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## Control of the geomorphology and gas hydrate extent on widespread gas emissions offshore Romania

### La géomorphologie des fonds marins et la présence d'hydrates de gaz contrôlent les émissions de gaz dans la Mer Noire au large de la Roumanie

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#### Abstract :

The Romanian sector of the Black Sea deserves attention because the Danube deep-sea fan is one of the largest sediment depositional systems worldwide and is considered the world's most isolated sea, the largest anoxic water body on the planet and a unique energy-rich sea. Due to the high sediment accumulation rate, presence of organic matter and anoxic conditions, the Black sea sediments offshore the Danube delta is rich in gas and thus shows Bottom Simulating Reflectors (BSR). The cartography of the BSR over the last 20 years, exhibits its widespread occurrence, indicative of extensive development of hydrate accumulations and a huge gas hydrate potential. By combining old and new datasets acquired in 2015 during the GHASS expedition, we performed a geomorphological analysis of the continental slope north-east of the Danube canyon compared with the spatial distribution of gas seeps in the water column and the predicted extent of the gas hydrate stability zone. This analysis provides new evidence of the role of geomorphological setting and gas hydrate extent in controlling the location of the observed gas expulsions and gas flares in the water column. Gas flares are today considered an important source of the carbon budget of the oceans and, potentially, of the atmosphere.

#### Résumé

Le secteur roumain de la Mer Noire est dominé par la présence du canyon du Danube et d'un des plus grands systèmes de dépôts de sédiment du monde. La Mer Noire est considérée comme la plus grande mer isolée du monde, la plus grande masse d'eau anoxique de la planète et une mer riche en énergie

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fossile. En raison d'un taux de sédimentation élevé, de la présence d'une grande quantité de matière organique et des conditions anoxiques, les sédiments de Mer Noire situés au large du delta du Danube sont riches en gaz et l'étude de données de sismique réflexion montre la présence d'un réflecteur sismique particulier appelé communément « Bottom Simulating Reflector ou BSR » qui marque la base de stabilité des hydrates de gaz. La cartographie du BSR au cours des 20 dernières années montre que les hydrates de gaz se seraient accumulés sur de vastes zones géographiques et que le secteur roumain de la Mer Noire a un fort potentiel d'hydrate de gaz. En combinant les anciens et les nouveaux jeux de données acquis en 2015 lors de la campagne océanographique GHASS, nous avons réalisé (1) une analyse géomorphologique de la pente continentale au nord-est du canyon du Danube, (2) une cartographie des panaches de gaz acoustiquement détectés dans la colonne d'eau et (3) le calcul et la cartographie de la zone de stabilité des hydrates de gaz. La comparaison de ces résultats fournit de nouvelles preuves du rôle de la géomorphologie et de la présence des hydrates de gaz sur la migration du gaz libre et la localisation des panaches de gaz dans la colonne d'eau. L'expulsion de gaz dans la mer est aujourd'hui considérée comme une source importante alimentant le budget carbone des océans et, potentiellement, de l'atmosphère.

**Keywords** : gas hydrates, free gas, gas flares, BSR, Black Sea, geomorphology

**Mots clés** : hydrates de gaz / gaz libre / panaches de gaz / BSR / Mer Noire / géomorphologie

## 21 **1. Introduction**

22 The Black Sea is considered as the world's most isolated sea, the largest anoxic water body on the planet  
23 and a unique energy-rich sea (Overmann and Manske, 2006). CH<sub>4</sub> seepage is extremely intense on the shelf  
24 and on the slope of the Black Sea (Dimitrov, 2002; Mert Küçük et al.; Vassilev and Dimitrov, 2003),  
25 especially along the Ukrainian (Greinert et al., 2010; Naudts et al., 2009; Naudts et al., 2008; Naudts et al.,  
26 2006) and Romanian margins (Popescu et al., 2007). Black Sea sediment abundantly contains GHs and H<sub>2</sub>S  
27 as CH<sub>4</sub> and hydrogen source, respectively (Demirbas, 2009). GH occurrence in the Danube fan has been  
28 known since the first hydrate discovery in shallow sub-bottom sediments at water depth of 1950 m in 1972  
29 (Ginsburg, 1998; Yefremova and Zhizhchenko, 1974). Recently, the presence of GHs in deep sediments  
30 was inferred from Bottom-Simulating Reflector (BSR) observations in the southern part of the Black Sea  
31 fan (Ion et al., 2002) and in the northwestern Black Sea (Lüdmann et al., 2004; Zillmer et al., 2005). Multiple  
32 BSRs occur on high-resolution reflection seismic data in the Danube deep-sea fan, associated with acoustic

33 features indicating free gas (Popescu et al., 2006). The shallowest BSR in the Black Sea exhibits its  
34 widespread occurrence, indicative of extensive development of hydrate accumulations and thus a huge gas  
35 hydrate potential (Merey and Sinayuc, 2016). The origin of CH<sub>4</sub> source for GHs is mainly biogenic (Hester  
36 and Brewer, 2009), and formed during diagenesis stage in the evolution of organic materials within  
37 sediment. CH<sub>4</sub> forms sI type of GH at hydrate forming conditions (Sloan and Koh, 2007).

38 The Romanian margin is composed by a wide continental shelf and a slope incised by several submarine  
39 canyons including the Danube canyon (Popescu et al., 2004). This canyon results from erosive sediment  
40 flows which fed the Danube deep-sea fan, one of the most developed deep-sea sediment depositional  
41 systems worldwide (Winguth et al., 2000; Wong et al., 1994), mainly during lowstand periods  
42 (Constantinescu et al., 2015). During the Late Quaternary, the Black Sea and the Mediterranean experienced  
43 several phases of connection and disconnection with relevant impact on the salinity of the Black Sea  
44 oscillating between freshwater lake and salt-water sea conditions (Zubakov, 1988). Since the last phase  
45 (9000 years ago), the Black Sea communicates with the Mediterranean Sea through the Bosphorus and  
46 Dardanelle straits (Ross et al., 1970). So, the Black Sea which was a freshwater lake, has become a salt-  
47 water sea. The salinity of the Black Sea reached its current value of 22 psu at ~2000 yr cal BP (Soulet et  
48 al., 2010). The Black Sea salinity is significant to appraise the extent of the Gas Hydrate Stability Zone  
49 (GHSZ). Indeed, the thermodynamic stability of the GHs primarily depends on temperature, pressure, gas  
50 composition and salinity (Sloan, 2003). GH deserve attention because their destabilization can provoke  
51 seafloor instability (Crémière et al., 2016; Nisbet and Piper, 1998), and release significant quantities of gas  
52 into the ocean, thus affecting gas inputs into the atmosphere (McGinnis et al., 2006; Solomon et al., 2009).

53 The study area, poorly known due to a lack of proper data resolution, is located in the Romanian sector of  
54 the Black Sea, north-east of the Danube canyon. The aim of this paper is to improve the knowledge about:  
55 (1) the seafloor morphology of the Romanian sector of the Black Sea, and (2) the influence of the  
56 geomorphology and the GH occurrence in the distribution of gas flares acoustically detected in the water  
57 column.

## 58 **2. Data and Method**

### 59 **2.1. Bathymetry and water column acoustic data**

60 The study is based on the analysis of bathymetry and water column acoustic data acquired during the 2015  
61 GHASS cruise on board the R/V Pourquoi Pas ? (doi:10.17600/15000500). Ship-borne multibeam surveys  
62 were conducted to map the external continental shelf and upper-mid slope adjacent to the Danube canyon  
63 and to detect and locate the presence of free gas in the water column (Figs. 1, 2, 3). The acoustic data were  
64 acquired with 1) a Reson seabat 7111 multibeam echo-sounder for shallow water from 5 to 500 m (100 kHz,  
65 301 beams, 1.8°x1.5° beam width, 0.17 to 3 ms pulse length, up to 20 pings per second), and 2) a Reson

66 seabat 7150 multibeam echo-sounder for mid and deep water from 200 to 2000 m (24 kHz, 880 beams,  
67 0.5°x0.5° beam width, 2 to 15 ms pulse length, up to 15 pings per second). The shelf and upper slope were  
68 surveyed with both echo-sounders, while the deepest area was only surveyed by the Reson seabat 7150.  
69 Bathymetric resolution of the whole study area is 20 metres. Water column processing was performed  
70 onboard with SonarScope and GLOBE softwares (© Ifremer).

## 71 2.2. High resolution seismic data

72 High-resolution reflection seismic data were acquired during the 1998 BLASON (doi 10.17600/98020030)  
73 and 2002 BLASON2 (doi 10.17600/2020070) surveys of IFREMER and GeoEcoMar (Figs. 4, 5). Data were  
74 obtained using consecutively two seismic sources: a GI gun (central frequency 70 Hz) and a mini GI gun  
75 (central frequency 150 Hz). The receiver was a 24-channel streamer, 300 m long. Data were processed using  
76 Landmark's ProMAX software. The conventional processing flow included CDP gather formation, velocity  
77 analysis, removal of noisy traces, normal moveout correction and stack, migration, and seabed mute. No  
78 amplitude corrections were applied. Analysis and interpretation of seismic data were conducted using  
79 Seismic Microsystems' Kingdom Suite software.

## 80 3. Results

### 81 3.1. Geomorphology

#### 82 3.1.1. Margin physiography

83 The continental shelf of the Romanian sector of the Black Sea has an average width of 160 km with a very  
84 subdued bathymetric gradient of 0.5° in the outer shelf. The shelf edge occurs between 180 and 190 m water  
85 depth with a local slope angle of 4° (Fig. 1). The continental slope has a regional slope of about 2°, but  
86 attains 4° between 200 and 500 m water depth. In correspondence of some geological features such as  
87 pockmark and canyon flanks, the slope locally reaches 35°. The outer shelf and slope are incised by two  
88 canyons which could act as zones of confined sediment transport/bypass (Figs. 1, 2). The upper slope shows  
89 many fluid expulsion features including pockmarks. The western part of the slope is affected by sediment  
90 gravity processes, while its central - eastern part by the presence of sediment wave fields and small mounts  
91 (Fig. 2).

#### 92 3.1.2. Canyons

93 The two major canyons presented above have incised the 1200 km<sup>2</sup> study area. In the 2.2 km wide Canyon  
94 1 (Fig. 2), flank height reaches 160-110 m in the head with, locally, a slope angle of 15°. Canyon 1 head  
95 reaches the shelf at about 180 m of water depth. Canyon 1 is narrower downstream with a width of 1.5 km  
96 at around 1500 m water depth. A single well developed thalweg with axial incision, 350 m wide and 50 m  
97 high on average, is well developed with a mean height of 100 m and an average width of 700 m starting  
98 from 1200 m water depth. Canyon 1 is wider between 200 and 700 m water depth probably due to the effect

99 of several submarine landslides (Fig. 2). Paradoxically, the slope value of the flanks, 15° in the upper part  
100 of the canyon, increases downstream to reach 25° where the axial incision is the most developed.

101 Canyon 2, located to the east, is less incised than Canyon 1. It is 1.5 km wide. Its thalweg reaches 70 m in  
102 the head with, locally, a 10° flank slope. Further seaward, the height increases to reach 100 m with a 25°  
103 flank slope. The axial incision, 15 m high, disappears at about 750 m of water depth (about 20 km from the  
104 canyon head). Two other small canyons/gullies incise the upper slope northward of Canyon 2.

### 105 *3.1.3. Mass transport Complexes*

106 The seafloor instabilities identified in the bathymetric map correspond to the morphologies of the headwall  
107 scarps and lateral margins of the translational domain of the Mass Transport Complex (MTC) described in  
108 Bull et al. (2009). They are observed between 200 and 900 m water depth, but most of them are detected  
109 between 500 and 750 m. Mainly on the both sides of the Canyon 1 (the scarp limits, in orange, are shown  
110 in Fig. 2), we distinguish the destabilisations associated with the canyon from open slope scarps. All the  
111 scarps disturb 20% of the surface of the seafloor of the study area. Their size is comprised between 1 and  
112 15 km<sup>2</sup> with an average slope of 12° that may locally reach 25°.

### 113 *3.1.4. Other seafloor features*

114 Some 50 pockmarks with diameters ranging from 100 to 150 m were detected at the seafloor in a region of  
115 3000 km<sup>2</sup>. The largest is 160 m wide and 9 m deep. The value of the slope of pockmark flanks is around  
116 10°, but it reaches up to 14°. Pockmarks were observed in the free gas area defined by Popescu et al. (2007),  
117 in a water depth range of 175-475 m. All pockmarks have the same morphology as those first documented  
118 in the literature (King and MacLean, 1970). They are mostly circular or oval in shape and have a conical or  
119 dish-shaped vertical section. The 20 m resolution of the bathymetric data prevented the detection of small  
120 pockmarks.

121 The central sector of the study area, between Canyons 1 and 2 at around 500-600 m water depth, is affected  
122 by seafloor undulations originated by sediment transport and/or, less likely, by creeping processes (Fig. 2).  
123 Some 63 sediment waves cover a region of 90 km<sup>2</sup>. The magnitude of the positive relief is around 2 m.

124 Further seawards, numerous small mounts are detected between 700 and 1000 m water depth. The 140  
125 mounts have an average diameter of 800 m and a positive relief of about 15 m with, locally, 12° flank slope  
126 (Fig. 2). The analysis of seismic profiles presented in Figure 5 suggests that the mounts are an inherited  
127 morphology resulting from a compressive bulge of a landslide deposit buried under 35 mbsf.

## 128 3.2. Free gas versus gas hydrates in the study area

### 129 3.2.1. Evidence of free gas in the water column

130 We identified some 1409 gas seeps within the water column acoustic records (15 days acquisition during  
131 GHASS cruise). The seepage activity does not appear homogenous, as the density of gas flares varies with  
132 bathymetry and laterally. Many of the numerous and widespread gas flares that were recorded at the scale  
133 of the Romanian sector of the Black Sea reach several hundreds of meters above the seafloor, attesting to a  
134 vigorous seepage activity with high fluid fluxes (Fig. 3) and questioning about the fate of the gas in the  
135 water column. Gas emissions may be particularly numerous within some sectors between 200 m and 800  
136 m. No gas flares were detected in deeper areas. Gas emissions can be classified into 6 groups based on their  
137 distribution and origin: (1) non-random gas seeps along the canyons/gullies; (2) non-random gas seeps along  
138 headwall scarps and lateral margin of the MTC (Fig. 3C); (3) non-random gas seeps along fault/ crest line  
139 (Fig. 3A); (4) non-random gas seeps at the landward termination of the GHSZ above small mounts; (5) non-  
140 random gas seeps right above pockmark (Fig. 3B); and (6) other random gas seeps (Figs. 2, 7).

141 The maximum density of acoustic anomalies was found along the canyon path. The 606 gas seeps detected  
142 at the break of slope of the canyon flanks represent 43% of the whole degassing sites. 495 gas seeps are  
143 localized right above the scarps of the MTC, 116 around the faults identified on the outer shelf, 30 right  
144 above the 50 pockmarks, 26 above a crest line at 750 mbsl inside the GHSZ, and 81 in the sector of the  
145 small mounts. Overall, 96% of the all gas seeps observed are above geomorphological structures: 78% are  
146 right above escarpment induced by sedimentary destabilizations inside or outside canyons and 60% of the  
147 pockmarks appear active. Only 4% (55) of the gas seeps appear randomly distributed in the study area.  
148 These gas seeps seem to be at location not affected by geomorphological structures.

### 149 3.2.2. Evidence of free gas in seismic data

150 The 2D HR seismic profiles show a relatively well preserved sedimentary stratification (Figs. 4, 5). Seismic  
151 facies is dominated by high amplitude parallel seismic reflectors. From the shelf down to the slope, a MTC  
152 is identified buried under 40 m of sediment. The source area of the MTC is delimited to the north in about  
153 200 m water depth by the shelf edge. The MTC is characterized by a transparent chaotic seismic facies. The  
154 thickness of the mass deposit, about 20 m at 300 m water depth, progressively increases seawards to attain  
155 75 m at 700 m water depth. The thickness is not homogeneous and varies in function of the inherited relief  
156 (Fig. 4B). The compressional domain of the MTC show many bulges draped by overlying sediment resulting  
157 from the presence of small mounts at the seafloor.

158 Under the MTC, the seismic signature of sediment shows anomalies interpreted as the localized  
159 accumulation of free gas (Figs. 4B, 4C). In marine sediments, free gas often yields anomalous seismic  
160 signatures, making seismic methods a useful tool for the identification and characterization of the sub-

161 seafloor gas charged body and the gas migrating system. Gas may appear as amplitude enhancement with  
162 an attenuation of the signal (Fig. 4) (Gay et al., 2007; Judd and Hovland, 1992; Netzeband et al., 2010). In  
163 Figure 4 we interpret the upward bending reflections observed right under the MTC as corresponding to a  
164 velocity pull-up artefact (Hustoft et al., 2007), and the inflection of seismic reflectors as corresponding to a  
165 velocity pull-down effect (Hustoft et al., 2010). The amplitude enhancement of sedimentary layers (i.e.,  
166 “bright spots”) under the MTC may occur when gas preferentially accumulates in highly permeable layers  
167 (Riboulot et al., 2013; Taylor et al., 2000; Tréhu et al., 2004).

168 The disruption of seismic reflections often referred to as “acoustic turbidity” (Gay et al., 2007; Jones et al.,  
169 2010; Judd and Hovland, 1992; Mathys et al., 2005; Schroot et al., 2005), and/or as “disturbed zones”  
170 (Schroot and Schuttenhelm, 2003) is observed right beneath the pockmarks above the MTC, where its  
171 thickness is reduced (Figs. 4A, 4B). These anomalies can be caused by the presence of vertical gas chimneys  
172 representing current migration of fluids in the sedimentary column (Heggland, 1997; Hempel et al., 1994).  
173 Moreover, the lack of reflection in such vertical conduits may occur due to physical disruption of  
174 sedimentary layering by migrating, gas-charged pore fluids (Davis, 1992; Gorman et al., 2002), or by highly-  
175 reflective overlying interfaces that significantly reduce the transmission of energy (Garcia-Gil et al., 2002;  
176 Judd and Hovland, 1992).

### 177 *3.2.3 Evidence of gas hydrates in seismic data*

178 In the Romanian sector, BSR observation from conventional High Resolution (HR) seismic profiles,  
179 acquired during the BLASON and BLASON2 cruises, provides indirect evidence of GH occurrence (Fig.  
180 4). It represents the base of the GHSZ that appears as strong, negative-polarity, high-impedance seismic  
181 reflections caused by free gas at the base of the phase boundary (Holbrook et al., 1996; Shipley et al., 1979).  
182 The BSR in the study area is characterized by a distinct seismic reflection, sub-parallel to the seafloor,  
183 showing reversed polarity, semi-continuous, crosscutting the sedimentary stratification and their position  
184 can also be inferred on the basis of aligned amplitude terminations as Bangs et al. (2005) described offshore  
185 Oregon (Fig. 5). Popescu et al. (2006) observed the same characteristics for multiple BSRs present in the  
186 Danube sea-fan zone. The appearance of a strong impedance contrast at the location of the BSR with an  
187 enhancement of the seismic reflection amplitude is an indication of the presence of gas beneath GHs (Dillon  
188 and Paull, 1983; Paull et al., 1995). The absence of gas signature on seismic data over the BSR, presented  
189 in Figure 5, provides useful information about the location of the gas, trapped beneath the BSR. The seal  
190 formed by GHs could be impermeable. At the landward termination of the GHSZ, the observed seismic  
191 hyperbola and deformation zone in the surficial sedimentary layers suggest gas migration or the presence of  
192 GHs close to the seafloor (Fig. 5-Inset).

193 *3.2.4 Predicted gas hydrate stability zone*

194 Theoretically determined phase equilibria allow to distinguish natural GHs from water ice, and can therefore  
195 be used to calculate the temperature and pressure at which hydrates form from a given gas composition  
196 (Sloan and Koh, 2007). The variations of water column temperature, pore pressure and geothermal gradient  
197 affect the thickness of the GHSZ.

198 Seafloor temperature was considered to be 8.9 °C at 850 m water depth, determined by Sippican  
199 measurements during GHASS cruise. A hydrostatic pore-pressure gradient of 0.1 bar/m was assumed to  
200 calculate the depth scale (Kvenvolden, 1993). The geothermal gradient was measured with 7 temperature  
201 sensors welded at regular intervals along a 12 m long core barrel. The geothermal gradient considered in  
202 this study is 24.5 °C/km. The composition of the gas enclathrated in hydrate form is a primordial parameter  
203 to estimate the boundaries of GHSZ (Sloan, 2003). It is known that the main component of gas from the  
204 Black Sea hydrates is CH<sub>4</sub> (93.3-99.7%: Vassilev and Dimitrov, 2003). As Poort et al. (2005) did, we  
205 assumed a composition of 100% methane for the composition of the hydrates (Judd et al., 2002), but heavier  
206 hydrocarbons could be present and would shift the hydrate stability curve towards higher temperatures  
207 (Sloan and Koh, 2007).

208 The calculation of the GH stability curve is complicated because it is usually performed for a system  
209 composed of water with a constant concentration of salt (0 psu to >35 psu). In the study area, Soulet et al.  
210 (2010) show a gradual fall in salinity from 21.9 psu at the seafloor level to near 2 psu at around 28 mbsf. In  
211 the case presented here (Fig. 6), we make the calculation using a salinity of 22 psu for the water column  
212 (850 m), a gradual fall of the salinity for the uppermost 28 m of sediment (the salinity of 22 psu at the  
213 seafloor reaching 2 psu in sediment at 28 mbsf) and a constant salinity of 2 psu for the rest of sedimentary  
214 column. The intersection of the GH stability curves with the water column temperature curve denotes the  
215 minimum water depth at which GHs are stable for a given water depth (Fig. 6), while the intersection with  
216 the geothermal gradient reveals the predicted base of the GHSZ (Kvenvolden, 1993).

217 The calculation to obtain a predicted GHSZ is made at different water depths. An example of the calculation  
218 for a water column of 850 m is shown in Figure 6. For this example, the intersection of the GH stability  
219 curves with the water column temperature curve at around 730 m indicates the water depth at which GHs  
220 are stable in this location of the Black Sea. The thickness of the GHSZ is 200 m. The predicted base of  
221 GHSZ is in agreement with the depth of the BSR observed in the study area (215 mbsf at 850 m water depth:  
222 Fig. 5). The minimum water depth where GHs begin to be stable is 660 m at around 20 mbsf and the  
223 thickness of the GHSZ would be 20 m. We theoretically find stable GHs at the seafloor starting from 720  
224 m water depth towards deeper waters.

## 225 **4. Discussion**

### 226 4.1. Impact of geomorphology in free gas expulsion

227 Overall, the distribution of gas flares observed in the water column of the study area are in agreement with  
228 the free gas areas defined in Popescu et al. (2007). However, in some cases, several gas flares are detected  
229 downward the areas defined in the literature: many gas flares are inside the BSR zone defined in (Popescu  
230 et al., 2006) close to the landward termination of the BSR (Fig. 7). The causes of this mismatch could be  
231 attributed to an evolution of the degassing zone in the water column over the last 10 years and/or to the  
232 variety of the data analysis. The free gas area described by Popescu et al. (2006) was derived from seismic  
233 data interpretation while our gas flare areas from analysis of acoustic data recently acquired. Indeed, the  
234 identification of seepage activity at continental margins, which is a relatively widespread phenomena (Judd  
235 and Hovland, 2007), is emphasized by the water column mapping and technological advances in the last  
236 decade (Dupré et al., 2015).

237 The distribution of the gas seeps in the Romanian sector of the Black Sea coincides in most cases with the  
238 presence at the seafloor of sediment deformation features. 96% of the gas flares are located above canyons  
239 (Fig. 7C), landslides (Figs. 3C, 7A), pockmarks (Figs. 3B, 7B), and fault/ crest line (Fig. 3A). These  
240 observations and interpretations coincide with: (1) the recent analysis made in the Sea of Marmara where it  
241 was demonstrated that gas emissions in the water column are spatially controlled by fault and fracture  
242 networks in connection with the Main Marmara Fault system (Dupré et al., 2015); (2) several studies  
243 offshore California showing active seeps right above a vertically faulted and fractured region along the walls  
244 of the Monterey Canyon (Barry et al., 1996; Paull et al., 2005); and (3) previous studies in the Dnepr paleo-  
245 delta (northwestern Black Sea), where seeps generally occur in association with pockmarks on the  
246 continental shelf, along crests of sedimentary ridges, canyon flanks and near submarine landslides on the  
247 continental slope (Naudts et al., 2006). Studies about the geomorphological control of the distribution of  
248 gas seepages finally show they follow the same pattern as the control of the distribution of pockmarks.  
249 Studies published during the last 20 years have demonstrated that the spatial organization of pockmarks  
250 (seafloor deformation due to fluid expulsion) may be the result of fluid seepage from underlying sedimentary  
251 structures such as fault systems (Pilcher and Argent, 2007), channels (Gay et al., 2003), mud volcanoes,  
252 mud diapirs, glaciogenic deposits (Forwick et al., 2009), and mass transport deposits (Riboulot et al., 2013).  
253 The spatial distribution of pockmarks suggests that all the discontinuities within the sedimentary column  
254 represent potential drains for fluid flow, and that simple diffusion through the sediments cannot explain the  
255 observed pattern of fluid expulsion. The spatial distribution of a large proportion of the gas flares in the  
256 study area seems to be associated with gas contained in underlying sediment using discontinuities formed  
257 by landsliding. The discontinuities resulted from mass wasting processes inside and outside the canyons are  
258 probably responsible for the gas seepages, by providing preferential migration pathways to gas as Riboulot

259 et al. (2013) demonstrated in the Niger delta where a buried landslide controls the distribution of the seafloor  
260 pockmarks.

#### 261 4.2. Impact of Gas Hydrates Stability Zone in free gas expulsion and sedimentary deformation?

262 We observed only 26 gas seeps of the 1409 detected in the study area really inside the GHSZ (Figs. 2, 3A,  
263 7). They are right above a crest line that represents 2% of the whole gas seeps detected in the study area.  
264 We interpret the crest line as the seafloor evidence of the presence of a fault affecting the underlying  
265 sedimentary sediments. If this is the case, as Gay et al. (2006) suggest in the Lower Congo Basin, we suppose  
266 the fluids accumulate under the base of the hydrate stability zone form a layer of free gas and the generation  
267 of excess pore fluid pressure in the free gas accumulation leads to the release of fluids along faults of the  
268 highly faulted interval responsible of the presence of free gas at the seafloor and in the water column.

269 Due to the concentration of gas seepages outside and at the landward termination of the GHSZ (98% of the  
270 whole degassing site) and the seismic anomalies observed under the BSR (Fig. 5), we suggest that the  
271 presence of GHs at the base of GHSZ constitutes an impermeable caprock over an accumulation of free gas.  
272 Indeed, GHs may fill pore spaces and reduce sediment permeability, so that in some cases hydrate-bearing  
273 sediment may act as seal and result in gas traps (Max and Dillon 1998). This interpretation is in agreement  
274 with the observations of Naudts et al. (2006) in the Dnepr paleo-delta area where the depth limit for 99.5%  
275 of the detected seeps coincides with the phase boundary of pure methane hydrate at 725 m water depth.  
276 They suggest GHs play the role of buffer for the upward migration of methane gas and thus prevent seepage  
277 of methane bubbles into the water column as it was proposed by Popescu et al. (2007) in the Danube Deep-  
278 Sea Fan area and by Westbrook et al. (2009) in the West Spitsbergen margin. This process may explain the  
279 lack of deformation of the overlying sedimentary layers (Fig. 5), the absence of gas flares in the water  
280 column inside the GHSZ and the possible deformation at the landward termination of the GHSZ around the  
281 small mounts.

282 Indeed, the analysis of the seafloor morphology inside the GHSZ combined with the seismic stratigraphy  
283 provide useful information on the impact of GHs on sedimentary deformation. The seafloor deformation,  
284 characteristics of the features named “gas-hydrate pockmarks” and described around the world (Macelloni  
285 et al., 2012; Riboulot et al., 2016; Simonetti et al., 2013; Sultan et al., 2014), are not observed in the study  
286 area. GH pockmarks characterize seafloors where GHs are present in the shallow sedimentary layers.  
287 Sediment deformation at the landward termination of the BSR may be induced by GH dynamics as it was  
288 described in the Niger delta by Riboulot et al. (2016) and Sultan et al. (2014). The presence of GHs close to  
289 the seafloor generate a disturbance of the sedimentary deposits and the loss of their original sedimentary  
290 structures. The small mounts, observed around this area and mentioned in Figures 2 and 5, rather seem to

291 be the result of an inherited morphology from the compressive bulge of an underlying landslide without  
292 implication of GH dynamics.

293 It may be noted that several headwall scarps are observed at around 650 m water depth. The landward  
294 termination of the GHSZ coincide with these escarpments. 35% of the gas seeps observed in the water  
295 column are localized right above scarps at the boundary with the GHSZ. It suggests GH dynamics may have  
296 an implication in sediment failure as it was interpreted by Westbrook et al. (2009). Further investigation  
297 will be needed to confirm this hypothesis.

## 298 **Conclusions**

299 The continental slope morphology of the Romanian sector of the Black Sea is incised by several landslides  
300 inside and outside canyons. It is a complex study area presenting sedimentary processes such as seafloor  
301 erosion and instability, mass wasting, formation of GHs, fluid migration, gas escape, where the imprint of  
302 geomorphology seems to dictate the location where gas seep occurs. We have detected 1409 active seeps  
303 within the 1200 km<sup>2</sup> of the shelf and slope north-east of the Danube canyon. Most gas seeps (96%) are not  
304 randomly distributed in this area. They occur along canyon flanks, scarps, crest lines, faults and in  
305 association with pockmarks and mounts.

306 Moreover the depth limit for 98% of the gas seeps coincides with the predicted landward termination of  
307 GHSZ. This suggest GHs formed at the base of the GHSZ act as an effective seal preventing gas to reach  
308 the seafloor and the water column. The extent and the dynamics of GHs have a probable impact on the  
309 sedimentary destabilization observed at the seafloor and the stability of the GHs is dependent on the salinity  
310 gradient through the sedimentary column and thus on the Black Sea recent geological history.

## 311 **Acknowledgment**

312 The support by officers and crew during the GHASS cruise on board R/V Pourquoi Pas ? (2015) is greatly  
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314 sincerely Alison Chalm for her revision of the English language.

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## 487 **Figure captions**

488 Figure 1: Bathymetric map, acquired during the 2015 GHASS cruise, showing the study area with the  
489 location of seismic profiles. Fine grey lines show the multibeam navigation where the presence of free gas  
490 in the water column was searched. The continental slope is dissected by the Danube canyon and the Canyons  
491 1 and 2 with several submarine landslide scars along canyon flanks.

492 Figure 2: Geomorphologic map of the study area with superposed: free gas and BSR areas detailed in  
493 Popescu et al. (2006, 2007), cartography of the gas bubbles acoustically detected in the water column, and  
494 the predicted landward limit of the predicted GHSZ (bold black line). The modern landward termination of  
495 the GHSZ correspond to the 660 mbsl bathymetric contour.

496 Figure 3: The 3D views of the seafloor and water column (GLOBE software © Ifremer), with processed  
497 polar echograms, show (A) a crest line inside the GHSZ, (B) a pockmark and (C) a headwall scarp. The  
498 acoustic anomalies recorded in the water column are echoes caused by escaping gas bubbles through the  
499 seafloor. The acoustic imprint of the plumes almost reaches a height of 300 m above the seafloor. The 3  
500 examples are localized on the figure 1.

501 Figure 4: Seismic reflection profile Bla 1-8 (BLASON cruise). Across the shelf break and the upper slope  
502 within the free gas area defined in Popescu et al. (2007; location in Fig. 1). The close up views (A, B and  
503 C) show how the occurrence of free gas affects seismic data. The most apparent free gas zones are identified  
504 under a mass transport complex (in orange). Several free gas zones coincide with the presence of gas  
505 chimneys and pockmarks (A and B) while when the seafloor depth is deeper we have a lack of seafloor fluid  
506 features. The gas seems to be trapped under the MTC.

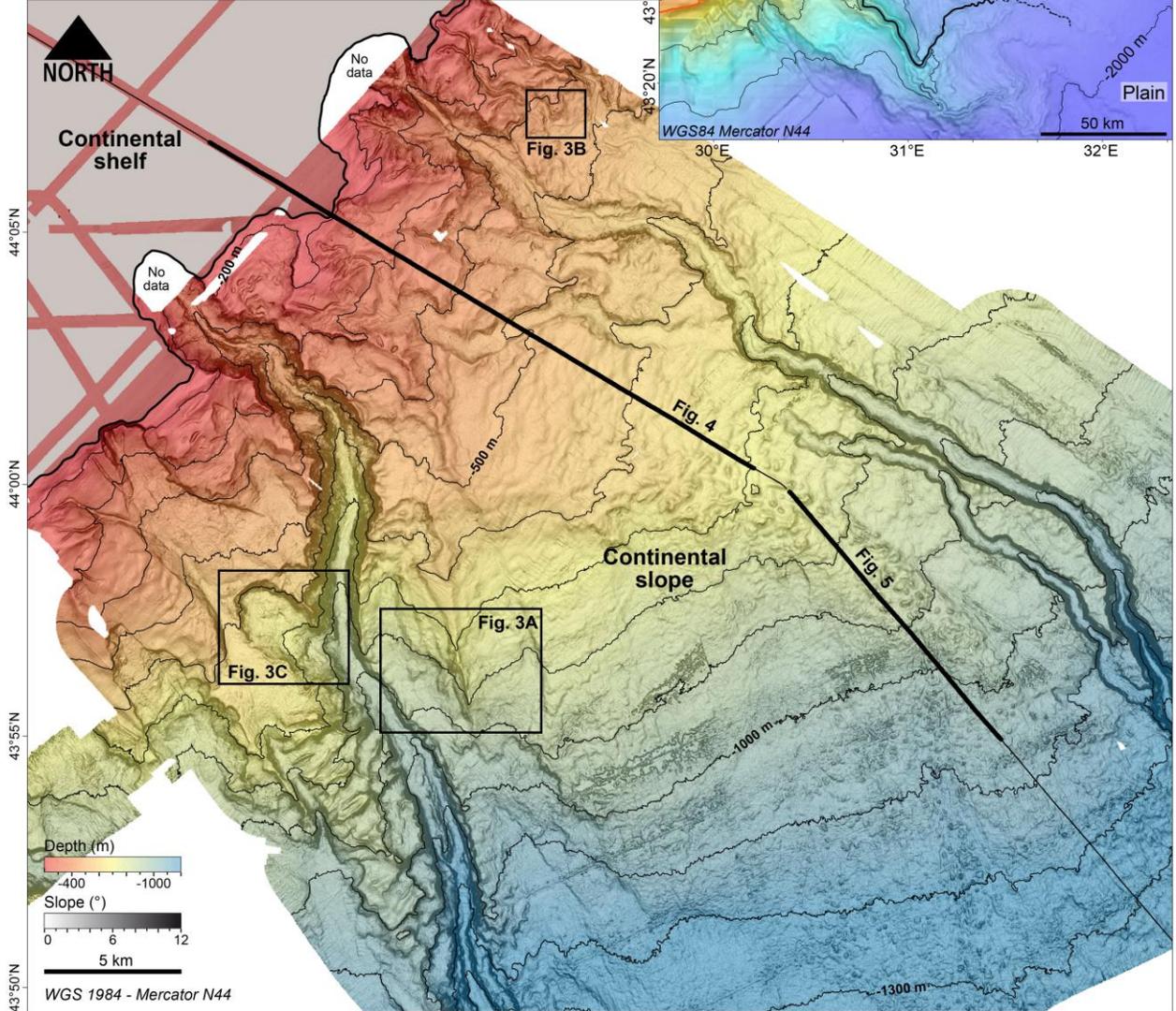
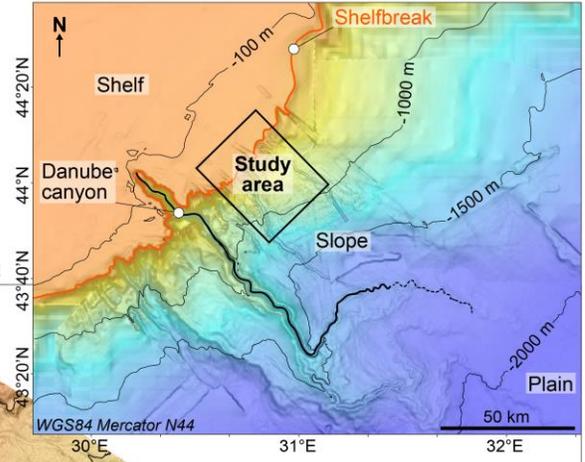
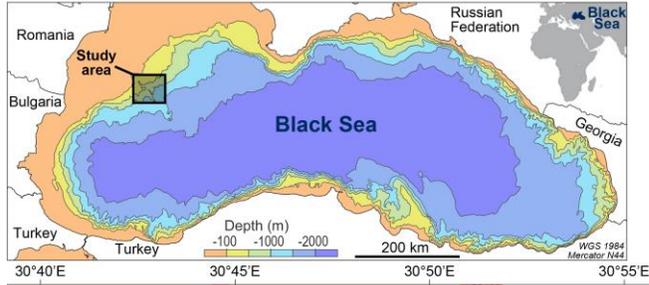
507 Figure 5: Seismic reflection profile Bla 1-7 (BLASON cruise): across the slope partly within the BSR area  
508 defined in Popescu et al. (2006; location in Fig. 1). The presence of a BSR is suggested by a strong and

509 negative polarity reflector associated to an increase in the attenuation and amplitude anomalies (seismic  
510 signature of the free gas – green arrows). Within the predicted GHSZ, right above the BSR, we do not  
511 observed seismic signature of the presence of free gas. The free gas seems to be trapped under the MTC.  
512 The black rectangle indicates the area of inset. The inset highlights the location of the supposed GH  
513 occurrence within a deformed sedimentary layers at the landward termination of the BSR.

514 Figure 6: Gas hydrate stability using pure s-I methane hydrate and the water column ( $S = 22$  psu) and  
515 porewater ( $S=2$ ; in depth higher than 25 mbsf) salinities. For this example used to illustrate the calculation  
516 (Seafloor: 850 m water depth), the minimum water depth where GHs are stable is 720 mbsl. The bottom  
517 water temperature used is 8.9 °C. For the regional observed geothermal gradients of 24.5 °C/km, the base  
518 of GHSZ is 200 mbsf. These results are calculated in 2D and change with depth of the seafloor due to the  
519 evolution of the salinity within the sediment.

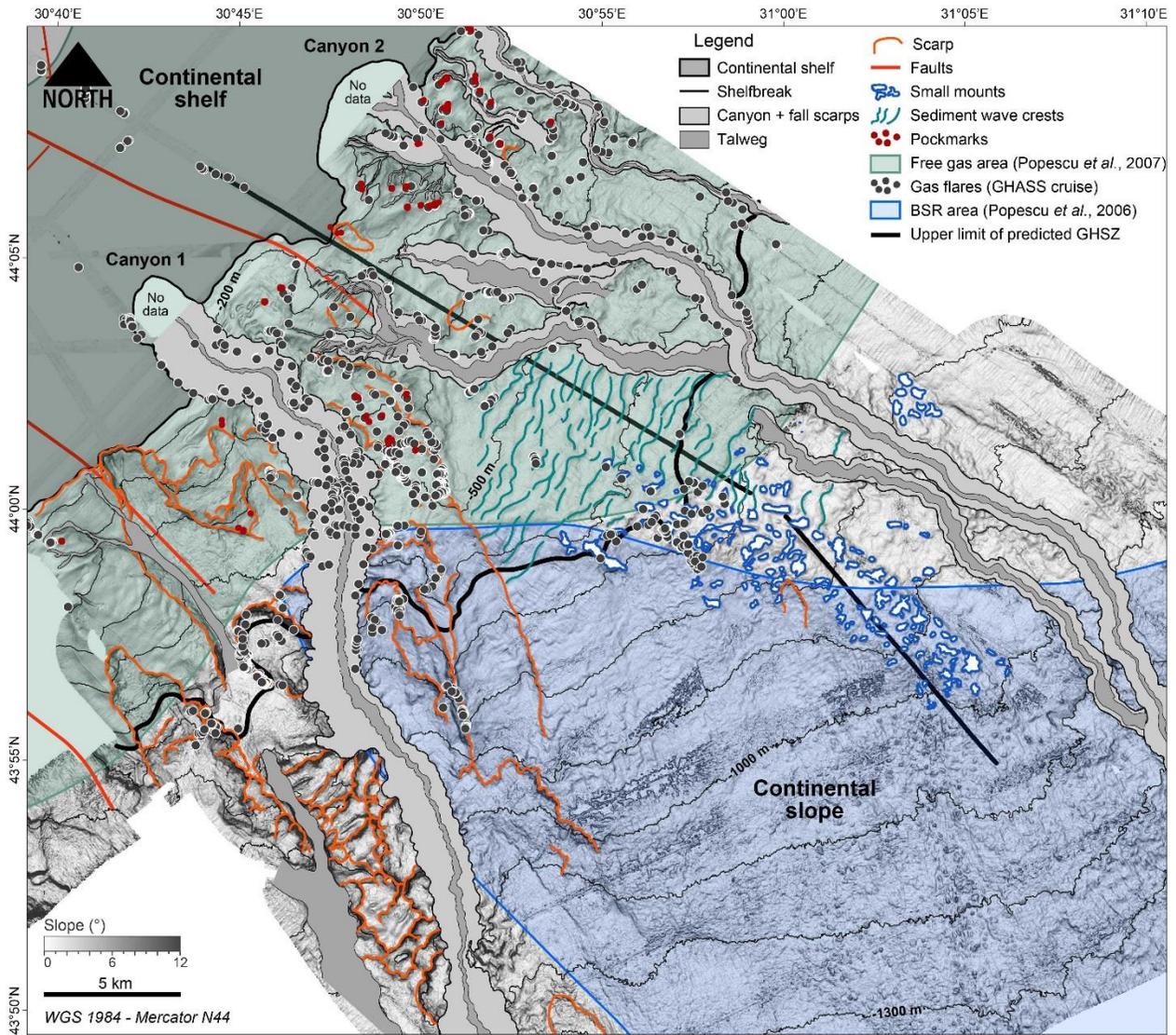
520 Figure 7: Dip map derived from the bathymetric map of the study area with superimposed (1)  
521 geomorphological features/zones, (2) limits of the GHSZ and (3) presence of measured gas bubbles in the  
522 water column. Red marks stand for water column acoustic anomalies recorded from Sept 1<sup>st</sup> to 15<sup>th</sup>, 2015.  
523 The white rectangles indicate the areas of inset. The insets highlight key zones showing the spatial  
524 distribution of bubbles along headwall scarps (A), pockmarks – destabilized zones (B), canyon flanks (C),  
525 and the landward termination of the GHSZ (D).

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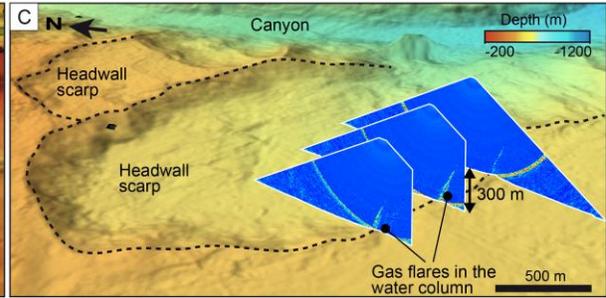
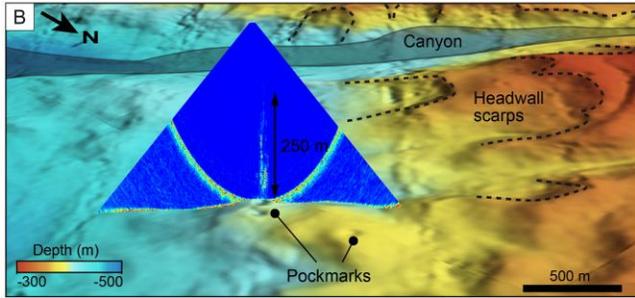
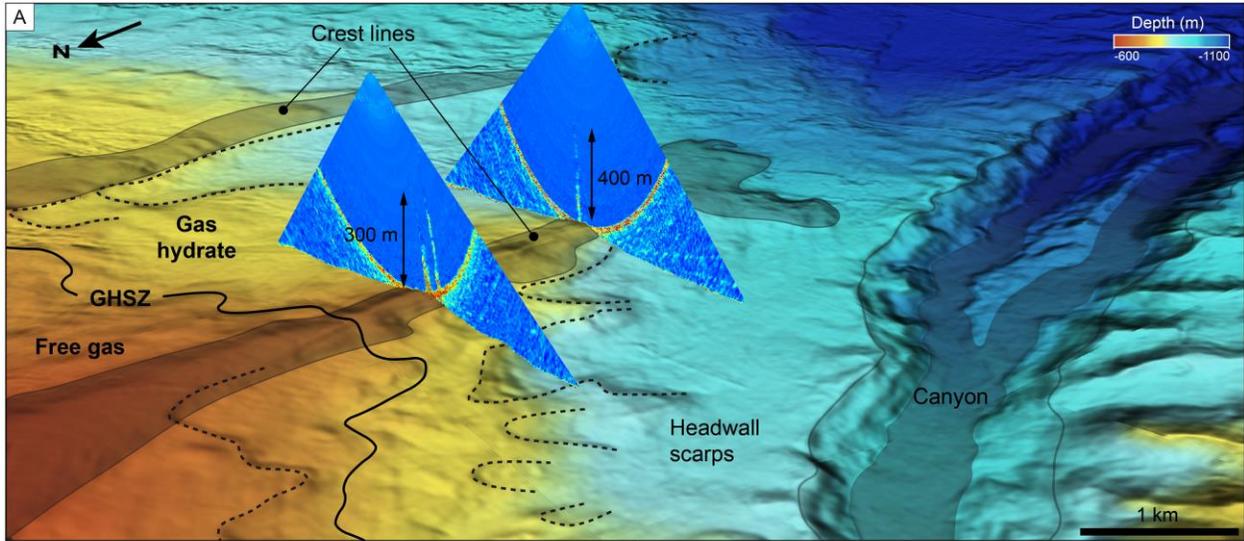
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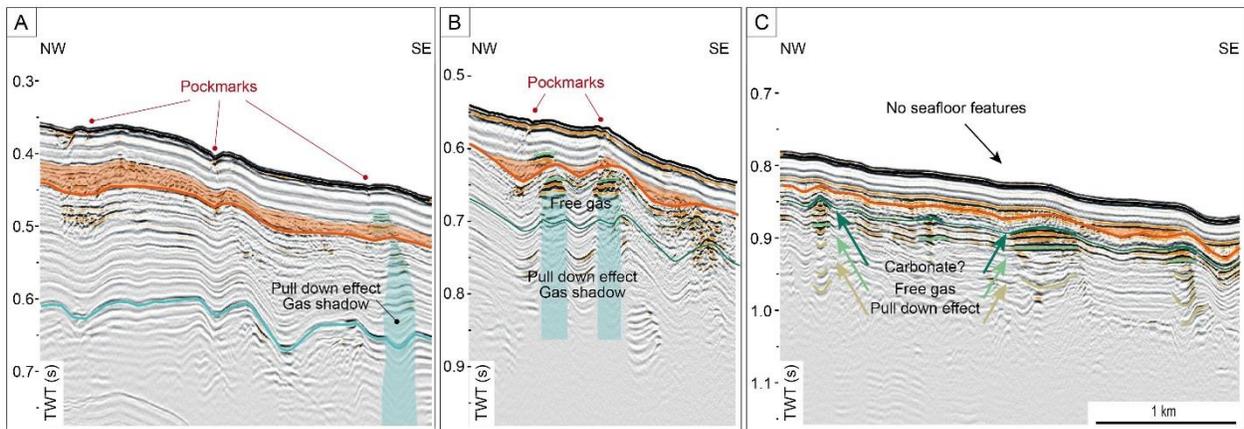
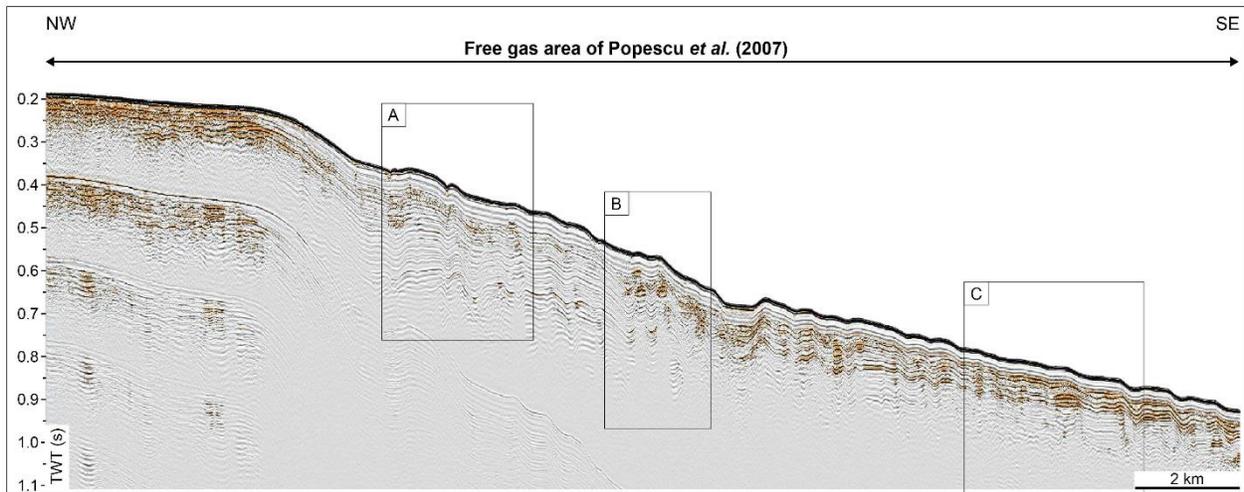
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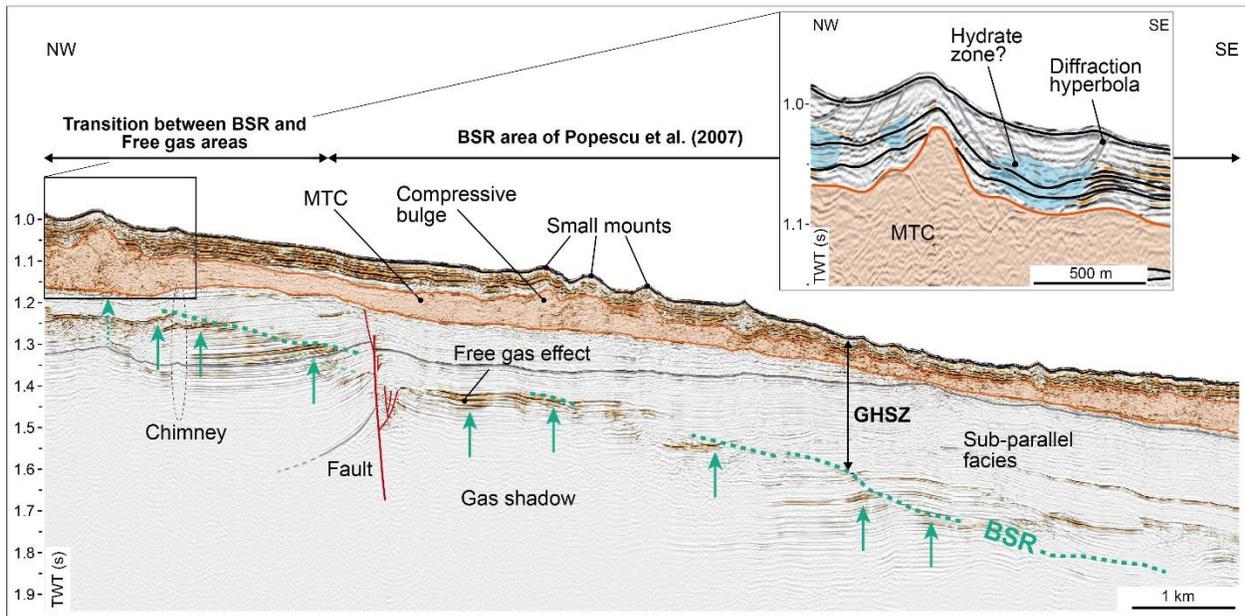
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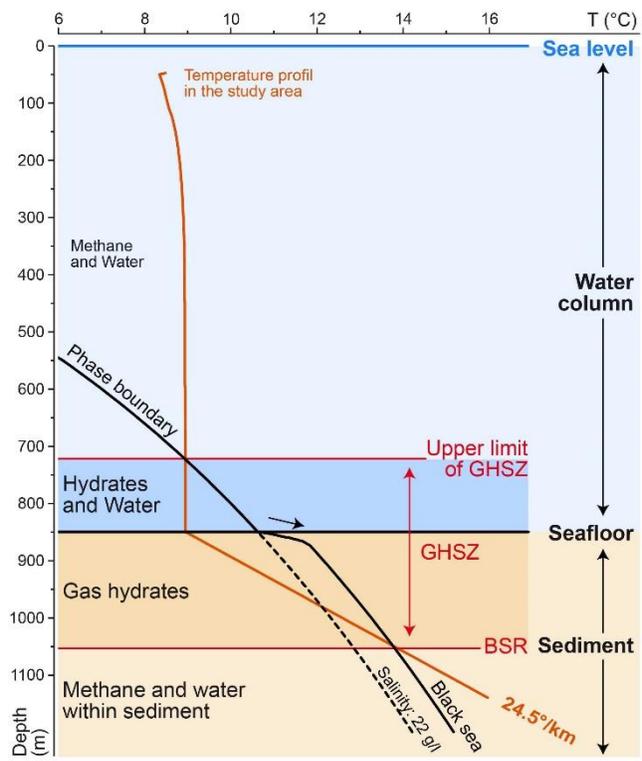
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