



Tromsø, 18th of March, 2015

The Editorial Office,
Climate of the Past

Dear Editor,

We would like to submit the 2nd version of the revised manuscript of the paper cp-2014-127, entitled “Carbon isotope ($\delta^{13}\text{C}$) excursions suggest times of major methane release during the last 14 ka in Fram Strait, the deep-water gateway to the Arctic”, authored by C. Consolaro, T. L. Rasmussen, G. Panieri, J. Mienert, S. Bünz, and K. Sztybor.

We thank again the two referees G. Dickens and L. J. de Nooijer for their final remarks and especially G. Dickens for his comments and suggestions. We have addressed all the final comments and followed his advices. A point-by-point response is included below (reviewers' comments in **black bold**, our answers in **blue**) and a marked-up version of the manuscript showing all the changes is included.

Yours sincerely,

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Reply referee G. Dickens

General Comment: I would remove the Sun and Turchyn (2014) reference from text and caption, as the authors do not consider methane and AOM. There are, however, much better papers on the topic of methane derived authigenic carbonate (e.g., Greinert et al., 2001), including some that even suggest that it represents a significant sink of CaCO₃ (e.g., Snyder et al., DSR, 2007).

The Sun and Turchyn (2014) reference has been removed from the text and caption and substituted with the references suggested.

-- Page 2 --

Line 19: This is awkward. Probably should be “emissions from the seafloor through time, especially in relation to past climate change and with a focus on periods of climate warming.”

The phrase has been modified as suggested.

Line 21: Change to “... along continental margins within a gas ...”

Changed as suggested.

Line 22: This is not entirely true and should be rewritten. In my mind, a BSR only occurs when pore space with free gas is in contact with pore space with gas hydrate; certainly, there are locations with gas hydrate and no BSR. (see Dickens, Org. Geochem., 2001 as to discussion of the GHSZ and its relation to gas hydrate distribution).

The phrase has been rewritten following the suggestions.

Lines 23-27: This text and referencing is somewhat misleading. To my knowledge, the community is unclear on how gas passes through a REGIONAL GHSZ (for example, see discussion in Milkov et al., EPSL, 2004).

This phrase has been removed.

-- Page 3 --

Lines 1-3: This is awkward as written, because Equation 2 does not consume methane; that is, the second equation comes after the next sentence. In any case, neither reference is very good, as these authors did not originally describe the reactions.

The Equation 2 has been moved as suggested and the references have been removed from next to the Equations and relocated in the sentences.

Line 5: Avoid Latin abbreviations in the middle of sentences, because it is sloppy writing. Such abbreviations should only come in parentheses.

The Latin abbreviation e.g. has been removed and “like” has been used instead.

Line 6: Whiticar 1999 is not a good reference here, as the paper is not really about authigenic carbonate. Greinert et al., in Natural Gas Hydrates, 2001 is a much better reference for this statement. There are numerous other papers, too.

Whiticar 1999 has been removed and Kulm and Suess, 1990; Greinert et al., 2001; Snyder et al., 2007 have been used instead.

Line 7: Should be plural (“tests”).

Ok, it has been corrected.

Line 9: Should be “the pore space of surrounding sediments”.

Corrected as suggested.

Line 11: Remove second “the”.

Second “the” has been removed.

Line 12: Borowski et al. (1996) is not a great reference here, as while a good paper on the topic, does not pertain to seeps and show this effect nicely. Other papers, such as Paull et al., GML, 2005; Castellin et al., Chem Geol., 2006, etc. are much better.

References have been changed as suggested.

-- Page 4 --

Line 27: “Induced” is the wrong word choice here. Rewrite the sentence.

The sentence has been rewritten.

-- Page 5 --

Line 26: What reference core? This should be shown on Figure 1 and stated.

The location of the reference core for western Svalbard slope is now shown in Fig. 1 and stated in the text.

-- Page 8 --

Line 20: Presumably should be “no negative oxygen isotope excursions”
Corrected as suggested.

-- Page 9 --

Line 31: I think the original argument for this concept was in Dickens, G3, 2001.
This reference has been added to the text.

-- Page 10 --

Line 1: Here and elsewhere, I would avoid adding the MDAC acronym, as this is not common usage.

The abbreviation has been deleted here and elsewhere.

Line 23: Change “are” to “is” or somehow pluralize silica.

Changed to “is”.

Line 26: The authors might want to modify the statement, because AOM does not increase pH per se.

This phrase has been deleted.

Line 28: Avoid starting sentences with abbreviations (e.g., Mg).

The start of the sentence has been changed.

-- Page 11 --

Lines 22-25: There are multiple papers that now clearly show this effect, including across seeps. In other words, it is beyond “according to Borowski” - the ideas he presented are really obvious.

The reference to Borowski has been deleted in this sentence.

-- Page 12 --

Lines 9-12: Authigenic carbonate precipitation at or near the seafloor happens at many seep locations, not just Hydrate Ridge.

The sentence has been modified accordingly.

Line 19: Avoid MDAC and should not be plural given the verb.

The sentence has been modified accordingly.

-- Page 13/14 --

Lines 23 (page 13) – 4 (Page 14): I do not follow the first paragraph. First, I think it needs to state “... registered also in benthic foraminiferal records at sites in the region away from seep sites.” It then needs rewording at the end so as to emphasize that this is consistent with the idea that CH₄ is rapidly oxidized in the water column such that any addition of CH₄ derived carbon is diluted across a broader region. The problem here is that this now conflicts with the next paragraph. Bottom line, this part needs some rewriting to clarify.

The whole paragraph has been rearranged.

1 **Carbon isotope ($\delta^{13}\text{C}$) excursions suggest times of major
2 methane release during the last 14 ka in Fram Strait, the
3 deep-water gateway to the Arctic**

4

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13

14 **Abstract**

15 We present results from a sediment core collected from a pockmark field on the Vestnesa
16 Ridge (~80°N) in the eastern Fram Strait. This is the only deep-water gateway to the Arctic,
17 and one of the northernmost marine gas hydrate provinces in the world. Eight ^{14}C AMS dates
18 reveal a detailed chronology for the last 14 ka BP. The $\delta^{13}\text{C}$ record measured on the benthonic
19 foraminiferal species *Cassidulina neoteretis* shows two distinct intervals with negative values
20 termed carbon isotope excursion (CIE I and CIE II, respectively). The values were as low as -
21 4.37‰ in CIE I correlating with the Bølling-Allerød interstadials and as low as -3.41‰ in CIE
22 II correlating with the early Holocene. In the Bølling-Allerød interstadials, the planktonic
23 foraminifera also show negative values, probably indicating secondary methane-derived
24 authigenic precipitation affecting the foraminiferal shells. After a cleaning procedure designed
25 to remove authigenic carbonate coatings on benthonic foraminiferal tests from this event, the
26 ^{13}C values are still negative (as low as -2.75‰). The CIE I and CIE II occurred during periods
27 of ocean warming, sea level rise and increased concentrations of methane (CH_4) in the
28 atmosphere. CIEs with similar timing have been reported from other areas in the North
29 Atlantic suggesting a regional event. The trigger mechanisms for such regional events remain
30 to be determined. We speculate that sea-level rise and seabed loading due to high sediment
31 supply in combination with increased seismic activity as a result of rapid deglaciation may

1 have triggered the escape of significant amounts of methane to the seafloor and the water
2 column above.

3

4 1 Introduction

5 Methane hydrate is an ice-like compound that exists in sediments at high pressures and low
6 temperatures with sufficient supply of water and gas (Sloan, 1998). Methane hydrate
7 provinces are widespread in the Arctic region, but their stability and longevity through time,
8 and the significance of their contribution to the global carbon budget are still poorly
9 understood (e.g., Biastoch et al., 2011). The Arctic region is highly sensitive to climate change
10 and the effects of on-going global warming are probably more extreme in the Arctic than
11 elsewhere (e.g., Screen and Simmonds, 2010; Spielhagen et al., 2011). Recent discoveries
12 suggest that the stability of gas hydrates in the Arctic Ocean in water depths up to about 400 m
13 is already affected by on-going ocean warming (e.g., Shakova et al., 2010; Ferré et al., 2012;
14 Berndt et al., 2014). Methane emissions offshore west Svalbard from pockmarks in water
15 depths greater than 800 m were recently recorded in the eastern part of the Vestnesa Ridge
16 (Figs. 1, 2), where several additional gas plumes have been detected in 2010 (Bünz et al.,
17 2012) and in 2012 (Smith et al., 2014), compared to the 2008 survey (Hustoft et al., 2009a),
18 possibly indicating an increase in methane release activity. It is critical, therefore, to
19 investigate the frequency of methane (CH_4) emissions from the seafloor through time,
20 especially in relation to past climate change and with a focus on periods of climate warming.

21 Gas hydrates occur in marine sediments along continental margins within a gas hydrate
22 stability zone (GHSZ). The base of the GHSZ can be marked in seismic data by a bottom-
23 simulating reflector (BSR) when sediments with free gas below and sediments with gas
24 hydrate above this boundary are in contact, generating a negative acoustic impedance contrast
25 (Shipley et al., 1979). Pockmarks are seafloor craters formed in soft, fine-grained sediments,
26 where localised seepage of gas and pore fluid occur (Judd and Hovland, 2007). The CH_4
27 originating from free gas below the BSR or released from gas hydrate dissociation may
28 dissolve in pore waters, remain trapped as gas, or rise toward the seafloor as bubbles. In the
29 near-seafloor sediments up to 90% of the methane can be consumed by anaerobic oxidation of
30 methane (AOM) according to the Equation (1): $\text{CH}_4 + \text{SO}_4^{2-} \rightarrow \text{HCO}_3^- + \text{HS} + \text{H}_2\text{O}$ by a
31 consortium of methanotrophic archaea and sulfate-reducing bacteria within the sulfate-methane
32 transition zone (SMTZ) (e.g. Barnes and Goldberg, 1976; Borowski et al., 1996; Boetius et al.,

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1 2000; Hinrichs and Boetius, 2002; Treude et al., 2003; Reeburgh, 2007). The production of
2 bicarbonate from AOM can induce the precipitation of calcium carbonate according to the
3 Equation (2): $\text{Ca}^{2+} + 2\text{HCO}_3^- \rightarrow \text{CaCO}_3 + \text{CO}_2 + \text{H}_2\text{O}$. These so-called methane-derived authigenic
4 carbonates can precipitate in different shapes like slabs, crusts, nodules, chimney, and pipes,
5 with typical negative $\delta^{13}\text{C}$ values (e.g. Kulm and Suess, 1990; Greinert et al., 2001; Snyder et
6 al., 2007). Benthonic foraminifera are often common in methane seep environments and it has
7 been demonstrated that their calcium carbonate tests can register the low $\delta^{13}\text{C}$ values of
8 ambient dissolved inorganic carbon (DIC) derived from the oxidation of methane in the pore
9 space of surrounding sediments (Wefer et al., 1994; Rathburn et al., 2003; Hill et al., 2004;
10 Martin et al., 2004; Panieri et al., 2009, 2012, 2014a).

11 At high flux seep sites the SMTZ is typically very shallow and methane may escape directly
12 into the water column (Paul et al., 2005; Castellini et al., 2006). Most of the methane emitted
13 at the seafloor is then consumed by methanotrophic aerobic microbes in the water column
14 (Niemann et al., 2006; Reeburgh, 2007). For this reason planktonic foraminifera do not
15 normally register the negative methane-derived $\delta^{13}\text{C}$ values in their tests. After deposition at
16 the seafloor, both planktonic and benthonic foraminifera can be affected by alteration due to
17 the precipitation of AOM-derived authigenic carbonates on their tests (Torres et al., 2003,
18 2010; Millo et al., 2005a; Panieri et al., 2009). In order to distinguish the isotopic records of
19 the secondary overgrowth from the primary tests, it is necessary to repeat the isotopic analyses
20 on specimens cleaned with a specific procedure designed to remove all authigenic carbonate
21 coatings (Pena et al., 2008; Panieri et al., 2012, 2014b).

22 Here we present a detailed data analysis from a sediment core that was taken in a pockmark
23 from the western part of the Vestnesa Ridge (Fig. 2). The core has been investigated for stable
24 isotopes ($\delta^{18}\text{O}$ and $\delta^{13}\text{C}$), together with the distribution of planktonic foraminifera and
25 sedimentological parameters, in order to reconstruct past changes in emission of methane in
26 the area. Our results of the ^{14}C AMS (accelerator mass spectrometry) dates suggest an
27 undisturbed sedimentary record for the last 14 ka BP. Negative carbon isotope excursions
28 (CIEs) during the Bølling-Allerød interstadials and during the early Holocene provide
29 important records of past methane release events. This study is part of an ongoing research
30 project at the Centre of Excellence for Arctic Gas Hydrate, Environment and Climate (CAGE)
31 at the Arctic University of Norway, where different sediment cores from the Vestnesa Ridge
32 pockmark field are being investigated in order to reconstruct past methane emissions from the
33 seafloor (Panieri et al., 2014b).

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1 **2 Study Area**

2 The Vestnesa Ridge is an elongated sediment drift at ~80°N at the northwestern Svalbard
3 margin in the eastern Fram Strait. The Fram Strait is dominated by two main surface currents:
4 the warm West Spitsbergen Current (WSC) and the cold East Greenland Current (EGC)
5 (Aagaard et al., 1987) (Fig. 1). The WSC is the northernmost branch of the North Atlantic
6 Current (NAC) and brings relatively warm, saline water along the western Svalbard margin
7 and through the Fram Strait into the Arctic Ocean. This current is the major source of heat and
8 salt to high northern latitudes and is very important for the generation of deep water in the
9 Nordic seas (Aagaard et al., 1985). The warm Atlantic water overlies the Greenland Sea
10 Intermediate Water, which is generated from convection in the Nordic Seas (Aagaard et al.,
11 1987). The EGC carries cold polar water and sea ice from the Arctic Ocean southwards along
12 the East Greenland margin through the Denmark Strait and into the North Atlantic Ocean (Fig.
13 1).

14 The Vestnesa Ridge is located on hot, thin and young (<20 Ma) oceanic crust (e.g. Hustoft et
15 al., 2009a) (Figs. 2a, b). It belongs to the eastern spreading segment of the Molloy Fracture
16 Zone that is connected to the northernmost extension of the Mid-Atlantic Ridge system: the
17 ultra-slow spreading Knipovich Ridge (Engen et al., 2008; Hustoft et al., 2009a) (Fig. 2a). The
18 sediment crest of the Vestnesa Ridge is pierced with pockmarks (Fig. 2b) (e.g. Vogt et al.,
19 1994; Hustoft et al., 2009a), and seismic data show a BSR 160–180 m beneath the seabed
20 indicating the presence of gas hydrate (Hustoft et al., 2009a; Petersen et al., 2010; Bünz et al.,
21 2012). Active gas venting has recently been observed in the eastern part of the ridge, where
22 thermogenic free gas migrates to the crest of the BSR anticline and further upward to the
23 Vestnesa Ridge pockmark field (Hustoft et al., 2009a; Bünz et al., 2012; Smith et al., 2014).
24 Seismic data beneath the pockmark field show vertical gas migration pathways (chimneys)
25 that form conduits allowing the gas to bypass the hydrate stability zone (HSZ) and escape
26 from the seafloor (Bünz et al., 2012). The pockmark fields in the deeper (1300 mwd), western
27 part of the ridge are most probably inactive because no acoustic gas flares have been observed
28 so far (Bünz et al., 2012).

29
30 **3 Material and methods**
31 Gravity core JM10-330GC (79.13°N, 5.6°E; 420 cm long) was collected from about 1300 m
32 water depth (mwd) in a pockmark located on the inactive western part of the Vestnesa Ridge

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1 (Figs. 1 and 2b). Before opening the core, magnetic susceptibility was measured with a
2 Bartington MS2 loop sensor (Fig. 3). Afterwards the core was split longitudinally, one half
3 was X-rayed and colour imaged with a Jai L-107CC 3 CCD RGB Line Scan Camera installed
4 on an Avaatech XRF core scanner (Fig. 3). The other half was sampled at 5 cm intervals in 1
5 cm thick slices, weighed and subsequently freeze-dried. Dry samples were weighed and wet
6 sieved over mesh sizes of 63 µm, 100 µm, and 1 mm. The residues were dried at 40°C.
7 Benthonic and planktonic foraminifera were picked from the >100 µm size fraction, counted
8 (at least 300 specimens for each sample when possible) and identified to species level for
9 assemblage analysis. In this paper, we only present data on the two most dominant planktonic
10 species *Neogloboquadrina pachyderma* sinistral (s) and *Turborotalita quinqueloba*, which
11 together constitute 88–98% of the assemblage. Ice Rafted Detritus (IRD) was counted in the
12 >1 mm size fraction (Fig. 3).

13 Eight ^{14}C AMS dates were performed on monospecific samples of *N. pachyderma* (s) (Table
14 1) at the Chrono Centre of Queen's University, Belfast, UK. The radiocarbon dates were
15 calibrated to calendar years using the Calib 7.0 program (Stuiver et al., 2014) and the marine
16 calibration curve Marine13 (Reimer et al., 2013) that operates with a standard reservoir
17 correction of -400 years (Mangerud and Gulliksen, 1975). A regional correction of $\Delta R=7\pm11$
18 years was applied, following the recommendations for planktonic foraminiferal dates by
19 Bondevik and Gulliksen (in Mangerud et al., 2006). The ages were calculated as the mid-point
20 value from the calibrated age range ($\pm 2\sigma$). Calibrated dates are presented in years before
21 present (BP) AD 1950 with standard deviation 2σ . The age model was constructed assuming
22 linear sedimentation between the calibrated dates (Fig. 3). The reservoir effect is probably not
23 constant through time, and especially during the Younger Dryas it was probably larger (e.g.,
24 Bard et al., 1994; Bondevik et al., 2006; Austin et al., 2011). However, the comparison of the
25 stratigraphy and the magnetic susceptibility data between core JM10-330GC and core JM03-
26 373PC2 (Fig. 1), which has been used as reference core for western Svalbard slope (Jessen et
27 al., 2010), indicate that a standard reservoir correction age is appropriate. In particular, the
28 presence of a diatom rich layer that was attributed to the early Holocene by Jessen et al.
29 (2010), together with a marked shift in the species composition of the planktonic foraminiferal
30 assemblage (previously dominated by the polar species *N. pachyderma* (s) and subsequently
31 by the subpolar species *T. quinqueloba*), and the decrease in oxygen isotope values in the
32 same interval, confirm an early Holocene age for these events, in accordance with our age
33 model (Fig. 4).

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1 Stable isotopes (oxygen and carbon; Table S1) were measured on the planktonic foraminifera
2 species *N. pachyderma* (s) and on the benthonic species *Cassidulina neoteretis* both picked
3 from the >100 µm size fraction. Stable isotopes were performed at the Leibniz-Laboratory for
4 Radiometric Dating and Isotope Research in Kiel, Germany, using a Finnigan MAT-253 mass
5 spectrometer with Kiel IV system (analytical precision of ±0.05‰ for δ¹³C and ±0.1‰ for
6 δ¹⁸O estimated by measuring the certified standard NBS-19). All isotope results are reported
7 in standard delta notation relative to Vienna PeeDee Belemnite (VPDB). The δ¹⁸O isotopic
8 values were corrected for ice volume effect (δ¹⁸O_{IVC}), using the Fairbanks (1989) sea-level
9 curve as dated by Bard (1990) with a correction of 0.11‰ δ¹⁸O per ten meters sea level
10 change (subtracted from the measured δ¹⁸O values; Table S1).

11 Additional stable isotope analyses of cleaned benthonic foraminifera samples from the lower
12 part of the core (418–370 cm) were performed on a Thermo Finnigan MAT252 mass
13 spectrometer coupled with a CarboKiel-II carbonate preparation device (analytical precision
14 ±0.03‰ for δ¹³C and ±0.08‰ for δ¹⁸O estimated by measuring the certified standard NBS-19;
15 results are reported in standard delta notation relative to VPDB) at the Serveis Científico-
16 Tècnics of the University of Barcelona (Table S1; Fig. 5). The benthonic foraminiferal
17 samples have been cleaned following the protocol of Pena et al. (2005), which is adapted from
18 Boyle and Rosenthal (1996). Prior to cleaning, the foraminifera were gently crushed between
19 clean glass plates to break open individual chambers. The cleaning steps comprise: 1) removal
20 of clays, Mn-Fe oxides and other mineral phases by a reductive cleaning step; 2) oxidative
21 cleaning to eliminate organic matter; 3) weak acid leaching to remove remaining impurities
22 from the shell surfaces. This protocol has proven to be efficient in removing the diagenetic
23 carbonates attached to the tests of foraminifera (Pena et al., 2008; Panieri et al., 2012, 2014b).

24 The preservation state of the foraminifera tests were examined by Scanning Electron
25 Microscope (SEM) in order to identify presence of secondary overgrowth of methane-derived
26 authigenic carbonates in selected representative specimens of *N. pachyderma* (s) and *C.*
27 *neoteretis* before the cleaning procedure (Figs. 6 and 8). Qualitative estimates of the trace
28 metal content were obtained from the test surface (coated with gold/palladium) with Energy
29 Dispersive X-ray Spectroscopy (EDS) (Fig. 7). SEM secondary electronic images and EDS
30 spectra were acquired on a JEOL 6610 tungsten SEM equipped with an Oxford Instrument
31 AzTEC EDS system at the Electron Microscopy Centre, Plymouth University, UK.

32

1 **4 Results**

2 **4.1 Chronology and lithology**

3 Based on the calibrated ^{14}C dates, the distribution patterns of polar and subpolar planktonic
4 foraminifera species (Fig. 4a), and the benthonic and planktonic $\delta^{18}\text{O}$ records (Figs. 4b–d), we
5 have correlated core JM10-330GC to the Greenland ice core event stratigraphy, applying the
6 new Greenland Ice Core Chronology 2005 (GICC05) of Rasmussen et al. (2006). The GICC05
7 time scale is b2k (before 2000 yrs), therefore to compare it with the calendar ages (AD before
8 1950) used in this paper, we have subtracted 50 yrs from the GICC05 time scale. In this new
9 chronology the different periods are defined as followed: end of Bølling interstadial: 14.025
10 ka; onset of Younger Dryas (YD) stadial: 12.85 ka; YD-Holocene transition: 11.65 ka.

11 Our age model shows that the core contains postglacial sediments covering the last 14 ka,
12 spanning from the upper part of the Bølling-Allerød (B-A) interstadial periods to Recent (Fig.

13 | 4). The lithology (Fig. 3) is very similar to the reference core of the western Svalbard slope
14 (Jessen et al., 2010), with the lower part (418–335 cm; 14.1–11.1 ka; B-A and YD interval)
15 characterized by high concentration of ice-raftered debris (IRD) and common pyritized burrows
16 indicative of bioturbation, with a greenish sandy layer at the beginning of the YD (360 cm;
17 12.7 ka). There is a fine-grained, structureless, silty mud interval with high abundance of
18 diatoms in the middle part (335–225 cm; 11.1–8.8 ka, labelled diatom rich mud in Fig. 4f). A
19 homogeneous hemipelagic, grey clay with very little amount of IRD is present in the upper
20 interval (225–0 cm; 8.8 ka to present).

21 The sedimentation rate is higher during the B-A period (~38 cm/ky) and in the early Holocene
22 (42–50 cm/ky), but lower during the Younger Dryas (YD: ~18 cm/ky) and after 7.5 ka (19–26
23 cm/ky) (Fig. 3; Table 1).

24

25 **4.2 Carbon isotope excursions (CIEs)**

26 During the Bølling-Allerød interstadials the $\delta^{13}\text{C}$ record of the infaunal benthonic foraminifera
27 *C. neoteretis* shows values considerably lower than the average core value of -1.10‰ (CIE I).
28 The low values occur at about 13.9 ka (-2.87‰) and at 13.5 ka (-4.37‰), and another but less
29 pronounced excursion at 12.9 ka (-2.21‰; Fig. 4e). This first interval with depleted $\delta^{13}\text{C}$ has
30 been termed carbon isotope excursion I (CIE I). The stable isotope analyses were repeated for
31 this interval on *C. neoteretis* specimens cleaned with a procedure designed to remove

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1 authigenic carbonate coatings (Pena et al., 2008; Panieri et al., 2012, 2014b). The $\delta^{13}\text{C}$ values
2 obtained are higher (by up to 1.6‰) compared to the tests cleaned with the standard protocol,
3 but are still lower (as low as -2.75‰) than the average value of the record (-1.10‰) (Table S1;
4 Fig. 5).

5 The planktonic $\delta^{13}\text{C}$ values during the CIE I interval are lower than the average core value of
6 0.07‰, and show one prominent excursion (-2.61‰) at 13.5 ka (Fig. 4c).

7 Another interval with low benthonic $\delta^{13}\text{C}$ values occurs in the early part of the Holocene (CIE
8 II; Fig. 4e). This event lasted approximately 500 years (c. 10.5–10 ka) and is characterized by
9 benthonic $\delta^{13}\text{C}$ values that are lower than -2‰, with the most prominent excursion at 10.3 ka
10 (-3.41‰). CIE II is not recorded in the planktonic record, where the $\delta^{13}\text{C}$ values are very close
11 to the average core value and within the normal range of the marine environment (Fig. 4c).

12 The $\delta^{13}\text{C}$ values in both benthonic and planktonic records between the two events (from about
13 12.8 to 10.5 ka) are very close or slightly lower than average values, whereas after 9 ka they
14 are higher compared to the average values (Figs. 4c–e).

15 The $\delta^{18}\text{O}_{\text{IVC}}$ benthonic and planktonic records present little variability if compared to the
16 average values of 4.35‰ and 3.12‰, respectively (Figs. 4b and 4d). Light isotope excursions
17 are present in both the benthonic (<0.7‰ more depleted than the average) and planktonic
18 record (about 1.2‰ more depleted than the average) during the Younger Dryas and are
19 probably related to a melt water event, which is also documented by a sandy layer (Fig. 4f).

20 No negative oxygen isotope excursions are associated with CIE I, and in the early Holocene
21 only two relatively light (<0.7‰ more depleted than the average) excursions exist in the $\delta^{18}\text{O}$
22 benthonic record at the beginning and at the end of CIE II (Fig. 4d).

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23 The SEM images of *N. pachyderma* (s) from the most negative excursion of CIE I (-2.61‰,
24 390 cm bsf at 13.5 ka) show evidence of surface alteration (Figs. 6A–B) with a thin coating
25 (<1µm) covering unevenly parts of the test (Fig. 6C). The Energy Dispersive X-ray
26 Spectroscopy (EDS) estimates show that the deposited layer is highly enriched in Si,
27 moderately enriched in Mg with minor traces of Fe and S (EDS 1 and 2 in Figs. 6C and 7).
28 The internal part of the wall looks unaltered with pristine crystal palisades consisting of
29 CaCO_3 (EDS 3 in Figs. 6C and 7).

30 The *N. pachyderma* (s) tests from CIE II (-0.02‰, 295 cm bsf at 10.3 ka) and from the late
31 Holocene (0.68‰, 80 cm bsf at 4.1 ka) are well preserved with pristine shell structure (Figs.

1 6D–E and G–H) and walls characterized by crystal palisades typical of this species (Figs. 6F–
2 I).

3 The SEM images of *C. neoteretis* (not cleaned specimens) are more difficult to interpret. No
4 clear coatings have been detected on the test surface of the specimens from both depleted
5 intervals CIE I ($\delta^{13}\text{C}$: -4.37‰, 13.5 ka, 390 cm bsf; Figs. 8A–C) and CIE II ($\delta^{13}\text{C}$: -3.41‰,
6 10.3 ka, 295 cm bsf; Figs. 8D–F). In both cases, the surface of the tests appears altered with
7 enlarged and irregular pores, probably due to dissolution and possibly associated with
8 recrystallization in the internal part of the test. The specimens from CIE II look even more
9 altered compared to the ones from CIE I. Specimens of *C. neoteretis* were very rare in this
10 interval and almost all the specimens have been used for isotope analyses. The specimens used
11 for the SEM pictures were the only examples left, were very small (note the scale in Fig. 8C)
12 and very poorly preserved.

13 The *C. neoteretis* tests from an interval with normal ^{13}C values (-0.44‰, 80 cm bsf at 4.1 ka)
14 are well preserved with pristine shell structure and small (<1µm), round pores (Figs. 8Q–R).

15

16 5 Discussion

17 5.1 CIEs: secondary overgrowth vs. primary tests

18 The benthonic foraminiferal (*C. neoteretis*) $\delta^{13}\text{C}$ record shows negative excursions in the
19 Bølling-Allerød interstadials (CIE I) and in the early Holocene (CIE II). These values are 1–
20 3.3‰ lower than the average value of -1.10‰ in the sediment core, which is comparable to
21 values of *C. neoteretis* recovered from sites unaffected by methane seepage from the same
22 region (ca. -1 to 0‰ in the northern Barents Sea: Wollenburg et al., 2001; -1.15‰ in a control
23 site away from the Håkon Mosby mud volcano in the Barents Sea: Mackensen et al., 2006).

24 The planktonic foraminiferal $\delta^{13}\text{C}$ record shows a similar negative trend only during CIE I
25 with one prominent negative excursion (-2.61‰) at 13.5 ka (Fig. 4c). Here the planktonic
26 values are generally lower compared to the normal $\delta^{13}\text{C}$ range of *N. pachyderma* (s) in the
27 same region (between ca. -0.5‰ and 1‰: Volkmann and Mensch, 2001; Nørgaard-Pedersen
28 et al., 2003; Sarnthein et al., 2003; Jessen et al., 2010).

29 Planktonic foraminifera are not expected to record a signal from methane in the water column
30 because most of the methane that have escaped from the seafloor should be consumed by
31 methanotrophic bacteria in the sediment and water column (Dickens, 2001; Reeburgh, 2007).
32 The negative values of *N. pachyderma* (s) can be attributed to diagenetic alteration that may

1 stem from methane-derived authigenic carbonates on the foraminiferal tests after their
2 deposition to the seafloor (Torres et al., 2003, 2010; Millo et al., 2005a; Panieri et al., 2009).

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3 Therefore, in order to distinguish the isotopic records of the secondary overgrowth from the
4 primary tests, we have repeated the isotopic analyses on cleaned *C. neoteretis* specimens from
5 the CIE I interval (see above). The repetition of the isotopic analyses for *N. pachyderma* (s)
6 and for *C. neoteretis* from the CIE II interval was impossible because of lack of material. Thus
7 a detailed SEM investigation was carried out on both species (see below).

8 The $\delta^{13}\text{C}$ values obtained after the cleaning procedure (as low as -2.75‰) are still significantly
9 lower than the core average (-1.10‰; Table S1; Fig. 5) and also lower compared to the $\delta^{13}\text{C}$
10 records of *C. neoteretis* of the last deglaciation from the same region with values ca. -0.5 to
11 0.3‰ in the Kara Sea (Lubinski et al., 2001), ca. -1 to 0‰ in the N Barents Sea (Wollenburg
12 et al., 2001) and ca. 0 to 1.5‰ in the SW Barents Sea (Aagaard-Sørensen et al., 2010). Dead
13 specimens (empty tests) of *C. neoteretis* from surface samples at the active Håkon Mosby mud
14 volcano in the Barents Sea show very similar $\delta^{13}\text{C}$ negative values (from -1.65‰ to -2.82‰)
15 (Mackensen et al., 2006). We suggest that the low $\delta^{13}\text{C}$ values obtained after the cleaning
16 procedure could be the result of calcification in presence of ^{13}C -depleted DIC and probably
17 ingestion of ^{13}C -depleted methanotrophic microbes on which foraminifera feed (e.g. Rathburn
18 et al., 2003; Hill et al., 2004; Panieri et al., 2014a). We interpret the ^{13}C -depleted values of
19 diagenetic overgrowth on the *C. neoteretis* shells as cumulatively added to the already
20 negative values of the primary tests.

21 The SEM pictures of *N. pachyderma* (s) from the most negative excursion during CIE I (390
22 cm, 13.5 ka) show clear evidence of surface alteration with a thin coating enriched in SiO_2 ,
23 Mg and traces of FeS (Figs. 6C and 7). Authigenic silica is common in ancient limestones
24 from seep sites (Smrzka et al., 2015) and similar SiO_2 enrichments have been observed in
25 secondary coatings on *N. pachyderma* (s) from depleted ^{13}C intervals in the southwestern
26 Greenland Sea (Millo et al., 2005a). The precipitation of silica phases would occur after
27 AOM-derived mineral phases ceased to form, but before the precipitation of late diagenetic
28 calcite cements (Kuechler et al., 2012; Smrzka et al., 2015). Enrichments in Mg in the
29 foraminiferal tests are typical of methane-derived authigenic carbonates, as previously
30 observed by Torres et al. (2003, 2010). The production of bicarbonate and hydrogen sulfide
31 increases carbonate alkalinity at the SMTZ, inducing the precipitation of carbonate minerals
32 and pyrite (e.g. Ritger et al., 1987; Peckmann et al., 2001; Sassen et al., 2004).

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1 *N. pachyderma* (s) in the early Holocene (CIE II) exhibit values in the normal marine range
2 ($>-0.5\text{\textperthousand}$) and the tests from this interval do not show any sign of surface alteration or coatings
3 (Figs. 6D–F)

4 No clear indications of coatings have been observed on the benthonic foraminiferal tests from
5 both depleted intervals (CIE I and CIE II; Figs. 8A–F). The surface alteration in this case
6 mainly consists of dissolution features probably due to an increase in CO₂ during AOM in
7 marine sediments (Eq. (2)). It is also possible that the precipitation of methane-derived
8 authigenic carbonates, in this case have occurred inside the benthonic tests. This is probably
9 because the smooth and imperforate test of *C. neoteretis* is less likely to accommodate
10 contamination and crystalline overgrowth than the *N. pachyderma* (s) test with higher porosity
11 and surface area, as already observed by Cook et al. (2011).

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12 The results of the repeated isotope analyses on the cleaned specimens from CIE I interval
13 seem to support this hypothesis since the cleaning procedure is meant to remove the authigenic
14 carbonate overgrowth and the values obtained are indeed less negative compared to the un-
15 cleaned specimens. However, these aspects are not fully understood and require additional
16 analyses and investigations.

17

18 **5.2 CIEs: evidence for methane release**

19 In order to explain the different results found for CIE I and CIE II, we have used a schematic
20 diagram modified after Borowski et al. (1996) and suggest two different scenarios depending
21 on the strength of the methane flux at the seep site (Fig. 9).

22 The intensity of upward methane flux can control sulfate (SO₄²⁻) profiles and depth of the
23 sulfate methane transition zone (SMTZ), if the sulfate diffusion from the seawater into the
24 sediment and the sediment characteristics are considered constant (Fig. 9B).

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25 The first scenario (Fig. 9A) represents CIE II, where only benthonic foraminifera show
26 negative $\delta^{13}\text{C}$ values. In this case the methane flux is high and the SMTZ is very close to the
27 seafloor. The oxidation of methane is less efficient causing lower rates of AOM and higher
28 methane flux into the bottom waters. The methane escaped is then partially oxidized by
29 methanotrophic aerobic microbes in the water column (Niemann et al., 2006). In this scenario,
30 the negative $\delta^{13}\text{C}$ values in the benthonic foraminiferal tests are likely to be the result of
31 calcification in presence of ¹³C-depleted DIC and probably ingestion of ¹³C-depleted

1 methanotrophic bacteria. In high advective flow settings, the surface communities are often
2 dominated by bacterial mats (Treude et al., 2003; Boetius and Wenzhöfer, 2013) on which
3 benthonic foraminifera can prey (Rathburn et al., 2003; Panieri et al., 2014a). Similar negative
4 $\delta^{13}\text{C}$ values (from -1.5 to -4.0 ‰) have been previously observed in benthonic foraminifera
5 living at methane seep sites (Sen Gupta and Aharon, 1994; Wefer et al., 1994; Sen Gupta et
6 al., 1997; Keigwin, 2002; Hill et al., 2003, 2004; Rathburn et al., 2003; Martin et al., 2007,
7 2010; Panieri et al., 2009, 2012, 2014a). However, we cannot exclude that early authigenic
8 carbonates were precipitated on the foraminiferal tests, when they were still alive, as a product
9 of AOM near the seafloor (e.g. Bohrmann et al. 1998).

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10 The second scenario (Fig. 9C) represents CIE I, where both benthonic and planktonic
11 foraminifera exhibit negative values of $\delta^{13}\text{C}$. We interpreted these anomalies in $\delta^{13}\text{C}$ as due to
12 secondary carbonate precipitation after the benthonic and planktonic foraminifera were buried.
13 The precipitation of carbonate occurs when the pore water is oversaturated with respect to
14 carbonate minerals. The SMTZ is typically a narrow zone of just a few centimetres (e.g.,
15 Iversen and Jørgensen, 1985; Niewöhner et al., 1998; Treude et al., 2005) resulting in the
16 sharp negative shift in $\delta^{13}\text{C}$ recorded by both benthonic and planktonic foraminifera (see Figs.
17 4 and 5). According to Borowski et al. (1996) methane-derived authigenic carbonate
18 precipitation occur when the methane flux is low and all the methane is oxidised by AOM
19 within the SMTZ. It is also possible that the primary test of the benthonic foraminifera was
20 already depleted in ^{13}C (orange benthonic foraminifera in Fig. 9C), and the negative values of
21 the secondary carbonate overgrowth were cumulative added (see above).

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22 These observations arise several questions and hypotheses about the possibility by benthonic
23 foraminifera to record past methane emissions. Further measurements on foraminifera from
24 methane seep sites and experiments are necessary to better understand how methane emissions
25 from different sources can affect the foraminiferal shells and the sort of information that can
26 be obtained from analysis of the isotope composition of their tests.

27

28 5.3 North Atlantic deglacial CIEs

29 Similar negative excursions in foraminiferal $\delta^{13}\text{C}$ records have been reported in several
30 Quaternary isotope records and have been interpreted as evidence for methane release
31 (Kennett et al., 2000; Smith et al., 2001; Keigwin, 2002; Millo et al., 2005b; Cook et al., 2011;
32 Hill et al., 2012). In particular, low $\delta^{13}\text{C}$ values during the deglaciation have also been

1 reported by Smith et al. (2001) in stable isotope records from the East Greenland continental
2 shelf (cores JM96 in Fig. 1). In order to compare these events with the Vestnesa Ridge record
3 (Fig. 10), the original radiocarbon ages of Smith et al. (2001) have been re-calibrated with
4 Calib 7.0 program (see above). No regional correction has been applied for the deeper cores
5 (JM96-1214: 574 mwd and JM96-1215: 668 mwd) as suggested by Jennings et al. (2006),
6 whereas in the shallow core (JM96-1207: 404 mwd) a correction of $\Delta R = -150$ years has been
7 applied, following Jennings et al. (2002). The first (14.4–14.2 ka; $-3.5\text{\textperthousand}$ for *C. neoteretis*) and
8 second excursions (12.9 ka; $-4.8\text{\textperthousand}$ for *N. pachyderma* (s)) isotopic excursions in the East
9 Greenland record are close in time to the beginning and the end of CIE I, respectively (Fig.
10 10b). The third excursion (10.9–9.7 ka; $-6.4\text{\textperthousand}$ for *N. pachyderma* s, $-3.8\text{\textperthousand}$ for *Cibicides*
11 *lobatus*) is coeval with CIE II, but lasted longer (Fig. 10b).

12 Negative $\delta^{13}\text{C}$ excursions during glacial and deglacial times in the Nyegga pockmark
13 field (800–1000 mwd, NPF in Fig. 1) have been recently reported (Hill et al., 2012). The
14 seepage in this area was particularly active between 15 and 13 ka, with several $\delta^{13}\text{C}$ negative
15 excursions (as low as $-6\text{\textperthousand}$) in both benthonic and planktonic records, while around 9 ka a
16 negative $\delta^{13}\text{C}$ excursion is present in the benthonic record ($-2.7\text{\textperthousand}$), but not reflected in the
17 planktonic record (Hill et al., 2012).

18 The similarity in timing of these carbon isotope excursions over long distances and
19 wide water depth ranges (Fig. 1) is remarkable and suggests that the CIEs could be regional at
20 the scale of the North Atlantic Ocean to the Fram Strait.

21 Both the Bølling-Allerød interstadials and the early Holocene are periods of climate warming
22 during the deglaciation (NGRIP Members, 2004) (Fig. 10c) and are characterized by increased
23 CH_4 concentration in the atmosphere (Brook et al., 2000; GISP2) (Fig. 10d). They also occur
24 during periods of rapid sea-level rise (melt water pulse-mwp-1A: 14.3–12.8 ka; mwp-1B:
25 11.5–8.8 ka; Fig. 10e) (Peltier and Fairbanks, 2006; Stanford et al., 2011). These findings
26 suggest an apparent correlation between methane events in the North Atlantic and the Fram
27 Strait, and climatic events at global or regional scale.

28 Since the methane escaped from the seafloor is rapidly oxidized in the water column, we do
29 not expect a short-term change in stable carbon isotope composition of the North Atlantic deep
30 water. The foraminiferal records published so far (e.g. Volkmann and Mensch, 2001;
31 Wollenburg et al., 2001; Nørgaard-Pedersen et al., 2003; Sarnthein et al., 2003; Aagaard-
32 Sørensen et al., 2010; Jessen et al., 2010) confirm this interpretation. However, the presence of
33 local carbon isotope perturbation at the same time in multiple locations (Fig. 1) is remarkable

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1 and possible common triggering mechanisms need to be considered.

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2 We are currently working on several cores from the Vestnesa Ridge in order to understand if
3 the negative carbon anomalies recorded by foraminifera are due to local or global processes
4 and if the apparent correlation with climatic events can be proved.

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5 ▲ 5.4 Possible triggering mechanisms and connection with climate change?

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Moved up [2]: Both the Bølling-Allerød interstadials and the early Holocene are periods of climate warming during the deglaciation (NGRIP Members, 2004) (Fig. 10c) and are characterized by increased CH₄ concentration in the atmosphere (Brook et al., 2000; GISP2) (Fig. 10d). They also occur during periods of rapid sea-level rise (melt water pulse-mwp-1A: 14.3–12.8 ka; mwp-1B: 11.5–8.8 ka; Fig. 10e) (Peltier and Fairbanks, 2006; Stanford et al., 2011). These findings suggest an apparent correlation between methane events in the North Atlantic and the Fram Strait, and climatic events at global or regional scale.

7 It is still not possible to determine if present-day gas emissions on the eastern part of the
8 Vestnesa Ridge are sourced from below the GHSZ (Gas Hydrate Stability Zone), directly from
9 dissociation of gas hydrates, or from a combination of deeper and shallower processes (Bünz
10 et al., 2012; Smith et al., 2014). It is also unclear whether focused fluid flow pathways from
11 the base of the GHSZ have been established recently at the end of the last glaciation, or
12 whether they have existed for much longer and have been reactivated multiple times. Even
13 though this study has only documented two former events of increased emission at the western
14 end of the Vestnesa Ridge, it favours a model of reactivation of chimney structures. The
15 timing of the gas emissions and reactivation seem to occur during periods of climate change
16 similar to observations from the mid-Norwegian margin, where the generation and initiation of
17 focused fluid flow is most likely related to an overpressure due to a combined effect of loading
18 of glaciogenic sediments and shelf ice glaciation (Hustoft et al., 2009b; Plaza-Faverola et al.,
19 2011). However, the initial generation of such chimney structures and their reactivation might
20 each be related to different geological processes. Chimneys are usually conceived as a network
21 of connected small-scale fractures originating from natural hydraulic fracturing (Arntsen et al.,
22 2007). They represent pre-existing zones of weakness and open pathways that might facilitate
23 fluid flow much more easily than during the initial generation of the fracture network. In deep-
24 water areas these fractures might be filled with gas hydrate (Kim et al., 2011).

25 The climate forcing of gas emission in the western part of the Vestnesa Ridge documented
26 herein could be the result of the individual or combined effect of sea-level rise, increased
27 seismicity, elevated sedimentation rates and/or gas hydrate dissociation. From the above
28 mentioned processes it seems unlikely that gas hydrate dissociation and loading of glaciogenic
29 sediment have played a major role given that our study area is in deep-water, far away from
30 the shelf edge and that the timing of the deposition of large amount of glaciogenic sediments
31 (Ottesen et al., 2005; Mattingdal et al., 2014) does not coincide with the venting periods
32 documented herein.

1 Both CIEs occur at times when deep convection and generation of cold bottom water is strong
2 (McManus et al., 2004; Ezat et al., 2014). The high $\delta^{18}\text{O}_{\text{ICV}}$ values of the benthonic record
3 during CIE I confirm that the area was bathed in cold water during the B-A interval (Fig. 4d).
4 During CIE II, the marked shift in the planktonic assemblage (previously dominated by the
5 polar species *N. pachyderma* (s) and subsequently by the subpolar species *T. quinqueloba*, Fig.
6 4a) indicates a surface water warming. The presence of two relatively light excursions of
7 benthonic $\delta^{18}\text{O}_{\text{ICV}}$ ($<0.7\text{\textperthousand}$; Fig. 4d) indicate a small warming of the bottom water (see
8 Rasmussen et al., 2007, 2014; Ezat et al., 2014; Groot et al., 2014), but certainly too small to
9 start gas hydrate dissociation. Hydrate dissociation at such depth would require a substantial
10 warming before any gas can escape from deeper buried sediments (>1000 mwd) (Reagan and
11 Moridis, 2007), although hydrates buried at shallower depth within chimneys could be
12 affected to some extent and release gas at the seabed (Smith et al., 2014).

13 The present sedimentation rate on the Vestnesa Ridge is about 19 cm/ka whereas, during the
14 deglaciation, it was considerably higher (40–50 cm/ka; Fig. 3 and Table 1). Around 14.6 to
15 14.3 ka sedimentation rates on the western Svalbard margin increased over large areas to $>$
16 5m/ka (Jessen et al., 2010 and references therein). It is however, not known if such a relatively
17 small loading alone could have significantly increased fluid overpressure in the subsurface
18 resulting in methane release at the seabed.

19 Sea level, on the other hand, has risen considerably after the last glaciation and the two
20 documented CIEs correlate well with two major melt water pulses (mwp-1A and -1B; Peltier
21 and Fairbanks, 2006; Stanford et al., 2011) (Fig. 10e). Global sea level rise has been
22 implicated in triggering of landslides by causing an increase in excess pore pressure in the
23 sub-seafloor during the deglaciation and the Holocene in the northern North Atlantic sector
24 (Owen et al., 2007; McGuire and Maslin, 2012). However, the hydrostatic pressure increase
25 would only affect excess pore pressures if impermeable sediments or a complex subsurface
26 structure traps the pores. These trapping mechanisms might be provided by gas hydrates that
27 clog the pore space along the BSR or fill the fractures of gas chimneys (Nimblett et al., 2003;
28 Kim et al., 2011). Potentially a cumulative effect of sedimentation and sea-level rise has
29 elevated excess pore pressure to initiate gas migration along the fracture network within a
30 chimney.

31 Moreover, sea level rise, elevated sedimentation rates in the whole study area and the isostatic
32 rebound following the retreat of the glaciers (Landvik et al., 1998; Forman et al., 2004;
33 Bungum et al., 2005) may foster other processes, such as lithospheric stress changes resulting

1 in increased seismicity. Modeling studies (Wallmann et al., 1988; Nakada et al., 1992) have
2 demonstrated that sea-level changes are capable of triggering or modulating tectonic activity.
3 More specifically, Luttrell and Sandwell (2010) have shown that lithospheric flexure due to
4 ocean loading caused by post-glacial sea-level rise was sufficient to promote failure through
5 the reduction of normal stress thereby reactivating faults.

6 Increased seismicity might have had a two-fold effect on the fluid flow system in the Vestnesa
7 Ridge. It might have led to an increased supply of gas along deep-seated faults leading to an
8 increase in pore pressures beneath the hydrate-bearing sediments. Furthermore, it might have
9 led to an excitation and dilation of fractures within the chimney structures leading to the
10 leakage of gas, similar to what has been observed on the Bear Island Fan (Franek et al., 2014).

11 We cannot conclusively distinguish which factors triggered gas venting in the two periods at
12 the end of and shortly after the last glaciation, but suggest that a combined effect of sea-level
13 rise, sedimentation and seismicity might have led to increased pore pressures in an already
14 over-pressured system, thereby promoting and initiating gas venting from the seabed at the
15 western part of the Vestnesa Ridge.

16

17 **6 Conclusions**

18 We have presented new data from a sediment core collected at the Vestnesa Ridge in the
19 eastern part of the Fram Strait, the deep-water gateway to the Arctic Ocean. Here, a sediment
20 core was retrieved from an apparently inactive pockmark in the western, deeper part of the
21 Vestnesa Ridge. The results show a surprisingly undisturbed sedimentary record that allows
22 establishing a detailed methane release event chronology for the last 14 ka. The benthonic
23 $\delta^{13}\text{C}$ record shows negative carbon isotope excursions (CIEs) as low as -4.37‰ during the
24 Bølling-Allerød interstadials (CIE I) and as low as -3.41‰ in the early Holocene (CIE II). The
25 persistence of negative values (as low as -2.75‰) after a thorough cleaning procedure, which
26 was designed to remove all authigenic carbonate coatings on benthonic foraminiferal tests
27 from CIE I samples, indicate that ^{13}C -depleted values of diagenetic overgrowth of shells of *C.*
28 *neoteretis* are cumulatively added to the already negative values of the primary test.

29 The planktonic foraminifera $\delta^{13}\text{C}$ record shows a similar negative trend during the CIE I (with
30 values as low as -2.61‰), but during CIE II the values are within the normal marine range (-
31 0.5‰ to 1‰). SEM investigations confirm the presence of a thin AOM-derived coating on the
32 *N. pachyderma* tests exclusively from the CIE I interval.

1 During CIE I in the Bølling-Allerød the negative ^{13}C values in both benthonic and planktonic
2 foraminifera have been interpreted as due to methane derived authigenic carbonates. During
3 CIE II in the early Holocene, only benthonic foraminifera exhibit negative ^{13}C values, which
4 in this case have been interpreted as the result of the incorporation of ^{13}C -depleted carbon
5 from methane emissions during the primary biomineralization of the tests and probably
6 ingestion of ^{13}C -depleted methanotrophic microbes on which foraminifera feed. It is also
7 possible that early authigenic carbonates, as AOM products in surface sediments, were
8 precipitated on the benthonic foraminiferal tests when they were still alive.

9 Methane release events with similar timing have been reported in several locations in the
10 North Atlantic and together with our new observation point to a more regional event, showing
11 an apparent correlation to northern climatic events.

12 We suggest that a combined effect of sea-level rise, high sediment loading and increased
13 seismicity during the deglaciation could have led to increased pore pressures, and therefore
14 promoting and initiating gas venting from the seafloor in the Vestnesa Ridge, eastern Fram
15 Strait. The methane contribution from the ocean floor to the water column and to the
16 atmosphere remains to be quantified.

17

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28

29 **Figure captions**

- 30 Figure 1. Bathymetric map of the Nordic seas showing the major surface currents and the
31 location of studied core JM10-330GC (yellow circle). *Other cores are indicated with a white*

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1 circle: JM96 are the cores described by Smith et al. (2001); JM03 represents core JM03-
2 373PC2 used as reference core for the western Svalbard slope (Jessen et al., 2010); NPF are
3 the cores described by Hill et al. (2012) in the Nyegga pockmark field next to the Storegga
4 Slide (dark grey area) on the mid-Norwegian margin. Abbreviations: NAC: North Atlantic
5 Current; WSC: West Spitsbergen Current; ESC: East Spitsbergen Current; EGC: East
6 Greenland Current.

7

8 Figure 2. A) Bathymetric map of the W-Svalbard margin and eastern Fram Strait.
9 Abbreviations: MFZ: Molloy Fracture Zone; MD: Molloy Deep; MR: Molloy Ridge; SFZ:
10 Spitsbergen Fracture Zone; YP: Yermak Plateau. B) Overview swath bathymetry map of the
11 Vestnesa Ridge. Locations of studied core JM10-330GC and location of active sites with gas
12 flares observed in June 2010 (Bünz et al., 2012) and in 2012 (Smith et al., 2014) are indicated.
13 Figures are modified after Hustoft et al. (2009a).

14

15 Figure 3. Left: lithology and colour scan of core JM10-330GC, together with magnetic
16 susceptibility and concentration of ice-raftered debris (IRD) per gram dry weight sediment.
17 Arrows indicate positions of original radiocarbon dates. Right: age model and calculated
18 sediment accumulation rates. Red triangles indicate radiocarbon dates.

19

20 Figure 4. Planktonic foraminifera and geochemical data of core JM10-330GC plotted versus
21 calibrated (cal) ka before present (BP) AD 1950; (a) relative abundance of *Neogloboquadrina*
22 *pachyderma* (s) (black line) and *Turborotalita quinqueloba* (purple line); (b) ice volume
23 corrected (IVC) $\delta^{18}\text{O}_{\text{IVC}}$ record of *N. pachyderma* (s); (c) $\delta^{13}\text{C}$ record of *N. pachyderma* (s);
24 (d) $\delta^{18}\text{O}_{\text{IVC}}$ record of *Cassidulina neoteretis* before (red line) and after (green line) the
25 cleaning protocol of Pena et al. (2005); (e) $\delta^{13}\text{C}$ record of *C. neoteretis* before (red line) and
26 after (green line) the cleaning protocol of Pena et al. (2005); (f) stratigraphy obtained for
27 JM10-330GC with chronological subdivisions (dashed lines). Dotted vertical lines indicate
28 average core values. White diamonds on the y axis indicate AMS ^{14}C dating points for JM10-
29 330GC. Shaded horizontal bars indicate carbon isotope excursions CIE I and CIE II.
30 Abbreviations: B-A: Bølling-Allerød; YD: Younger Dryas; sl: sandy layer.

31

32 Figure 5. Detail of the $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ record of *C. neoteretis* from CIE I interval, before (red
33 line) and after (green line) the cleaning protocol of Pena et al. (2005).

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1 Figure 6. Representative SEM images of *N. pachyderma* (s) from different intervals: A, B and
2 C from 390 cm bsf; D, E and F, from 295 cm bsf; G, H, and I from 80 cm bsf. A) *N.*
3 *pachyderma* (s) test from the most depleted interval within CIE I ($\delta^{13}\text{C}$: -2.61‰, 13.5 ka, 390
4 cm bsf), showing a test with surface alteration. B) Detail of picture A with clearly altered
5 external surface. C) Broken test of a different specimen of *N. pachyderma* (s) from the same
6 interval, where a thin deposited layer (methane-derived coating) unevenly cover the external
7 test, whereas the foraminiferal wall shows a pristine structure with pristine crystal palisades.
8 The numbers in the picture indicate EDS analyses in Fig. 7. D) *N. pachyderma* (s) from CIE II
9 interval ($\delta^{13}\text{C}$: -0.02‰, 10.3 ka, 295 cm bsf) showing an unaltered test. E) Detail of picture D
10 with pristine shell structure. F) Detail of the wall structure with pristine crystal palisades from
11 a different specimen in the same interval. G) *N. pachyderma* (s) from interval with normal ^{13}C
12 values ($\delta^{13}\text{C}$: 0.68‰, 4.1 ka, 80 cm bsf) showing a well-preserved test with pristine shell
13 structure. H) Detail of picture G with pristine crystals shape. I) Detail of the wall structure
14 with pristine crystal palisades from a different specimen in the same interval. Picture
15 magnifications and length of the white bar are indicated in each picture.

16
17 Fig. 7. 1–2) Energy Dispersive X-ray Spectroscopy EDS spectra of outer layer on the test of
18 *N. pachyderma* (s) from the most depleted interval within CIE I (Fig. 6C) showing
19 enrichments in Si and Mg and traces of Fe and S. 3) EDS spectra of the internal part of the
20 wall of the same specimens (fig. 6C) showing a pristine composition consisting of CaCO_3 .

21
22 Figure 8. Representative SEM images of not cleaned *C. neoteretis* specimens from the
23 depleted intervals (A–C from 390 cm bsf; D–F from 295 cm bsf;) and from an interval with
24 normal ^{13}C values (G–I from 80 cm bsf). A) *C. neoteretis* test from the most depleted interval
25 within CIE I ($\delta^{13}\text{C}$: -4.37‰, 13.5 ka, 390 cm bsf), showing a test with surface alteration by
26 dissolution features. B) Detail of picture A where the external test shows enlarged pores (5–
27 7µm) with irregular shapes. C) Detail of picture A with a different magnification showing the
28 irregular profile of the test wall. D) *C. neoteretis* from CIE II interval ($\delta^{13}\text{C}$: -3.41‰, 10.3 ka,
29 295 cm bsf) showing a test with surface alteration characterized by dissolution features. Note
30 the smaller scale of the picture. E) Detail of picture D, where the enlarged and irregular shaped
31 pores are widespread. F) Detail of the test of a different specimen from the same interval,
32 where the surface alteration is less evident, but the profile of the test walls is quite irregular.
33 G) *C. neoteretis* from an interval with normal ^{13}C values ($\delta^{13}\text{C}$: -0.44‰, 4.1 ka, 80 cm bsf)
34 showing a well-preserved test with pristine shell structure. H) Detail of picture G showing

1 small (<1µm) and round shape pores with no alteration features. I) Detail of picture G with a
2 different magnification showing a well preserved test and regular profile of the test wall.
3 Picture magnifications and length of the white bar are indicated in each picture.
4
5 Fig. 9. A) Cartoon representing CIE II, where only benthonic foraminifera exhibit negative
6 $\delta^{13}\text{C}$ values. In this scenario, methane activity is sufficiently high as to surpass the SMTZ and
7 oxidized less efficiently with the consequence of lower AOM rates and higher methane fluxes
8 into the bottom waters. The methane escaped is then partially oxidized by methanotrophic
9 aerobic microbes in the water column. In this scenario the negative $\delta^{13}\text{C}$ values are likely to be
10 the result of calcification in presence of ^{13}C -depleted DIC and probably ingestion of ^{13}C -
11 depleted methanotrophic microbes on which foraminifera feed. B) Schematic diagram
12 showing how methane flux controls the SMTZ, if the flux of sulfate (SO_4^{2-}) from the seawater
13 is constant and the characteristics of the sediments are constant. Arrow size is proportional to
14 upward methane flux. Modified after Borowski et al. (1996). C) Cartoon representing CIE I,
15 where both benthonic and planktonic foraminifera exhibit negative $\delta^{13}\text{C}$ values. In this case
16 the methane flux is low and upward fluxes of methane and downward fluxes of sulfate
17 (coming from the sea water) meet within the SMTZ and the anaerobic oxidation of methane
18 (AOM) can take place (e.g. Boetius et al., 2000; Knittel and Boetius, 2009). Here, sulfate and
19 methane may be consumed simultaneously according to the Equation (1). The production of
20 bicarbonate from AOM can induce the precipitation of calcium carbonate, so-called methane-
21 derived authigenic carbonates, on the benthonic and planktonic foraminifera tests after burial
22 through the Equation (2).

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23
24 Fig. 10. Isotopic data of core JM10-330GC and other records plotted versus calibrated (cal) ka
25 before present (BP) AD 1950; (a) $\delta^{13}\text{C}$ record of *N. pachyderma* (s) (blue line) and of *C.*
26 *neoteretis* (red line) from JM10-330GC; (b) $\delta^{13}\text{C}$ negative excursions from east Greenland
27 continental shelf (Smith et al., 2001): event 1 from core JM96-1215, event 2 from core JM96-
28 1214, and event 3 from core JM96-1207; (c) NorthGRIP ice core $\delta^{18}\text{O}$ record (North
29 Greenland Ice Core Project Members, 2004); (d) Greenland Ice Sheet Project 2 (GISP2)
30 atmospheric CH₄ record (Brook et al., 2000); (e) melt water pulse mwp-1A and mwp-1B
31 (Peltier and Fairbanks, 2006; Stanford et al., 2011); (f) stratigraphy obtained for JM10-330GC
32 with chronological subdivisions (dashed lines). White diamonds on y axis indicate dating
33 points for JM10-330GC (next to plot a) and black diamonds next to plot (b) for cores of Smith

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1 et al. (2001). Shaded horizontal bars indicate carbon isotope excursions CIE I and CIE II.

2 Abbreviations: B-A: Bølling-Allerød; YD: Younger Dryas; sl: sandy layer.

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