	23200	64938	354	465	27200	35462
692	70.62	31.72	252	2	5	7280	45885	190	232	10900	17556
St.1.	72.00	22.00	367	6	7	19300	61762	604	852	24700	32454
St.2.	72.02	20.92	371	7	8	21900	63932	1040	1317	28300	35532
St.3.	72.03	19.85	324	9	11	18800	50966	725	929	23000	29377
St.4.	72.02	18.77	315	9	12	17200	47208	770	1084	20700	27348
St.5.	72.03	17.70	296	8	7	13200 9300	45250 41757	668 366	929 542	17500 14500	23991 20844
St.6. St.7.	72.02 72.02	16.62 15.52	362 767	10	12	8390	43345	369	620	15900	24551
St.8.	72.02	14.73	1260	7	6	18000	65573	336	465	25700	33084
St.9.	72.01	14.62	1317	7	6	13500	51707	873	1239	17000	25740
St.11.	73.17	12.94	1499	8	6	14600	46467	854	1162	18300	24760
St.12.	73.17	14.09	1030	5	3	10300	44668	555	852	13600	22312
St.13.	73.17	15.23	485	8	11	8800	48426	386	542	12900	18815
St.14.	73.17	16.38	475	8	11	12500	54088	434	542	18000	24411
St.15.	73.17	17.54	460	9	12 18	14300 12700	54935 52977	459 421	542 542	21300 21900	26229 26929
St.16. St.17.	73.17 73.17	18.82 19.86	423 441	7	8	14400	55941	350	465	20500	25949
St.17.	73.17	20.95	463	12	15	17900	62768	642	774	27000	33713
St.19.	73.17	22.01	444	15	21	18300	62662	938	1084	27900	34832
St.20.	74.82	18.02	296	18	25	11100	41969	326	387	20900	25810
St.21.	74.82	17.00	280	7	7	8120	35353	212	310	14700	18465
St.22.	74.82	16.03	356	8	7	10100	42763	275	387	16900	20354
St.23.	74.82	14.79	1507	9	10	19200	52448	1120	1471	23600	30636
St.24.	75.64	12.92	1500	10 18	14 20	18900 13100	51971 54512	1260 518	1626 697	24000 24300	31055 30426
St.25. St.26.	75.75 75.83	13.84 14.77	807 370	18	25	19200	66949	800	929	31600	36511
St.27.	75.95	15.72	369	45	59	21500	71236	3650	4337	42300	49031
St.28.	76.05	16.67	328	21	25	17200	63509	942	1084	31300	36441
St.29.	76.16	17.62	309	45	57	20500	67108	1340	1549	40200	44625
St.30.	76.22	18.58	257	31	33	20200	67849	577	697	36800	41058
St.31.	76.31	19.57	258	53	66	19400	70019	2160	2556	42100	49801
St.32.	76.38	20.58	228	54	68	19600	69278	787	929	44000	48891
St.33.	76.47	21.60	262	91	117	20200 18800	71447 61127	1050 504	1239 697	46900 24200	53927 32804
St.34. St.35.	71.75 71.62	22.00 21.07	356 319	5	6	13900	51813	490	697	17800	24970
St.36.	71.60	20.86	320	7	7	14400	48902	646	852	18800	25390
St.37.	71.60	21.19	335	5	6	14300	53347	464	620	18600	25460
St.38.	71.49	20.82	310	6	3	15800	46520	651	929	19700	25880
St.39.	71.34	20.19	234	5	5	13800	39958	695	1007	16900	23431
St.40.	71.18	19.56	225	6	3	12700	39111	718	1007	15700	21963
St.41.	71.03	18.95	199	1	3	6390	35830	266	465	9060	15598
St.42.	70.87	18.34	173	3	3	5860	23657	219	387	8090	13499
St.43.	70.72	17.75	273	7	5 7	11300 7290	41651 39746	606 274	929 620	14700 9690	22382 19794
St.44. St.45.	70.55 70.44	17.14 16.75	706 1500	6	7	9350	53136	517	929	14000	26859
JL.45.	70.44	10./5	1300	U U	<u>'</u>	5550	00100	317	323	14000	20000

Table 2. Methods accuracy for measuring As by XRF.

Parameter	UB-N	MESS-1	JLK-1
Sample type	Serpentine	Marine sediment	Lake sediment
Average, 3 replicates, mg/kg	10.94	7.9	26.91
Standard deviation, 3 replicates,	2.5	0.77	3.43
mg/kg	2.3	0.77	3.43
%RSD	23.20 %	9.80 %	12.80 %
Certified value, mg/kg	10*	10.6	27.7
Uncertainty, mg/kg	4.22	1.2	
References GeoReM	GeoReM 149	GeoReM 5021	
Bias, mg/kg	0.94	-2.7	-0.79
Relativ bias	9.40 %	-25.50 %	-2.90 %

* Compiled value.

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457 Table 3. Methods accuracy for measuring Al by XRF.

Parameter	PACS-1	MESS-1	JLK-1
Sample type	Marine sediment	Marine sediment	Lake sediment
Measured, %	12.06	12.05	16.77
Certified value, %	12.23	11.03*	16.73
Uncertainty, %	0.22	0.38	0.184
References GeoReM(Jochum et al., 2005)	GeoReM 5021	GeoReM 5021	GeoReM 659
Bias, mg/kg	- 0.17	1.03	- 0.04
Relativ bias	-1.37%	9.26%	-0.23%

458 * Compiled value.

460 **Table 4.** AMS¹⁴C dates and calibrated dates for core JM10-10GC as published by Rasmussen

461 and Thomsen (2014, 2015).

Core ID	Depth (cm)	14C Age	Calendar Age	Lab. Code	Species
JM10-10GC	44.5	832±21	473±20	UB-17204	Nucula sp.
JM10-10GC	102.5	1491±22	1029±43	UB-17205	Nuculana sp.
JM10-10GC	136.5	3008±27	2770±32	UB-17206	Astarte sp.
JM10-10GC	210.5	4182±41	4278±73	UB-18845	N. labradorica
JM10-10GC	250.5	4573±28	4805±34	UB-18946	N. labradorica
JM10-10GC	324-326	6065±31	6482±49	UB-17207	Bivalve
JM10-10GC	325	5990±43	6398±57	UB-21198	N. labradorica

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Table 5. Downcore variability of IP₂₅ concentrations in JM10-10GC.

Depth_cm	Age_a_BP1950	IP25com μg/gSed	IP25com μg/gTOC
2.5	26.57	0.00799	0.467
4.5	47.83	0.00759	0.440
6.5	69.09	0.00862	0.499
8.5	90.35	0.00826	0.492
10.5	111.61	0.00804	0.496
12.5	132.87	0.00789	0.507
14.5	154.12	0.00777	0.509
16.5	175.38	0.00527	0.362
18.5	196.64	0.00706	0.409
20.5	217.90	0.00612	0.375
24.5	260.42	0.00781	0.460
26.5	281.67	0.00961	0.553
28.5	302.93	0.00782	0.442
30.5	324.19	0.00794	0.454
32.5	345.45	0.00518	0.298
34.5	366.71	0.00822	0.473
36.5	387.97	0.00660	0.377
38.5	409.22	0.00655	0.389
40.5	430.48	0.00617	0.349
42.5	451.74	0.00589	0.332
44.5	473.00	0.00709	0.402
46.5	492.17	0.00782	0.449
48.5	511.34	0.00585	0.337
68.5	703.07	0.00558	0.371
80.5	818.10	0.00560	0.367
104.5	1131.41	0.00553	0.379
110.5	1438.65	0.00566	0.329
118.5	1848.29	0.00576	0.353
126.5	2257.94	0.00642	0.370
130.5	2462.76	0.00636	0.359
132.5	2565.18	0.00782	0.446
142.5	2892.27	0.00747	0.429
152.5	3096.05	0.01052	0.605
156.5	3177.57	0.00801	0.457
170.5	3462.86	0.00719	0.427
176.5	3585.14	0.01502	0.849
178.5	3625.89	0.00837	0.472
190.5	3870.43	0.00867	0.492
192.5	3911.19	0.00690	0.396
208.5	4237.24	0.00683	0.394
210.5	4278.00	0.00686	0.401
224.5	4462.45	0.00890	0.487
246.5	4752.30	0.00874	0.466
250.5	4805.00	0.00790	0.424
266.5	5165.16	0.00777	0.414
280.5	5480.30	0.00932	0.502
284.5	5570.34	0.00811	0.436
298.5	5885.48	0.00760	0.421
324.5	6470.74	0.00622	0.383

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