

1 **Foraminiferal $\delta^{18}\text{O}$ reveals gas hydrate dissociation in Arctic and North Atlantic oceans**
2 **sediments**

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14

15 **Abstract**

16 Paleooceanographic investigations in the Arctic and north Atlantic are crucial to understanding
17 past and current climate change, in particular considering amounts of pressure-temperature
18 sensitive gas stored in marine sediments of the region. Many paleooceanographic studies are
19 based on foraminiferal oxygen and carbon stable isotope compositions ($\delta^{18}\text{O}$, $\delta^{13}\text{C}$) from
20 either planktonic specimens, benthic specimens or both. However, in seafloor regions
21 proximal to high upward methane fluxes, such as where seafloor gas emission and shallow
22 gas hydrate-bearing sediment occur, foraminiferal $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ display a wide range of
23 values. Our study focuses on foraminiferal stable isotope signatures in shallow sediment at
24 core sites in the Arctic affected by significant upward flow of methane. This includes cores
25 with shallow sulfate methane transitions that are adjacent to seeps and containing gas hydrate.
26 We place emphasis on potential effects due to gas hydrate dissociation and diagenesis. Gas
27 hydrate dissociation is known to increase pore-water $\delta^{18}\text{O}$, but our results indicate that
28 precipitation of methane-derived authigenic carbonate (MDAC) also affects the foraminiferal
29 $\delta^{18}\text{O}$ of both planktonic and benthic species. In addition to this post-depositional overprint, we
30 investigate the potential bias of the stable isotope record due to ontogenetic effects. Our data
31 show that the size fraction does not impact the isotopic signal of planktonic and benthic
32 foraminifera.

33

34 Keywords: Foraminiferal stable isotopes, Arctic Ocean, gas hydrates, authigenic carbonates

35

36 **1. Introduction**

37 The Arctic is particularly sensitive to climate change (e.g., IPCC13; Screen and
38 Simmonds, 2010; Serreze and Barry, 2011) and multiple oceanographic parameters are
39 rapidly changing (Jakobsson et al., 2008). The Arctic is a fundamental component of the
40 climate system because of its role in global carbon cycling (e.g., McGuire et al., 2009). First,
41 the Arctic Ocean sequesters carbon dioxide that enters North Atlantic Deep Water. Second,
42 the Arctic modulates carbon exchange with the atmosphere because of seasonal sea-ice
43 coverage (e.g., McGuire et al., 2009). Third, the region contains very large amounts of
44 methane in permafrost and gas hydrates, both which are sensitive to temperature change
45 (Corell et al., 2008).

46 Along the Arctic continental shelves and slopes, probably between 30 and 170 Pg of
47 methane exists as gas hydrate (e.g., Kvenvolden 1988; McGuire et al., 2009; James et al.,
48 2016). Gas hydrates are crystalline solids that consist of gas (mostly methane) trapped in a
49 lattice of hydrogen-bonded water molecules (Sloan and Koh, 2007). Hydrates are stable at
50 relatively low ($< 10^{\circ}\text{C}$) temperatures and moderate ($>3\text{-}5\text{ MPa}$) pressures, which at high
51 latitudes generally correspond to water depths greater than 300 m (James et al., 2016).
52 However, warming of intermediate- and deep-water masses or depressurization because of
53 isostatic rebound might trigger hydrate dissociation (Thomas et al., 2002; Yao et al., 2019).
54 Already, it has been estimated that dissociation of gas hydrates located on Arctic shelves
55 contributes 0.08-0.13 Tg of methane per year to the atmosphere (McGuire et al., 2009).

56 Methane in marine sediment is significantly depleted in ^{13}C , often having a stable
57 carbon isotope composition ($\delta^{13}\text{C}$) less than $<-40\text{ ‰}$ (Whiticar, 1999). In areas characterized
58 by upward methane seepage, consumption of this methane by aerobic and anaerobic processes
59 and the stable carbon isotope composition ($\delta^{13}\text{C}$) of DIC in bottom water and especially pore
60 water and can become greatly depleted in ^{12}C . Benthic foraminifera living on or just below
61 the seafloor precipitate carbonate shells (or tests) using ambient dissolved inorganic carbon
62 (DIC) (McCorkle et al., 1990). Benthic foraminifera have been widely used to reconstruct
63 methane seepage at cold seeps and gas hydrate-rich sediments (e.g., Wefer et al., 1994;
64 Kennett et al., 2000; Hill et al., 2003; Barbieri and Panieri, 2004; Martin et al., 2007 and
65 2010, Panieri et al., 2009, 2012, 2014 and 2016; Consolaro et al., 2015; Schneider et al.,
66 2018). However, it has been demonstrated that both benthic and planktonic foraminifera can
67 be affected by diagenetic processes, particularly the secondary overgrowth precipitation of

68 methane derived authigenic carbonate (MDAC). While it is now accepted that MDAC
69 overgrowth alters the $\delta^{13}\text{C}$ of the foraminiferal isotope record (Torres et al., 2003; Panieri et
70 al., 2016, 2017a; Schneider et al., 2017 and 2018; Consolaro et al., 2018; Wan et al., 2018),
71 the impact of such diagenesis on foraminiferal $\delta^{18}\text{O}$ is less clear. In studies involving hydrate
72 stability reconstructions, both the stratigraphy and climate variations are usually interpreted
73 based on the foraminiferal $\delta^{18}\text{O}$ record (e.g., Dickens et al., 1995; Kenett et al., 2000; Thomas
74 et al., 2002). Nevertheless, the climate reconstruction approach is problematic in cold seeps
75 releasing methane from gas hydrate source, as one would expect considering that the water
76 trapped in gas hydrates is more enriched in ^{18}O compared to the adjacent pore water
77 (Davidson, 1983). During gas hydrate dissociation, ^{18}O -enriched water is released, and this
78 signal can potentially be incorporated in the shell precipitated by living benthic foraminifera.
79 In addition, this signal can be captured by the MDAC precipitating on the foraminiferal shell
80 after the death of the organisms (secondary overgrowth).

81 In this study, we investigate living and fossil foraminiferal specimens from different
82 cold seeps in the Arctic Ocean and the Norwegian Sea to understand if and how methane
83 release and/or hydrate dissociation can affect the foraminiferal $\delta^{18}\text{O}$ signature. We generate
84 new foraminiferal $\delta^{18}\text{O}$ data sets using living (Rose Bengal stained) and fossil benthic and
85 planktonic foraminiferal species (fractions >63 and >125 μm) from an active pockmark
86 currently releasing methane at Vestnesa Ridge, from gas hydrate mounds from Storfjordrenna
87 (south Svalbard) and from two canyons offshore the Lofoten islands (Northern Norway)
88 characterized by methane-rich sediments. We interpret the results obtained in the context of
89 the sampling environment (i.e, presence/absence of gas hydrates and methane seepage), but
90 also of the foraminiferal ontogeny and ecological preferences (i.e., microhabitat). Finally, we
91 compare the results obtained examining the $\delta^{18}\text{O}$ data together with $\delta^{13}\text{C}$ data from the same
92 samples. This study represents a significant advancement in the application of the
93 foraminiferal $\delta^{18}\text{O}$ in paleoclimatic reconstructions conducted at sites of methane release and
94 gas hydrate dissociation.

95

96 **2. Study area**

97 This study is based on sediment samples from push cores collected at three geographic
98 locations: 1) Vestnesa Ridge, western Svalbard margin (79°N , 6°E , 1200 m water depth), 2)
99 Storfjordrenna, south Svalbard margin (76°N , 16°E , ~ 390 m water depth); and 3) an area of
100 the Norwegian margin east of the Lofoten Islands (68°N , 10°W , ~ 750 m water depth) (Figs. 1

101 A-D; Table 1). Vestnesa Ridge is a 100 km-long sediment drift oriented SE-NW to E-W
102 (Talwani and Eldholm, 1977; Thiede et al., 1998; Bünz et al., 2012) characterized by gas
103 hydrate in the subseafloor and methane emitting pockmarks (Bünz et al., 2012; Panieri et al.,
104 2017b). Storfjordrenna is a channel characterized by several mounds (~500 m in diameter and
105 ~10 m in height above the seafloor) constituted by hemipelagic sediments with gas hydrate
106 and carbonate layers of, referred to as gas hydrate mounds (GHM) (Hong et al., 2017, 2018)
107 or gas hydrate pingos (Serov et al., 2017). The third geographic location comprises two
108 canyons situated north of the Trænadjupet slide, on the southern part of the continental
109 Lofoten-Vesterålen slope (Rise et al., 2013). These canyons are ~1.3 km long and 50 m deep
110 relative to surrounding seafloor. Within the canyons, the seafloor is characterized by active
111 methane seepage and microbial mats (Sen et al., 2019); however, there is no evidence for gas
112 hydrates in this area (Rise et al., 2013; Hong et al., 2019).

113

114 **3. Materials and Methods**

115 **3.1 Sediment core collection**

116 Push cores from Vestnesa Ridge were collected in July 2016 using the R/V *G.O. Sars*
117 and the ROV *Ægir 6000*. The sampling was conducted within the two most active pockmarks
118 (Lunde and Lomvi; Figs. 1 A and B). In particular, we collected 7 push cores within the
119 Lunde pockmark (cores V-15, V-16, V-17, V-18, V-19, V-21 and V-25) and 3 push cores
120 within the Lomvi pockmark (cores V-7, V-8 and V-9) (Table 1). The sampling targeted
121 whitish microbial mats (Figs. 1 F and G), indicative of active methane seepage. One push core
122 was collected in the Lunde pockmark in sediments devoid of microbial mats and with no
123 rising methane bubbles (core V-20; Fig. 1E). At this site, head space analysis confirmed the
124 absence of methane. Because of this, we consider this a non-seep reference core.

125 Multicores from two gas hydrate bearing mounds (GHM) in Storfjordrenna (GHMs 1
126 and 5) were collected in June 2017 using the R/V Helmer Hanssen and a multicorer equipped
127 with a video camera system (cores 898, 900, 902, 916, 917, 918, 919, 920, 921, and 922; Figs.
128 1 A and C; Table 1). The push cores from the Lofoten-Vesterålen (LV) canyons were
129 collected in August 2017 during a cruise on board the R/V *G.O. Sars* using the ROV *Ægir*
130 *6000* (cores L-8, L-19, L-31, L-32, L-35, L-52, L-56; Figs. 1 A and D; Table 1).

131

132 **3.2 Pore water analysis**

133 Pore water samples were collected in all cores considered in this study, with the
134 exception of cores V-15 and V-19 (Vestnesa Ridge), where we could not extract enough pore

135 water for sulfate analyses. All pore water samples were measured for sulfate (SO_4^{2-}) except
136 cores 916, 920, 921, and 922 (Storfjordrenna) because of the low yield. In the LV area, the
137 cores collected in the southern canyon were shared for macro-biology, geochemistry, and
138 micropaleontology investigations. Thus, sulfate profiles were obtained from push cores
139 adjacent to the cores studied for foraminifera. Sulfate concentrations were determined by a
140 Dionex ICS-1100 Ion Chromatograph equipped with a Dionex IonPac AS23 column at the
141 Norwegian Geological Survey (NGU, Trondheim, Norway; Sauer et al., 2016).

142 Chloride concentrations were measured on pore water samples from cores V-7, V-8,
143 V-9, V-16, V-20, 898, 900, 902, 917, 918, and 919. Chloride concentrations were also
144 determined from the LV canyons cores and reported in Hong et al., (2019). All chloride
145 concentrations were measured onshore also by ion chromatograph (see analytical details in
146 Yao et al. (2019).

147 The $\delta^{13}\text{C}_{\text{DIC}}$ was determined on every core analyzed for sulfate, with the exception of
148 cores L-52 and L-56. Measurements were conducted at EAWAG (The Swiss Federal Institute
149 of Aquatic Science and Technology) using an IRMS (Isotope Ratio Mass Spectrometer,
150 Isoprime) equipped with a Gilson 222XL Liquid Handler and a Multiflow unit (Isoprime).
151 Data are reported relative to the Vienna Pee Dee Belemnite (VPDB). The laboratory standard
152 deviation for the VPDB $\delta^{13}\text{C}$ was $\pm 0.1\%$, based on repeated measurements of the standard.
153 $\delta^{13}\text{C}_{\text{DIC}}$ from the LV area were measured at Oregon State University (see details for methods
154 in Torres et al., 2005). All pore water raw data are available in supplementary Table a.

155

156 **3.3 Foraminiferal stable isotope geochemistry**

157 All cores for micropaleontological analysis were sliced on board. Specifically, we
158 collected the first 5 cm of each core at 1-cm resolution at Vestnesa for foraminiferal analyses,
159 with the exception of cores V-15 and V-20 for which only the first 2 cm and first cm,
160 respectively, were available. The same has been done for the first sediment horizon (0-1 cm)
161 in Storfjordrenna and LV samples. All samples were stored in a 2 g L⁻¹ Rose Bengal solution
162 in 96 % ethanol, in order to identify living individuals (Walton, 1952). Stained individuals
163 were considered alive or recently alive following Corliss (1991). Prior to washing, samples
164 were stored at 4° C for at least 14 days, following the FOBIMO protocol (Schönfeld et al.,
165 2012), then wet sieved using 63 and 125 μm mesh sieves and dried at 40° C. Foraminifera
166 were picked from the fractions >63 and >125 μm using a stereo microscope.

167 Isotope measurements ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) were performed on Rose Bengal stained, dead
168 benthic and dead planktonic foraminifera from the 0-1 cm interval of all cores collected at the
169 three sampled geographic locations (Figs. 1 A-D). In addition, foraminiferal isotope data were
170 collected from the 3-4 cm interval of the Vestnesa Ridge cores. This allowed the comparison
171 of the foraminiferal isotope composition with the most superficial pore water $\delta^{13}\text{C}_{\text{DIC}}$ data
172 available. On cores V-8 and V-16, the isotopic composition of foraminifera was measured for
173 the entire length of the cores (i.e, 30 and 20 cm for cores V-8 and V-16, respectively).

174 Foraminiferal $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ measurements were conducted at the stable isotope
175 laboratory at UiT – The Arctic University of Norway in Tromsø (Norway) using a Thermo
176 Scientific MAT253 IRMS coupled to a Gasbench II. Species-specific analyses were done on
177 several benthic (*Cassidulina neoteretis*, *Cibicides wuellerstorfi*, *Melonis barleeanus*,
178 *Nonionellina labradorica* and *Trifarina earlandi*) and planktonic (*Neogloboquadrina*
179 *pachyderma*) foraminiferal species. Recently, it was reported that *M. barleeanus* is
180 characterized by sedimentary particles within its shell, but this mostly influence isotopic data
181 collected using in-situ, rather than bulk, techniques (Borrelli et al., 2018). Foraminiferal shells
182 were placed in 4.5 mL vials and flushed with He gas. Five drops of water-free H_3PO_4 were
183 added manually. After equilibration (>3 hours at 50°C), the samples were analyzed on a
184 Gasbench II and MAT253 Isotope Ratio Mass Spectrometer. Normalization to the VPDB for
185 carbon and oxygen isotopes was done using in-house standards (1.96 ‰, -10.21 ‰, and -
186 48.95 ‰ for $\delta^{13}\text{C}$ and -2.15 ‰ and -18.59 ‰ for $\delta^{18}\text{O}$). Analytical precision was estimated to
187 be better than 0.07 ‰ for $\delta^{13}\text{C}$ and 0.08 ‰ for $\delta^{18}\text{O}$ by measuring the certified standard NBS-
188 19. Foraminiferal $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ data are reported in supplementary Tables b, c, and d.

189 Selected specimens were examined by scanning electron microscopy (SEM) and
190 energy dispersive x-ray spectrometry (EDS) to investigate possible diagenetic alterations of
191 the shells. Analyses were performed on several benthic (*Cassidulina neoteretis*, *Melonis*
192 *barleeanus*, *Nonionellina labradorica* and *Trifarina earlandi*) and on one planktonic
193 (*Neogloboquadrina pachyderma*) foraminiferal species. 24 specimens have been analyzed and
194 we selected 12 of them to show in this study, based on the quality of the images obtained. The
195 analyzed specimens were chosen from different sediment depths (0-1 cm, 3-4 cm, and 29-30
196 cm) from cores collected at the three study areas. Specimens were mounted on a circular 25
197 mm diameter mold using adhesive tape. The mount was then carbon coated and examined
198 with a SEM Hitachi Tabletop Microscope TM-3030 equipped with a Bruker Quantax 70 EDS
199 Detector at UiT. Uncalibrated EDS analyses were performed on the same specimens imaged

200 by SEM to evaluate the elemental composition of the foraminiferal shells and secondary
201 overgrowth.

202

203 **4. Results**

204 **4.1 Pore Water profiles**

205 At Vestnesa Ridge, sulfate concentrations remain fairly constant for core V-20 (our
206 inactive, non-seep control core), and near those of seawater (~28 mM), ranging from 28.4 to
207 29.6 mM (Fig. 2). Sulfate concentrations in all other cores examined decrease sharply from
208 the seafloor to the bottom of the core. We note that sulfate concentrations in cores V-9 and V-
209 16 are much lower than seawater values, even close to the seafloor (3.6 mM at 2 cm depth and
210 8.1 mM at 1 cm depth for cores V-9 and V-16, respectively). Except for cores V-18 and V-21,
211 sulfate concentrations are undetectable by 10 cm sediment depth (Fig. 2).

212 In cores from Storfjordrenna GHM1 (Fig. 3), sulfate profiles at the tops of core have
213 sulfate concentrations similar to that of seawater, with the exception of core 902, where it is
214 20.2 mM. Sulfate concentrations remain almost constant with depth in cores 898, 917, and
215 919. A sharp decrease in sulfate concentration with depth is observed in core 902, where
216 sulfate reaches 0.4 mM at 24 cm depth. In cores 900 and 918, sulfate concentration decreases
217 with sediment depth, reaching 2.7 mM at 34 cm and 13.7 mM at 47 cm depth in core 900 and
218 918, respectively (Fig. 3).

219 In the LV area, sulfate concentrations were measured on cores from the northern
220 canyon (L-52 and L-56) and from the southern canyon (L-8, L-19, L-31, L-32 and L-35) (Fig.
221 4). In the southern canyon, cores available for pore water analysis were adjacent to cores
222 studied for foraminifera. Specifically, push core L-9 and push core L-12 were collected next
223 to push cores L-8 and L-19, respectively, whereas push core L-30 was collected next to cores
224 L-31, L-32, and L-35 (Table 1). Overall, the LV canyons cores are characterized by a sharp
225 decrease of sulfate with increasing sediment depth, with the exception of core L-12, which
226 shows constant sulfate concentrations similar to seawater values. In the northern canyon,
227 sulfate reaches very low concentrations around 12 cm depth (cores L-52 and L-56). In the
228 southern canyon, sulfate concentrations are lower than 1 mM below 2 cm in core L-9 and
229 below 6 cm in core L-30 (Fig. 4).

230 Chloride concentration ranges between 500 and 600 mM and it remains almost
231 constant regardless of sediment depth in all cores analyzed (Figs. 2 and 3). The $\delta^{13}\text{C}_{\text{DIC}}$ shows
232 values close to 0 in cores V-20 and 898, reflecting the $\delta^{13}\text{C}_{\text{DIC}}$ signature of normal marine

233 environment (-1 to 1‰; Tagliabue and Bopp, 2008). The $\delta^{13}\text{C}_{\text{DIC}}$ ranges between -20 and -50
234 ‰ in the cores collected at Vestnesa Ridge and Storfjordrenna GHM1, whereas the cores
235 collected at the LV canyons are characterized by $\delta^{13}\text{C}_{\text{DIC}}$ values between -10 ‰ and -70 ‰
236 (Figs. 2-4).

237

238 **4.2 Foraminiferal isotopic composition**

239 In this study, we primarily focus on samples collected at Vestnesa Ridge. The presence
240 of gas hydrates at the seafloor and associated gas seepages in the area (Vogt et al., 1994;
241 Hutsoft et al., 2009; Petersen et al., 2010; Bünz et al., 2012; Panieri et al., 2017b), together
242 with the the fact that the cores were collected on microbial mats using a remotely operated
243 vehicle (ROV), make these samples the ideal ones to study the potential effect of gas hydrate
244 dissociation on the foraminiferal $\delta^{18}\text{O}$ signature. Hence, at Vestnesa Ridge, we generated
245 complete foraminiferal stable isotope data sets using samples from cores V-8 (30 cm long)
246 and V-16 (20 cm long) (Figs. 1B and 5). We selected these two cores because of the high
247 number of living and dead individuals belonging to *N. pachyderma*, *M. barleeanus*, and *C.*
248 *neoteretis*. The foraminiferal isotopic composition of *N. pachyderma*, *C. neoteretis* and *M.*
249 *barleeanus* was also measured in core V-20, in the interval 0-1 cm (Vestnesa Ridge; Figs. 1B
250 and 5). These values represent the isotopic composition of foraminifera not affected by
251 methane release and oxidation.

252 The foraminiferal $\delta^{13}\text{C}$ values from core V-20 are similar to the foraminiferal $\delta^{13}\text{C}$
253 typical of normal marine conditions (-1 to 1‰; McCorkle et al., 1990), whereas the $\delta^{18}\text{O}$
254 values range from 2.7 to 2.8 ‰ (*N. pachyderma*) and from 4.3 to 4.5 ‰ (*C. neoteretis*), with
255 *M. barleeanus* recording values from 3.9 to 4 ‰, which are very close to benthic and
256 planktonic values measured in the area (e.g., Consolaro et al., 2017, Schneider et al., 2018).

257 In samples from cores V-8 and V-16, the *M. barleeanus* $\delta^{13}\text{C}$ values range from -2 to -
258 8 ‰, whereas *N. pachyderma* is characterized by values between 1.2 and -14 ‰. In core V-8,
259 we analyzed also *C. neoteretis* and its $\delta^{13}\text{C}$ values range from -1.5 to -16 ‰. In general, the
260 foraminiferal $\delta^{13}\text{C}$ decreases with increasing sediment depth at core V-8, whereas at core V-
261 16, the $\delta^{13}\text{C}$ values decreases from the core surface to a depth of 10 cm and increases
262 afterwards. The trend in foraminiferal $\delta^{18}\text{O}$ values is less straightforward. In core V-8, *N.*
263 *pachyderma* $\delta^{18}\text{O}$ ranges from 0.2 to 4.4 ‰, with higher values below 11 cm. On the other
264 hand, the *M. barleeanus* $\delta^{18}\text{O}$ ranges from 3.9 to 4.3 ‰, with no trend with increasing
265 sediment depth. The *C. neoteretis* $\delta^{18}\text{O}$ varies from 4.6 ‰ to 5.0 ‰ and shows a slight

266 increase with increasing sediment depth. In core V-16, relatively higher $\delta^{18}\text{O}$ values were
267 measured in *N. pachyderma* and *M. barleeanus* in samples close to the sulfate methane
268 transition (SMT). Below this depth, the $\delta^{18}\text{O}$ values decrease only slightly. The *N. pachyderma*
269 $\delta^{18}\text{O}$ ranges from 2.3 to 4.2 ‰, whereas the range of *M. barleeanus* $\delta^{18}\text{O}$ values is 4.0 to 4.6
270 ‰.

271 For cores V-8, V-16, and V-20, isotopic measurements were conducted on planktonic
272 and benthic species from the size fractions $>63\ \mu\text{m}$ and $>125\ \mu\text{m}$. Our results do not show a
273 size fraction effect on the $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ data. Specifically, a difference of less than 3 ‰ and 6
274 ‰ difference for $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$, respectively, is measured between the two size fractions
275 analyzed. The only exception is represented by *M. barleeanus* samples, for which a slight
276 difference between the fractions >63 and $>125\ \mu\text{m}$ is observed for the core V-16 $\delta^{13}\text{C}$ record.

277 Additional analyses were conducted on samples from all the other cores collected at
278 Vestnesa Ridge, Storfjordrenna, and the LV canyons (Figs. 1 and 6; Table 4). In superficial
279 samples (0-1 cm; Fig. 6A), planktonic foraminiferal $\delta^{18}\text{O}$ values are heavier in the GHM
280 Storfjordrenna samples (3-4 ‰) compared to the Vestnesa and LV ones (< 3 ‰). On the
281 contrary, *M. barleeanus* $\delta^{18}\text{O}$ values are heavier in cores from Vestnesa Ridge (> 4 ‰ in most
282 of the cores) than in cores from the other sampled areas. The range of $\delta^{18}\text{O}$ values is
283 significantly variable in these samples, with planktonic $\delta^{18}\text{O}$ values of 1.0 – 4.5 ‰ and
284 benthic $\delta^{18}\text{O}$ values of 3.7 – 5.6 ‰. The isotopic signal recorded on living individuals (*C.*
285 *wuellerstorfi*, *C. neoteretis* and *M. barleeanus*) from Vestnesa Ridge and GHM
286 Storfjordrenna cores does not clearly differ from the values measured in dead foraminifera
287 from the same samples (living-dead difference of 0.6 ‰ for $\delta^{13}\text{C}$ and 0.5 ‰ for $\delta^{18}\text{O}$).

288 We note that lower $\delta^{18}\text{O}$ values are measured in cores from the LV sites, where
289 heavier benthic (*T. earlandi*) and planktonic (*N. pachyderma*) $\delta^{18}\text{O}$ values (2.5 – 3 ‰)
290 coincide to more negative $\delta^{13}\text{C}$ values (down to -34.1 ‰). This relationship seems to be
291 present also in the deeper (3-4 cm) samples from Vestnesa Ridge (Fig. 6B), where heavier
292 benthic and planktonic foraminiferal $\delta^{18}\text{O}$ (4.5 – 5.5 ‰) coincide to $\delta^{13}\text{C}$ values of -11.6 ‰
293 (*C. neoteretis*) and -15.2 ‰ (*N. pachyderma*). In cores V-8, V-16, and V-20, a similar
294 correspondence between higher $\delta^{18}\text{O}$ values and lower $\delta^{13}\text{C}$ values is present as well (Fig. 5).

295

296 **4.3 Foraminiferal microscopy and spectroscopy analyses**

297 Scanning electron microscopy and EDS investigations revealed pristine shells (shells
298 not affected by diagenesis) in superficial samples (0-1 cm) from Vestnesa Ridge and

299 Storfjordrenna (i.e, *C. neoteretis*, *M. barleeanus*, *N. labradorica*, and *N. pachyderma*; Figs. 7
300 A, E, G, and I). Altered shells (shells characterized by carbonate secondary overgrowth) are
301 identified from the 3-4 cm interval of samples collected at Vestnesa Ridge (*C. neoteretis*, *M.*
302 *barleeanus*, *N. labradorica*, and *N. pachyderma*; Figs. 7 B, C, D, F, H, and J). Unexpectedly,
303 we found the presence of diagenetic alterations on foraminifera shells in superficial samples
304 (0-1 cm) from cores collected at the LV canyons (*N. pachyderma* and *T. earlandi*; Figs. 7 K
305 and L).

306 The EDS semi-quantitative analysis show low Mg concentrations in pristine shells
307 (Figs. 7 A, E, and G), even if the EDS map of *N. pachyderma* from core V-8 (0-1 cm; Fig. 7I)
308 reveals a region of high Mg content. Compared to pristine shells, shells affected by diagenesis
309 are characterized by higher Mg concentrations (Figs. 7B, C, D, F, H, and J), with the
310 exception of the superficial samples from the LV canyons where spots of high Ba, rather than
311 Mg, were observed (Figs. 7K and L).

312

313 **5. Discussion and conclusions**

314 **5.1 Geochemical characteristics of gas hydrate and methane-rich sediments**

315 Regions of the seafloor with significant methane are characterized by opposing
316 gradients of pore water sulfate and methane that intersect at a SMT. This is because upward
317 migrating methane can react with sulfate through microbially-mediated anaerobic oxidation of
318 methane (AOM; e.g., Boetius et al., 2000). At the broad scale, the depth of the SMT, though
319 affected by numerous factors, mostly relates to the flux of upward methane (Bhatnagar et al.,
320 2008).

321 The sharp decrease in sulfate concentration with sediment depth suggests that the SMT
322 is within the first 10 cm in most cores from Vestnesa Ridge except for the inactive, non-seep
323 control core (Fig. 2). The SMT is close to the seafloor in Vestnesa cores V-7, V-8, V-9 and V-
324 16 (less than 10 cm) indicating high methane fluxes putatively advective (Yao et al., 2019).
325 On the contrary, the reference core V-20 displays a sulfate profile typical for marine
326 environments, with no methane influence (Borowski et al., 1996). At Storfjordrenna GHM1,
327 sulfate profiles indicate a deeper SMT (tens of cm), with several cores unable to penetrate the
328 SMT (Fig. 3). In the LV canyons (Fig. 4), the SMT seems close to the seafloor (~5 cm) in the
329 southern canyon, with the exception of core L-12, deeper (10-15 cm) in the northern canyon
330 (core L-52).

331 The AOM reaction produces hydrogen sulfide and bicarbonate, the latter which
332 comprises most of DIC in marine settings. Universally, the DIC in sediment pore waters is

333 depleted in ^{13}C around SMT (e.g., Torres et al., 2003; Ussler et al., 2008), due to both the ^{13}C -
334 depleted methane as the carbon source and isotopic fractionation associated with this
335 microbial process (Borowski et al., 1997; Hong et al., 2013; Yoshinaga et al., 2014). The
336 actual $\delta^{13}\text{C}$ value of DIC at the SMT is complicated. Methane can derive from thermogenic
337 ($\delta^{13}\text{C}$: -55 ‰ to -40 ‰) or microbial ($\delta^{13}\text{C}$: -100 ‰ to -55 ‰) sources (Whiticar, 1999); DIC
338 enriched in ^{13}C and formed during methanogenesis can also migrate upward (Chatterjee et al.,
339 2011).

340 The $\delta^{13}\text{C}_{\text{DIC}}$ profiles (Figs. 2-4) reveal depleted values at cores V-7, V-8, V-9, V-16,
341 V-17, V-18, and V-21 from Vestnesa Ridge; cores 900, 902, 917, 918, and 919 from
342 Storfjordrenna, and cores L-9, L-12, and L-30 from the LV canyons. Core V-20 is the only
343 core at Vestnesa Ridge that records the $\delta^{13}\text{C}_{\text{DIC}}$ values similar to modern normal marine
344 environment (-1 to 1‰; Tagliabue and Bopp, 2008), confirming that this core is not affected
345 by methane seepage and oxidation. Core 898 also shows $\delta^{13}\text{C}_{\text{DIC}}$ values similar to the modern
346 normal marine environment. In all other cores investigated, the negative $\delta^{13}\text{C}_{\text{DIC}}$ values
347 suggest that cored sites are influenced by upward methane fluxes (e.g., Whiticar et al., 1999).
348 In GHM cores, we cannot exclude a signal reflecting organoclastic sulfate reduction, which
349 occurs between the seafloor and the SMT and is characterized by depleted but greater $\delta^{13}\text{C}_{\text{DIC}}$
350 than AOM (Chatterjee et al., 2011).

351 The chloride profiles available from Vestnesa Ridge and Storfjordrenna GHM1 (Figs.
352 2 and 3) do not vary with sediment depth, suggesting that the sampling sites are not affected
353 by fluid migration of a low-chloride fluid through the sediment column.

354

355 **5.2 Foraminiferal evidences of gas hydrate emission**

356 **5.2.1 Foraminiferal stable isotope compositions**

357 Overall, the isotopic compositions of Rose Bengal stained ('living') foraminifera do
358 not substantially differ from other values reported in literature. In fact, specimens of *C.*
359 *neotretis* and *N. pachyderma* from the top of the control core V-20 (0-1 cm; Fig. 5), which has
360 no evidence for upward methane migration, exhibit values consistent with other studies in the
361 region (e.g., Consolaro et al., 2015; Schneider et al., 2018). There is also a clear distinction
362 between planktonic and benthic $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values. Vital effects and ecological preferences
363 (e.g., microhabitat), but also environmental parameters (e.g., temperature, pH), all influence
364 the isotopic composition of the foraminiferal shells (Duplessy et al., 1970; Bemis et al., 1998;
365 Ravelo and Hillaire Marcel, 1999; Barras et al., 2010). In this context, we interpret the offset

366 between the $\delta^{18}\text{O}$ of *C. neoteretis* and *N. pachyderma* as a consequence of different habitats for
367 these species (e.g., Kennett et al., 2000; Consolaro et al., 2015). Ecological preferences
368 (Murray, 2006) might also explain the small differences in stable isotope compositions
369 between *C. neoteretis* (a shallow infaunal species) and *M. barleeanus* (an intermediate
370 infaunal species). Notably, the former has higher $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$.

371 However, of all foraminifera samples examined, the most depleted $\delta^{13}\text{C}$ value (-5.7 ‰)
372 was measured in Rose Bengal stained *N. labradorica*, a deep infaunal species (Racine et al.,
373 2018). We note that the depleted $\delta^{13}\text{C}$ value we measured in this species is similar to the $\delta^{13}\text{C}$
374 signature measured in another living deep infaunal foraminifer from Monterey Bay
375 (*Globobulimina pacifica*; -6 ‰; Bernhard et al., 2010). We believe that these $\delta^{13}\text{C}$ values
376 represent a ‘threshold’ of the isotopic composition of non-diagenetically altered foraminifera
377 inhabiting seep sites. It might be possible that the depleted $\delta^{13}\text{C}$ measured in our living *N.*
378 *labradorica* is a consequence of vital effects. However, considering the sampling location, we
379 think that the *N. labradorica* $\delta^{13}\text{C}$ is the result of the incorporation of methane derived ^{13}C -
380 depleted carbon during shell formation and likely ingestion of ^{13}C -depleted methanotrophic
381 microbes (Rathburn et al., 2003; Panieri, 2006; Bernhard and Panieri, 2018).

382 In gas hydrate bearing sediments, the $\delta^{18}\text{O}$ of pore water can be affected by gas
383 hydrate formation and dissociation because the water trapped in gas hydrates is more enriched
384 in ^{18}O compared to the adjacent pore water (Davidson et al., 1983; Tomaru et al., 2006; Ijiri et
385 al., 2018). With the data available, we cannot draw a firm conclusion regarding the possibility
386 that the $\delta^{18}\text{O}$ signature of living benthic foraminifera can record episodes of gas hydrate
387 dissociation. Our data indicate that living foraminifera cannot record gas hydrate dissociation
388 as shell formation is a discontinuous and brief process that might not be coeval with episodes
389 of gas hydrate decomposition, which are also transient in time (Thatcher et al., 2013;
390 Anderson et al., 2014). It might be equally possible that the $\delta^{18}\text{O}$ signature of living benthic
391 foraminifera does not reflect the influence of gas hydrate dissociation, because no dissociation
392 happened during the life span of the specimens analyzed. However, a different interpretation
393 can be drawn when considering foraminifera with diagenetic overgrowth.

394 Over relatively long time scales (Plio-Pleistocene), gas hydrate dissociation can alter
395 the carbonate system and the MDAC isotopic signature (Bohrmann et al., 1998; Crémière et
396 al., 2016), as revealed by a recent study showing a disequilibrium between carbonate crust
397 clumped isotopes and the expected temperature of formation (Lloyd et al., 2016). In cores V-8
398 and V-16, the slight downcore increase in the *C. neoteretis* and *M. barleeanus* $\delta^{18}\text{O}$ (up to 5

399 ‰) values compared to the reference-like values of core V-20 suggest the possible influence
400 of gas hydrate dissociation on the $\delta^{18}\text{O}$ signature of fossil foraminifera through sedimentary
401 overgrowth. The variability in the *N. pachyderma* $\delta^{18}\text{O}$ values (1.8 to 4.5 ‰) from cores V-8
402 and V-16 supports this hypothesis.

403 It was proposed that the planktonic foraminiferal isotopic composition can be
404 influenced by hydrate dissociation (Maslin et al., 2005). However, in this work, the
405 precipitation of authigenic carbonate around the foraminiferal shells was not discussed. Cores
406 V-8 and V-16 are located at ~1,200 m water depth. Thus, methane seepage at these locations
407 is rapidly dispersed in the ocean or microbially oxidized to CO_2 in the water column (Damn et
408 al., 2005; Steinle et al., 2015), making it improbable that living planktonic foraminifera can
409 record episodes of gas hydrate dissociation and methane oxidation (e.g., Consolaro et al.,
410 2018). However, evidence of gas hydrate dissociation can be recorded after the death and
411 burial of planktonic species, in particular at the depth of the SMT, where MDAC can
412 precipitate on the foraminiferal shells (Panieri et al., 2016; Schneider et al., 2018).

413 In this study, the wide range of $\delta^{18}\text{O}$ values measured in both planktonic and benthic
414 foraminiferal shells confirms that the isotopic composition of *N. pachyderma* and *C.*
415 *neoteretis* is a consequence of the precipitation of ^{18}O -rich authigenic carbonates (see also
416 next section). Our conclusion is in agreement with other studies conducted at different
417 geographic locations (Fig. 6). For example, Torres et al. (2003) estimated that the range of
418 $\delta^{18}\text{O}$ values (0-1.75 ‰) measured on foraminiferal calcite from the Hydrate Ridge was due to
419 22 wt.% (authigenic) carbonates precipitated around the shells. Heavier foraminiferal $\delta^{18}\text{O}$
420 (range from 2 to 4 ‰) were reported also for dead foraminifera from methane vents
421 environments in the Gulf of California (Herguera et al., 2014). Also in this case, the
422 foraminiferal isotopic composition was interpreted to be influenced by authigenic carbonates.
423 Those results are in agreement with the range of data measured in our study areas (Fig. 6A).
424 At Vestnesa Ridge, the identification of authigenic carbonate precipitated on foraminiferal
425 tests from deeper sediments (several meters of sediment depth) points out the same depleted
426 $\delta^{13}\text{C}$ and heavy $\delta^{18}\text{O}$ (Schneider et al., 2017; Fig. 6B).

427 Secondary overgrowth affected planktonic and benthic foraminifera alike, even if the
428 ^{18}O enrichment is more evident in *N. pachyderma* because of the lower $\delta^{18}\text{O}$ values recorded
429 by this species in marine environments not affected by methane (e.g. core V-20). In addition,
430 similar $\delta^{18}\text{O}$ enrichments to the ones we measured in foraminifera were reported for MDAC

431 from gas hydrate and cold seeps settings (Greinert et al., 2001, 2010; Eichhubl and Boles,
432 1998; Eichhubl et al., 2000; Naehr et al., 2007, 2009; Crémière et al., 2016).

433 Gas hydrate dissociation, and consequent release of ^{18}O -enriched water is a process
434 that takes place below the SMT, while ^{13}C -depleted methane is present within the SMT. This
435 would suggest that gas hydrate dissociation might not be associated with foraminiferal low
436 $\delta^{13}\text{C}$ and high $\delta^{18}\text{O}$ values. However, our results clearly show a relationship between depleted
437 $\delta^{13}\text{C}$ and enriched $\delta^{18}\text{O}$ values in foraminifera, in particular in cores V-8 and V-16, collected
438 at gas hydrate bearing sediments where the SMT is close to the seafloor (Fig. 5).

439 We recognize that the foraminiferal $\delta^{18}\text{O}$ can vary because of changes in
440 environmental parameters that are unrelated to gas hydrate dissociation. However, we do not
441 think that this is the case for the samples analyzed in this study for several reasons. First, we
442 observe a wide range of $\delta^{18}\text{O}$ values on both planktonic and benthic foraminifera even within
443 the first few centimeters of sediment (Figs. 5 and 6). At Vestnesa Ridge, this difference in
444 $\delta^{18}\text{O}$ values would translate in several degrees C difference in less than a millennium, if we
445 take into account the modern sedimentation rate at this location (~ 19 cm/kyrs; Consolaro et
446 al., 2015). We think that such a big temperature change in a short time frame is highly
447 unlikely. In fact, this change would be more abrupt than temperature shifts characterizing the
448 Heinrich events over the Pliocene (Cortijo et al., 1997). We note that a similar variability in
449 the foraminiferal $\delta^{18}\text{O}$ as recorded in the Vestnesa Ridge samples was measured in surface
450 sediments from the LV canyons (0-1 cm; Fig. 6A), confirming that a temperature change
451 cannot be a likely explanation of our data, within the same 1 cm-sediment layer.

452 Second, similar foraminiferal $\delta^{18}\text{O}$ ranges from cold seep sites as the ones recorded in
453 surface samples at Vestnesa Ridge (Fig. 6A) have not often been reported in literature, with a
454 few exceptions possible as a consequence of temperature changes among the sites analyzed
455 (e.g., 2 ‰ in Sen Gupta and Aharon, 1994; 1.35 ‰ in Burkett et al., 2018). Burkett et al.
456 (2018) observed a maximum $\delta^{18}\text{O}$ increase of +0.7 ‰ in cold seeps that they interpreted to be
457 the consequence of fluids impact. Considering our stable Chloride profiles together with the
458 heat flow estimated in the Vestnesa Ridge, leading to heat excess of less than 0.04°C at 50 cm
459 sediment depth (Bohrmann et al., 2016), we exclude this hypothesis.

460 Third, clay dehydration through the smectite transformation into illite can cause a ^{18}O
461 enrichment of pore waters. This diagenetic process occurs in deep sediments at temperature
462 ranging from 60° to 160° C (~ 1000 m below sea floor). However, the Chloride profiles at
463 Vestnesa Ridge and Storfjordrenna GHM do not support any clay dehydration, as observed

464 also in mud volcano systems (Hensen et al., 2004). Yet, we cannot exclude an impact of
465 meteoric fluids in the LV canyons, as documented by Hong et al. (2019), with a different $\delta^{18}\text{O}$
466 signature, even though the correlation between depleted $\delta^{13}\text{C}$ and heavy $\delta^{18}\text{O}$ points to the
467 presence of authigenic carbonate on foraminiferal shells. Consequently, we do not think that
468 clay dehydration, temperature, or salinity changes can affect the pore water of surface
469 sediments in our three study areas.

470 Finally, the size fraction analyzed might also cause biases in the isotopic records used
471 to interpret short time-scale processes because the isotopic composition of large foraminifera
472 ($>150\ \mu\text{m}$) can reflect a longer-term averaged calcification process that is not influenced by
473 episodic processes (i.e. eutrophic periods or seasonal changes; Fontanier et al., 2006). In
474 addition, ontogenic effects are reported in literature for both planktonic (e.g., Spero et al.,
475 1997; Elderfield et al., 2002) and benthic (e.g., Friedrich et al., 2006; Barras et al., 2010;
476 Schumacher et al., 2010) foraminifera. The analysis of different size fractions (150, 250, 350,
477 450, 550, 650 and $750\ \mu\text{m}$) by Corliss et al. (2002) did not reveal a seasonal influence on the
478 foraminiferal isotopic record, suggesting the absence of an ontogenetic effect on the
479 foraminiferal isotope incorporation. The foraminiferal isotope data we collected using
480 different size fractions (i.e., >63 and $>125\ \mu\text{m}$) are in agreement with the conclusions reached
481 by Corliss et al. (2002), demonstrating that the benthic and planktonic foraminiferal $\delta^{18}\text{O}$
482 values are very similar in both fractions analyzed and that they are not biased by ontogenetic
483 effects.

484

485 **5.2.2 Foraminiferal secondary overgrowth**

486 Our SEM analyses revealed significant visual diagenetic alteration on several of the
487 foraminiferal shells analyzed, but not on all of them. Living individuals of *N. labradorica* and
488 *M. barleeanus* from Storfjordrenna GHM1 (Figs. 7 E and G) were not affected by diagenetic
489 alteration, even if we measured relatively low $\delta^{13}\text{C}$ in these samples. This suggest that the
490 depleted $\delta^{13}\text{C}$ signature of these samples was incorporated in the primary foraminiferal
491 calcite (calcite deposited by the foraminifera during shell growth), as proposed in other
492 studies (e.g., Panieri et al., 2017a, Schneider et al., 2017; Wan et al., 2018).

493 Diagenetic features were confirmed by EDS maps of samples collected deeper in the
494 sediment column. In particular, we interpret the high Mg concentration to be the evidence of
495 the presence of secondary overgrowth on the shells analyzed (e.g., Panieri et al., 2017a;
496 Schneider et al., 2017). In the 3-4 cm sediment horizon, diagenetic alterations were observed

497 in different Vestnesa pockmarks (*C. neoteretis* from cores V-8 and V-16; Fig. 7 B and C),
498 suggesting that precipitation of MDAC on foraminiferal shells occurs at a regional scale. The
499 *C. neoteretis* collected at deeper sediment depths (core V-8, 29-30 cm; Fig. 7D) is
500 characterized by the highest Mg content.

501 The samples from the LV canyon south did not reveal high concentrations of Mg, but
502 an enrichment in Ba (Fig. 7 K and L). Ba-calcite has been described as a product of early
503 diagenesis (Dejonghe and Boulvain, 1993; Schroeder et al., 1997), which can affect
504 foraminiferal carbonate (Lea and Boyle, 1993). Barite in seafloor crusts as well as high
505 dissolved Ba concentration have been described in the LV area (Sen et al., 2019; Hong et al.,
506 2019) and EDS analysis of foraminiferal shells show an initial possible precipitation of
507 authigenic Ba-rich overgrowth. Further investigations are needed to understand if these
508 overgrowths can impact the foraminiferal isotopic composition in samples from the LV
509 canyons and other geographic locations. Overall, our microscopy and spectroscopy analyses
510 confirm the hypothesis that the isotopic composition measured in fossil foraminifera is
511 affected by secondary overgrowth precipitated on the foraminiferal shells at the SMT and that
512 this signal is related to methane oxidation and gas hydrate dissociation.

513 Nevertheless, isotopic composition of fossil foraminifera might also depend on the
514 secondary overgrowth mineralogy. In fact, the mineralogy of MDAC usually include
515 microcrystalline Mg-calcite, aragonite, and dolomite (e.g., Roberts and Aharon, 1994,
516 Bohrmann et al., 1998, Crémière et al., 2012; Sauer et al., 2017). Different types of carbonate
517 have different oxygen isotopic fractionation (e.g., Kim et al., 2007; Vasconcelos et al., 2005).
518 For example, dolomite is estimated to show an enrichment of 5 to 7 ‰ of ^{18}O compared to
519 synthetic calcite (O'Neil and Epstein, 1966). At methane cold seeps, a wide range of $\delta^{18}\text{O}$ has
520 been related to the mineralogy of authigenic carbonates (e.g., Orphan et al., 2004; Gieskes et
521 al., 2005), with $\delta^{18}\text{O}$ values up to 7.5 ‰ in high Mg-calcite, while aragonite displayed lighter
522 $\delta^{18}\text{O}$.

523 The ^{18}O enrichment in authigenic carbonate rocks has been interpreted as evidence for
524 gas hydrate dissociation at several methane seep and non-seep environments (e.g., Bohrmann
525 et al., 1998; Aloisi et al., 2000; Pierre et al., 2002; Conti et al., 2004). At these sites, the heavy
526 $\delta^{18}\text{O}$ of Mg-calcite was used as an evidence for gas hydrate dissociation, whereas the origin of
527 light aragonite $\delta^{18}\text{O}$ remains uncertain. It is possible that this difference is a consequence of
528 Mg-calcite and aragonite precipitating from two highly distinct pore-water conditions
529 (Bohrmann et al., 1998), even if the chemical controls on the carbonate phase that precipitates

530 are still not completely understood (Burton, 1993). Schneider et al. (2017) estimated that the
531 authigenic carbonate proportion on foraminiferal tests can be up to 58 %. A calcite containing
532 50 % of Mg-CaCO₃ is expected to be enriched in ¹⁸O by 3-4 ‰ (Tarutani et al., 1969; Fritz
533 and Smith, 1970).

534 Finally, in this study, we demonstrate that at methane seeps and gas hydrate bearing
535 sediments fossil planktonic and benthic foraminifera are characterized by relatively high δ¹⁸O
536 values. We propose that this ¹⁸O enrichment is related to past gas hydrate dissociation
537 event(s). Hydrate dissociation release ¹⁸O-enriched water, which affect the pore water isotopic
538 signature. This signal can be incorporated by authigenic carbonates precipitating around
539 foraminiferal shells at the SMT. In addition, we speculate that mineralogy of the secondary
540 overgrowth might also influence the isotopic signal measured in fossil foraminifera.

541

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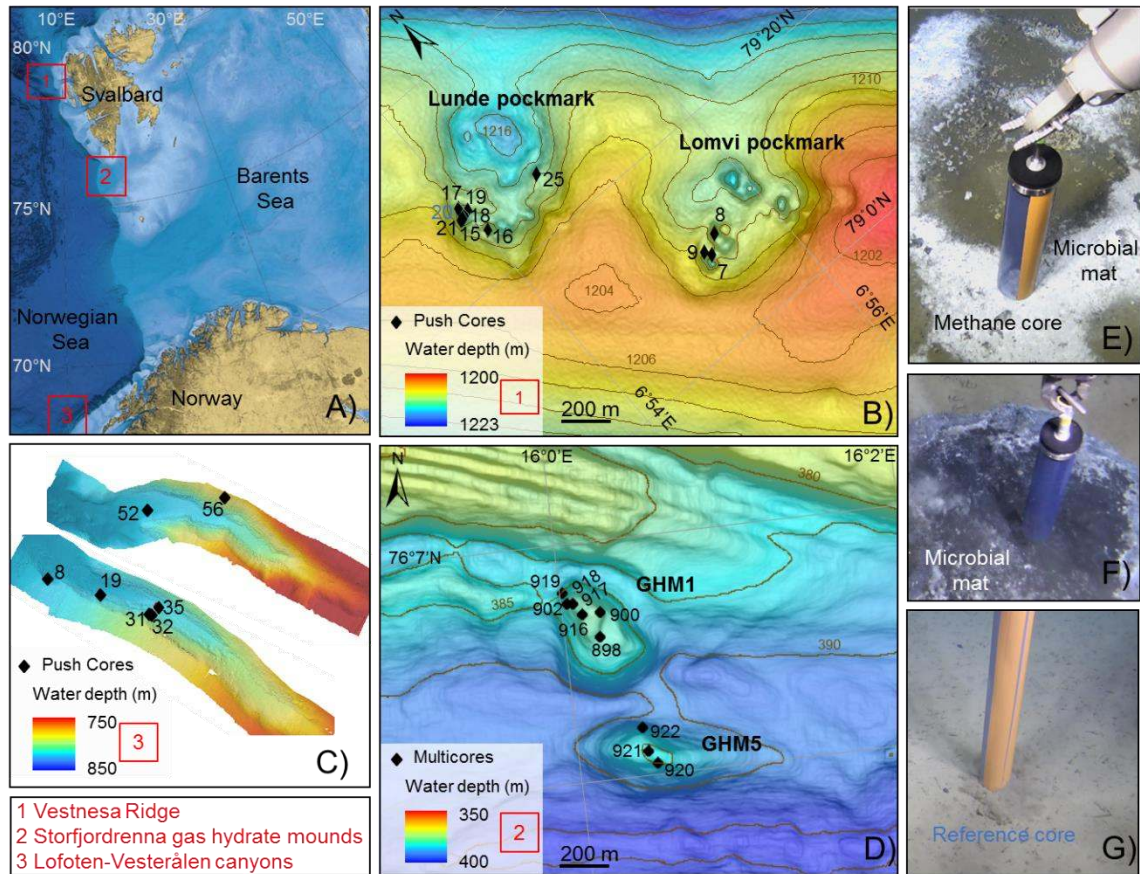
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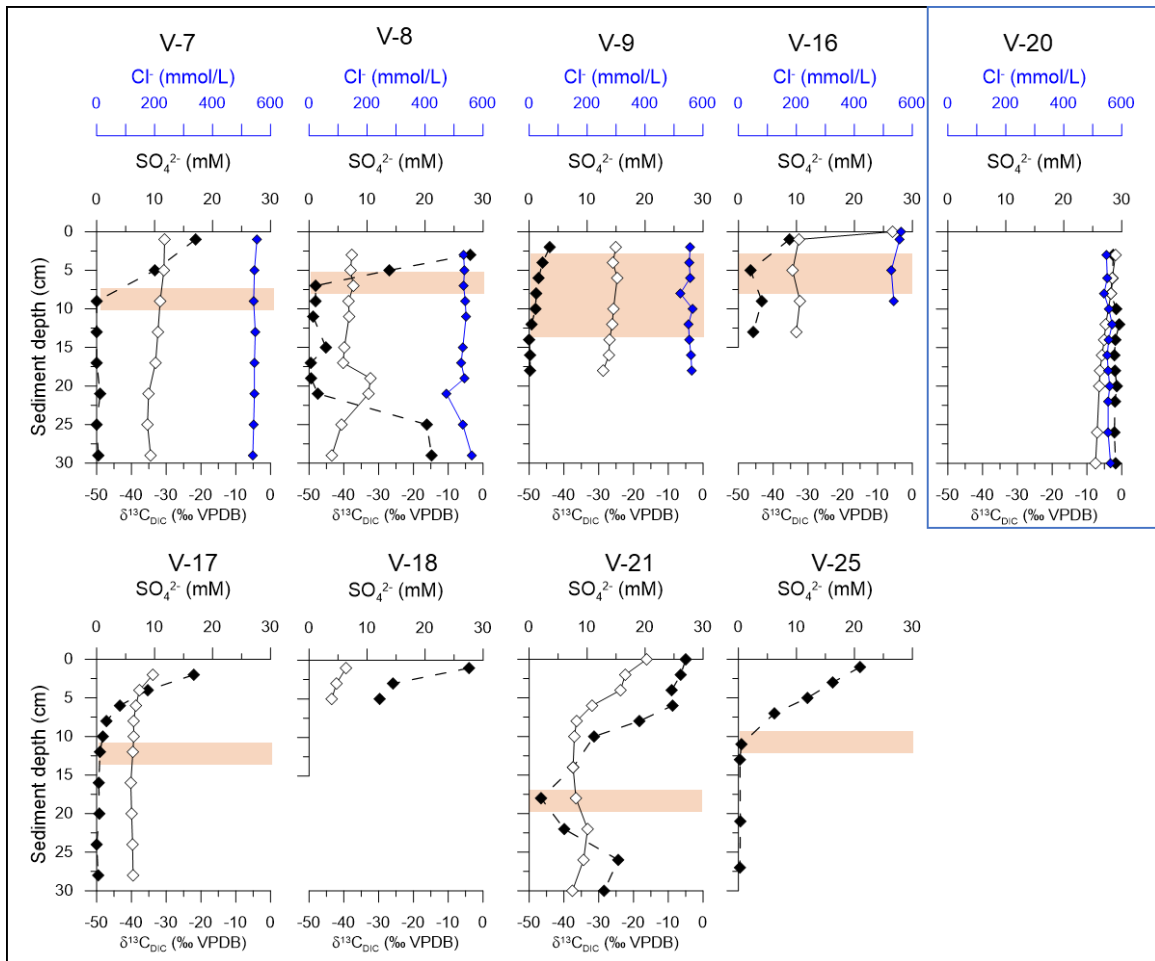
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Figure 1. A) Map of the study area with sample areas identified by red squares. B) Location of the push cores collected within two pockmarks (Lunde and Lomvi) on Vestnesa Ridge. C) Location of push cores collected within the Lofoten-Vesterålen canyon region (Canyons North and South). D) Location of multicores collected in Storfjordrenna (GHMs 1 and 5). E) and F) Examples of push cores collected on microbial mats covering methane-rich sediments (i.e., cores V-8 and V-16, respectively) and G) Example of a push core collected outside a microbial mat (i.e., core V-20).

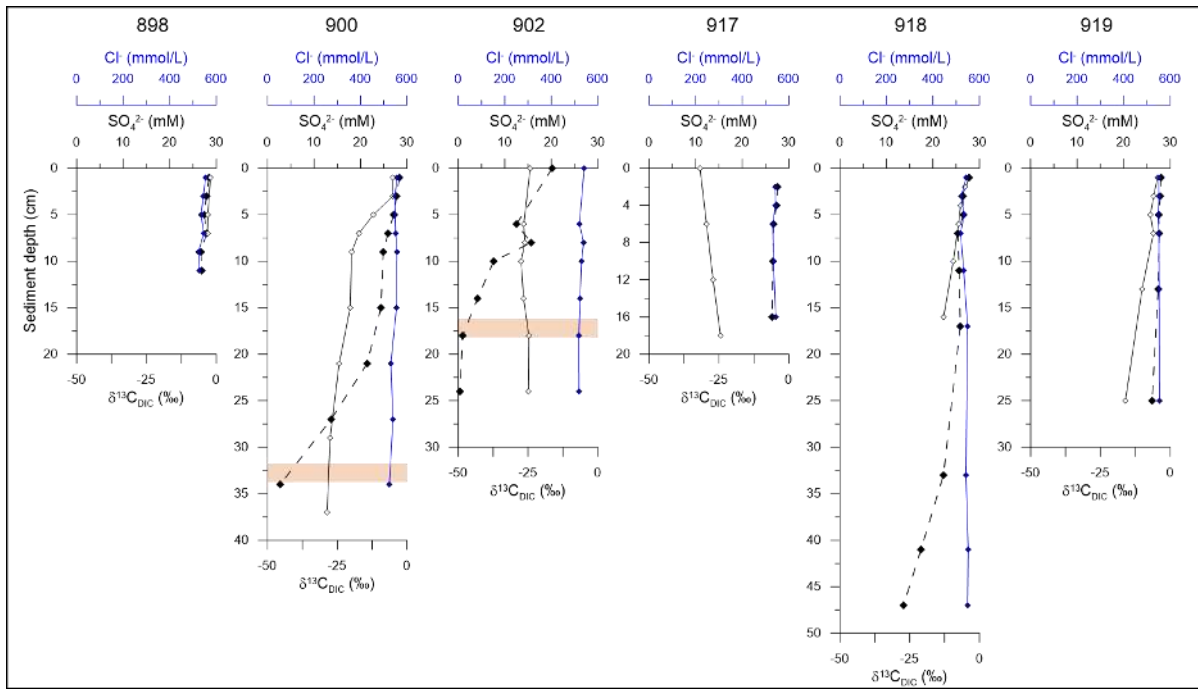


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932 **Figure 2.** Pore water sulfate (SO_4^{2-} , black), $\delta^{13}\text{C}_{\text{DIC}}$ (white), and Chloride (Cl^- , blue) profiles
 933 of push cores collected at Vestnesa Ridge. The numbers at the top of each graph identify the
 934 stations; the blue square indicates the core sampled outside a microbial mat (control core).

935 The estimated sulfate-methane transition is in orange.

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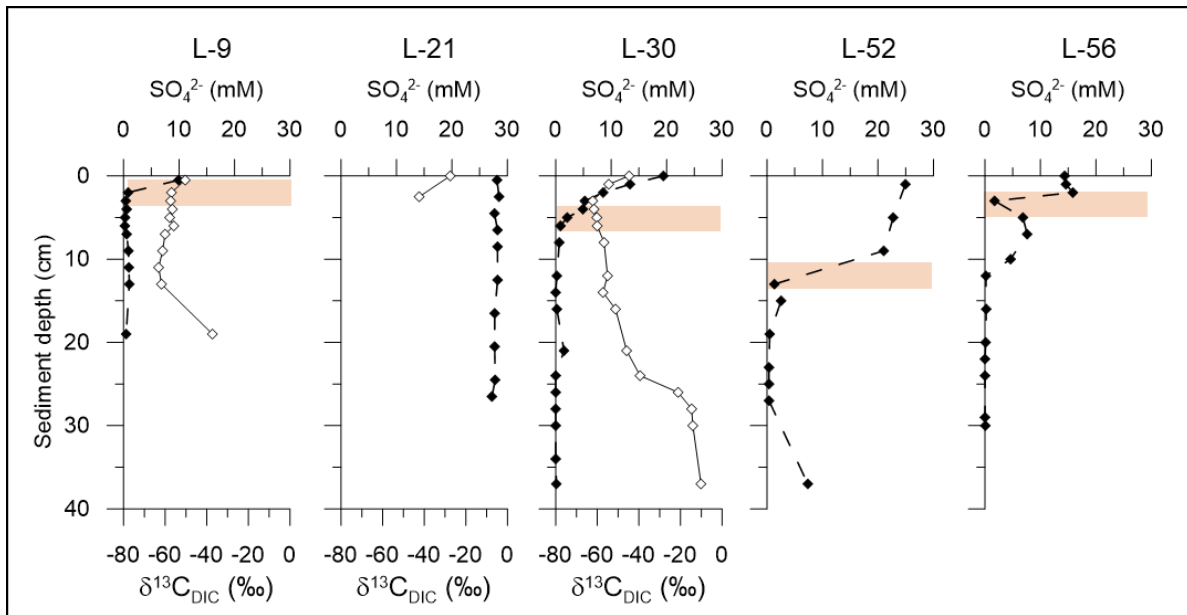
938 **Figure 3.** Pore water sulfate (SO₄²⁻, black), δ¹³C_{DIC} (white), and Chloride (Cl⁻, blue) profiles

939 of multicores collected at Storfjordrenna GHM1. The numbers at the top of each graph

940 identify the stations; the estimated sulfate-methane transition is in orange.

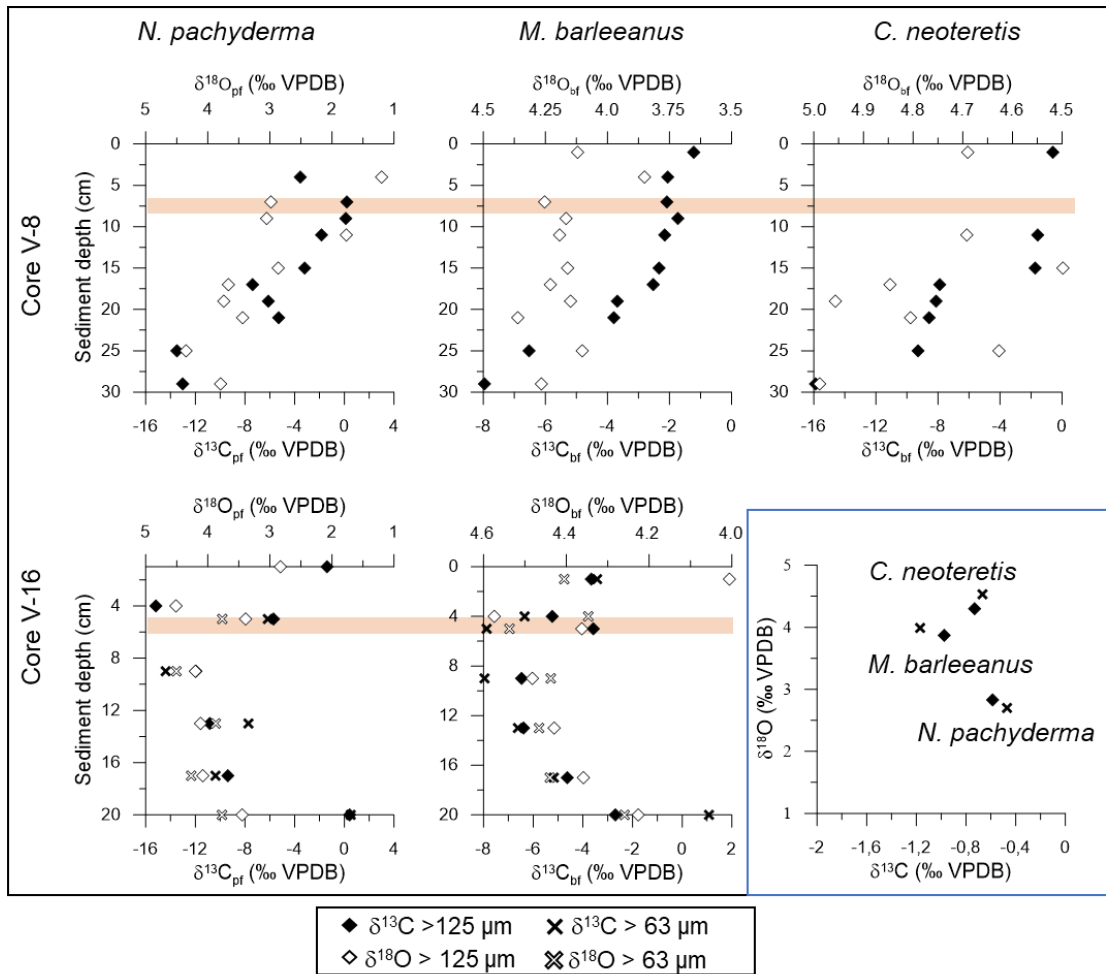
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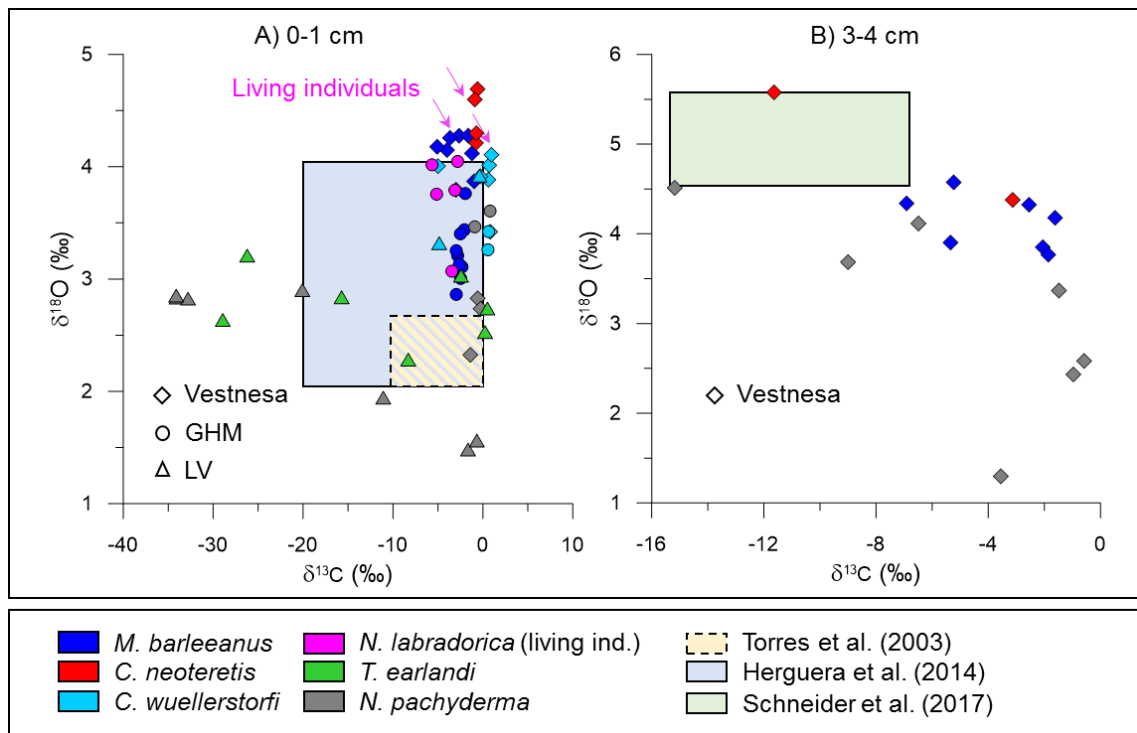
945 **Figure 4.** Pore water sulfate (SO_4^{2-} , black) and $\delta^{13}\text{C}_{\text{DIC}}$ (white) profiles of push cores
 946 collected at the Lofoten-Vesterålen canyons. The numbers at the top of each graph identify
 947 the stations; the estimated sulfate-methane transition is in orange.
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950 **Figure 5.** Cores V-8 and V-16 (Vestnesa Ridge) planktonic (pf) and benthic (bf) foraminiferal
 951 isotopes ($\delta^{13}C$ and $\delta^{18}O$) data. The blue square indicates the values measured on the surface
 952 sample (0-1 cm) of core V-20. Analyses were performed on two benthic (*C. neoteretis* and *M.*
 953 *barleeanus*) and one planktonic (*N. pachyderma*) foraminiferal species. Diamonds are used to
 954 identify data collected analyzing the fraction $>125 \mu m$. Crosses are used to identify data
 955 collected analyzing the fraction $>63 \mu m$. VPDB = Vienna Pee Dee Belemnite.

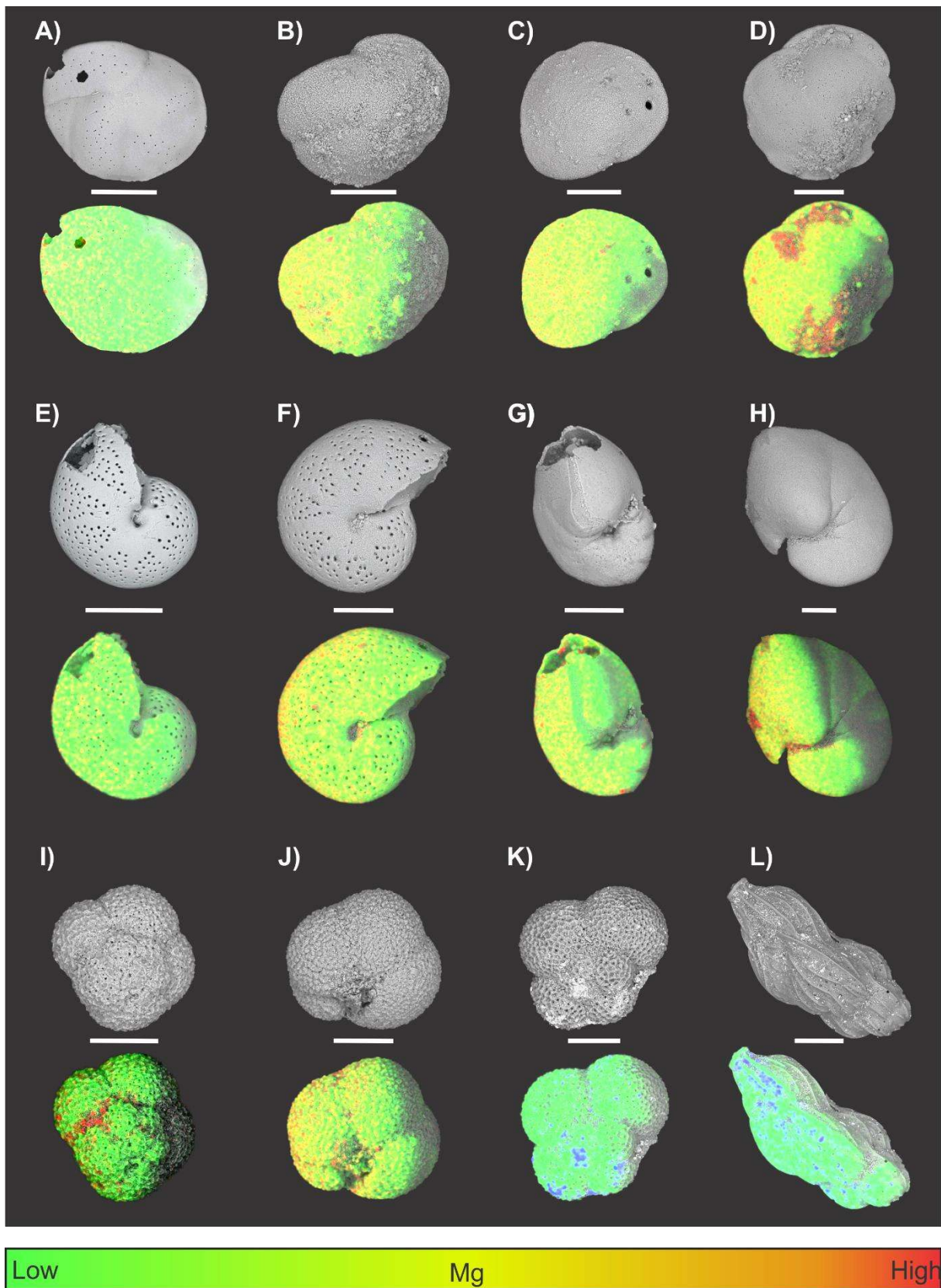
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958 **Figure 6.** Foraminiferal stable isotopes ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) measured on benthic (colored
 959 symbols) and planktonic (grey symbols) species. A) Data collected from surface samples (0-1
 960 cm) of cores from Vestnesa Ridge, Storffjordrenna (GHM), and the Lofoten-Vesterålen
 961 Canyons. Data from Rose Bengal stained ('living') individuals are identified by pink arrows.
 962 B) Data collected from the interval 3-4 cm of cores collected at Vestnesa Ridge. Colored
 963 areas in (A) show the range of isotopic values of benthic foraminifera from methane seeps and
 964 gas hydrate-rich sediments from different geographic locations (Hydrate Ridge (in yellow)
 965 and Gulf of California (in blue); Torres et al., 2003, Herguera et al., 2014). All the
 966 foraminifera analyzed were picked from superficial sediments (0-5 cm) and were affected by
 967 secondary overgrowth. The green square in panel (B) shows the range of values measured on
 968 benthic foraminifera from Vestnesa Ridge (Schneider et al., 2017). The foraminifera analyzed
 969 were picked deeper in the sediment column (meters below the seafloor) and were affected by
 970 secondary overgrowth. VPDB = Vienna Pee Dee Belemnite.

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Figure 7. Backscatter-scanning electron microscopy images and corresponding energy-dispersive x-ray spectroscopy (EDS) maps of selected foraminiferal shells. *Cassidulina*

975 *neoteretis* from core V-16 0-1 cm (A) and 3-4 cm (B), and core V-8, 3-4 cm (C) and 29-30 cm
976 (D). *Melonis barleeanus* from core GHM1 0-1 cm (Rose Bengal stained specimen) (E), and
977 core V-8 29-30 cm (F). *Nonionella labradorica* from core GHM1 0-1 cm (Rose Bengal
978 stained specimen) (G), and core V-8 29-30 cm (H). *Neogloboquadrina pachyderma* from core
979 V-8 0-1 cm (I), core V-8 29-30 cm (J), and core L-8 0-1 cm (K). *Trifarina earlandi* from
980 Lofoten-Vesterålen L-19 0-1 cm (L). Scale bars are 100 μ m. Grey areas on EDS maps
981 represent portions of the shells where the analysis was not possible, due to the angle of the
982 detector and shell morphology.

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985 **Table 1.** Summary of the cores collected at Vestnesa Ridge (Lomvi and Lunde),
 986 Storfjordrenna (GHMs 1 and 5), and the Lofoten-Vesterålen canyons (Canyons South and
 987 North) and used in this study. The analyses performed on each core are specified. Chloride
 988 data are not available for the LV Canyons cores. The data collected are reported in
 989 Supplementary Tables 1, 2 and 3. Nd = no data.
 990

Core reference	name	Location	latitude °N	longitude °E	water depth (m)	pore water SO ₄ ²⁻ , δ ¹³ C _{DIC} , Cl ⁻ (cm)	foraminifera stable isotopes (cm)
P1606-007	V-7	Lomvi	79.0023	6.225	1204	0-30	0-5
P1606-008	V-8	Lomvi	79.0027	6.9248	1208	0-30	0-30
P1606-009	V-9	Lomvi	79.0025	6.922	1205	0-20	0-5
P1606-015	V-15	Lunde	79.0076	6.9003	1208	0-30	0-2
P1606-016	V-16	Lunde	79.0068	6.9006	1206	0-15	0-20
P1606-017	V-17	Lunde	79.0078	6.8994	1205	0-30	0-5
P1606-018	V-18	Lunde	79.0075	6.899	1207	0-7	0-5
P1606-019	V-19	Lunde	79.0075	6.8986	1207	nd	0-5
P1606-020	V-20	Lunde	79.0075	6.899	1207	0-30	0-1
P1606-021	V-21	Lunde	79.0075	6.8989	1207	0-30	0-5
P1606-025	V-25	Lunde	79.0071	6.9111	1200	0-30	0-5
P1702-898	898	GHM1	76.1144	16.0033	380	0-12	0-1
P1702-900	900	GHM1	76.1150	16.0036	380	0-35	0-1
P1702-902	902	GHM1	76.1153	16.0011	377	0-25	0-1
P1702-916	916	GHM1	76.1150	16.0019	377	nd	0-1
P1702-917	917	GHM1	76.1153	16.0011	377	0-20	0-1
P1702-918	918	GHM1	76.1153	16.0006	389	0-50	0-1
P1702-919	919	GHM1	76.1156	16.0003	378	0-25	0-1
P1702-920	920	GHM5	76.1114	16.0075	379	nd	0-1
P1702-921	921	GHM5	76.1117	16.0067	380	nd	0-1
P1702-922	922	GHM5	76.1122	16.0064	386	nd	0-1
P1710-008	L-8	Canyon South	68.1583	10.4607	794	nd	0-1
P1710-009	L-9	Canyon South	68.1583	10.4606	794	0-20	nd
P1710-012	L-12	Canyon South	68.1591	10.4559	804	0-30	nd
P1710-019	L-19	Canyon South	68.1587	10.4561	794	nd	0-1
P1710-030	L-30	Canyon South	68.1578	10.4654	770	0-40	nd
P1710-031	L-31	Canyon South	68.1578	10.4653	769	nd	0-1
P1710-032	L-32	Canyon South	68.1578	10.4654	769	nd	0-1
P1710-035	L-35	Canyon South	68.158	10.4658	778	nd	0-1
P1710-052	L-52	Canyon North	68.1665	10.4633	807	0-40	0-1
P1710-056	L-56	Canyon North	68.1671	10.4702	782	0-30	0-1

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