

1 Abiotic methane from ultraslow-spreading ridges can charge

2 Arctic gas hydrates

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11 **ABSTRACT**

12 Biotic gas generation from the degradation of organic carbon in ocean sediments supplies
13 and maintains gas hydrates throughout the world's oceans. In nascent, ultra-slow spreading
14 ocean basins, methane generation can also be abiotic, occurring during the high temperature
15 (>200 °C) serpentinization of ultramafic rocks. Here, we report on the evolution of a growing
16 Arctic gas and gas-hydrate charged sediment drift on oceanic crust in eastern Fram Strait, a
17 tectonically controlled, deep-water gateway between the subpolar North Atlantic and Arctic
18 Ocean. Ultraslow-spreading ridges between NW Svalbard and NE Greenland permit the
19 sustained interaction of a mid-ocean ridge transform fault and developing sediment drift, on both
20 young (<10 Ma) and old (>10 Ma) oceanic crust, since the Late Miocene. Geophysical data
21 image the gas-charged drift and crustal structure and constrain the timing of a major 30 km
22 lateral displacement of the drift across the Molloy Transform Fault. We describe the build-up of

23 a 2 Ma, long-lived gas hydrate/free gas-charged drift system on young oceanic crust that may be
24 fed and maintained by a dominantly abiotic methane source. Ultra-slow spreading, sedimented
25 ridge flanks represent a previously unrecognized carbon reservoir for abiotic methane that may
26 supply and maintain deep-water methane hydrate systems throughout the Arctic.

27 INTRODUCTION

28 Marine sediments contain large quantities of biotic methane formed by microbial
29 degradation of organic matter occurring at high temperature ($>80^{\circ}\text{C}$) (Claypool and Kvenvolden,
30 1983). About 500 -10000 Gt of methane carbon are stored in marine gas hydrate systems
31 (Kvenvolden, 1988; Buffet and Archer, 2004). Abiotic methane has been recently recognized as
32 a new fundamental source generated in slow to ultra-slow spreading mid-ocean ridge
33 environments during the serpentinization of ultramafic rocks (Proskurowski et al., 2008; Cannat
34 et al., 2010). Serpentinization rates in ultramafic rocks are highest at temperatures between 200°
35 and 350°C (Martin and Fyfe, 1970) and occur within a permeability zone in the upper crust that
36 may not exceed 3–4 km in depth (Cannat et al., 2010). In magma-limited slow and ultra-slow
37 ridges, serpentinization is focused along large detachment faults (e.g. Escartín et al., 2008; Sauter
38 et al., 2013), which can accommodate the majority (nearly 100%) of relative plate motion (e.g.,
39 Sauter et al., 2013), and are often well developed at the inside corners of ridge-transform
40 intersections (Tucholke et al., 1998). Slow to ultra-slow spreading ridge detachment faults form
41 near the ridge axis and are believed to be active over a period of 1–4 m.y. (Tucholke et al., 1998;
42 Tani et al., 2011), thus limiting active serpentinization and methane venting to the youngest crust
43 near the ridge axis.

44 In the Arctic, (Figure 1) low angle detachment faults and exhumed serpentinized
45 peridotites have been observed and sampled on Gakkel Ridge (Dick et al., 2003; Michael et al.,

46 2003), serpentinite and peridotite sampled on Lena Trough and Molloy Ridge (Snow et al.,
47 2001), and black smokers and vent fauna observed at the junction of the Mohn and Knipovich
48 Ridges on an axial high that is flanked by exhumed detachment surfaces or oceanic core
49 complexes (Pedersen et al., 2010). BSRs (bottom simulating reflectors) that indicate the base of
50 the gas hydrate stability zone (GHSZ), identified in seismic sections above interpreted
51 serpentinized ultramafic diapirs are also documented on the sediment covered eastern flank of
52 the Knipovich Ridge (Rajan et al., 2012). These observations establish the possibility for gas
53 delivery for gas hydrates from an abiotic, serpentinized mantle source of methane throughout
54 sediment covered portions of the Arctic Ocean ultra-slow spreading ridges.

55 In the Arctic Fram Strait, the tectonic controlled oceanographic gateway to the Arctic
56 Ocean, deep-water sediment drifts have formed over millions of years along the continental
57 margins by interactions with geostrophic currents (i.e. Heezen et al., 1966). The Vestnesa Ridge,
58 a >100 km long and 50 km wide sediment drift, developed under such geostrophic currents
59 within Fram Strait (**Figure 1**). Prior to the Oligocene (33 Ma), Fram Strait was closed along a
60 major continental transform plate boundary between the Arctic and Norwegian/Greenland Sea
61 basins (Engen et al., 2008). A change in plate motion at 33 Ma resulted in oblique rifting across
62 this continental transform and initiated the opening of Fram Strait (Talwani and Eldholm, 1977).
63 As a result, a narrow oceanographic gateway slowly developed due to the ultraslow-spreading
64 Molloy and Knipovich Ridges, the last ridges created between the Gakkel Ridge/Lena Trough in
65 the Arctic Ocean and the Mohns Ridge in the Norwegian Sea (Engen et al., 2008; Ehlers and
66 Jokat, 2013). The active Molloy Transform Fault (MTF) and Spitsbergen Transform Fault (STF)
67 clearly offset these spreading ridges by ~120 and ~150 km, respectively (**Figure 1**). This tectonic
68 development controlled initial exchanges of Arctic and Norwegian sea surface and deep-water

69 masses through Fram Strait, which likely occurred at the earliest during the late Early Miocene
70 (~17 Ma; Jakobsson et al., 2007; Ehlers and Jokat, 2013) or at the latest, during the Late
71 Miocene (~10 Ma; Engen et al., 2008).

72 Such a plate-tectonic development of Fram Strait, and subsequent water mass exchanges
73 between the Arctic and subpolar North Atlantic, created an environment for the formation of
74 sediment drifts throughout Fram Strait (Eiken and Hinz, 1993; Gebhardt et al., 2014). The
75 Vestnesa sediment drift, between the NW Svalbard margin and the MTF, evolved within the
76 West Spitsbergen Current (WSC, continuation of the Atlantic Current flowing north through the
77 eastern side of Fram Strait), and grows on oceanic crust at the North American-Eurasian plate
78 boundary and within the stability field for marine gas hydrate, an ice-like substance that forms
79 under low temperature and high pressure conditions if enough microbial, thermogenic, and/or
80 abiotic gas and water coexist in the pore space of sediments. A gas hydrate reservoir and active
81 free gas system within the Vestnesa sediment drift north of the MTF (Figure 1) creates vents that
82 release gas through the seafloor and into the ocean (Hustoft et al., 2009; Petersen et al., 2010;
83 Bünz et al., 2012; Smith et al., 2014).

84 In this study, we reconstruct the tectono-sedimentary evolution of an Arctic gas hydrate
85 system through time by integrating existing data with newly collected high resolution P-Cable 2-
86 D seismic and swath bathymetry data to: (1) reconstruct both the build-up and break-up of the
87 Vestnesa drift along the MTF with a significant 30 km offset; (2) constrain the age of an Arctic
88 gas/gas hydrate system that concurrently developed within the growing sediment drift; and (3)
89 image the link between crustal structure and gas migration pathways that suggest the gas hydrate
90 system south of the MTF is likely charged by a significant portion of abiotic gas.

91 **Stratigraphy of the Vestnesa Sediment Drift**

92 The stratigraphy of Vestnesa Ridge in the area north of the MTF has been divided into
93 three seismostratigraphic units (YP-1, YP-2, and YP-3) (Eiken and Hinz, 1993; Hustoft et al.,
94 2009), with age control based on correlation to ODP Leg 151 holes (Geissler et al., 2011;
95 Mattingsdal et al., 2014). The YP-1 sequence shows syn-rift and post-rift sediments deposited
96 directly on oceanic crust. Magnetic anomaly chrons 6 (19.6 Ma), 5 (9.8 Ma) and 2A (2.8 Ma)
97 constrain the age of the ocean crust beneath Vestnesa Ridge (Engen et al., 2008 (**Figure 1**). The
98 YP-2 sequence exhibits contourites and YP-3 encompasses glaciomarine contourites and
99 turbidites (Howe et al., 2008). The boundary between YP-2 and YP-3 lies at an estimated age of
100 2.7 Ma (Knies et al., 2009). The basal age of the YP-2 sequence beneath Vestnesa Ridge could
101 be at least 11 Ma (Mattingsdal et al., 2014) and as old as 14.6 Ma (Geissler et al., 2011); this
102 range in age may be a consequence of the time transgressive nature of a migrating sediment
103 drift.

104 **OFFSET ALONG THE MOLLOY TRANSFORM FAULT**

105 We discovered a new major southern extent of the Vestnesa sediment drift that rests on
106 significantly younger oceanic crust, between magnetic anomaly chrons 5 and 2A, compared to
107 the drift north of the MTF, and lies offset to the west across the MTF (**Figure 1 and DR Figure**
108 **1**). The offset of the drift is significant (30 km) and is accurately measured using the lateral
109 displacement of the faulted and pockmarked apex of the drift bodies, imaged by seismic and
110 seabed mapping, both north and south of the transform. Restoration of the two portions of the
111 drift to their original position when they first encountered the transform fault, is based on the
112 most recently published half-spreading rates from the eastern side of the Molloy Ridge, 6.5
113 mm/yr, and the western side of the Knipovich Ridge, 8 mm/yr (Ehlers and Jokat, 2009). These
114 half spreading rates yield a full plate slip rate on the MTF of 14.5 mm/yr. This slip rate implies

115 that the 30 km offset of the Vestnesa sediment drift by the MTF would take ~2 Myr to reach the
116 present configuration.

117 Our reconstruction of the entire drift suggests that the southern part of the Vestnesa
118 sediment drift must have started to grow just prior to the beginning of its break-up phase at 2 Ma.
119 If the drift south of the MTF was much older than ~2 Ma, then the offset distance between the
120 faulted and pockmarked apexes from south to north of the drift would increase with age and the
121 observed separation would be significantly larger. To explain the age contrast in deposition of
122 the drift across the MTF, we propose a two phase evolution for the drift in space and time
123 (**Figure 2**). First, the >2km thick sediment accumulation of Vestnesa Ridge, its onset during the
124 Middle to Late Miocene, and its accumulation above old crust (~10–20 Ma) suggest that a N-S
125 oriented depocenter (i.e., Eiken and Hinz, 1993) developed north of the MTF, building the drift
126 here from at least 11 Ma to 3 Ma (**Figure 2**). Second, the intensification of Northern Hemisphere
127 glaciation at 2.7 Ma (Knies et al., 2014) and the subsequent increase in continental-shelf-edge
128 glaciation of the Svalbard margin caused a rapid increase in sedimentation rates (twofold)
129 throughout eastern Fram Strait (Mattingsdal et al., 2014). The increased sedimentation rates
130 promoted drift growth throughout Fram Strait (Gebhardt et al., 2014), including both sides of the
131 MTF (**Figure 2**), where continued seafloor spreading resulted in the 30 km offset of the Vestnesa
132 drift during the last 2 Ma.

133 **GAS HYDRATE SYSTEMS ON YOUNG AND OLD CRUST ACROSS THE MTF**

134 A well-documented gas hydrate and free gas system on Vestnesa Ridge, north of the
135 transform, (Hustoft et al., 2009; Petersen et al., 2010; Bünz et al., 2012) indicated also
136 thermogenic gas hydrates (Smith et al., 2014). South of the MTF, our new high-resolution
137 seismic data reveal an equally well-developed gas hydrate and free gas system, including a

138 strong bottom simulating reflection (BSR), representing the base of the GHSZ (**Figure 3; DR**
139 **methods**). Noteworthy, the water depth is ~450 m deeper at the crest of the drift south of the
140 MTF, but the depth of the BSR is shallower (~140 m) compared to the north (~200 m) (**Figure**
141 **3**). This 60 m difference is consistent with younger and hotter crust as indicated by higher
142 measured heat flow in the south ($139 \pm 2 \text{ mW/m}^2$) versus the north ($103 \pm 3 \text{ mW/m}^2$) (Crane et
143 al., 1991) (**Figure 3 insets**). To confirm the BSR is gas hydrate related, we calculate the heat
144 flow based on the BSR depth both north (95 mW/m^2) and south (141 mW/m^2) of the MTF, and
145 document remarkable agreements with the measured heat flow in each region (**Figure 3 insets**).

146 **GAS SOURCES NORTH AND SOUTH OF THE TRANSFORM**

147 Scientific drilling on the Vestnesa sediment drift on both sides of the MTF has not yet
148 been accomplished, but shallow gas hydrates in the Vestnesa sediment drift north of the MTF are
149 derived from thermogenic gas sources (Smith et al., 2014). Biotic gas producing source rocks do
150 exist in older Miocene age sediments (Knies and Mann, 2002) recovered from the base of ODP
151 Site 909 ~50 km to the west (**Figure 1**), which also may exist in equivalent age sediments
152 beneath Vestnesa drift, north of the MTF. However, the absence of this age sediment south of the
153 MTF may exclude comparable biotic gas sources here, although one cannot rule out
154 contributions from lateral gas migration via undiscovered stratigraphic/structural conduits. Given
155 an Arctic tectonic setting in a magma-limited ultra-slow spreading environment, we also do not
156 expect *in situ* thermal maturation of organic carbon driven by shallow magmatic sources (e.g.,
157 Gulf of California, Lizarralde et al., 2007). The well constrained age (~2 Ma) and thickness
158 (~700 m) of the drift deposit south of the MTF compared to the north (~11 Ma, >2 km), yet
159 showing a similar extensive gas hydrate/free gas system, suggests an additional gas source,
160 beyond biotic sources.. Seismic data south of the MTF image large offset normal faults in the

161 oceanic crust that are interpreted as detachment faults (**Figure 4**). Above one of these
162 detachments, high amplitude reflectors, a gas wipe out zone, and a large free gas accumulation
163 are visible directly beneath a BSR (**Figure 4**), suggesting a likely contribution from abiotic
164 methane sources, formed by the serpentinization of these exhumed ultramafic rocks. We suggest
165 that the preservation of the drift deposit south of the MTF in (1) a region of asymmetric ultra-
166 slow spreading, (2) at the elevated inside corner of a ridge transform discontinuity (**Figure 1**),
167 (3) above relatively young underlying crust (2.8–9.8 Ma), (4) with large offset faults imaged
168 beneath the sedimentary cover (**Figure 4**), and (5) the onset of its deposition in this region 2–3
169 Ma, creates a very high potential for abiotic methane production from the serpentinization of
170 ultramafic rocks. This production may provide a significant source of abiotic methane to the
171 overlying sediments in this Arctic ultra-slow spreading ridge environment within the Fram Strait
172 deep-water gateway.

173 **EARLY GAS CHARGE TO DEEP-WATER SEDIMENT DRIFT**

174 We propose an early gas charge, and thus a long lived (~2 Ma) gas hydrate system, at the
175 portion of the drift south of the MTF for two reasons. First, if abiotic gases are a dominant
176 source for methane in this region, its production is likely to have been ongoing during the last
177 ~1-4 m.y., during active detachment faulting and when the seafloor crust was young, sufficiently
178 warm, and infiltrated with seawater to drive serpentinization (**Figure 4B**). This early-formed
179 methane would have likely escaped into the water column until significant sediment
180 accumulation began (at 2.7 Ma) south of the MTF, and the drift sediments became an available
181 reservoir for this methane and subsequent gas hydrate. Second, the close association of high
182 angle faults and fluid escape features (chimneys and seafloor pockmarks) both north (e.g.,
183 Hustoft et al., 2009) and south (e.g., **DR Figure 1**) of the MTF, suggests faults play a critical role

184 as conduits for methane produced at depth and transferred upward into the GHSZ. South of the
185 MTF, these faults are syn-depositional (**Figure 4; DR Figure 1**) and thus developed early during
186 drift sedimentation, forming effective conduits for advective methane delivery to the overlying
187 sediment drift as it grows through time. South of the MTF, early-formed abiogenic methane would
188 have encountered a gas hydrate stability zone that expanded progressively with continued
189 translation of the drift into deeper water above a cooling crust.

190 **CONCLUSIONS**

191 Our geophysical results suggest that mixed biotic and abiogenic gas/gas hydrate systems can
192 initiate, develop, and survive on tectonic timescales near young, sedimented, ultraslow-
193 spreading, mid-ocean ridge transform intersections. These active tectonic environments may not
194 only provide an additional, serpentinized crustal source of methane for gas hydrate, but serve as
195 a newly identified and stable tectonic setting for the long term storage of large amounts of
196 methane carbon in deep marine sediments. Future scientific ocean drilling and isotopic
197 characterization of the recovered gases is necessary to quantify the proportion of biotic and
198 abiogenic gases stored in these deep-water reservoirs throughout the ultra-slow spreading Arctic
199 Ocean ridges. .

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324 **FIGURE CAPTIONS**

325 Figure 1. **[[No “A” label seen]]** Tectonic setting of the Vestnesa Sediment Drift. Bathymetry
326 from IBCAO grid; magnetic anomaly chrons (in red) 2A: 2.8 Ma, 5: 9.8 Ma, 6: 19.6 Ma (Engen
327 et al. 2008); half-spreading rates from Ehlers and Jokat (2009); pockmark fields in white; seismic
328 lines in black-bold where shown; ODP core sites as numbered; seismic line X shown in Vanneste
329 et al. (2005). KR-Knipovich Ridge, MR-Molloy Ridge, LT-Lena Trough, GR-Gakkel Ridge,
330 MhR-Mohns Ridge, MTF-Molloy Transform Fault, STF-Spitsbergen Transform Fault, YP-
331 Yermak Plateau, KF-Kongsfjorden Trough, PKF-Prins Karls Forland, ISF-Isfjorden Trough.

332 Figure 2. Tectonic reconstruction of the Vestnesa sediment drift during the last 11 Ma. MTF-
333 Molloy Transform Fault, MFZ-Molloy Fracture Zone, WSC-West Spitsbergen Current, magnetic
334 anomaly chrons as in Figure 1. An early crest of the growing drift (black axis in middle and right
335 panel) across the MTF is observed today (Vanneste et al., 2005, and Supplemental Figure 1),
336 faulted and pockmarked, and serves as a strain marker for the measured 30 km offset. Continued
337 eastward growth of the drift north of the MTF within the WSC resulted in the development of a
338 younger crest (white axis), which is also faulted and pockmarked today.

339 Figure 3. High-resolution p-cable seismic profiles across (A) the crest of Vestnesa Drift, north
340 (A) and south (B) of the MTF. Map locations shown bold in Figure 1. Insets: Theoretical heat
341 flow data (Stein and Stein, 1992) and measured heat flow data (Crane et al., 1991) vs age for the
342 east flank of Molloy Ridge (inset in A) and west flank of Knipovich Ridge (inset in B). BSR-
343 derived heat flow values shown as yellow boxes. Black bars designate the age of the crust
344 beneath the Vestnesa drift on each side of the MTF.

345 Figure 4. A. High-resolution seismic profile across the drift south of the MTF. Oceanic crustal
346 structures on the western flank of the Knipovich Ridge are shown and interpreted as large offset
347 detachment faults that exhume probable serpentinized ultramafic rock (**DR Crustal Structure**).
348 A broad gas wipeout zone, high amplitude (gas charged) reflectors, free gas zone and observed
349 BSR are present above the largest offset detachment fault. The observed BSR is restricted to the
350 crest of the drift and shows a vertical offset with the modeled BSRs (parameters as in **Figure 3**),
351 consistent with advection-driven shoaling of the BSR. B. Conceptual diagram of an abiotic
352 methane window for serpentinized oceanic crust in an ultra-slow spreading ridge environment.
353 Temperature and crustal age constraints described in the text. Position of the sediment drift south
354 of the MTF pre- and post-offset are shown as purple age brackets. The drift clearly evolved
355 initially above crust within the abiotic methane window and, as we propose developed its gas
356 hydrate system early. Continued offset into deeper water will eventually result in diminished
357 serpentinization sources of methane, but much of the early-formed abiotic methane could be
358 retained in long-lived, deep-water gas hydrate.

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