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Environmental challenges related to methane hydrate decomposition from climate change scenario and anthropic activities: State of the art, potential consequences and monitoring solutions

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Abstract

Natural gas hydrate deposits (NGHD) have been investigated for decades and represent one of the major methane reservoirs on Earth. They are encountered in sediment of both the continental margins and the permafrost region; areas considered to host amongst the most climate-sensitive ecosystems on Earth. With worldwide temperature increases affecting continental margins and the permafrost, it is important to raise concern about the fate of the NGHD in the coming centuries. Thus, this review presents an overview of the potential consequences of hydrate decomposition on its surrounding areas. It compiles and discusses hydrate-derived methane fluxes measured or inferred from *in situ* data at several sites by considering both dissociation and dissolution. Depending on the magnitude and the duration of hydrate decomposition, the amounts of methane released can affect to varying degrees the seafloor and the microbial communities that sustain the methane cycle and regulate its transfer from the sediment to the water column; and that aspect is addressed in this review. Here, we also considered the transfer of methane from NGHDs and more broadly from marine emissions to the atmosphere, as it is assumed that such transfer will likely increase in the future. Finally, multi-scale monitoring in space and time is a key element to evaluate the impacts of natural and anthropic perturbations

39 on NGHDs. We thus propose potential engineering solutions for the monitoring of
40 NGHD, mainly based on the long-term deployment of sensor systems.

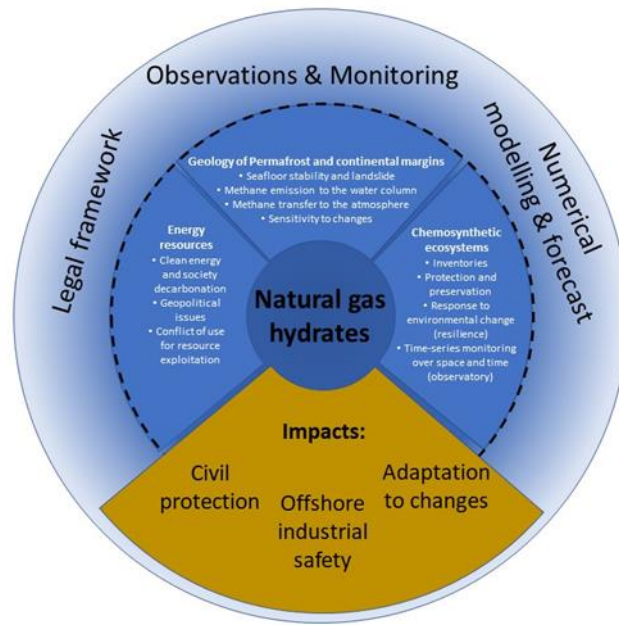
41 **Keywords:** Continental margins, gas emissions, methane flux into the atmosphere,
42 natural gas hydrate deposits, permafrost and slope stability

43

44 **1. Introduction**

45 Natural gas hydrate deposits (NGHD), one of the major methane (CH₄) reservoirs
46 on Earth, are mainly located on continental margins, generally at water depth beyond
47 ~500 m (Max, 2003; Max and Lowrie, 1996; Mienert et al., 2022; Milkov, 2004; Paull
48 and Dillon, 2001; Ruppel, 2011). They are found in the upper sedimentary interval where
49 the temperature is rather low (below 4 °C at the sediment-water interface for open seas)
50 to meet their stability requirements. On continental margins, NGHD are generally
51 encountered within the sediment at cold seep areas, which are specific ecosystem areas
52 characterized by natural gas emissions at the seafloor (Sassen et al., 1993; Suess, 2014;
53 Villar-Muñoz et al., 2021). Outcropping hydrates are less common but can also be
54 observed at sites characterized by intense gas venting (MacDonald et al., 1994; Olu et al.,
55 2009; Sassen et al., 1999). The amount of methane stored within the marine hydrate-
56 bearing sediments ranges between 3-1469 GtC (Burwicz et al., 2011; Lee et al., 2022;
57 Pinero et al., 2013; Wallmann et al., 2012). Methane released from hydrates promotes the
58 development of chemosynthetic communities. At the higher latitude, in Polar Regions
59 where the temperature is almost permanently below freezing point, natural gas hydrates
60 are found in the permafrost at shallower water depth. The mind map in Figure 1 illustrates
61 the large variety of disciplines the studies of the dynamics of NGHDs can collate: from
62 field work including observations, sampling and monitoring, to laboratory study with
63 analyses, modeling and forecast. Such studies are also relevant for societal issues such as
64 civil protection and offshore industrial safety when dealing with seafloor stability, gas
65 emissions, climate change adaptation, meeting energy needs, and preservation of
66 chemosynthetic ecosystems. Tackling these issues requires the need of legal framework.

67



68 Figure 1: Mind-map representing the principal hydrate-driven scientific researches with
 69 the societal issues.

70 The hydrate-bound gases either come from deep thermogenic reservoirs that contain
 71 methane, and usually a significant amount of heavier hydrocarbons (often called C₂₊), or
 72 are generated by the microbial degradation of organic matter at shallower sedimentary
 73 depth, that produces mainly methane (Kvenvolden, 1995; Milkov, 2005; Milkov and
 74 Etiope, 2018; Whiticar, 1994, 1999). The presence of hydrocarbons other than methane
 75 in the hydrate-bound gases expands the hydrate stability field towards lower pressure and
 76 higher temperature. For instance, thermogenic hydrates are found in the Sea of Marmara
 77 at ~660 m water depth and ~15 °C (Bourry et al., 2009), whereas the microbial gas hydrate
 78 deposit present in the Romanian sector of the Black Sea is located at >700 m water depth
 79 and 9 °C (Burwicz and Haeckel, 2020; Chazallon et al., 2020; Ker et al., 2019; Riboulot
 80 et al., 2018; Zander et al., 2017).

81 Most of the NGHD inventoried on Earth contain microbial gases. Thus, they are more
 82 sensitive to changes in temperature, pressure and fluid chemistry, especially salinity. Any
 83 changes in one of these physicochemical parameters can trigger their decomposition (also
 84 called breakdown), leading to the discharge of the hydrate-bound gases into the water
 85 column, with potential input into the atmosphere (Schmale et al., 2005; Shakhova et al.,
 86 2010; Solomon et al., 2009; Thatcher et al., 2013; Westbrook et al., 2009). Here, the term
 87 decomposition refers to either dissolution with release of methane-enriched water, or
 88 dissociation when the NGHD is brought outside its stability field and the hydrates are
 89 fully decomposed into water and free gases. Previous studies have shown that other
 90 natural geological processes such as tides, seismic activity, seawater infiltration, change
 91 in the Gulf Stream current, or the decrease and cease in gas supply may also trigger
 92 hydrate decomposition (Lapham et al., 2013; Phrampus and Hornbach, 2012; Riboulot et
 93 al., 2018; Sultan et al., 2014). Less frequently observed in nature, hydrate-piece
 94 detachment has been proposed as a possible mechanism for causing the degradation of

95 NGHD and allowing the transfer of significant amounts of methane into the hydrosphere
96 (Pape et al., 2011a).

97 Furthermore, hydrate decomposition changes the mechanical properties of the subsoil and
98 affects the seafloor stability (Fig. 1). Such decomposition can enhance the formation of
99 seafloor structures like pockmarks and carbonate build-up, and/or can also trigger
100 sediment sliding (Hovland et al., 2002; Maslin et al., 2010; McConnell et al., 2012; Sultan
101 et al., 2014; Yan et al., 2020). Climate change and/or anthropogenic activities are amongst
102 the two major processes that may cause NGHD decomposition in the future (Dickens,
103 2001; Dickens and Quinby-Hunt, 1997; Kennett et al., 2003; Marcelle-De Silva and
104 Dawe, 2011; Moridis and Collett, 2003; Moridis et al., 2009; Ruppel, 2011; Ruppel and
105 Kessler, 2017; Schicks et al., 2011; Vargas-Cordero et al., 2020b). Seabed deformation
106 and prolonged release of methane in the water column are the two main hazards related
107 to hydrate decomposition (Maslin et al., 2010), and they may have severe consequences
108 on the dynamics of the surrounding ecosystems (Fig. 1). Thus, NGHD decomposition
109 may be considered as a threat to the biodiversity of these ecosystems. However, they also
110 represent a potential energy resource (Fig. 1) and several countries have undertaken
111 comprehensive programmes for hydrate inventory on continental margins located within
112 their Exclusive Economic Zone (EEZ), with the aim of estimating the amount of methane
113 that can be recovered (Boswell et al., 2017; Collett et al., 2014; Collett et al., 2012; Konno
114 et al., 2017; Li et al., 2018; Ruppel et al., 2008). It is worth noticing that current solutions
115 for methane production from NGHD are not economically viable, and our increasing
116 awareness of climate change requires us to advance our researches on the understanding
117 of natural-gas hydrate dynamics and its relevance in the global carbon cycle. Such
118 research studies are key to developing sustainable solutions for the aforementioned
119 societal issues.

120 Furthermore, it is worth emphasizing that the time scale of hydrate decomposition can
121 extend from a few years to millennia, depending on the nature of the disturbance. Thus,
122 here we define disturbances leading to short-term decomposition as resulting from
123 processes that significantly affect the NGHD after few years from their onset; whereas
124 disturbances leading to long-term decomposition refer to processes for which the impacts
125 on the NGHD appear after few decades, centuries or millennia. For instance, the influence
126 of temperature change on hydrate stability is different when considering hydrate
127 production (short-term human-induced disturbances to recover the methane as quick as
128 possible) and climate change (long-term human-induced disturbances). Indeed, as an
129 example of disturbance leading to long-term decomposition, modeling of the West
130 Svalbard by Thatcher et al. (2013) and Marín-Moreno et al. (2015), respectively,
131 suggested that the delay between the onset of warming and the inception of gas emissions
132 due to this warming could amount to ~20-30 years. Such a time lag is not conceivable for
133 hydrate production induced by thermal stimulation for instance. The magnitude of the
134 decomposition and its duration also raise the question of the resilience and adaptation of
135 the ecosystems associated with these deposits.

136 In this paper, an analysis of the environmental challenges associated with the
137 decomposition of NGHD is proposed. It starts with a detailed description of the possible
138 hazards related to the decomposition of NGHD and their consequences on the marine
139 ecosystems considering disturbances leading to both short-term and long-term
140 decomposition. Finally, it discusses possible monitoring solutions to assess the state of
141 decomposition of a deposit and quantify the gas discharge over time.
142

143 2. Prerequisites and main factors governing hydrate dynamics

144 The necessary thermodynamic conditions to form and sustain hydrate growth
145 within sediments are the saturation of pore water with gas in coexistence or not with free
146 gas, low temperature and high pressure. A failure of one of these three prerequisites will
147 systematically trigger hydrate decomposition. However, several other factors govern the
148 formation, distribution and extent of gas hydrates in nature. Amongst them, the
149 physicochemical properties of the hydrate-forming geofluids and the sediment (*e.g.* gas
150 generation and transport processes, gas fluxes, pore-water salinity, sediment mineralogy
151 and permeability) and the geological characteristics of the setting (*e.g.* tectonics and fluid
152 transport pathway, sedimentation regime and lithology, geothermal gradient and heat flux
153 pattern) are key points. These factors affect both the formation and the decomposition of
154 hydrates to varying extents as stressed in previous review papers (Hassanpouryouzband
155 et al., 2020; Malagar et al., 2019; Ruppel and Waite, 2020; Waite et al., 2009), making
156 the prediction of the consequences of changing any of them on the surrounding geosphere
157 as well as on the hydrosphere, and potentially the atmosphere, a challenging task. Such a
158 prediction becomes even more demanding once we consider that the accumulation of
159 natural gas hydrates within the sediments is a time and space-evolving process due to the
160 continuous changes in the deposit permeability and thermal properties that, in turn,
161 continuously modifies the hydrate growth kinetics, the gas flow regime and pathway, and
162 thus reroutes their distribution pattern (Schmidt et al., 2022). Such a behavior explains
163 the wide varieties of hydrate habits encountered in nature, from disseminated through lens
164 to massive nodules, and its heterogeneous distribution within the sediment from
165 microscale to kilometer scales (Daigle and Dugan, 2011a, b; Egeberg and Dickens, 1999;
166 Guo et al., 2020; Kossel et al., 2018; Naudts et al., 2012; Pape et al., 2011b; Paull and
167 Dillon, 2001; Simonetti et al., 2013; Solomon et al., 2014; Stern and Lorenson, 2014;
168 Stern et al., 2011; Wang et al., 2020). Thus, the knowledge of how all the aforementioned
169 properties and factors affect the hydrate dynamics is often site-specific and depends on
170 the geological history of the setting. Nevertheless, these properties are critical to
171 predicting and quantifying the macroscale responses of hydrate-bearing sediments to
172 environmental changes at different time scales. The environmental challenges are
173 primarily related to gas hydrate decomposition, which also leads to severe and time-
174 evolving changes in the permeability field of the deposit. The magnitude of the
175 decomposition and how it happens strongly depend on the nature and magnitude of the
176 disturbances applied to the NGHD. The following section focuses on the environmental
177 challenges related to anthropic (human-induced) disturbances and climate change. As

178 explained above, the first corresponds to short-term perturbations with localized
179 consequences, while the second induces disturbances spanning over a larger timescale
180 and with broader impacts.

181

182 **3. Environmental challenges related to hydrate decomposition**

183 **i. Impact of hydrate decomposition on the seafloor** 184 **morphology and stability**

185 Methane hydrate formation and decomposition induce the build-up or loss of solid
186 materials that have a density significantly different from that of pore water or free gas
187 (Lee et al., 2010; Loreto et al., 2011; Pape et al., 2011a). Density difference is one of the
188 physical mechanisms that drives seafloor deformation. Several studies have demonstrated
189 that hydrate formation and decomposition modifies the regime and pathway of free gas
190 migration through the sediments, and can strongly affect the morphology of the seafloor
191 by creating or enhancing the formation of pockmarks, mounds or pingos (Andreassen et
192 al., 2017; de Prunelé, 2015; Gupta et al., 2022; Hovland and Svensen, 2006; Loreto et al.,
193 2011; Paull et al., 2007; Riboulot et al., 2016; Serié et al., 2012; Sultan et al., 2014; Sultan
194 et al., 2010; Taleb et al., 2020; Vargas-Cordero et al., 2020a; Waage et al., 2020; Zander
195 et al., 2020). Besides the formation and decomposition of hydrates, dislodgement of
196 hydrate pieces has been proposed to explain the rough seafloor topography encountered
197 in the Eastern Black Sea (Pape et al., 2011a). While the hydrate formation stores methane
198 within the sediment, the melting of hydrates triggers its release, and this is accompanied
199 by the development of excess pore pressure that, in turn, reduces the mechanical strength
200 of the sediment (Le et al., 2020; Nguyen-Sy et al., 2019; Sultan et al., 2007). In the rest
201 of this section, we will focus on the consequences of hydrate decomposition as this
202 process can trigger sediment mass flow (Sultan et al., 2004), which can be prodigious in
203 scale and generate damaging tsunamis on open continental slopes (Talling et al., 2014).

204

205 In nature, hydrate dissolution is more widespread than dissociation because dissolution
206 does not require the significant amount of energy needed to take the system outside its
207 stability field (Ruppel and Waite, 2020). It can result from the shutdown or the slowdown
208 of the gas flux supplying the NGHD, or an increase in either the pore water salinity or the
209 bottom water temperature. Very few works mention explicitly the consequences of
210 hydrate dissolution on the seafloor. Sultan et al (2010, 2014) showed that the formation
211 of pockmarks in the Nigeria deep-water and the evolution of their morphology are
212 strongly related to hydrate formation and dissolution. The observed pockmarks can be 40
213 m depth and nearly a kilometer in diameter, making the installation of offshore oil and
214 gas production facilities in their close vicinity a risky task. The dissociation of natural
215 hydrates requires severe and more rapid changes in the (T, p) conditions. It can result
216 from an increase in the bottom water temperature or a decrease in the hydrostatic pressure
217 (Andreassen et al., 2017; Marin-Moreno et al., 2015). Most of the existing articles on the

218 impact of gas hydrate decomposition on the seafloor mentioned hydrate dissociation
219 explicitly.

220
221 The role of gas hydrate dissociation on submarine slope instability has been investigated
222 over decades but controversy persists (Collett et al., 2014; Hassanpouryouzband et al.,
223 2020). Dillon and Max (2000) have argued that dissociation of gas hydrates can be
224 considered as the main cause of submarine slope failure only when the three following
225 criteria are met: (i) gas hydrates are widespread; (ii) slides are initiated in areas within the
226 hydrate phase boundaries; and (iii) sediment of low permeability (mainly fine-grained
227 sediments) is present at the base of the gas hydrate occurrence zones. There are two well-
228 studied submarine slope failures that happened in interglacial periods that have been
229 potentially attributed to hydrate dissociation, although debate still remains: Storegga
230 Slide and Hinlopen Slide. The Storegga Slide is one of the largest submarine slope failures
231 known to have happened 8000 years ago (Bugge et al., 1988). The slide was most likely
232 triggered by a strong earthquake and developed as a retrogressive slide, where massive
233 sediments of about 150-200 m thick and up to 10.30 km wide slid down over ~200 km,
234 causing huge tsunamis (Bryn et al., 2005). Besides, data presented few years ago (Geissler
235 et al., 2016) strongly indicates that gas hydrates, free gas and excess pore pressure may
236 have been the trigger or at least supportive factors for the slope instabilities north of
237 Svalbard, in the vicinity of the Hinlopen Slide. Considering other locations, Lee argued
238 that hydrate dissociation might have played a role in the large submarine landslides on
239 the US Atlantic margin (Lee, 2009). Gas hydrate dissociation during deglaciation phases
240 was also mentioned as one of the possible climate-driven changes that significantly
241 affected slope stability in the Mediterranean Sea (Urgeles and Camerlenghi, 2013). In
242 another recent work, by considering a seafloor temperature increase of 2 or 4 °C,
243 Alessandrini et al (2019) simulated the impact of future global warming over the next 50
244 and 100 years on the Gas Hydrate Stability Zone (GHSZ) off the Chilean margin. They
245 suggested that, despite an increase in hydrostatic pressure due to sea level rise, seafloor
246 temperature increase would induce the dissociation of the entire NGHD located along the
247 upper slope in the next century. The dissociation will cause slope instability in some areas,
248 and could seriously damage coastal cities if this dissociation triggers tsunamis. Using a
249 large geophysical dataset including bathymetry and 2D/3D P-cable seismic data, Hillman
250 et al. (2018) investigated the potential relationship between gas migration pathways, gas
251 vents observed at the seafloor and submarine slope failures near the S2 Canyon in the
252 Danube fan (Northwestern Black Sea). They found abundant evidence for gas migration
253 in the area that suggests a connection between slumps and gas migration, although it is
254 not clear whether gas migration facilitates slope failure through pressure and pre-
255 conditioning of the sediments, or vice versa. From 2D seismic reflection profiles along
256 the active margin of northern Colombia in the western Caribbean Sea, Leslie and Mann
257 defined a series of three giant, previously unidentified, submarine landslides (Leslie and
258 Mann, 2016). They suggested that an over-pressurized zone of weakness located at the
259 boundary of the hydrate stability has likely facilitated slope failure.

260

261 Since the early works of McIver (1982), several studies have argued for a causal link
262 between gas hydrate dissociation and the occurrence of submarine landslides (Bünz et al.,
263 2005; Dawson et al., 2011; Elger et al., 2018; Grozic, 2010; McIver, 1982; Mienert et al.,
264 2005). Such a link was supported by theoretical considerations based on the loss of
265 cementation coupled with the release of free gas, giving rise to the development of excess
266 pore pressure when changes in environmental conditions are no longer favorable for
267 stable gas hydrates (Sultan et al., 2004; Tinivella and Giustiniani, 2013). Indeed, the
268 dissociation of hydrates at the basis of their stability field could liberate free gas and
269 increase the pore pressure just beneath the Bottom Simulating Reflector (BSR), and
270 consequently decrease the effective normal stress (Hornbach et al., 2004). Thus, such
271 focused dissociation weakens the strength of sediments due to loss of hydrate cementation
272 between grains (Brown et al., 2006; Tinivella and Giustiniani, 2013). In the last two
273 decades, the role of excess pore pressure (overpressure) on submarine slope instability
274 has been very often put forward. The overpressure could potentially (i) facilitate or trigger
275 submarine slides, (ii) form nearly vertical columns of focused fluid flow and gas
276 migration, and (iii) determine the failure of a permeable layer in presence of low-
277 permeability zone (Xu and Germanovich, 2006). Recent analyses aiming to assess
278 quantitatively the role of gas hydrates on submarine slope instability have highlighted the
279 importance of sediment lithology (Handwerker et al., 2017) and drainage conditions
280 (Priest and Grozic, 2016), or fracture formation (Yang et al., 2018) on the development
281 of excess pore pressures during hydrate dissociation. Mountjoy et al. (2014) analyzed
282 seismic reflection data of the Tuaheni landslide complex (New Zealand East Coast) and
283 proposed that, gas hydrates can directly destabilize the seafloor via three mechanisms
284 (Mountjoy et al., 2014): (1) excess pore pressure induced by gas hydrate dissociation; (2)
285 hydro-fracturing in the gas hydrate zone; (3) and time-dependent plastic deformation of
286 hydrate-bearing sediment. Recently, unlike previous studies, Gross et al. (2018)
287 performed a 3D analysis of the distribution of gas and gas hydrate indicators and did not
288 show evidence of gas hydrates within the upper landslide unit. However, their
289 observations support the hypotheses of Mountjoy et al. (2014) that argue that build-up of
290 overpressure below low permeability gas hydrate-bearing sediments could cause hydro-
291 fracturing in the gas hydrate zone and valving excess pore pressure into the landslide
292 body. The hypotheses also suggested that gas is migrating into the critical mechanical
293 zone at the base of the landslide, and thus building up overpressure that can lead to the
294 reactivation of the landslide. Results obtained by Horozal *et al.* (2017) in the Ulleung
295 Basin also suggested that excess pore pressure caused by gas hydrate dissociation could
296 have contributed to slope failures. Such an increase in the interstitial pore pressure
297 generated by gas hydrate dissociation was equally reported by Ker et al. (2019) in the
298 Western Black Sea. They suggested that values of excess pore pressure of 1-2 MPa are
299 probably the result of the low permeability of hydrate-bearing sediments. This excess
300 pore pressure is expected to drastically reduce sediment stability and could be at the origin
301 of some of the observed landslides in the area. Such assumptions agree with the study of
302 Elger et al. (2018) who used reflection seismic data from the Arctic Ocean and numerical
303 modeling to show a direct link between hydrates and slope instability. Their study showed
304 that hydrates reduce the sediment permeability and causes the build-up of overpressure at

305 the base of the GHSZ. This overpressure reaches the shallow permeable beds, through
306 hydro-fracturing forms like pipe structures, and then it is transferred laterally within the
307 sedimentary column, weakening the slope sediment and leading to its destabilization.

308
309 Hovland and Gudmestad discussed the engineering significance of gas hydrates and the
310 possible engineering consequences on seabed installations (Hovland and Gudmestad,
311 2001). Offshore petroleum exploration and production involves operations that alter
312 ambient subsurface conditions (changes in stress, temperature and compositions of pore
313 fluids), which in turn, may induce gas hydrate dissociation. For this reason, gas hydrates
314 may be considered as a geohazard for this sector of activity. The consequences of gas
315 hydrate-related seafloor instability to engineering structures would be very damaging
316 such as causing hydrocarbon release from producing wells, or seafloor slumping
317 obliterating subsea oil and gas production facilities. The following activities would also
318 trigger gas hydrate dissociation in the sediment:

- 319 - Drilling into the sea bottom produces heat that could dissociate gas hydrates in the
320 sediment and release gas. If the free gas released during drilling forms gas
321 hydrates around the wellhead, it could cause a loss of mud density, resulting in
322 wellbore instability and even mechanical system failure. More important gas
323 releases could have consequences on the stability of the drilling rig. In addition,
324 excess local pressure caused by gas hydrate dissociation in the sediment can
325 induce casing collapse.
- 326 - Heat from a warm pipeline will propagate in the surrounding sediments. This
327 could cause gas hydrate dissociation, inducing sediment instability and slumping.
328 Under such conditions, the pipeline may rupture.
- 329 - Suction anchors usually used in deep water to keep floating structures anchored
330 to the seafloor would reduce pressure and cause local gas hydrate dissociation,
331 disturbing the friction forces along the walls of the anchor and thus threatening
332 the safety of the operation.

333 McConnell et al. reviewed the history of gas hydrate shallow hazard assessment and
334 found that low-saturation, mud-hosted hydrate layers can be safely drilled using existing
335 industry protocols (McConnell et al., 2012). For highly saturated hydrate-bearing sand
336 reservoirs, they will likely continue to be avoided, unless they are the specific targets of
337 the well for methane production.

338
339 Mining of hydrates may also cause submarine landslides. Song et al. (2019) applied a
340 thermal-fluid-solid-stress coupling model to NGHD to investigate the failure process of
341 submarine landslides induced by hydrate decomposition. Their results show that hydrate
342 mining may cause settlement of the seafloor and slippage of submarine sedimentary
343 layers to the mining center, and thus can potentially trigger submarine landslides. Based
344 on geophysical and geotechnical data, Zander et al. (2017) investigated the potential
345 impacts of gas hydrate exploitation on slope stability in the Danube deep-sea fan in
346 northwestern Black Sea. Their 2D slope stability model applied to a hypothetical gas
347 hydrate reservoir suggests that the area is relatively safe against slope failure under static

348 conditions, but probably not sufficiently safe to allow the implementation of
349 infrastructure for hydrate production at the seabed without taking specific mitigation
350 measures into account (*e.g.* relocation options, design criteria).

351 **ii. Impact of hydrate decomposition on permafrost**

352 It has been generally accepted that global warming tends to destabilize gas hydrates
353 present in permafrost through different mechanisms, such as directly melting top layers
354 of gas hydrates (Mienert et al., 2005), thermal shock led by sea level rise (Maslin and
355 Thomas, 2003), and reduction in lithostatic pressure hence hydrostatic pressure due to
356 shrinking of subsea permafrost (Paull and Dillon, 2001; Tinivella and Giustiniani, 2016;
357 Tinivella et al., 2019; Waage et al., 2020; Wallmann et al., 2018). However, the
358 environment in which hydrates are found is different from that encountered on continental
359 margins. In permafrost regions, gas hydrates and ice may coexist with salts, water, and
360 free gas in the offshore sediments, which leads to unique characteristics of the ice-
361 hydrate-bearing sediments compared to those of the other marine sediments. However,
362 little work has been done to understand such properties of permafrost sediments
363 containing gas hydrates, although the mechanical properties of frozen soil and rock in the
364 absence of gas hydrates and unfrozen gas hydrate-bearing sediments were extensively
365 investigated (*e.g.* (Arenson et al., 2007; Tsyтович, 1975; Waite et al., 2009)). The
366 mechanical properties of frozen sand, silt and clay containing tetrahydrofuran (THF)
367 hydrate were determined at -10 °C (Yun et al., 2007), and they found that shearing results
368 in particle rotation, slippage, and rearrangement in the absence of THF hydrates. In
369 contrast, shear force leads hydrates to detach from the mineral surface in the presence of
370 low saturation (<40%) of THF hydrates, or to debond from the sediment grains in the
371 presence of high saturation (>40%) of THF hydrates. The mechanical properties of kaolin
372 clay-ice powder mixtures were investigated in the presence of carbon-dioxide (Liu et al.,
373 2013) and methane hydrates (Li et al., 2016; Li et al., 2019; Luo et al., 2018; Yun et al.,
374 2007). The determined shear strength was unexpectedly low compared to the sediments
375 containing gas hydrates only, and this was mainly explained by the lack of cementation
376 between hydrate crystals, ice powders and clay grains. Recently, Yang et al. (2019)
377 reported a set of comprehensive experiments for methane hydrate-bearing frozen
378 sediments. Their results showed that gas hydrates can form micro frames or network
379 structures, resulting in distinctive effects on the mechanical properties (higher shear
380 strength and less brittle under compressional loading) of the simulated hydrate-bearing
381 permafrost sediments compared to frozen sediments in the absence of gas hydrates (Yang
382 et al., 2019).

383 Old estimation provided huge volumes (up 9.2 to 22.3x10³ Pg (1 Pg = 10¹⁵ g)) of methane
384 trapped as hydrates in permafrost of the Arctic regions (Max and Lowrie, 1996).
385 However, more recent studies provided most lower estimates ranging from 27 (~1% of
386 global hydrate estimate) to 800 Pg of methane (Knoblauch et al., 2018; Miesner et al.,
387 2023; Ruppel, 2015). Gas hydrate-bearing permafrost sediments (GHBPS) are sensitive
388 to temperature changes that could be induced by global warming, seasonal changes,
389 geothermal fluxes, and human activities. Particularly, global warming is causing the

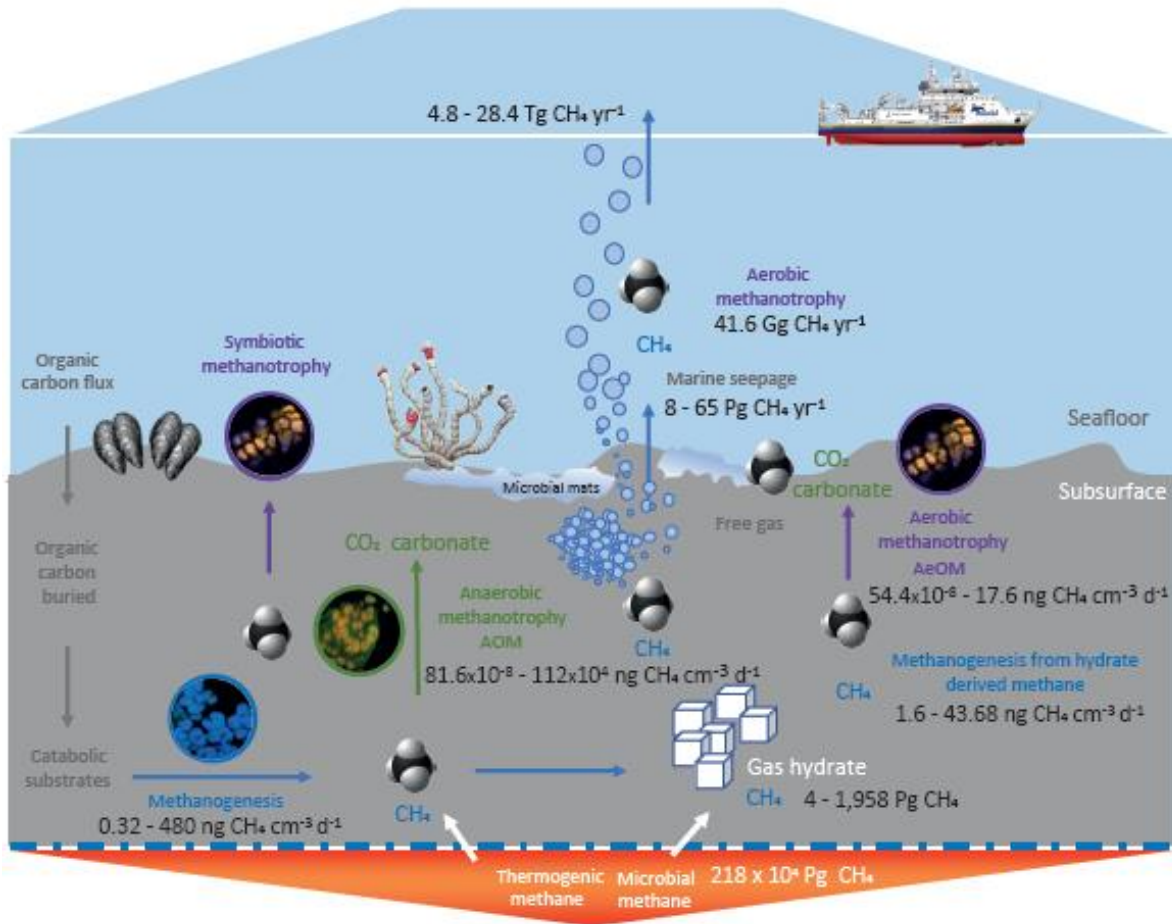
390 shrinking of both terrestrial and subsea permafrost, leading to a reduction in lithostatic
391 pressure and hence hydrate decomposition (Maslin et al., 2010). Scientific research has
392 revealed that global warming has accelerated the rate of ice thawing and gas hydrate
393 decomposition in permafrost regions, and, consequently, led to the release of methane
394 and carbon dioxide into the atmosphere, exacerbating global warming (Andreassen et al.,
395 2017; Portnov et al., 2016). A more detailed review on gas release follows in the next
396 section. This adverse effect of greenhouse gases reaching the atmosphere becomes even
397 more severe in the Arctic regions due to the effect of polar amplification. In other words,
398 the atmosphere or an extensive ocean circulation can lead to heat transport toward the
399 poles. Consequently, climate warming would affect the areas adjacent to the Earth poles
400 at relatively greater magnitude compared to the rest of the Earth surface (Polyakov et al.,
401 2002), accelerating decomposition of gas hydrates in permafrost regions such as the
402 Arctic. Observations of natural catastrophic events have demonstrated that this increasing
403 negative feedback could tip over the mechanical stability of GHBPS, and trigger a chain
404 of large-scale land and/or seafloor failures, and subsequently induce an abrupt greenhouse
405 gas venting from subsea permafrost in the East Siberian Arctic Shelf (Ruppel and Kessler,
406 2017) and life-threatening events such as tsunamis that could reach the shore of the UK
407 based on evidence of historic events (Long et al., 2016; Tinivella et al., 2019). The
408 instability of GHBPS has been attributed to landslides in Yukon Territory in Canada
409 (Maslin et al., 2010), the permafrost lava observed on the Tien Shan Mountains in China
410 (Marchenko et al., 2007), the formation of massive gas blow-out craters in the northern
411 Barents Sea (Andreassen et al., 2017) and in the Yamal and Gydan Peninsulas in northern
412 Siberia in Russia (Bogoyavlensky et al., 2020). These destructive consequences in recent
413 years are also attributed to the global warming of anthropogenic activities. Based on
414 numerical modeling, Andreassen et al. (2017) developed an understanding of how the
415 craters and mounds were formed in the northern Barents Sea and postulated that climate
416 warming leads to the ice sheet retreating, and this is accompanied with catastrophic
417 reduction of the sediment pore pressure. Consequently, gas hydrates initially in
418 permafrost or sediments dissociate. The dissociation of gas hydrates at depth, particularly
419 those underlying impermeable layers, results in building up of overpressure in pores.
420 Finally, when the overpressure reaches a certain threshold, gas hydrate pingos could be
421 formed, and their collapse creates craters in permafrost regions. Furthermore,
422 composition analysis of the gas from such craters showed particularly high concentration
423 of methane (Kvenvolden and Field, 1981). Such high concentration of methane only
424 allows formation of structure I hydrate which makes the gas hydrate-bearing permafrost
425 more vulnerable to global warming. Furthermore, the occurrence of methane hydrates in
426 permafrost layers gives GHBPS more complex characteristics than the sediments
427 containing either ice (*e.g.* hydrate-free frozen soils) or gas hydrates (*e.g.* unfrozen
428 hydrate-bearing sediments). Such characteristics include fluid distribution in pores,
429 unfrozen water content, and ice/hydrate morphologies such as pore filling or grain
430 cementing, load bearing or micro frame structures. At present, there is little knowledge
431 about the physical properties of permafrost sediments that are saturated with variable ice,
432 hydrate, gas, and saline water (Farahani et al., 2021a, b; Hassanpouryouzband et al., 2020;
433 Yang et al., 2019).

434

435 4. Gas release from hydrate decomposition

436 Figure 2 illustrates the functioning of cold seep areas where the main methane-involving
437 biogeochemical processes are presented. It provides some numbers related to methane
438 transport from one compartment to another, storage, and transformations (genesis and
439 degradation) that will be discussed throughout this paragraph.

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445 Fig.2: Conception scheme of biogeochemical processes taking place at hydrate-bearing
446 setting on continental margins with estimated fluxes and stocks. Ranges of microbial
447 methane fluxes (methanogenesis, AOM and AeOM) were converted in mole-to-mass
448 from microbial rates reported and estimated from cited references in Table 2.

449

450 Methane venting at the seafloor widely occurs on shelves and continental margins. There,
451 methane and hydrogen sulfide are generated, transported and transformed within the
452 sediment, and part of the former is discharged into the water column (Suess, 2014). At
453 hydrate-bearing settings, the amount of methane that can potentially be released from

454 hydrate decomposition processes is variable and site-specific (Baumberger et al., 2022;
455 Braga et al., 2020; de la Cruz Vargas-Cordero et al., 2021; Ketzer et al., 2020; Marcon et
456 al., 2022; Marin-Moreno et al., 2015; Merle et al., 2021; Meurer et al., 2021; Riboulot et
457 al., 2018; Thatcher et al., 2013). However, judging from the number of articles published
458 in recent years, methane discharge from hydrate decomposition is not a sporadic event
459 (Fu et al., 2021; Hautala et al., 2014; Johnson et al., 2015; Phrampus et al., 2014;
460 Westbrook et al., 2009), and it is likely that this process will further increase in the
461 decades and centuries to come due to climate change.

462 Once released from the hydrates, methane undergoes several physical and
463 biogeochemical processes, starting from its dissolution in the pore water followed by its
464 oxidation (Figure 2). The remaining methane is transferred into the hydrosphere where it
465 undergoes similar processes. These processes act as efficient barriers preventing methane
466 transfer to the atmosphere. However, the efficiency of these barriers depends on multiple
467 factors (*e.g.* water depth, upwelling flowrate, local current, water stratification, *etc.*); and,
468 at the end, only a small portion of the total amount of methane released from NGHD
469 decomposition may reach the atmosphere (de Garidel-Thoron et al., 2004; Ruppel, 2011;
470 Ruppel and Kessler, 2017; Westbrook et al., 2009). Such a transfer occurs particularly in
471 the case of catastrophic releases.

472 In the following sections, we discuss in detail the variability of the seafloor methane from
473 hydrate decomposition, followed by the impact of such a leakage on the seafloor
474 ecosystems, seawater chemistry, and the conditions that lead to its transfer to the
475 atmosphere.

476 **i. Fluxes of methane discharged at the seafloor**

477 Due to the sensitivity of the NGHD to environmental changes, concerns have been
478 raised regarding a massive and prolonged release of methane from hydrate decomposition
479 and its consequences in terms of climate feedback, ocean acidification and ecosystem
480 vulnerability (Archer, 2007; Biastoch et al., 2011; Garcia-Tigreros et al., 2021). To
481 provide some answers to these societal issues, it is necessary to consider the fluxes of
482 methane released from hydrate decomposition. These fluxes are related to the rate of
483 hydrate decomposition, which depends on multiple factors. For outcropping hydrates, the
484 main factors are the flow velocity of the contacting methane-free water, the extent of
485 partial-sediment cover, and the presence of oil film or biofilms and non-methane guest
486 molecules. Considering hydrate-bearing sediments, the porosity, thickness and thermal
487 properties of the sediment are key factors affecting build-up and flow of methane-charged
488 fluids until its release at the seabed; and these flow-driving factors depend on both the
489 free-gas and hydrate saturation levels that evolve over the course of the decomposition.

490 Although hydrates are widespread on continental margins and in the permafrost,
491 and were intensively investigated in the former location, there are few natural sites where
492 hydrate decomposition has been quantified (Tables 1 and 2). The rate of hydrate
493 dissolution has been determined in the Cascadia Margin (Heeschen et al., 2005; Hester et
494 al., 2009; Lapham et al., 2010; Linke et al., 1994; Riedel et al., 2010; Römer et al., 2016;
495 Suess et al., 1999; Torres et al., 2002; Wilson et al., 2015), the Gulf of Mexico (Lapham
496 et al., 2014; Solomon et al., 2008) and the Monterey Bay (Rehder et al., 2004). The data,
497 either measured or calculated from other measured parameters, revealed high variabilities

498 not only from one region to another, but also within a given geological setting (Table 1).
499 Thus, for studies undertaken in the Cascadia Margin the dissolution rate ranges between
500 29 to 7.9×10^6 mmol.m⁻².yr⁻¹. Lowest values were obtained for deposits characterized by
501 thermogenic hydrates trapping multiple guest molecules, and/ or oil film covering the
502 outcropping specimens as the presence of such cover significantly mitigates the
503 dissolution rate. Lapham et al. (2014) showed that the aforementioned range of
504 dissociation rates is similar to previously reported laboratory measurements (Bigalke et
505 al., 2009). In the Gulf of Mexico, Lapham et al. (2014) measured a dissolution rate of
506 6.6×10^6 mmol.m⁻².yr⁻¹ for thermogenic hydrates. Such a rate for thermogenic hydrates is
507 in the same order of magnitude than the value of 4.7×10^6 mmol.m⁻².yr⁻¹ obtained by Hester
508 et al (2009) at Barkley Canyon, and it is in the same range than the rate range of 8- 9.5×10^6
509 mmol.m⁻².yr⁻¹ (141-167 cm.yr⁻¹) measured from synthetic methane hydrates brought at
510 the seafloor for *in situ* decomposition experiments (Rehder et al., 2004). Furthermore,
511 Bigalke et al. (2009) performed laboratory-based experiments at temperature and pressure
512 conditions within the hydrate stability field to investigate the dissolution of methane
513 hydrates in undersaturated seawater. The dissolution rate was found to be strongly
514 dependent on the friction velocity and the temperature of the seawater turbulent flow,
515 showing that hydrate dissolution in undersaturated seawater is a diffusion-controlled
516 process. They suggested that additional experimental work is required to address the
517 dissolution kinetics of non-pure methane hydrates, as both, the solubility limit and the
518 understanding of the molecule transfer processes at the phase boundary during
519 dissolution, are less well constrained. It is also important to note that the measurement
520 principles, the instruments and the computing approaches used for these aforementioned
521 studies of *in situ* hydrate-dissolution may differ from one research group to another,
522 leaving bias in the comparison.

523 Moreover, methane hydrate dissolution rate depends on the thickness of the
524 diffusion boundary layer and the methane concentration in water, and this second
525 parameter is in turn controlled by the solubility of methane. Rehder et al. (2004) showed
526 that gas hydrate dissolution rate depends on the difference between the local solubility in
527 the presence of gas hydrates and the local dissolved-phase concentration of the hydrate-
528 forming gas. Lapham et al. (2010 and 2014) measured the methane concentration in
529 sediments immediately surrounding outcropping deep-sea gas hydrates for two sites in
530 Barkley Canyon and the Gulf of Mexico, respectively. Their results showed that pore-
531 fluids are significantly under-saturated with respect to expected values at equilibrium
532 with methane hydrates, indicating that the hydrates should continue to dissolve. They also
533 showed that dissolution rates calculated from *in situ* methane concentration gradients are
534 significantly lower than those predicted for methane-undersaturated pore water in direct
535 contact with pure methane hydrates if equilibrium methane concentrations exist
536 immediately adjacent to the hydrate surface. Lapham et al. (2010) suggested that the
537 factors that retard hydrate decomposition in sediments here could be oil coating or
538 biofilms. This is supported by Wilson et al. (2015) who investigated how that sediment
539 cover can affect hydrate dissolution by means of field (at Barkley Canyon) and laboratory
540 measurements. The dissolution rate of outcropping thermogenic hydrates covered with
541 sediment was around 3.4×10^3 mmol.m⁻².yr⁻¹ (<1 cm.yr⁻¹), whereas that of synthetic
542 hydrates exposed directly to bulk water was an order of magnitude higher. Their results

543 are in agreement with dissolution-rate values calculated by Riedel et al (2010) for buried
544 hydrates at sites on the Clayoquot Slope of the Cascadia margin. Thus, as hydrate
545 dissolution is mainly controlled by diffusion, the presence of sediment slows the rate of
546 molecular diffusion via porosity/tortuosity effects.

547
548 For its part, hydrate dissociation has been investigated at the high latitude regions such as
549 the Western Svalbard (Ferre et al., 2020; Marin-Moreno et al., 2015; Marin-Moreno et
550 al., 2013; Thatcher et al., 2013; Westbrook et al., 2009), the U.S. Beaufort margin
551 (Phrampus et al., 2014) and the Laptev Sea (Chernykh et al., 2020); as these regions are
552 very sensitive to temperature increase due to climate change (Hassol, 2004; Hassol and
553 Corell, 2006). Investigations on the U.S. Atlantic margins also suggest that multiple
554 methane emission sites are partly related to hydrate dissociation due to the sea-bottom
555 temperature increase (Phrampus and Hornbach, 2012; Skarke et al., 2014; Weinstein et
556 al., 2016). However, as it is relatively usual to observe gas emissions associated with
557 hydrates at cold seeps, this coexistence makes it difficult to accurately assess the
558 contribution of hydrate dissociation in the total gas discharge. The dissociation process
559 requires bringing the pressure and temperature conditions of the deposits to the boundary
560 of the GHSZ to allow the formation of gas bubbles, and the common factor amongst the
561 aforementioned sites is the occurrence of intense gas emissions at the upper limit of the
562 GHSZ. Nevertheless, like for hydrate dissolution, Table 2 shows that the estimated
563 methane fluxes and the potential discharge trajectory are highly heterogeneous. In one of
564 the first estimates, Westbrook et al (2009) estimated that $\sim 20 \text{ Tg.yr}^{-1}$ of methane can be
565 released from hydrates over an area of $22,300 \text{ km}^2$ corresponding to a swath within 360-
566 400 m water depth nearly all around the Svalbard archipelago. Marin-Moreno et al (2013
567 and 2015) also investigated the sensitivity to climate change of the hydrate deposit present
568 in the Western Svalbard continental margin by considering a temperature-increase path
569 following either the lower (2.6) or the upper (8.5) cases of the Representative
570 Concentration Pathways (RCP). Their projection indicated that $0.029\text{-}0.053 \text{ Tg.yr}^{-1}$ of
571 methane can be released from hydrate dissociation over an area of $\sim 134 \text{ km}^2$ during the
572 next three centuries. Such a projection is also in agreement with the estimates of 22.4 Tg.
573 yr^{-1} proposed by Ferré et al (2020) along the 360 m isobaths at the Norwegian–western
574 Svalbard Margin. By extending to the swath width of Svalbard archipelago corresponding
575 to 400-500 m water depth (swath width complementary to the one considered by
576 Westbrook et al., 2009), Marin-Moreno et al. (2013 and 2015) showed that the methane
577 discharge may reach $1.7\text{-}9 \text{ Tg.yr}^{-1}$ over $41,400 \text{ km}^2$. Thus, their estimates of vulnerable
578 hydrates are more than twice smaller than the estimate obtained by Westbrook et al (2009)
579 for a surface area three times larger but located at deeper depth. Their results also showed
580 that hydrates remain stable beyond 500 m water depth. Such studies illustrate the
581 importance of the water depth (hydrostatic pressure) on the stabilization of hydrates. It is
582 very interesting to mention that they found that the choice of the RCP scenario has a small
583 impact on the total discharge. The calculated methane fluxes off Svalbard ranges from an
584 estimated average of $2 \text{ mmol.m}^{-2}.\text{yr}^{-1}$ (Thatcher et al., 2013) to $13.5 \text{ mol.m}^{-2}.\text{yr}^{-1}$
585 calculated by Marin-Moreno et al (2013). The fluxes calculated by Phrampus et al. (2014)
586 fall within this range of values for the U.S. Beaufort margin ($0.88\text{-} 4.4 \text{ mol.m}^{-2}.\text{yr}^{-1}$),
587 whereas values up to two and three order of magnitude higher were obtained by Skarke

588 et al (2014) and Chernykh et al (2020) on the U.S. Atlantic margin and in the Laptev Sea,
 589 respectively.

590 Moreover, it is important to recall that these methane fluxes are not constant over the
 591 time; and therefore, there is a need to take into account processes and seasonal changes
 592 that induced short to long-term variations that affect annual fluxes such as tides (Römer
 593 et al., 2016; Sultan et al., 2020), seismic activities (Fischer et al., 2013; Franek et al.,
 594 2017; Lapham et al., 2013) and temperature (Berndt et al., 2014; Ferre et al., 2020).

595 Table 1: Dissolved methane fluxes measured from field study of hydrate dissolution

Hydrate dissolution			
Location	Dissolution rate/cm.yr ⁻¹	Dissolution rate/mm ² .yr ⁻¹	Reference
Monterey Bay (Monterey Canyon- MontC)	187 [*] -220 [*]	1.07 – 1.26 x 10 ⁷	Rehder et al. (2004)
Gulf of Mexico (Green Canyon- BC and Mississippi Canyon- MC)	15 [#] (GC)	8.57 x 10 ⁵	Lapham et al. (2014)
	0.001 ^{#π} (MC)	63	Lapham et al. (2010)
Northern Cascadia Margin: (Barkley Canyon- BC), South Hydrate ridge, Hydrate ridge (regional average- RA), Clayoquot Slope (CS)	62 [#] and 104 (BC)	4.7 x 10 ^{6#} and 7.9 x 10 ⁶	Hester et al. (2009); Lapham et al. (2014)
	0.0005/ 0.03 ^{#π} (BC)	29/ 1800	Lapham et al. (2010)
	3.5 [#] (BC)	0.2 x 10 ⁶	Lapham et al. (2010)
	0.06- 0.33 (CS)	3.43 x 10 ³ - 1.88 x 10 ⁴	Romer et al. (2016) Riedel et al. (2010)
	0.06 [#] (BC)	3.4 x 10 ³	Wilson et al. (2015)

597 # for complex thermogenic hydrates, * for synthetic hydrates, ^π for buried hydrates (otherwise it is outcropping hydrates)

598
 599
 600 Table 2: Free methane fluxes measured from field study of hydrate dissociation and over a hydrate deposit

Hydrate dissociation		
Location	Methane flux from gas discharged at hydrate sites	Reference
Svalbard Continental margin; West Svalbard	0.027 Tg yr ⁻¹ (30-km-long plume-area) potentially 20 Tg yr ⁻¹ (22300 km ²)	Westbrook et al. (2009)
	2 mMol.m ⁻² .yr ⁻¹	Thatcher et al (2013)
	0.0053-0.029 Tg yr ⁻¹ over the next 300 years; e.g 2.5–13.5 mol yr ⁻¹ m ⁻²	Marín-Moreno et al (2013)

	0.0004–0.0015 Tg yr ⁻¹ and 0.0017-0.0045 Tg yr ⁻¹ over the next 100 years	Marín-Moreno et al (2015)
	22.4 Gg yr ⁻¹ (along the 360 m isobaths at the Norwegian–western Svalbard Margin); e.g 2120 mol m ⁻¹ yr ⁻¹	Ferré et al (2020)
Hudson Canyon	4.375- 17.5 Mmol yr ⁻¹ for the extended area	Weinstein et al (2016)
From Cape Hatteras to Georges Bank	0,938- 5,625 Mmol yr ⁻¹ e.i. 25-150 mol m ⁻² yr ⁻¹	Skarke et al (2014)
Carolina rise off the east coast of North America	2.5 Gt of methane stored as GH would be destabilized	Phrampus and Hornbach (2012)
U.S. Beaufort margin	0.44-2.2 Gt of methane would be released e.i. 0.88- 4.4 mol m ⁻² yr ⁻¹	Phrampus et al. (2014)
Laptev Sea	9360- 11 170 mol.m ⁻² .yr ⁻¹	Denis Chernykh et al (2020)

ii. Microbial processes and methane discharge at the seafloor

Currently global methane emissions are about 500-600 Tg per year (Ehhalt et al., 2001; Saunio et al., 2020) (see section 4.iv “Transfer to the atmosphere”), with around 70% (350-400 Tg) of which is due to methanogenesis from a wide variety of habitats including anoxic seafloor ecosystems (Conrad, 2009). Around 40% of the methane produced by methanogenesis escapes to the atmosphere (Lyu et al., 2018). In marine sediments, microbial methanogenesis produces methane through the terminal anaerobic breakdown of organic matter deposited on the seafloor (Parkes et al., 2014; Wellsbury et al., 2000) (Fig.2). Estimations suggest that 3-18% of the buried organic carbon within the seafloor is converted to CH₄ by methanogenesis in continental margin (Egger et al., 2018; Xu et al., 2022). Recent estimates from Xu et al. (2022) indicates that the global methanogenesis budget approximates 13 Tg of methane per year. Methanogenic activities have been observed from surface to deep sub-seafloor sediments (Table 3).

Table 3: Rates of methanogenesis, anaerobic methanotrophy (AOM) and aerobic methanotrophy (AeOM) in various microbial marine habitats.

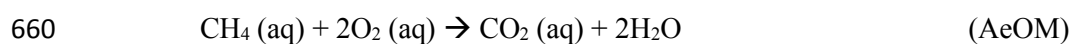
	Methanogenesis	Anaerobic methanotrophic oxidation	Aerobic methanotrophic oxidation
Sediments:	0.02 to 30 nmol cm ⁻³ d ^{-1*****}	5.1 x 10 ⁻⁸ to 7.0 x 10 ⁴ nmol cm ⁻³ d ^{-1****}	3.4 x 10 ⁻⁸ to 1.1 nmol cm ⁻³ d ^{-1****}
-with hydrate	0.1-2.73 nmol cm ⁻³ d ^{-1***} < 4.25 x 10 ⁻⁶ nmol cm ⁻³ d ^{-1*}	10 ² to 5x10 ³ nmol cm ⁻³ d ^{-1*****}	0-12 mmol m ⁻² d ^{-1*****}
-without hydrate	0.015-0.62 nmol cm ⁻³ d ^{-1**}	0.001 to 10 ⁴ nmol cm ⁻³ d ^{-1*****}	0-289 mmol m ⁻² d ^{-1*****}

*Estimation of microbial methane production rates in deep marine sediment at hydrate ridge is based on representative species of methanogen *Methanoculleus submarinus* starved in culture with Biomass

621 Recycle Reactor coupled to quantitative PCR of *mcrA* gene abundances (Colwell et al., 2008). **Vigneron
622 et al., 2015, Cold seeps at Guaymas Basin in Sonora margin; ***Wellsbury et al., 2000, Blake Ridge; ****(Gao
623 et al., 2022). ***** (Parkes et al., 2007) and (Newberry et al., 2004) ***** Boetius and Wenzhöfer, 2013. The
624 < sign indicate underlying uncertainties.

625 The formation of methane involves a group of strictly anaerobic archaea called
626 methanogens. However, smaller amounts of methane are produced from
627 methylphosphonates in oxic waters by both phytoplanktons with mainly Cyanobacteria
628 and Thaumarchaea (Bižić et al., 2020; Klintzsch et al., 2019; Metcalf et al., 2012).
629 Methanogens use various low-molecular-weight catabolic substrates for biosynthesis of
630 methane, and thus the rate of methanogenesis can be difficult to estimate. The Redox
631 reaction of dihydrogen (H₂) with carbon dioxide (CO₂) and/or with formate are
632 considered as the main substrates for methanogenesis in marine sediments (Newberry et
633 al., 2004; Parkes et al., 2007). However, it has been shown that acetate can become the
634 dominant substrate in deeper sediments (Reeburgh, 2007). Methanogens also performed
635 methanogenesis with methylated compounds. Methylophilic methanogenesis may be an
636 important methane source in coastal shallow sediment (Conrad, 2020). Acetate and other
637 potential substrates for microbial methanogenesis could derive from either thermal
638 alteration or biological degradation of organic matter. Recent developments in high
639 throughput sequencing techniques have revealed an unexpected larger diversity within
640 the methanogens and suggest that this metabolic pathway is one of the most ancient on
641 Earth (Vanwonterghem et al., 2016; Wang et al., 2021). Metagenome assembled genomes
642 (MAGs) of potential methanogens suggest the acquisition of both novel functions in
643 activation of alternative catabolic substrates such as short and medium chain of alkanes
644 and volatile fatty acids (Laso-Pérez et al., 2019; Wang et al., 2021), and in derivation of
645 cellular energy from methoxylated aromatic compounds from lignin occurring in coal bed
646 (Inagaki et al., 2015; Kurth et al., 2021). The overall rates of methanogenesis in marine
647 sediments range from 0.02 to 30 nmol cm⁻³ d⁻¹ (Table 3) (mole-to-mass conversion is 0.32
648 to 480 ng cm⁻³ d⁻¹ reported in Figure 2). However, microbial rates are limited and
649 restricted to discrete habitats and mostly obtained in incubation with radiotracers in
650 laboratory conditions and ambient pressure. Thus, the extrapolation to estimates of global
651 microbial methane production and consumption per year may be subjected to high
652 uncertainties. All the methanogenesis pathways lead to the production rate of 5-33 Tg of
653 methane yr⁻¹ in continental marine sediment at global scale (Wallmann et al., 2012).
654 However, the transfer of this amount of methane generated from the seabed along
655 continental margins to the atmosphere is efficiently buffered and controlled by both
656 aerobic (AeOM) and anaerobic oxidation of methane (AOM), respectively at and below
657 the oxic-anoxic interface of the sediment and water columns (Reeburgh, 2007) (Fig.2).
658 These two oxidation processes can be expressed as follows:

659



662 AeOM occurs into the geosphere at cold seeps and hydrothermal fields from a symbiotic
663 association between specific methane-oxidizing bacteria with marine animals (Petersen
664 and Dubilier, 2009). In the water column, AeOM represents the final sink for methane
665 before reaching the atmosphere. Within the anoxic sediment, AOM is a microbial-
666 mediated process that occurs within the geochemical horizon commonly called the
667 sulfate-methane transition zone (SMTZ) where upward diffusion of methane reacts with
668 downward diffusing seawater sulfate (Borowski et al., 1996, 1999; Gao et al., 2022;
669 Hinrichs and Boetius, 2002; Pohlman et al., 2008). Laboratory experiments from natural
670 samples have shown that the overall rates of AOM in marine sediments coupled to sulfate,
671 metal oxides and nitrite/nitrate as electron acceptors range from 5.1×10^{-8} to 7.0×10^4
672 $\text{nmol cm}^{-3} \text{d}^{-1}$ (Gao et al., 2022, Table 3) (mole-to-mass conversion is 81.6×10^{-8} to $112 \times$
673 $10^4 \text{ ng cm}^{-3} \text{d}^{-1}$ reported in Figure 2). Predicted global rates of AOM based on the
674 regression model with diffusive fluxes of CH_4 suggest that 45-61 Tg of methane are
675 oxidized annually; corresponding to ~3-4% of the organic debris deposited on the seabed
676 (Egger et al., 2018). On a global scale, 8 to 65 Tg of methane per year bypasses the two
677 aforementioned sedimentary biofilters and is released in the water column (Saunois et al.,
678 2016; Steinle et al., 2015). This estimate is not restricted to hydrate decomposition only
679 as it is difficult to decipher, but involves several methane pools, including free methane
680 generated either from thermal cracking at deep depth or shallow methanogenesis.

681 Methane released from hydrate decomposition affects the transport of chemical species
682 within the sediment overlaying the deposits by generating a continuous carbon flux that
683 represents an energy supply for the benthic chemosynthetic assemblages (Niemann et al.,
684 2006). Such an energy supply stimulates the seepage-associated ecosystems located in
685 the sediment as well as in the water column (Boetius and Suess, 2004; Marcon et al.,
686 2014; Sahling et al., 2002; Suess, 2014) and gives rise to the following consequences:

- 687 • **Formation of interface horizons shaped by redox processes.** The microbial
688 processes and the community diversity that characterized them are often
689 stimulated at discrete subsurface geochemical interfaces (Parkes et al., 2014),
690 creating a zonation within the sedimentary column. From a biogeochemical
691 viewpoint, anaerobic methane oxidation, methanogenesis from acetate and
692 Hydrogen/ Carbon Dioxide, acetate oxidation, sulfate-reduction, and bacterial
693 productivity were stimulated in both the hydrate and the free-gas sediment
694 horizons beneath compared to hydrate-free sediments (from 1.5 to 15 times
695 higher) at the Black Ridge setting in NW Atlantic Ocean (Wellsbury et al., 2000)
696 and surface sediment of the Cascadia Margin in the North Pacific Ocean (Cragg
697 et al., 1995); and distinct microbial communities dominated the former
698 sedimentary environments. Estimations of microbial methane production rate
699 ranged from 0.015 to $0.62 \text{ nmol cm}^{-3} \text{d}^{-1}$ in sediments without hydrate to 0.1 to
700 2.73 in methane hydrate bearing sediments (Table 2) (mole-to-mass conversion
701 of methane production rates in hydrate bearing sediments were 1.6 to 43.68 ng
702 $\text{cm}^{-3} \text{d}^{-1}$ reported in Figure 2). Thus, the cell numbers as well as the microbial
703 activities involved in the methane cycle (methanogenesis and anaerobic

704 methanotrophy) strongly increase within the sedimentary geochemical-horizons,
705 thus increasing the amount of hydrates and associated free gas (Dickens and
706 Quinby-Hunt, 1997; Wellsbury et al., 2000). At the Black Ridge hydrate-setting,
707 the total microbial number is significantly higher around the Bottom Simulating
708 Reflector (BSR), especially beneath it within the free-gas or dissolved-gas
709 occurrence zone (Wellsbury et al., 2000). Indeed, the potential rates of microbial
710 activity increase at the interface between the BSR and the free-gas zone beneath,
711 mainly towards the free-gas zone (Cragg et al., 1996; Cragg et al., 1995;
712 Wellsbury et al., 2000). Thus, the presence of free gas associated with hydrate
713 deposits rather than free gas-depleted hydrates, impacts on the microbial activities
714 including methane and acetate metabolisms. At the SMTZ, pore water methane
715 and sulfate are consumed through the AOM by syntrophic consortia of anaerobic
716 methanotroph archaea (ANME) and sulfate reducing bacteria (SRB). The depth
717 of the SMTZ is strongly affected by the intensity of the methane flux, while its
718 state (steady or transient) is rather linked to the flux variations over time
719 (Borowski et al., 1999; de Prunelé et al., 2017; Jørgensen and Kasten, 2006;
720 Kasten et al., 2003). A recent investigation of site located at the rim of a hydrate-
721 bearing zone off Svalbard has shown that nonvertical methane transport can
722 significantly affect the gas migration pathway, leading to the occurrence of double
723 SMTZs and unusual microbial distribution within the sediment (Treude et al.,
724 2020). Previous studies have shown that the settlement of the SMTZ starts around
725 100 to 1000 years after the inception of gas hydrate dissociation (Argentino et al.,
726 2021; de Prunelé, 2015; de Prunelé et al., 2017; Hong et al., 2016; Pape et al.,
727 2020; Sultan et al., 2016). The SMTZ depth is controlled by the diffusive fluxes
728 of methane and sulfate, which depend on continuous decrease in the reactivity of
729 organic matter with the increasing sediment depth and therefore the age of the
730 sediment (Borowski et al., 1999). The sedimentation rate and the associated
731 organic matter burial flux are the key control factors to define the depth of the
732 SMTZ (Egger et al., 2018). These two factors depend in turn on the water depth,
733 the distance from land and surface biological productivity in the overlying water
734 column. Human alteration of nutrient loading and oceanographic regimes due to
735 global warming can also impact on the SMTZ distribution in sediment (Egger et
736 al., 2018).

- 737 • **Ecosystems-hosted methanotrophy.** Microbial consortia of anaerobic
738 methanotroph archaea and sulfate-reducing bacteria are not restricted to gas
739 hydrate-bearing sediments (Boetius et al., 2000), but are widely distributed in
740 other marine environments including cold seeps (Hinrichs et al., 1999; Vigneron
741 et al., 2013), mud volcano (Lazar et al., 2011), deep hypersaline sediments (La
742 Cono et al., 2011) and hydrothermally impacted sediments (Holler et al., 2011;
743 Teske et al., 2002). The major controlling factors for the ANME population
744 distribution are the availability of methane and sulfate, or known alternative
745 electron acceptors such as nitrate, iron or manganese (Wallenius et al., 2021).
746 ANME is believed to use a process of “reverse methanogenesis” biochemical
747 pathway and are phylogenetically related to methanogenic lineages. The doubling

748 time of ANME/SRB consortia, estimated to approximately seven months under
749 controlled *in vitro* conditions (Nauhaus et al., 2007), suggests that the set-up of an
750 efficient AOM microbial filter in sediments would be on the order of decades
751 (James et al., 2016). The coupled process of AOM and sulfate-reduction increases
752 alkalinity, promoting the precipitation of authigenic carbonate by reacting with
753 the seawater calcium (Luff and Wallmann, 2003). This authigenic carbonate
754 precipitation results in a decreasing of the sediment porosity that changes the fluid
755 flow pattern, and in turn affects the supply of methane at the SMTZ (Steeb et al.,
756 2014). At Hydrate Ridge in the Cascadia margin subduction zone, carbon volume
757 estimations suggest that 14% of the oxidized methane is sequestered into this
758 seafloor mineral. Thus, carbonate mounds may represent a significant methane
759 sink in deep-sea (Marlow et al., 2021; Marlow et al., 2014). Natural and
760 anthropogenic methane seepages into an oxygenated water column stimulate the
761 distribution, abundance and activity of aerobic methanotrophic bacterial
762 populations (Rogener et al., 2018; Steinle et al., 2015). The AeOM reaction
763 hinders the methane transfer to the atmosphere by converting it into biomass or
764 dissolved inorganic carbon; which in turn could affect the pH in the ocean
765 (Biaostoch et al., 2011). Estimations of aerobic methanotrophic oxidation rates in
766 oxygenated sediments ranged from 3.4×10^{-8} to $1.1 \text{ nmol of CH}_4 \text{ cm}^{-3} \text{ d}^{-1}$ (Table
767 2, Gao et al., 2022) (mole-to-mass conversion is 54.4×10^{-8} to $17.6 \text{ ng of CH}_4 \text{ cm}^{-3} \text{ d}^{-1}$
768 reported in Figure 2).

- 769 • **Temporal and spatial dynamics of (micro)-biological communities.** The
770 methane concentration and flux strongly affect the diversity of the chemosynthetic
771 communities such as sulfur bacterial mats, tubeworms, and clams that are settled
772 on the seafloor (Fischer et al., 2012; Pop Ristova et al., 2012). These
773 chemosynthetic seep fauna are therefore bioindicators of AOM activity and
774 methane flux. Thus, macrofauna colonies such as vesicomyid clam populating the
775 *Beggiatoa* mats (filamentous microbes) settled on methane hydrate-bearing
776 sediments consist nearly exclusively of endemic species and are found in higher
777 density at seeps and hydrothermal vents compared to non-seep environments
778 (Sahling et al., 2002). The methane fluxes may vary on very large spatial scales
779 as a consequence of factors such as the variation of the rate of hydrate
780 decomposition or carbonate precipitations, or after a submarine mud eruption, a
781 continental slope sliding, and even an earthquake. Thus, large expulsions of fluids
782 and mud from active submarine volcanos may strongly affect the distribution of
783 communities; sometimes causing their extinction. However, long-term
784 observation after such large expulsions of fluids also indicates a resilience of the
785 ecosystem to recover the main microbial function. For instance, the re-
786 colonization of complex pioneer microbial populations composed of aerobic and
787 anaerobic methanotrophs were observed several years after a subsurface mud
788 eruption at the active center of the Håkon Mosby mud volcano (Ruff et al., 2019).

790 Predicting how climate change will affect the aforementioned microbial processes
791 remains elusive. Several studies have claimed that microbial methane production may be

792 enhanced by global warming. In fact, methanogens are active in most habitats where
793 temperature ranges from 0 to 122°C (Hoehler et al., 2018). In marine sediments, the
794 methanogenesis process is restricted to either the sediment layer deep below the SMTZ
795 (meter to tens of meters) and used acetate and hydrogen, or in surficial sediment (<1 m)
796 where non-competitive substrates such as methanol and other methylated compounds are
797 available. Interestingly, the coexistence of sulfate-reduction and methanogenesis also
798 occur in shallow sediment when methylotrophic pathway to produce methane takes place
799 (L'Haridon et al., 2014). Methanogen distribution may be limited by competition for
800 acetate and dihydrogen and are rapidly out-competed by sulfate-reducers for these same
801 substrates. In shallow sulfate-rich sediments, methanogens utilizing non-competitive
802 substrates such as methylated amines could meet the substrate requirements of sulfate-
803 reducing bacteria. Thus, variation of *in-situ* temperature in marine sediments can strongly
804 impact methane production pathways via biochemical, bioenergetic and/or ecological
805 mechanisms (Conrad, 2020; Hoehler et al., 2018). Temperature increase positively
806 impacts methanogenesis activity (Maltby et al., 2018), resulting in higher methane
807 production with potential higher emission budget to the hydrosphere. In fact, a warming
808 of bottom water may result in increasing the amount of methane contained within the
809 shallow sediments and eventually lead to the shoaling of the SMTZ in vast areas of the
810 continental shelf. Even if representative isolated microbes grow within a wide range of
811 temperatures in the laboratory, changing global temperatures may also shift the diversity
812 of the microbial community towards more responsive and efficient microbial
813 metabolisms at higher *in situ* temperature.

814 Moreover, the lifetime of methane in the water column is longer when it is transferred
815 from the seafloor as bubbles, partly because microbes use dissolved methane. The aerobic
816 oxidation of methane by free-living and symbiont-associated bacteria occurs on the
817 seafloor and within water column at sites where free methane is released (Levin, 2018;
818 Steinle et al., 2015; Sweetman et al., 2017). Accordingly, hydrate dissociation may affect
819 the water column even more than hydrate dissolution. If the methane hydrate dissociation
820 is fast enough and releases large quantities of gas, part of it will escape the function of
821 the microbial filter, as it will not be accessible by the microorganisms. However,
822 additional methane loss prior to reaching the atmosphere occurs as bubble stripping, *i.e.*
823 dissolution in the surrounding water, or replacement by oxygen, nitrogen or other
824 dissolved gases in the water masses. Such a gas exchange may not be complete over a
825 short distance, and thus a significant methane fraction would be transferred to the
826 atmosphere at shallow water depth. The amount of methane bypassing sedimentary
827 benthic biofilters, entering in the water column, and transfer to the atmosphere can vary
828 widely depending on the type of water and specific environmental conditions. For coastal
829 and marine environments, estimation is challenging due to complex interactions between
830 sediments, water column and the atmosphere. Therefore, it is difficult to provide depth
831 threshold from which sedimentary methane is transferred to the atmosphere. McGinnis et
832 al. (2006) studied methane emissions in the Black Sea and showed that significant transfer
833 to the atmosphere occurs at water depth shallower than 100 m. Other studies highlighted
834 the coastal region located in water depth ranging between ~0 and 50/ 75m, where gas

835 hydrates are not stable, as being by far the more important contributor of sedimentary
836 methane to the atmosphere (Mao et al., 2022; Michel et al., 2021; Römer et al., 2021;
837 Weber et al., 2019). In a recent study, Joung et al. (2022) investigated the transfer of
838 methane from the seafloor to the sea surface along the US margins at seep areas including
839 hydrate-bearing sites, both in the Atlantic and Pacific sides. Their results showed no
840 occurrence of such a vertical transfer at water depth deeper than 430m, which is shallower
841 than the upper boundary of the hydrate-stability field (Joung et al., 2020; Joung et al.,
842 2022). In addition, during the aerobic oxidation of methane in the water column, bacteria
843 produce biomass and/or carbon dioxide, which latter is dissolved in the seawater. Large
844 production of carbon dioxide may contribute to the decrease of the water pH, and lead to
845 local ocean acidification as mentioned in section 4.i. Biastoch et al (2011) estimated that
846 the oxidation of methane discharged in the Arctic Ocean within the next centuries will
847 result in a drop of 0.25 pH units and a local decrease of up to 25% of the oxygen
848 concentration. A bloom of aerobic methanotroph communities in response to the large-
849 scale methane release may happen, creating a vertical stratification of aerobic
850 methanotrophs consuming the dissolved oxygen and depleting the amount of oxygen in
851 the water column. Such a microbial bloom has been observed from the Deep Water
852 Horizon blowout in the northern Gulf of Mexico, which has increased methane oxidation
853 rates in the water column compared to natural methane seepage (Rogener et al., 2018).

854 **iii. Marine methane transfer to the atmosphere**

855 After CO₂, atmospheric methane is the second most important climate-forcing trace gas
856 driven by anthropogenic sources. Its burden in the atmosphere has drastically increased
857 by 150% since the pre-industrial era and is currently rising at a high rate (Nisbet et al.,
858 2014; Nisbet et al., 2016; Platt et al., 2018). Our understanding on the respective
859 contribution from its different natural sources and sinks remains incomplete (Kirschke et
860 al., 2013; Saunio et al., 2016; Saunio et al., 2017; Saunio et al., 2020). Top-down and
861 bottom-up studies show that natural processes emit between 218 and 371 Tg CH₄ yr⁻¹ on
862 average, respectively (Saunio et al., 2020).

863 Aquatic methane enters the atmosphere either as bubbles rising from the seafloor or by
864 diffusion from saturated waters across the sea-air interface. Early estimates by Lambert
865 & Schmidt (1993) provided a marine source to the atmosphere of 3.5 Tg CH₄ yr⁻¹ and
866 predicted that the open ocean might become a sink at atmospheric concentrations
867 equivalent to today's concentrations (Lambert and Schmidt, 1993). That prediction did
868 not materialize. Survey of the global ocean ΔCH₄ data combined with estimates of
869 ebullitive fluxes found a range of 6-12 Tg CH₄ yr⁻¹ (Weber et al., 2019). It is generally
870 admitted that seafloor emissions reach the atmosphere only at shallow depth (continental
871 shelf and upper slope), whereas in the open ocean methane flux correlates with net
872 primary production and would therefore be mainly produced *in situ* by organic matter
873 cycling (Weber et al., 2019). Rosentreter et al. (2021) further extended the study of
874 Weber et al (2019) to sources from coastal ecosystems, and estimated a median flux of
875 8.4 Tg CH₄ yr⁻¹ (interquartile range 4.8–28.4 Tg CH₄ yr⁻¹) (Rosentreter et al., 2021)
876 (Fig.2). These estimates can separate emissions from seafloor reaching the atmosphere

877 on one side with *in situ* production on the other side. However, atmospheric estimates are
878 based on measurements that do not discriminate whether sedimentary methane has been
879 buffered in methane hydrates that eventually decomposed or is a direct emanation of
880 methane from seeps.

881 Focusing on submarine seeps only, Etiope et al. (2019), compiling the available literature,
882 estimated the methane transfer from the geosphere to the atmosphere at 3.9 Tg CH₄ yr⁻¹
883 with a maximum potential value of 9 Tg CH₄ yr⁻¹. In this synthesis, all sources were
884 located at depths shallower than 500 m. The main emitting area is the East Siberian Arctic
885 Shelf with 2 Tg CH₄ yr⁻¹ (range 0-4 Tg CH₄ yr⁻¹, (Berchet et al., 2016)). Etiope et al.'s
886 estimate is nearly twice lower than a previous geological estimates of 20 Tg CH₄ yr⁻¹
887 (Judd, 2004; Kvenvolden et al., 2001). There, offshore direct sources account for 5-20 Tg
888 CH₄ yr⁻¹ and hydrate dissociation around 0-5 Tg CH₄ yr⁻¹.

889 Numerous investigations on the consequence of methane release from hydrate
890 decomposition during past climate change such as the Paleocene-Eocene thermal
891 maximum argued that these events have contributed to an increased injection of methane
892 into the atmosphere that moderately accelerated climate change (Dickens et al., 1997;
893 Kennett et al., 2003; McInerney and Wing, 2011; Norris and Röhl, 1999). Such an impact
894 involves a huge amount of methane passing through the entire water column to reach the
895 atmosphere. Several modelling and simulation studies explored the interactions between
896 hydrate dissociation and climate change (Archer, 2007; Ruppel and Kessler, 2017 and
897 references therein). Most of these studies found moderate release of methane to the
898 atmosphere under the worst climate scenario (RCP 8.5) (Biajoch et al., 2011; Hunter et
899 al., 2013; Kretschmer et al., 2015). Kretschmer et al. (2015) estimated that up to 4.73 Tg
900 CH₄ yr⁻¹ could be released on average, ignoring potential oxidation in the water column.
901 This is comparable to today's global ocean methane sources, a minor term in the global
902 annual methane budget. They also noted the importance of the uncertainty associated with
903 the inventory of hydrates in the sediment within the GHSZ. Serov et al. (2017) highlighted
904 the importance of understanding the post-glacial history of present-day gas hydrate
905 provinces to better predict the trajectory of their destabilization. It is also important to
906 assess the role of anthropic activities on the vulnerability of NGHDs. Indeed, although
907 several studies investigated the monitoring of marine methane emissions from oil and gas
908 wells (Böttner et al., 2020), there is no study related to human-induced destabilization of
909 NGHD from seafloor or sub-seafloor resource extraction. To our knowledge there is no
910 literature dealing with the quantification of methane leaks during production tests from
911 hydrates and its fate, nor on its impact on the associated ecosystems over time. As
912 mentioned previously, the water column acts as a relatively efficient but still poorly
913 characterized oxidative-barrier to prevent seafloor methane from reaching the
914 atmosphere. Therefore, continuous efforts driven by field-based quantitative approaches
915 are needed to achieve an in-depth understanding of the transfer and fate of gases in the
916 different Earth compartments, as well as to identify under which circumstances marine
917 methane reaches the atmosphere. Thus, the assessment of the physical and
918 biogeochemical processes related to methane in the water column and its probability to
919 reach the atmosphere remains timely and challenging.

1 920 Besides, like seafloor methane emissions, methane transfer from the ocean to the
2 921 atmosphere is influenced by environmental factors such as seasonality. Experiments
3 922 showed that strong vertical density gradients in the water column developed during the
4 923 warm season might limit the ascent of methane gas to the sea surface and its release into
5 924 the atmosphere. During winter, the vertical mixing in the water column is enhanced and
6 925 consequently more methane is released into the atmosphere (Gentz et al., 2014; von
7 926 Deimling et al., 2011). Such a seasonal effect could also explain variations in the extent
8 927 of the gas hydrate stability zone (GHSZ) on the Svalbard margin and the distribution of
9 928 gas flares in the water column (Ferre et al., 2020; Riedel et al., 2018), but the net methane
10 929 budget still needs to be estimated. Apart from seasonal shifting, meteorological events
11 930 such as storms (Shakhova et al., 2014), chemical gradients and geological site-specific
12 931 parameters (Ryerson et al., 2011; Schmale et al., 2010a; Schmale et al., 2010b; Yvon-
13 932 Lewis et al., 2011; Zhang, 2003) can also facilitate methane transfer from the sea to the
14 933 atmosphere.
15 934 As mentioned in section 4.ii, the methane injected in seawater is then submitted to
16 935 dissolution, gas exchange within the bubbles and oxidation. Modeling of gas bubble
17 936 ascent can predict the evolution of bubble sizes and the fractions of methane gas reaching
18 937 the sea surface, and then transferred to the atmosphere under idealized conditions
19 938 (McGinnis et al., 2006; Ruppel and Kessler, 2017). For a water depth of 100m, methane
20 939 from only bubbles with a diameter larger than 2cm at the seafloor can reach the
21 940 atmosphere (Ruppel and Kessler, 2017). Furthermore, the internal dynamics of large-
22 941 scale bubble plumes and the interactions with ambient bottom currents and density
23 942 stratification adds further complexity that is not yet fully resolved by numerical models
24 943 (Leifer et al., 2015; Schneider von Deimling et al., 2015). Such an achievement
25 944 necessarily requires a sound knowledge, not only of its micro-biological state of the local
26 945 water column, but also its the physical properties (*i.e.* currents, temperature). Hence,
27 946 hydro-acoustic measurements and more additional field data are required to monitor and
28 947 forecast the fate of methane gas released at the seabed to improve the existing models.
29 948 Thus, in order to reliably evaluate methane contribution from hydrate decomposition to
30 949 the global atmospheric budget, it is necessary to (1) develop new bio-geochemical tools
31 950 to better identify and decipher all oceanic sources and understand the processes that
32 951 degrade this molecule in the water column, and (2) intensify observations and monitoring
33 952 of sites by increasing the number of devoted cruises as well as the deployment of
34 953 autonomous or remotely operated systems to achieve reliable estimates of the methane
35 954 fluxes across the interfaces sediment (geosphere)/ seawater (hydrosphere)/ atmosphere.
36 955 Once reaching the uppermost water layer, the diffusive transfer to the atmosphere is
37 956 strongly affected by ocean state and weather conditions (James et al., 2016; Wanninkhof
38 957 et al., 2009). Wind speed modifies the exchange velocity across the interface. Wave
39 958 breaking further enhances gas exchange through bubble formation. Strong winds during
40 959 storms combine effects of increased water column vertical mixing and gas exchange. At
41 960 the high latitudes, sea ice acts as a barrier preventing the transfer of methane to the
42 961 atmosphere (Kort et al., 2012), and accordingly the reduction in sea ice is expected to
43 962 significantly increase the annual total methane flux to the atmosphere (He et al., 2013).

1 963 Currently, attempts to accurately measure atmospheric methane inputs from NGHDs,
2 964 especially in the Arctic where hydrate-bearing sediments are encountered at shallow
3 965 locations, failed due to widely varying numbers of factors that need to be considered.
4 966 Using a diffusive model (Wanninkhof, 1992), Thornton et al., (2016) estimated a
5 967 maximum flux range of 68-87 mmol.m⁻².yr⁻¹ in the Laptev Sea and East Siberian Arctic
6 968 Shelf. Comparably, using inverse modelling of atmospheric measurements Myhre et al.
7 969 (2016) found a source not higher than 120 mmol.m⁻².yr⁻¹ off the coast of Svalbard on a
8 970 known methane seep area (Myhre et al., 2016). When including the impact of storm,
9 971 Shakhova et al. (2014) proposed higher annual average estimate reaching up to 506
10 972 mmol.m⁻².yr⁻¹ for the East Siberian Arctic Shelf. For the same area, Berchet et al. (2016)
11 973 used an atmospheric inversion method and propose that atmospheric observations in the
12 974 Arctic are compatible with a mean source ranging between 0-134 mmol.m⁻².yr⁻¹.
13 975 The estimates are different from each other from one site to another as well as from one
14 976 method to another for the same site, and such a result indicates that several challenges
15 977 still remain for a better quantification of these fluxes at the sea-air interface. Indeed, the
16 978 aforementioned studies assumed an atmospheric diffusive flux and estimated it through
17 979 the gradient method, which does not represent adequately ebullitive fluxes. Atmospheric
18 980 inversions as top-down methods are unable to unambiguously quantify specifically
19 981 methane released from hydrate dissociation distinctly from that originating from other
20 982 marine sources. They are further limited by model transport error, embedded sources and
21 983 atmospheric data coverage. However, top-down methods may provide valuable upper
22 984 constraints to marine emissions (Berchet et al., 2016). Estimates from closed chamber
23 985 accumulation could be more accurate but is restricted to a very small spatial coverage,
24 986 weakening the spatial representativity and therefore remain difficult to operate in open
25 987 ocean and large areas. The eddy covariance technique deployed on ships is technically
26 988 adequate (Thornton et al., 2020) but challenging in marine environment, especially
27 989 because it requires a careful correction of the measured high-frequency, three-
28 990 dimensional wind for ship motion and attitude (Edson et al., 1998). Measurements are
29 991 also needed to characterize the sub-seafloor origin of methane, and ground-checked on
30 992 whether or not it has been buffered in hydrates.
31 993

32 994 **5. Strategy to tackle environmental challenges related to hydrate** 33 995 **decomposition**

34 996 In the previous sections, we have shown the scarcity of methane flux measurements at
35 997 hydrate-bearing sites and the need to intensify such measurement campaigns. We also
36 998 stressed the need to undertake investigations at different spatial and temporal scales to
37 999 gain insights into the impacts of hydrate decomposition on the seafloor stability and the
38 1000 fate of the permafrost. The outcomes of such studies will help to clarify the evolution path
39 1001 of NGHDs and its impact upon climate change or/ and human activity, and how this
40 1002 evolution path affects the ecosystems colonizing the sediment as well as the water
41 1003 column. Addressing the aforementioned environmental challenges requires both the
42 1004 definition of an environmentally sound monitoring strategy and the assessment of the
43 1005 legal framework regulating the release of gases and particles in the water column. The

1006 current legal framework must be adjusted to account for the specific environmental risks
1007 associated with natural gas hydrate dynamics. Therefore, the following actions should be
1008 considered:

- 1009 ● Assess how slope stability may be compromised by either global warming or gas
1010 production from hydrates under different geological boundary conditions;
- 1011 ● Identify suitable precursors/changes for slope failure to be targeted in monitoring
1012 program;
- 1013 ● Develop a generic strategy for environmental baseline studies and the
1014 environmental monitoring of hazards related to gas hydrate dynamics;
- 1015 ● Evaluate whether national and international legal frameworks are suitable to
1016 protect the environment.

1017
1018 In the previous sections, the hazards related to gas hydrate decomposition have been
1019 identified and discussed, whereas this section is devoted to the risk analysis related to
1020 hydrate decomposition. Risk analysis is essential to ensure safety of operations. Although
1021 the terms “risk” and “hazard” are often used interchangeably, “risk” and “hazard” have
1022 well defined meanings in risk assessment procedure. Hazard is a potential source of harm
1023 or damage. Harm or damage is always the consequence of an accident, whereas risk is a
1024 combination of the likelihood (L) of the harm occurring and the severity of that harm
1025 (damage D). Thus, risk can be formulated as follows:

$$R = f(D, L) = D \times L$$

1026
1027 Where R represents the risk matrix.

1028

1029 **a. Generic strategy related to gas production from NGHDs**

1030 The production of gas hydrates, whether or not coupled with carbon dioxide
1031 sequestration, can create environmental hazards by affecting seafloor stability and
1032 delicate microbial/benthic ecosystems and by releasing methane and particles into the
1033 water column. Therefore, countries that are considering such resources are committed to
1034 developing an appropriately focused environmental baseline survey prior to gas hydrate
1035 production operations to quantify those risks. An environmental risk assessment of gas
1036 hydrate exploration and production (E&P) operations will be generated from the baseline
1037 survey and will include proposed mitigation measures to minimize the impacts of the
1038 E&P operations. During E&P operations, an appropriate monitoring program to assess
1039 the effectiveness of mitigation measures is necessary. There are precedents for baseline
1040 and monitoring studies from oil and gas E&P (IOGP, October 2017) and CO₂ storage
1041 (<https://www.quintessa.org/case-studies/coupled-process-modelling-for-co2-storage>).

1042 A lightweight drillship using coiled tubing drilling over a short period of time may be
1043 used in NGHD exploration because the NGHD prospect will be at shallow depths in
1044 unconsolidated marine sediments. Production of NGHD is likely to be carried out by

1045 subsea infrastructure on the seafloor with real-time monitoring and control from a remote
1 1046 location (Max and Johnson, 2019). Hydrate is stable and effectively inert in its reservoir
2
3 1047 host sediment. During depressurization, which seems the most promising method of
4 1048 production, pressure in the reservoir is lower than normal formation pressure which
5 1049 means that a blow-out is physically impossible and the likelihood of leakage of gas is
6
7 1050 highly reduced. If gas leakage does occur methane gas bubbles are dissolved in ambient
8 1051 seawater during their ascent through the water column and oxidized by aerobic
9 1052 microorganisms using dissolved oxygen as terminal electron acceptor (Atlas and Hazen,
10 1053 2011) with the potential consequences of oxygen depletion in the water column as
11 1054 detailed in section 4.ii.

14 1055 Prior to drilling an NGHD exploration borehole, a site survey to establish baseline
15 1056 environmental conditions and identify drilling hazards at the proposed drill site is needed.
16 1057 Site surveys are performed to minimize the risk of harm to personnel and equipment, and
17 1058 to protect the natural environment. Therefore, the site survey must acquire sufficient data
18 1059 over an appropriate period of time to adequately describe the physical, chemical,
19 1060 geological and biological environments in the operational area. Initially a desktop study
20 1061 is carried out to inform the design of the site survey. With respect to the specific hazards
21 1062 presented by NGHDs the integrated analysis of exploration 3D and/or 2D seismic data,
22 1063 offset well data (logs, operation reports, industry databases, etc.), geotechnical borehole
23 1064 information, offset site surveys and relevant public domain data will allow an initial
24 1065 ground model of the seabed and shallow section to be developed. This geological
25 1066 characterization will include detailed bathymetry, geochemical, mineralogical and
26 1067 petrophysical characteristics of the sediment and geological structures. The
27 1068 biogeochemical characteristics of the seafloor and its surroundings (bottom water, near
28 1069 seafloor sediment characteristics as well as pore water composition, together with the
29 1070 dissolved benthic fluxes) must be measured. Further measurements will be required to
30 1071 establish baseline environmental conditions. These include the physical and chemical
31 1072 characteristics of the entire water column especially in relation to oxygenation and
32 1073 acidification, *i.e.* dissolved gases such as O₂, CO₂, and CH₄ as well as other parameters
33 1074 of the carbonate system (pH, DIC, alkalinity), nutrients, SO₄²⁻, H₂S, Ca²⁺, and total
34 1075 Hardness. Potential gas emissions from the seafloor into the water column in the target
35 1076 area should be known, and qualitatively and quantitatively mapped (Bayrakci et al., 2014;
36 1077 Greinert et al., 2006; Greinert and Nützel, 2004; Jerram et al., 2015; Leblond et al., 2014)
37 1078 as well by appropriate acoustic devices and sensors (*e.g.* single-, split- and multi-beam
38 1079 echo sounder systems, dissolved gas sensors). The biological environment must also be
39 1080 described including the biological community of the seafloor and near seafloor sediments
40 1081 (biocenosis) with reference to conservation of protected areas, habitats and species.
41 1082 Economic activity that might be impacted by NGHD production operations should be
42 1083 described including archaeological heritage.

56 1084 The desktop study will confirm if the shallow geology and the environmental hazards
57 1085 generated by the production of gas hydrates is well understood. Invariably supplemental
58 1086 data acquisition will be required, and a site survey will be designed to fill the data gaps

1087 identified by the desktop study. Because NGHD E&P operations will involve deploying
1088 a seabed coring tool from a workboat, a seabed clearance survey will be required. An
1089 indicative site survey area is 200 m below seabed and 500 m radius around the borehole
1090 site location.

1091 The baseline studies can be carried out by an autonomous underwater vehicles (AUV)
1092 and a seafloor and water column generic instrument module or lander deployed from a
1093 site survey vessel with seabed sampling equipment and a towed magnetometer (cf. Klar,
1094 2019 for information on monitoring technology). The seabed sampling using box cores
1095 and piston cores deployed from the site survey vessel will acquire geotechnical
1096 information on the strength of near seabed sediments and biological habitats. The
1097 magnetometers will provide archaeological information. The AUV and lander should be
1098 equipped with suitable instruments to monitor the water column, the water-sediment
1099 interface and partially the sub-seafloor as well (*e.g.* side scan sonar and synthetic aperture
1100 sonars (SAS), echo sounders, sub-bottom profilers, hydrophones, physical sensors,
1101 chemical sensors, still camera, sediment traps, benthic chambers, etc.). It may be
1102 necessary to deploy an autonomous, fully instrumented lander for several months at the
1103 proposed site to acquire adequate baseline data and capture variabilities related to season,
1104 tide, storm or other aforementioned environmental and anthropogenic events.

1105 Geological characterization is necessary to assess the risk and likely impacts of seafloor
1106 deformation caused by NGHD E&P operations. The geological characterization will
1107 include the aforementioned of surveys and analyses proposed for the site surveys,
1108 together with data from OBS (Ocean-Bottom Seismometer) or OBC (Ocean-Bottom
1109 Cable), as well as high-resolution multibeam bathymetry/backscatter mapping of the
1110 seabed and hydro-acoustic imaging of shallow gas accumulations in the seabed if they are
1111 not already available from site surveys. A paleo-geological history study will also be
1112 required. The combination of multibeam backscatter data with the results of the analyses
1113 of box cores will allow the characterization of the benthic biotopes of the operational area.

1114 Likewise, to assess the likely impacts of methane gas release on the environment, several
1115 parameters will need to be measured in the baseline site survey. In addition to all
1116 aforementioned measurements, these include detection and quantification of gas bubbles
1117 in the water column through acoustic systems; sampling and analyses, including both
1118 isotopic and molecular composition determination of any ascending gases; video/photo
1119 imaging of biota at the seabed; biological integrity; oceanographic measurements to
1120 model the transport of sediments and gases; chemical detection of dissolved gases;
1121 chemical and isotopic compositions of key pore-water elements and dissolved gases;
1122 habitat mapping. Several of these measurements can be achieved by deploying an
1123 autonomous seafloor and water column generic instrument module, water column
1124 moorings and/ or lander from the site survey vessel to collect the data. However, although
1125 some gas (*e.g.* methane, carbon dioxide) sensors can be implemented on these platforms,
1126 most of the geochemical analyses of the gas bubbles and pore water will be performed
1127 onshore after sampling with appropriate tools such as corer and bubble sampler (Chen et
1128 al., 2007; Ruffine et al., 2018a; Ruffine et al., 2018b; Wu et al., 2013; Wu et al., 2014).

1129 The environmental impact assessment will also need to assess the impact of NGHD E&P
1130 operations on human economic activity. Therefore, the site survey will also need to
1131 acquire information on the physical infrastructures, pipelines, telecommunication cables,
1132 fishing activity, etc., in the vicinity of the NGHD E&P operation area. A stakeholder
1133 engagement plan may also be necessary based on the human activity that may be
1134 impacted.

1135 With respect to the environmental monitoring plan for long-term NGHD production
1136 operations, it is advisable to deploy an instrumented lander and/ water column moorings
1137 to monitor essential environmental variables at an undisturbed reference site within a
1138 suitable distance from production operations where a similar lander/ and or water column
1139 moorings acquires information on the same essential environmental variables for
1140 comparison (Klar et al., 2019). The results of the baseline study based on the site survey
1141 data will form the input for modelling (geotechnical, geochemical, fluid flow, etc.) to
1142 predict reservoir conditions during production and to compare after decommissioning. In
1143 addition, the modelling can be used as a tool to plan the monitoring regime and to define
1144 the area that may be influenced both temporarily and spatially by the NGHD production.
1145 The modelling can help to understand if an additional dataset is necessary to complete the
1146 baseline study. Commercial software is only available for partially modelling site
1147 evolution, so one suggestion would be to evaluate the possibility to develop new software
1148 to enable complete modelling of site evolution.

1149 **b. Generic strategy to monitor gas seepages from hydrates**

1150 After the baseline study has delivered the comprehensive characterization of the site,
1151 monitoring is necessary in order to identify any anomalies relative to the anthropic- (*e.g.*
1152 pre-production state) or the climate change-related impacts (Liu et al., 2019). Indeed, the
1153 strategy detailed in this section applied for assessing the impacts of disturbances due to
1154 both human activities and climate change. However, as mentioned previously the impacts
1155 due to climate change may be smoother and expand over a longer time period. Thus, the
1156 monitoring period should be adapted accordingly.

1157 The first step towards a suitable monitoring strategy is to generate a detailed project
1158 description based on the baseline study and modelling, in order to identify the main risks
1159 for the environment and assess the potential impacts. From the environmental impact
1160 assessment, a recommendation of mitigation measures and a plan for environmental
1161 management and spatiotemporal monitoring should be established. The next step is to
1162 plan the management and monitoring strategy based on the risk and impact assessment.
1163 Surveys carried out during the baseline study should be repeated. It could include
1164 autonomous underwater vehicles (AUVs), monitoring vessels, water column moorings
1165 and lander deployments, as used for the baseline study (Klar et al., 2019). Multiple
1166 surveys should be conducted for full areal coverage. However, each survey should be
1167 conducted at a specific height above the seabed, along with permanent monitoring on and
1168 of the seabed by fixed installations such as observation wells and deployed sensors, to
1169 achieve optimal results. Additional targeted studies will have to be conducted, if active

1 1170 formation water seeps, gas seeps, geological features such as mud volcano and
2 1171 pockmarks, etc., connected to the NGHD producing reservoir are observed at the seabed.
3 1172 These sites must be revisited on a regular basis to determine emission rates of gases and
4 1173 fluids. Such periodic visits are key to check for instance, if any seepage has strengthened
5 1174 and/or seafloor geological structure formation is (re-)activated by the NGHD production
6 1175 operation. If new seeps develop during the production operational phase, they must be
7 1176 investigated and sampled in detail to determine the origin and chemical composition of
8 1177 the seeping fluids and gases and their emission rates.

11 1178 Especially during the NGHD production it is also important to accurately monitor seabed
12 1179 deformation, since deformation and subsidence processes around the center of activity
13 1180 could already be an imminent hazard precursor. This type of monitoring relies on lander-
14 1181 deployed high precision, high stability pressure sensors together with inclination
15 1182 measurements at several positions within the production field. Acoustic Long Baseline
16 1183 (LBL) methodology can be used to monitor relative positions in between the measuring
17 1184 nodes at the seafloor (Klar et al., 2019).

22 1185

25 1186 **CONCLUSION AND PERSPECTIVES**

27 1187 Natural gas hydrates have been investigated for multiples purposes for decades, whether
28 1188 to understand their role in the oceanic methane (carbon) cycle, as a geohazard, or as a
29 1189 potential energy resource. Whatever the purpose of their study, the question of their
30 1190 decomposition, -the main factors that trigger this process, its speed and duration, as well
31 1191 as the methane flux released-, are highly relevant. In the present review, the
32 1192 environmental impacts related to hydrate decomposition have been discussed and
33 1193 monitoring strategies have been proposed to identify and better characterize those
34 1194 impacts.

39 1195 Hydrate decomposition can be relatively fast, from a few months/ years to few decades,
40 1196 if it is voluntarily caused by human activities. On the other hand, the changing climate of
41 1197 our planet may also trigger the decomposition of vulnerable NGHDs located in the
42 1198 permafrost and the shallow sediments on the ocean margins. The kinetics of such a
43 1199 decomposition will likely be slower but will last over a longer time period. There are
44 1200 natural biogeochemical processes that degrade methane within both the sediment and the
45 1201 water column, preventing any large accumulation of it in the water masses and mitigating
46 1202 its transfer to the atmosphere. However, the question of the efficiency of these processes
47 1203 under a changing climate needs to be understood. How will the increase of sea bottom
48 1204 temperature affect the microbial communities that perform the aerobic and anaerobic
49 1205 oxidation of methane, and accordingly the amount of methane that will be released from
50 1206 hydrates? What will be the consequences of such releases over decades and centuries?
51 1207 These questions remain to be answered.

58 1208 Coastal regions are amongst the most populated areas on Earth. The decomposition of
59 1209 hydrates, mainly due to climate change, may reduce seafloor stability and cause

1210 submarine landslides with associated tsunamis impacting coastal communities. The
1211 potential occurrence of such catastrophic socio-economic events that threaten human life,
1212 natural resources and key infrastructure in populated coastal regions highlight the need to
1213 monitor the dynamics of NGHD. This review has shown that field observations and
1214 measurements are limited in space and time. Field observations are crucial to achieving
1215 an in-depth and comprehensive understanding of the operating environment, and to
1216 developing an appropriate environmental monitoring and management plan. Currently,
1217 what we observe on a given site is not representative for other sites and an intensification
1218 of the measurements of key parameters, both spatially and temporally, is urgently needed.

1219

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