Supplementary Material

Ice sheet melt drove methane emissions in the Arctic during the last two interglacials

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Supplementary methods

1. Micropaleontology.

The micropaleontological work has been done on the basis of the taxonomy, ecology and geochemistry previously studied on living/modern benthic foraminifera in the study area (Dessandier et al., 2019 and 2020). We have briefly summarized the applied methods below:

a. Foraminiferal taxonomy

Taxonomy of all foraminiferal species of this study was based on Ellis and Messina (1940–1978) and Loeblich and Tappan (1987) taxonomic references. The benthic foraminiferal species *Cassidulina neoteretis* has been morphologically defined as the modern (Pleistocene) bio-indicator that differs from *Cassidulina teretis* (Tappan, 1951) by a change in its aperture corresponding to an adaptation of the tooth for modern environmental conditions (Seidenkrantz et al., 1995). Lazar et al. (2016) demonstrated that the limit between the two species is probably more progressive and depends on the geographic location. According to Cronin et al. (2019), both *C. neoteretis* and *C. teretis* have been used in paleoceanographic investigations in the Arctic, however, not always were the different wall structure or apertural morphology considered. In the present study, we did not make this difference between the two species and consider potential specimens of both *C. neoteretis* and *C. teretis* under the name *C. neoteretis*, to be consistent with the local literature (Schneider et al., 2017; 2018).

b. Micropaleontological analyses

Fifty-three samples of benthic foraminifera were handpicked from the core 126 and 50 samples from cores 125 and GC3, all from the >125 µm size fraction. A minimum of 300 specimens per samples has been considered. Only major species (>5%) are discussed in this study. Faunal number and specific richness correspond to the number of individuals and of different species found in each sample. Shannon index has been calculated following the equation 1.:

$$S = -\sum_{i=1}^N p_i \log_2 p_i \quad 1.$$

Where N is the specific richness, the number of species in samples, p_i indicates the relative abundance of a species that means the number of individuals of one species, relative to the total number of all the individuals. Tables of counting are provided in the spreadsheet foraminiferal counting.

c. Stable isotopes

$\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ isotopic ratios were measured on the planktonic species *Neogloboquadrina pachyderma* (Ehrenberg, 1861) and on the benthic species *Cassidulina neoteretis* (Lazar et al., 2016). Ten to twelve specimens were hand-picked from each sample, and the isotopic analyses were conducted at the stable isotope laboratory SIL at CAGE - UiT The Arctic University of Norway, using Gasbench II and MAT253 Isotope Ratio Mass Spectrometers. Foraminiferal tests were placed in 4.5 mL vials then flushed with Helium. 5 drops of water-free H_3PO_4 were added manually with a syringe. After equilibration (>3 hours at 50°C), the samples were analyzed. Normalization to Vienna Pee Dee Belemnite (VPDB) is done with 2-3 in-house standards (1.96 ‰, -10.21 ‰ and -48.95 ‰ for $\delta^{13}\text{C}$ and -2.15 ‰ and -18.59 ‰ for $\delta^{18}\text{O}$), which had been normalized by several international standards as well.

2. Dating

The chronology of gravity cores GC3 and GC3 is based on the correlation of $\delta^{18}\text{O}$ records with nearby sediment core HH-13-212 (Fig. 1). Schneider et al. (2018) established the age model for HH-13-212 through correlation of several AMS ^{14}C dating points and tie points from previously dated sediment cores in the study area. All AMS ^{14}C datings from these cores are summarized in Table 1. We used ages obtained by Ambrose et al. (2015) and Szybor and Rasmussen (2017) in cores JM10-333GC and JM10-335GC where the $\delta^{13}\text{C}$ signature is characteristic of normal marine conditions. The ages were converted into calendar years using the calibration program Calib 7.1 (Stuiver et al., 2014) with a marine reservoir age of 400 years that was incorporated within the Marine13 calibration curve (Reimer et al., 2013). In addition, a regional reservoir age correction DR of 7 ± 11 was applied (Mangerud et al., 2006). The age model is based on the calibrated ages obtained from the peaks of the probability curves within the 2s range.

Supplementary Table 1. AMS ^{14}C data.

Core	Depth (cm)	Dated material	Laboratory code	Age ($^{14}\text{C} \pm 1\sigma$)	Calibrated age $\pm 1\sigma$
<i>Jessen et al. 2010-TP4</i>	18	<i>N. pachyderma</i> sin	–	12,250 $\pm$ 150	14,070 $\pm$ 210
JM10-333GC	20	Mixed benthic foraminifera	Beta-4133010	12,920 $\pm$ 40	14,442 $\pm$ 119 _a
<i>Jessen et al. 2010-TP5</i>	50	Bivalve	–	12,840 $\pm$ 150	14,300 $\pm$ 260
<i>Jessen et al. 2010-TP6</i>	110	Bivalve	–	13,140 $\pm$ 150	14,780 $\pm$ 220
JM10-333GC	137	<i>N. pachyderma</i> sin	UBA-23419	14,475 $\pm$ 61	16,385 $\pm$ 142
JM10-333GC	147	<i>N. pachyderma</i> sin	UBA-23420	15,228 $\pm$ 61	17,677 $\pm$ 190
<i>Jessen et al. 2010-TP7</i>	258	<i>N. pachyderma</i> sin	–	20,150 $\pm$ 130	23,550 $\pm$ 185
<i>Jessen et al. 2010-TP8</i>	281	<i>N. pachyderma</i> sin	–	20,580 $\pm$ 130	24,080 $\pm$ 150
JM10-333GC	359	<i>N. pachyderma</i> sin	Beta-412,869	37,420 $\pm$ 340	42,118 $\pm$ 339
<i>Jessen et al. 2010-TP2</i>	10	Planktonic foraminifera	\	9240 $\pm$ 100	9840 $\pm$ 200
JM10-335GC	20	<i>N. pachyderma</i> sin	UBA-18137	9302 $\pm$ 38	9981 $\pm$ 111
<i>Jessen et al. 2010-TP3</i>	35	Planktonic foraminifera	\	9390 $\pm$ 100	10,100 $\pm$ 150
JM10-335GC	56	<i>N. pachyderma</i> sin	UBA-23417	10,829 $\pm$ 43	12,245 $\pm$ 107
JM10-335GC	57	Mixed benthic foraminifera	UBA-23418	10,820 $\pm$ 43	12,225 $\pm$ 106
JM10-335GC	75	<i>N. pachyderma</i> sin	UBA-18138	11,830 $\pm$ 44	13,236 $\pm$ 69
<i>Jessen et al. 2010-TP5</i>	198	Bivalve	–	12,840 $\pm$ 150	14,300 $\pm$ 260
<i>Jessen et al. 2010-TP6</i>	240	Bivalve	–	13,140 $\pm$ 150	14,780 $\pm$ 220
JM10-335GC	278	<i>N. pachyderma</i> sin	UBA-23197	15,486 $\pm$ 73	18,197 $\pm$ 189
JM10-335GC	278	Bivalve	UBA-18142	15,283 $\pm$ 56	17,789 $\pm$ 182
JM10-335GC	290	<i>N. pachyderma</i> sin	UBA-18139	16,830 $\pm$ 71	19,505 $\pm$ 90
JM10-335GC	369	<i>N. pachyderma</i> sin	UBA-18140	19,327 $\pm$ 77	22,462 $\pm$ 95
<i>Jessen et al. 2010-TP7</i>	380	<i>N. pachyderma</i> sin	–	20,150 $\pm$ 130	23,550 $\pm$ 185
<i>Jessen et al. 2010-TP8</i>	405	<i>N. pachyderma</i> sin	–	20,580 $\pm$ 130	24,080 $\pm$ 150
JM10-335GC	460	<i>N. pachyderma</i> sin	UBA-18141	27,132 $\pm$ 150	31,977 $\pm$ 221

3. Drilling and Coring

The MeBo drilling system is a mobile drilling rig (Freudenthal and Wefer, 2013) remotely operated from RV “Maria M. Merian” during expedition 57 (Bohrmann et al., 2016, cruise report). MeBo 126 recovered a total cored length of 2465 cm from a total drilled length of 6250 cm. MeBo 125 recovered a total cored length of 905 cm from a total drilled length of 2280 cm. Gravity cores showed continuous sediment recovery of 765 and 584 cm for GC2 and GC3, respectively. All these data are available in the cruise report (Bohrmann et al. 2016), which is available here (: <http://nbn-resolving.de/urn:nbn:de:gbv:46-00105895-15.>)

Supplementary notes

a. Chronology

The age model of reference core MeBo 126 is based on intervals where stable oxygen and carbon isotopes of planktonic foraminiferal species provided identifiable glacial and interglacial cycles and MIS boundaries (sup. Table 2). The MIS 3/2 and 6/5 boundaries in MeBo 125 were identified by correlating its isotopic record with high-resolution 3D seismic data between MeBo 126 and 125 (Fig. 3, main text).

The chronology of methane emission over the last deglaciation differs from what has been recently suggested in Dyonisius et al., (2020). The record of these authors starts from 18 ka and they particularly pay attention to the Oldest-Dryas-Bølling transition and Younger-Dryas Preboreal periods, respectively at ca 14.5 and 11.5 ka while the main negative excursion of $\delta^{13}\text{C}$ in the core GC2 is recorded at 19.5 ka. This may question the small contribution of methane emissions from hydrates in the global budget for the last deglaciation and even more for the Eemian interglacial for which no other record of seepage exist. Differences of timing for methane emissions between our record in the Arctic and those of Dyonisius et al. (2020) from the Antarctica may also be explained by different geological processes. Ice Sheet instabilities most likely triggered gas hydrate dissociation in Arctic marine sediments. This methane leakage was potentially episodic considering the impact of faults re-activation with a time lag of hundreds of years and its consequence in atmospheric methane budget is probably significantly delayed as well.

Supplementary Table 2. Marine Isotope Stage (MIS) boundaries and ages according to Lisiecki and Raymo (2005).

MIS Boundary	LR04 Age (ka)
1/2	14
2/3	29
3/4	57
4/5	71
5.1	82
5.2	87
5.3	96
5.4	109
5.5	123
5/6	130

6/7	191
7/8	243
8/9	300
9/10	337
10/11	374
11/12	424

Supplementary fig. 1. Chronological correlation for the Eemian period between the MeBo cores. Orange and yellow bars, corresponding to MIS boundaries 3/2 and 6/5, respectively, are based on seismic correlation. A) full isotopic profiles from the pockmark MeBo 125, B) zoom on MIS 5e (i.e the Eemian) from MeBo 125, C) zoom on MIS 5e (i.e. the Eemian) from MeBo 126 and D) full isotopic profiles out from the reference, non-seepage site MeBo 126.

b. Foraminiferal preservation and distribution

We analyzed benthic foraminiferal assemblages to support the chronology, perform the stable isotope measurement and evaluate the impact of methane release on the fauna. The latter is reached through the assessment of variable preservational states and precipitation of methane-derived authigenic carbonate as secondary overgrowth in and around foraminiferal shells. We support evidence for secondary overgrowth by using SEM images (sup. Fig. 2). SEM results reveal pristine tests with dominantly calcite (Ca) in specimens of diagenetically unaltered intervals during MIS 2 (i.e. 375 cm) in MeBo

125 (Fig. 2A, B, F). In contrast, secondary calcification on tests of *C. neoteretis* and *N. pachyderma* are recorded during intervals characterized by depleted $\delta^{13}\text{C}$ values (last deglaciation, Eemian/penultimate deglaciation) (Fig. 2D, G, I). The latter is supported by EDS maps highlighting the abundance of Mg on specimens affected by the precipitation of methane-derived authigenic carbonate (Fig. 2E). The foraminifera in the reference core also revealed a general better preservation during glacial cycles that matches with high values of XRF-Ca/Ti ratio (sup. Fig. 3). Benthic foraminiferal distribution supported our interpretations for methane impact, especially regarding the distributions of *Cassidulina neoteretis* and *Melonis barleeanus*. The different phases of seepage (sup. Fig. 4), are characterized by a decrease of the *C. neoteretis* and an increase of *M. barleeanus* over the main excursion of $\delta^{13}\text{C}$ during the MIS 5e (Eemian) (sup. Fig. 5).

Supplementary Fig. 2: Backscatter-scanning electron microscopy (SEM) images of selected foraminiferal tests shells. A) *Cassidulina neoteretis* from MeBo 125 (375 cm depth), B) Energy-dispersive X-ray spectroscopy (EDS) map of the specimen (Ca 100% in green, Mg 100% in red) A) and C) zoom of the aperture of A). D) *Cassidulina neoteretis* from MeBo 125 (last deglaciation, 250 cm), E) Energy-dispersive X-ray spectroscopy (EDS) map of the specimen D). F) *Neogloboquadrina pachyderma* from MeBo 125 (375 cm). G) *Cassidulina neoteretis* from MeBo 125 (Eemian/penultimate deglaciation, 1600 cm), H) zoom of the aperture of G). I) *Neogloboquadrina pachyderma* from MeBo 125 (Eemian/penultimate deglaciation, 1600 cm). The precipitation of

methane-derived authigenic carbonate is visible as secondary overgrowth around the tests of D), G) and I), corresponding to the two periods of deglaciation during which methane emission events have been interpreted in our record.

Supplementary fig. 3. Relative contributions of the major (>5%) benthic foraminiferal species, stable isotopes and Ca/Ti-XRF ratios of the reference MeBo 126.

Supplementary fig. 4. Relative contributions of the major (>5%) benthic foraminiferal species in the GC2 (light grey) and in the MeBo 125 (dark grey) cores.

Supplementary fig. 5. Relative contributions of the major (>5%) benthic foraminiferal species from the section 7 of the MeBo 125, corresponding to a shell bed similar to the one observed by Ambrose et al. (2015) over the last deglaciation.

Supplementary references

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