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The Origin of Gas Seeps in the Northern Adriatic Sea

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ABSTRACT

A multidisciplinary approach has been used for the first time to study the widespread occurrence of hydrocarbon seeps in the northern Adriatic Sea. Geological, geophysical and geochemical analyses were performed to identify and characterize the gas-charged fluids occurring throughout the Plio-Quaternary succession, and to date the shallow gas seeping at three leakage sites. The analysis of CHIRP, morpho-bathymetric and multichannel seismic data allowed us to identify different types of gas-related features, which occur within the whole Plio-Quaternary succession up to the seafloor and to the water column. Quantitative analyses of CHIRP data were conducted to better define, characterize and quantify the gas occurrence within the uppermost stratigraphic succession. CHIRP data also allowed identifying the gas leakage sites. Three gas seepage areas were sampled with the aim to determine the gas composition and origin. The isotopic analyses revealed that seep gases are microbial in origin, and are primarily composed by methane, mostly formed within relatively laterally persistent Late Pleistocene peat layers, which are widely distributed throughout the northern Adriatic Sea and represent the main source of organic matter feeding the seeping gases.

Key words: *gas seeps, methane, fluid flow in porous media, isotope geochemistry, northern Adriatic Sea.*

INTRODUCTION

Natural gas seeps in marine environments can release considerable amounts of carbon into the water column and the atmosphere (Etiope, 2012); in some cases, individual seeps can release CO_2 and CH_4 emissions on the order of tens to hundreds of tons per year (e.g., the Katakolo seep in Greece, the Tommeliten seepage in the North Sea, the seeps off of Panarea Island in the Southern Tyrrhenian Sea; see Etiope *et alii*, 2014 and the references therein). The occurrence of seeps and their distribution are controlled by a number of factors, including the availability of a gas source, the presence of faults and fracture systems, the vertical and lateral changes in sediment properties (i.e., porosity and permeability) and in their depositional history, the sedimentation rates, together with hydrocarbon and methane hydrate exploration (Berndt, 2005; Kessler *et alii*, 2005; Cartwright, 2007; Gay *et alii*, 2007; Moerz *et alii*, 2007; *Van* Rensbergen *et alii*, 2007; Anka *et alii*, 2012 and references therein; Toth *et alii*, 2014; Zhao *et alii*, 2015; Kramer *et alii*, 2017).

Understanding the mechanisms for natural gas seeps is considerable also because little is still known about the contributions of CO_2 and CH_4 from natural emissions into the atmosphere, where they play a major role as greenhouse gases, and whose drastic reduction by 2050 would strongly contribute to limiting warming to 1.5°C (Rogelj, Shindell, Jiang *et alii*, 2018); especially in shallow (<100 m) marine environments, the potential transfer of gas from sediments into the water column and subsequently into the atmosphere may occur (Etiope *et alii*, 2014). In fact, according to McGinnis *et alii* (2006), most marine sources of the atmospheric CH_4 are located in water depth less than 100 m, as testified by the great amounts of CH_4 fluxes in the order of 200 μmol m^{-2} d^{-1} recorded in the Black Sea (Schmale *et alii*, 2005).

In the northern Adriatic Sea, gas seeps are widespread. Both the presence of gas seeps and the occurrence of gas-related seabed and sub-seabed features have been widely documented (Newton & Stefanon, 1975; Stefanon, 1980; Colantoni *et alii*, 1998; Conti *et alii*, 2002; Panieri, 2006; García-García *et alii*, 2007; Gordini, 2009; Gordini *et alii*, 2012; Donda *et alii*, 2013 and 2015). Multi-resolution geophysical data collected by the National Institute of Oceanography and Applied Geophysics (OGS) in 2009 and 2014 revealed the widespread occurrence of acoustic anomalies associated with the presence of gas in the form of sub-vertical zones interpreted as gas chimneys, through which gas can migrate up to the seafloor (Donda *et alii*, 2015). These gas seeps appear to be locally associated with distinct rock outcrops, represented by bio-concretionned carbonate rocks, irregularly distributed on the seafloor, that are commonly named "tegnue" or "trezze". Since the past century, different hypotheses regarding the origin of these lithified deposits have been invoked. Recent geological and geophysical data support two possible genetic models: the first one has been developed for a group of rocky outcrops named "Tegnue di Chioggia", which are interpreted as coralligenous buildups growing along paleo tidal channels (Tosi *et alii*, 2017); the second model involves the role of the gas seeps, thus leading to interpret such deposits as methane-derived carbonates (Gordini, 2009; Gordini *et alii*, 2012; Donda *et alii*, 2013 and 2015). In fact, the occurrence of both deep and shallow gases throughout the Plio-Quaternary succession is well known in the study area. The former are microbial gases occurring within Pliocene-to-Pleistocene turbiditic sands, as multiple pools within thin sand beds at approximately 1200-1500 mbsf (Casero, 2004; Bertello *et alii*, 2008; Casero & Bigi, 2013). They have been exploited during the 60's, while no hydrocarbon-

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related activities are currently underway in the study area. Shallow gases permeate the Late Pleistocene and Holocene succession, with gas peaks between 15 and 25 m below the seafloor (Curzi *et alii*, 1997; Curzi *et alii*, 1998; Calderoni *et alii*, 1998; Donda *et alii*, 2008). The current hypothesis is that gases most likely originate from laterally persistent peat layers, which are widely distributed throughout the northern Adriatic Sea and formed between 16,000 and 24,000 years BP in an alluvial plain environment (Gordini, 2009; Correggiari *et alii*, 1996; Zecchin *et alii*, 2011) during phases of climate improvements of the last glacial phase (i.e., the Laugerie Interstadial; Zecchin *et alii*, 2011).

Here, we present the results of a multidisciplinary approach that led us to provide an integrated assessment of gas occurrence in the study area, which represents an excellent natural laboratory to study offshore gas seeps because they are widespread and easily accessible, i.e., they are close to the coast at shallow depths. In particular, quantitative analyses of CHIRP data were performed to determine the presence of gas within the uppermost stratigraphic succession, and geochemical analyses of three leakage areas led to define the origin of the seeping gases.

REGIONAL SETTING

The northern Adriatic Sea is a shallow-water (mean water depth: 25 m) sedimentary basin that is a part of the Padano-Adriatic foreland-foredeep domain, which comprises three orogenic chains, i.e., the northern Apennines, the Southern Alps and the Dinarides. Its present-day configuration is the result of two different tectonic stages: an extensional phase spanning the Mid-Late Jurassic into the Early Cretaceous and a complex Cenozoic compressional phase (Fantoni & Franciosi, 2010; Ghielmi *et alii*, 2010). Following the Cenozoic compressional events, the Po Plain and the northern Adriatic areas underwent crustal flexure, giving rise to deep foredeep basins filled with clastic sequences (Royden *et alii*, 1987). A turbidite succession with a thickness of up to 8 km was deposited in the Apennine foredeep basin and records four Late Miocene-to-Pleistocene compressional tectonic phases (Fantoni & Franciosi, 2010; Ghielmi *et alii*, 2010). The Miocene sequence is mostly composed of hemipelagic deposits belonging to the Gallare Marls, whereas the Plio-Quaternary succession is characterized by alternations of sand, clay and silt (Donda *et alii* 2013; Zecchin *et alii*, 2017 and references therein). Littoral and shelf sands, clayey sands and sandy clays compose the majority of the surficial sediments of the study area (Brambati *et alii*, 1988).

The oceanographic setting of the study area is characterized by a microtidal regime dominated by cyclonic circulation that is driven by thermohaline currents (Malanotte Rizzoli & Bergamasco, 1983; Bondesan *et alii*, 1995). In the Venice area, the monthly distribution of high tides recorded between 1872 and 2017 shows tides peaks during winter time, with oscillation in the order of 100 cm, but peaks of 140-150 cm also occur (Previsioni delle altezze di marea per il bacino San Marco e delle velocità di corrente per il Canal Porto di Lido - Laguna di Venezia). The frequency of such episodes reaches maximum during Autumn-Winter, when sea-level setup induced by winds blowing from southeast coincides with a low atmospheric pressure (Gacic *et alii*, 2004 and references therein).

MATERIALS AND METHODS

Data used in this study were collected on board the R/V OGS Explora in 2009 and 2014 in the framework of the "STENAP" (Seismostratigraphic and Tectonic Evolution of the northern Adriatic in the Plio-Quaternary) and "GANDI" (GAs emissions in the northern ADriatIc Sea) projects. The data set consists of approximately 1300 km of multichannel seismic lines, CHIRP sub-bottom profiles and swath morpho-bathymetry data (Fig. 1).

The acquisition parameters of the multichannel seismic data and the processing sequence is described in Donda *et alii* (2015). The seismostratigraphic interpretation has been calibrated with well logs obtained from hydrocarbon exploration boreholes drilled in the study area (Fig. 1), where several gas fields were discovered and exploited. Well data are available through the ViDEPI "Visibility of Petroleum Exploration Data in Italy" database (http://www.videpi.com); it consists in a huge data set comprising well and seismic data, in conjunction with technical reports concerning exploration permits collected by oil companies since the 50's.

The CHIRP data were collected through a hull-mounted Chirp Benthos system with 16 transducers and a sweep modulated bandwidth from 2 to 7 kHz. The ping rate was set to 0.125-0.25 s (with a vessel speed of 2 m/s, which corresponds to one trace per second). The data were stored in a SEGY format, and positioning data from a GPS system was automatically incorporated into the data headers.

Morpho-bathymetry data were collected with a Reson Seabat 8111 multibeam echosounder with a frequency of 100 kHz, simultaneously with the seismic and CHIRP data, and were stored and processed within Teledyne RESON's PDS2000 Multibeam software package.

Geophysical analysis of the CHIRP data

We performed a quantitative analysis of some key CHIRP profiles in order to constrain the gas occurrence within the uppermost stratigraphic succession. The chosen CHIRP data (see Fig. 2 for location) were acquired from different sectors of the study area; we have selected them based on their sedimentological and acoustic characteristics, i.e., clayey-predominant vs sandy-predominant seafloor (based on Brambati *et alii*, 1988), acoustic blanking, occurrence of gas flares, presence of a rock outcrop.

Seismic reflection coefficients are related to the acoustic impedance contrasts produced by the different petrophysical properties of sediments in the subsurface. The values of reflection coefficients typically decrease at the interfaces of gas-bearing sediments (Tinivella, 2002). In our estimation, we considered two reflecting surfaces, i.e. the seafloor and the gas front, where detectable as a high amplitude reflector on top of the blanking zone. We extracted the reflection coefficient between the seafloor and the gas front by using the method presented by Warner (1990) and Bull *et alii* (1998) that is based on the amplitudes of the primary seafloor reflection, the first multiple of the seafloor and the gas front. The reflection coefficients extracted from the CHIRP data were converted into P-wave velocities by using the following available information regarding the seawater and the shallow sediments: seawater - P-wave velocity: 1520 m/s, density: 1.03 g/cm^3; shallow sediments - grain density: 2.60 g/

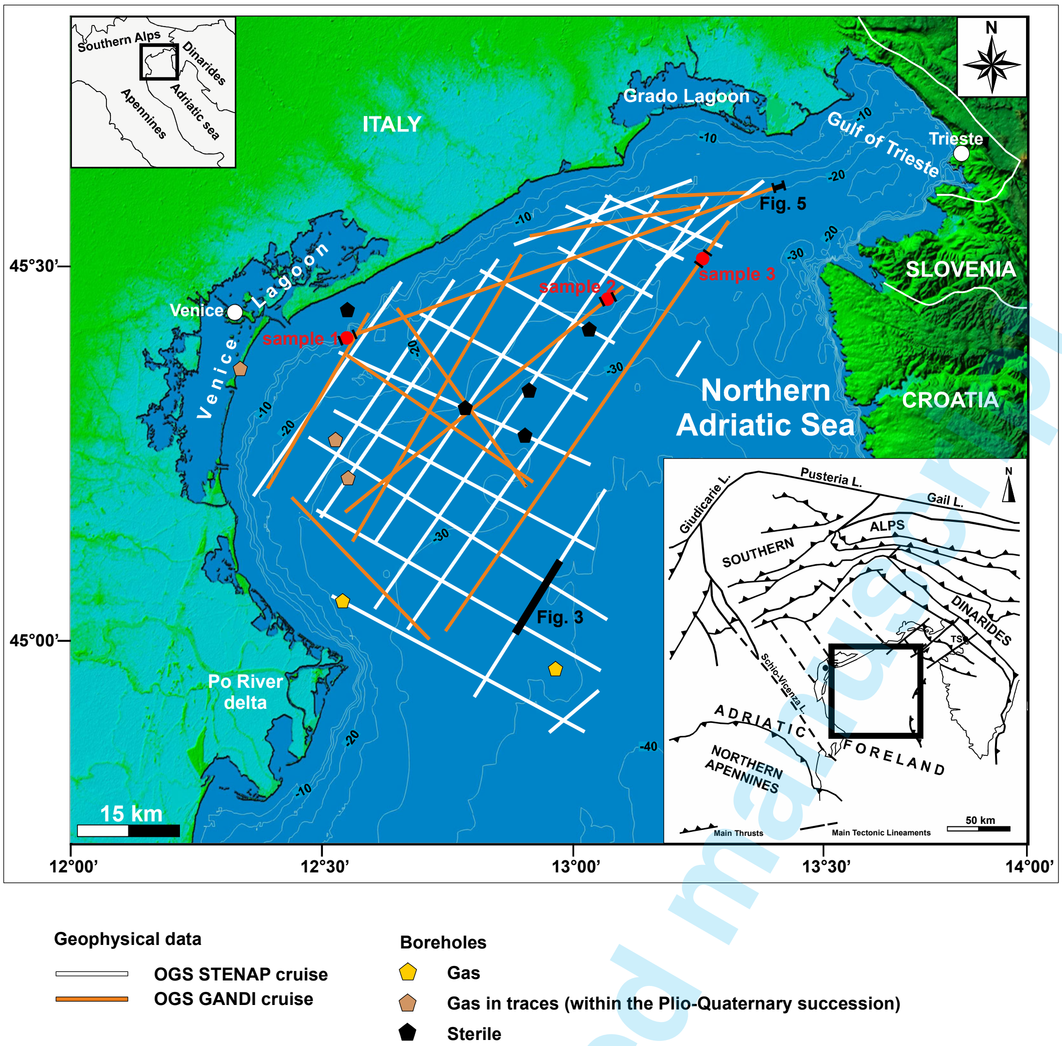


Fig. 1 - Location map of the geophysical data collected by OGS in the framework of the STENAP and GANDI cruises. The position and information concerning the hydrocarbon exploration boreholes is marked, based on the Videpi database (www.videpi.com). The location of the gas sampling sites and of the portions of CHIRP profiles of Fig. 4 (black ticks) are also shown (modified from Donda *et alii*, 2015).

cm³ (i.e., Schön, 1996), shallow-sediment porosity (in the selected areas): 38-58%.

The sediment porosity is a parameter needed for the quantitative analyses. The spatial distribution of the surface porosity in the northern Adriatic Sea is shown in Fig. 2, whereas the empty circles indicate the sample location; measurement-related values were achieved from the dbSEABED Colorado database: http://instaar.colorado.edu/~jenkinsc/dbseabed/coverage/adriaticsea/adriatico.htm.

The map was made using DIVA Interpolation (Data-Interpolating Variational Analysis): http://modb.oce.ulg.ac.be/mediawiki/index.php/DIVA.

Gas sampling and analyses

We have selected the seep sites for gas sampling based on the interpretation of our CHIRP data and the numerous scuba dives performed in the study area in the framework of several research projects (see also Gordini, 2009). We have tried to sample the seep sites three times: during the first two dives, seeps were scarce, intermittent and sometimes absent. Then, because gas flares are clearly imaged on the CHIRP data, we hypothesized that tides should have a role in the gas seeps occurrence, as suggested by Mikolaj & Ampaya (1973) and Boles & Clark (2011). Variations in seepage due to tidal forcing have been documented in a number of shallow (<20 m) environments, where pressure changes due to tide oscillations cause 5 to 20% changes in seepage rates (Mikolaj & Ampaya, 1973). We then noticed that, during the first two dives, high tide conditions occurred. Taking into consideration the above mentioned studies, where a straight correlation between high tides and reduced flows, and between low tides and increased flows is highlighted (Boles & Clark, 2011), we planned the third scuba diving campaign at low tide conditions: the gas was seeping from the seafloor and gas bubbles were clearly identifiable in the water column. Gas samples were then collected in May 2016 from the R/V Castorino II, using a self-built device for the fluid accumulation and its transfer into suitable containers. The conveyor device consists of a funnel having a base of 0.1 square meters (0.35 m of diameter), mounted on a ballasted steel support that allows to distance the funnel of about 0.15 meters from the surface on which it is placed. Two divers placed the funnel conveyor/accumulator just above the selected gas seeps. After about 30/60 minutes, the gas accumulated in the funnel was removed and subsequently stored inside glass vacuum vials (250 ml of volume).

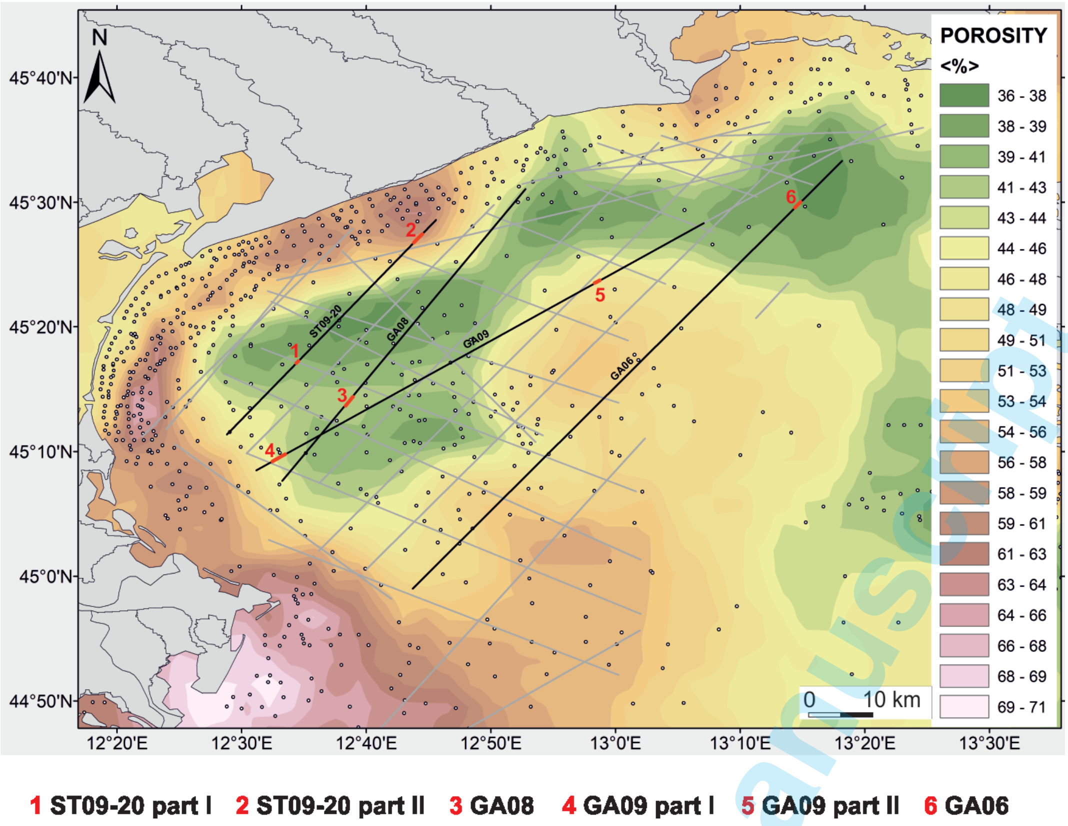


Fig. 2 - Spatial distribution of the surface sediment porosity throughout the northern Adriatic Sea (see text for details). Red ticks indicate the portions of the selected CHIRP profiles where the quantitative analyses have been performed (see also Table 1), whereas the empty circles indicate the sample location; measurement-related values were achieved from the dbSEABED Colorado database.

The gas samples were analyzed at Isotech Laboratories Inc. (Illinois, USA) to determine the proportions of C_1-C_6+ hydrocarbons, He, H_2, O_2, Ar, CO_2, CO, and N_2 (Shimadzu 2010 Gas Chromatograph (GC) with thermal conductivity (TCD) and flame ionization (FID) detectors; accuracy/precision +/-5% (C1-C4) and +/-10% (C_5-C_6+)) and the isotopic compositions of $d^{13}C_{CH4}$ (Finnigan Delta Plus XL mass spectrometer, accuracy/precision ± 0.1‰(1 s) for ^{13}C) and dD_{CH4} (Finnigan Delta V Plus mass spectrometer, accuracy/precision +/-4.0‰(1 s) for 2H). The gas samples were outsourced to Beta Analytic (Florida, USA) for the radiocarbon analysis of CH_4 via accelerator mass spectrometry. The ^{14}C1 pMC were converted into apparent age through the following equation: Age=-8034 ln (pMC)+37000, which provides an apparent age of ca. 32,000 to 34,000 yrs BP.

RESULTS

The occurrence of both deep and shallow gases is clearly imaged on the OGS dataset. On the multichannel seismic profiles, as also evidenced by Donda *et alii* (2013 and 2015), widespread acoustically wipe-out zones (Løseth *et alii*, 2009), interpreted as focused fluid flow pipes (Huuse *et alii*, 2010), indicate gas migration along sub-vertical pathways (Fig. 3). Gas chimneys commonly root at the base of the Pliocene succession and affect the whole Plio-Quaternary stratigraphic sequence; they are also locally associated with Cenozoic sub-vertical faults. High amplitude reflections at the chimney top (bright spots) are also often recognizable and are interpreted as due to local accumulations of upward migrating gas (Fig. 3).

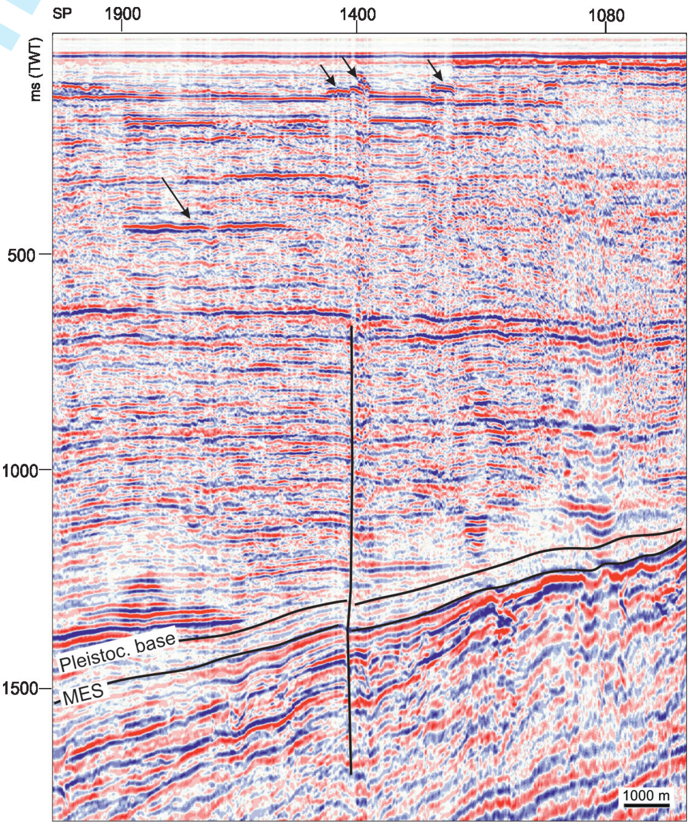


Fig. 3 - Part of the ST09-11b multichannel seismic profile (see Fig. 1 for location). Arrows indicate high amplitude reflections (bright spots), revealing the occurrence of local gas accumulations. A focused fluid flow pipe is recognizable just above of the subvertical fault affecting the Miocene-Pleistocene succession. MES: Messinian Erosional Surface.

On the CHIRP data, we observe diffuse acoustic blanking, which almost completely masks underlying reflections in the stratigraphic succession. The acoustic blanking is bounded at the top by a high amplitude, sub-horizontal to highly irregular acoustic reflection that represents the gas front (Fig. 4). Truncated reflections at the gas front indicate that gas-trapping horizons are not continuous. Focused acoustic blanking also occurs, i.e., gas chimney-like features (Fig. 4) that, in places, reach the seafloor. In the northernmost sector of the study area, we have identified pockmarks either on CHIRP profiles and at the coincident multibeam data; they display a maximum width and depth of 15 m and 50 cm, respectively (Fig. 5). Rock outcrops are recognizable on the CHIRP data as rugged seafloor morphologies, acoustically opaque and up-bounded by a high amplitude reflection, with maximum elevations above the seafloor of ca 1 m (Fig. 4; they and reveal a wide range of sizes, shapes and distributions see also Gordini *et alii*, 2012). The findings of this study support the methane-related origin of at least part of the rock outcrops occurring in the northern Adriatic Sea, although we do not reject the hypothesis of Tosi *et alii* (2017) for the Tegnue di Chioggia outcrops, where, in fact, no gas seeps have been detected.

For a comprehensive description of these features, see Gordini (2009), Gordini *et alii* (2012), Donda *et alii* (2015), and Tosi *et alii* (2017) (Fig. 4). Several seep areas are recognizable on the CHIRP data. Based on the acoustic data, gas flares have variable heights that range from 4 m to approximately 20 m (Figs. 4 and 5). Flares occur along the water column just above focused fluid migration pathways, in areas of diffuse acoustic blanking and at rock outcrops.

To better constrain the data interpretation, we have then focused our analysis to the uppermost stratigraphic section through a quantitative analysis of key portions of CHIRP data.

In a first instance, knowing the acoustic impedance values of the sediments below the seafloor, we estimated the impedance contrast, and thus, the P-wave velocity at the top of the free gas layer. This allowed us to provide indications regarding the free gas content by using a

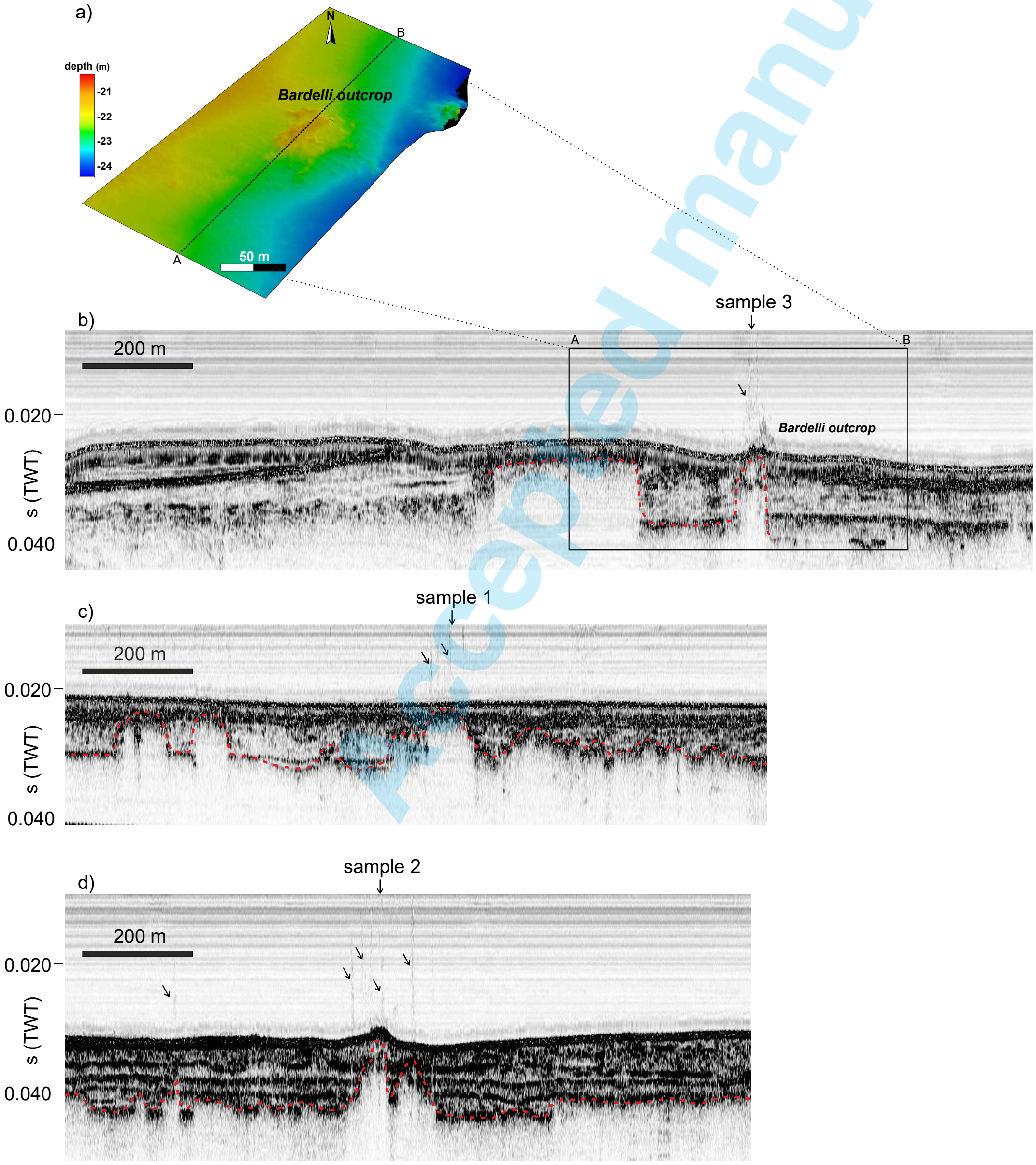


Fig. 4 - Parts of GA06 multibeam data (**a**) and of the coincident CHIRP profile (**b**) collected across the Bardelli outcrop, and of GA01 (**c**) and GA09 (**d**) CHIRP profiles collected on the leakage areas where the gas have been sampled (see Fig. 1 for location). The dotted red line represents the gas front. Arrows indicate gas flares in the water column. The quantitative analyses resumed in Fig. 8 have been performed on the same portion of GA06 profile shown here.

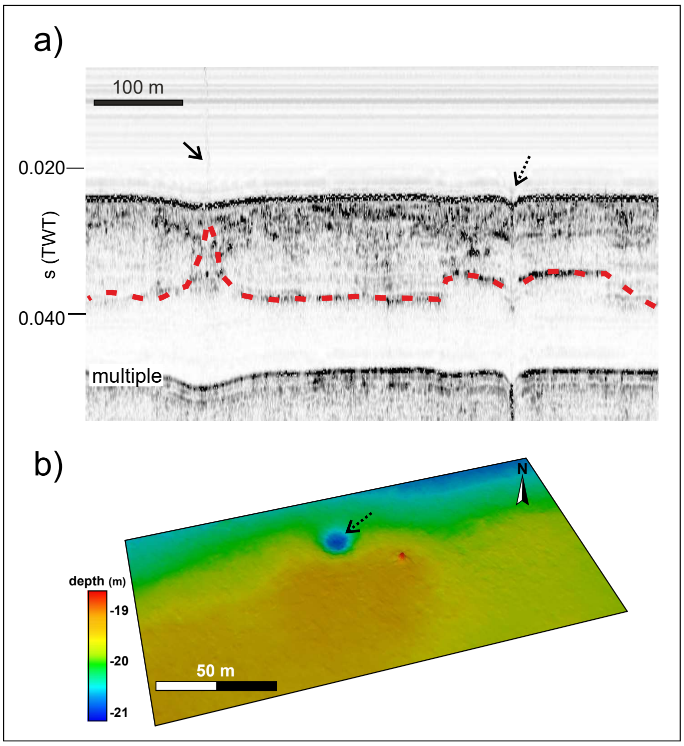


Fig. 5 - Part of GA01 CHIRP profile (**a**) and of the corresponding multibeam bathymetry data **(b)** collected in the northernmost sector of the study area (see Fig. 1 for location), where pockmarks have been identified (dotted arrows). The gas front and gas flares in the water column are also indicated by the red dotted line and by the arrow, respectively (Fig. 5a).

theoretical velocity model. Where possible, we selected a reflector between the seafloor and the top of the free gas layer to better constrain the amplitude and the extracted velocities. We used Biot's theory and its approximation in order to convert the velocity into an estimate of the free gas content (i.e., Tinivella & Carcione, 2001). Figure 6 shows an example of the behaviour of the compressional velocity versus the free gas saturation under the assumption that the water depth is 20 m (the average value extracted from the bathymetry data), the top of the free gas layer is 5 m below the seafloor (average value of the gas front depth based on the analysed CHIRP data), and the porosity ranges from 0.38 (dashed lines) to 0.58 (dotted lines).

We assumed two distributions of the free gas within the pore space, namely, a patchy (gray lines in Fig. 6) and a uniform distribution (black lines in Fig. 6). As expected, if we consider a patchy distribution, the velocity is higher relative to the velocity obtained considering a uniform distribution (i.e., Tinivella, 2002). The amplitude of the reflection interpreted as the top of the free gas layer is expected to be negative due to the velocity decrease resulting from the presence of gas. Fig. 6 shows that, if the porosity is equal to 0.58, the velocity is slightly smaller than that obtained by assuming a porosity of 0.38 in the case of a uniform distribution, while the velocity is constant for a patchy distribution. The relationship between the theoretical absolute reflection coefficient and the free gas saturation, which is evaluated using a viscoelastic single-phase constitutive model (i.e., Carcione & Tinivella, 2000 and 2001), is shown in Fig. 7.

Only the absolute value of the reflection coefficient is reported, because CHIRP data represents an amplitude envelope without phase information. These results, which are consistent with the velocity behaviour, indicate a drop in the reflection coefficient with a small concentration of free gas that is more pronounced if a uniform distribution is assumed. Supposing an increasingly high free gas content (with free gas on the order of approximately 10% and 20% in the pore space with a uniform and patchy distribution, respectively), the absolute value of the reflection coefficient strongly increases, thereby producing an ambiguity if only the reflection coefficient value is interpreted; thus, other information, such as geological data, are necessary to constrain the interpretation. For example, for a patchy distribution, an absolute value of the reflection coefficient equal to 0.2 can be interpreted as indicative of the presence of up to approximately 20% of free gas in the pore space.

The mean values of the reflection coefficient and P-wave velocity are reported in Table 1.

The sampled gases are mainly composed of methane (Table 2). The methane δ^{13}C values range between -73.7 and -64.7‰ VPDB, while the δD values range from - 264.2 to -223.6‰ VSMOW. The ^{14}C1 pMC values range from

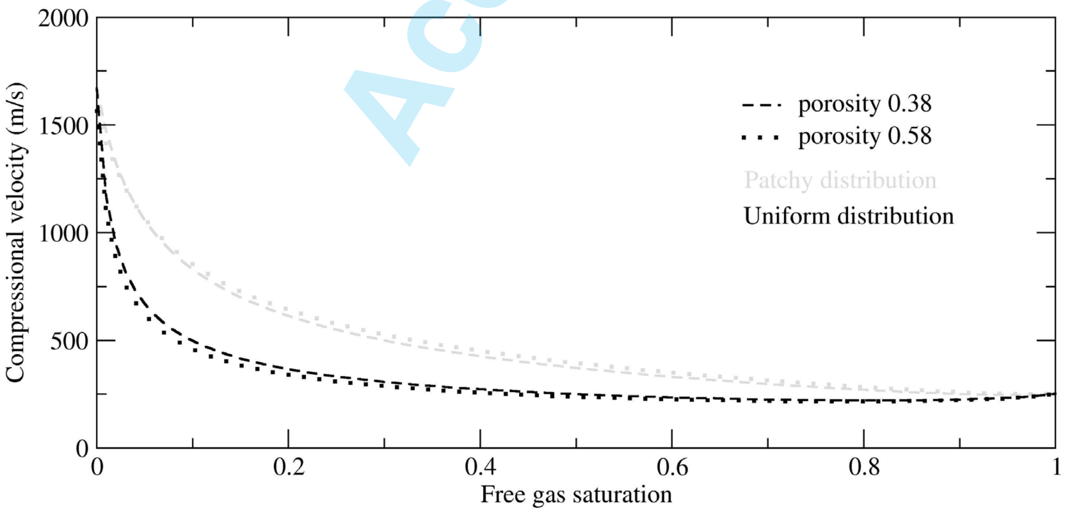


Fig. 6 - Compressional velocity versus free gas saturation, considering different porosity values (0.58: dashed lines; 0.38: dotted lines) and different distribution of free gas in the pore space (uniform: black lines; patchy: grey lines). See text for details.

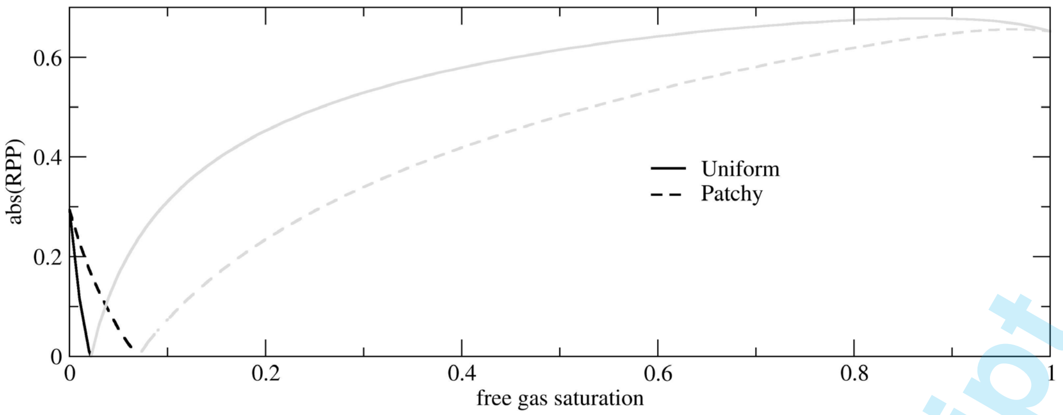


Fig. 7 - Theoretical absolute reflection coefficient versus free gas saturation at the seafloor, considering a uniform (solid line) and a patchy (dashed line) free gas distributions in the pore space. The water velocity and the water density are assumed equal to 1.5 km/s and to 1.03 g/cm³, respectively. A sediment density of 2.60 g/cm³ and a porosity of 0.50 have been considered respectively. Grey lines indicate negative reflection coefficients.

1.3 to 1.8, which indicates an apparent age of the organic material source for the microbial gas between ca. 32,000 to 34,000 yrs BP. The real age of the source organic material is equal to or less than the apparent age.

DISCUSSION

Quantitative analysis on the CHIRP data

An example of CHIRP data analysis performed on the GA 06 profile is shown in Fig. 8.

We estimated the P-wave velocity from reflection coefficient by using a layer stripping approach. In fact, knowing the reflection coefficient at the interface and the impedance of the upper layer, the bottom impedance can be easily evaluated. Knowing the seawater and the shallow sediment properties, the P-wave velocity at the seafloor and the gas front can be extracted from CHIRP data analysis (Fig. 8 B,D). To notice, in our analysis we assume that the free gas in the pore space is not present at the seafloor and the reflection coefficient is positive. In fact, as reported in Fig. 7, if the free gas presence is assumed at the seafloor, high values should result (about 10% and 25% if a uniform or

TABLE 1

Results of the CHIRP Data Analysis. The Error Corresponds to the Standard Deviation. The parts of CHIRP data where the analyses have been performed are also shown.

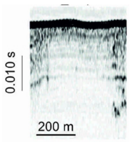
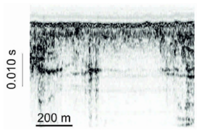
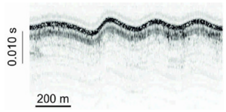
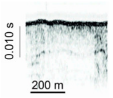
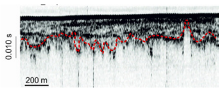
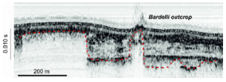
Acoustic Profile	**Porosity (%)/Density (g/cm³)**	**Average reflection coefficient at the seafloor**	**Average reflection coefficient at the gas front**	**Average P-wave velocity at the seafloor (km/s)**	**Average P-wave velocity at the gas front (km/s)**	**CHIRP**
ST09-20 (part I)	41/1.96	0.30±0.05	/	1.5±0.2	/	
GA 08	42/1.94	0.27±0.08	/	1.5±0.3	/	
GA09 (part I)	44/1.91	0.28±0.09	/	1.5±0.3	/	
GA 09 (part II)	48/1.84	0.28±0.06	/	1.5±0.2	/	
ST09-20 (part II)	58/1.69	0.28±0.08	-0.04±0.03	1.7±0.3	1.4±0.1	
GA 06	38/2.00	0.37±0.11	-0.07±0.08	1.8±0.5	1.5±0.3	

TABLE 2

Molecular and Isotopic Composition of the Gases Sampled at Three Leakage Sites (see Fig. 1 for location).

SAMPLE 1	Chemical mol. %	d13 (‰)	dD (‰)	14C conc. (pMC)	SAMPLE 2	Chemical mol. %	d13 (‰)	dD (‰)	14C conc. (pMC)	SAMPLE 3	Chemical mol. %	d13 (‰)	dD (‰)	14C conc. (pMC)
Carbon Monoxide	nd				Carbon Monoxide	nd				Carbon Monoxide	nd			
Helium	nd				Helium	nd				Helium	nd			
Hydrogen	nd				Hydrogen	nd				Hydrogen	nd			
Argon	0.431				Argon	0.0970				Argon	0.415			
Oxygen	9.70				Oxygen	0.98				Oxygen	7.67			
Nitrogen	35.92				Nitrogen	8.95				Nitrogen	31.36			
Carbon Dioxide	0.99				Carbon Dioxide	0.40				Carbon Dioxide	0.049			
Methane	52.96	-64.75	-264.2	1.3±0.1	Methane	89.57	-73.69	-237.6	1.8±0.1	Methane	60.50	-76.22	-223.6	1.8±0.1
Ethane	0.0003				Ethane	0.0021				Ethane	0.0011			
Ethylene	nd				Ethylene	nd				Ethylene	nd			
Propane	nd				Propane	nd				Propane	nd			
Propylene	nd				Propylene	nd				Propylene	nd			
Iso-butane	nd				Iso-butane	nd				Iso-butane	nd			
N-butane	nd				N-butane	nd				N-butane	nd			
Iso-pentane	nd				Iso-pentane	nd				Iso-pentane	nd			
N-pentane	nd				N-pentane	nd				N-pentane	nd			
Hexanes+	0.0003				Hexanes+	nd				Hexanes+	nd			

patchy distribution is assumed respectively), contrary to the gas concentration estimated in this area, which are in order of a few percent (García-García *et alii*, 2007). So, we draw the conclusion that local high values of the reflection coefficients of the seafloor should be associated to lithological changes, as confirmed by the absence of reversal phases along the corresponding seismic lines, i.e. absence of evidence of negative reflection coefficient at the seafloor (i.e., Fig. 8F). Therefore, we suggest that the diffuse acoustic blanking observed below the seafloor is associated with coarser-grained sediments (sand and coarse sand in the case of GA08, GA09-part I and ST20-part I) and/or relatively compacted sediments relative to adjacent areas (GA09-part II). These properties would serve to absorb the energy of seismic signals, thereby resulting in the observed acoustic blanking.

We conducted the same analyses in the sectors where gas flares in the water column confirmed the presence of active gas emissions (Fig. 4). This is the case for profiles ST020-part II and GA06, where the reflection coefficient and velocity were computed at the seafloor and at the potential gas front (Fig. 4; Table 1). The low reflection coefficient and acoustic velocity values at the latter reflector confirm the presence of gas within the sediments with an estimated mean volume concentration on the order of a few percent based on the velocity data reported in Table 1. These values agree with gas analyses performed on sediment cores collected in the southern portion of the study area, which reveal gas concentrations of at least 1-2% within the mud-dominated Holocene highstand system tract (García-García *et alii*, 2007). The occurrence of gas is represented within our CHIRP data by diffuse acoustic blanking bounded on the top by a high amplitude reflector (Fig. 4). The highest reflection coefficient values at the seafloor in profile GA06 are associated with a rock outcrop.

Gas analysis

CHIRP data show the occurrence of widespread gas escapes into the water column in the form of numerous gas bubble streams, which are imaged as 'acoustic flares' on echograms (Gordini, 2009; Donda *et alii*, 2013 and 2015). However, only a few gas samples have been collected and analyzed, the results of which revealed that the gases are mainly composed of methane (CH_4: 81-84%; N_2: 15-18%; O_2: 0.7-1.3%; Gordini, 2009; Gordini *et alii*, 2012). Further measurements where performed in the northernmost portion of the Adriatic Sea, i.e. the Gulf of Trieste, which was selected as a testing site for the implementation of techniques to monitor submarine gas fluxes and their ecological impacts in the framework of the European CO2GeoNet project. A data record spanning approximately one year revealed the occurrence of both CH_4 and CO_2 at low concentrations, i.e. ca 0.2% and 0.5%, respectively (Faber *et alii*, 2009). Except for these localized measurements, a whole characterisation of the gas seeps in the northern Adriatic Sea was still missing. In our samples, we discovered several pieces of evidence for the microbial origin of the methane: 1) the ratios of methane to ethane and propane (C1/(C2 +C3)) > 1000 (Bernard *et alii*, 1976); 2) the $\delta^{13}C\text{-}CH_4$ values were generally less than approximately -55‰, which is expected for biogenic gases (Bernard *et alii*, 1976; Whiticar *et alii*, 1986; Whiticar, 1999); and 3) the stable carbon and hydrogen isotope compositions of methane plotted in the field of biogenic gas (Whiticar, 1999; Fig. 9). These characteristics argue for a microbial origin through CO_2 reduction, which is the main primary methanogenic pathway in marine sediments (Claypool & Kvenvolden, 1983; Whiticar, 1999).

The near absence of hydrocarbons with densities greater than methane (C_2-C_6) from the gas composition

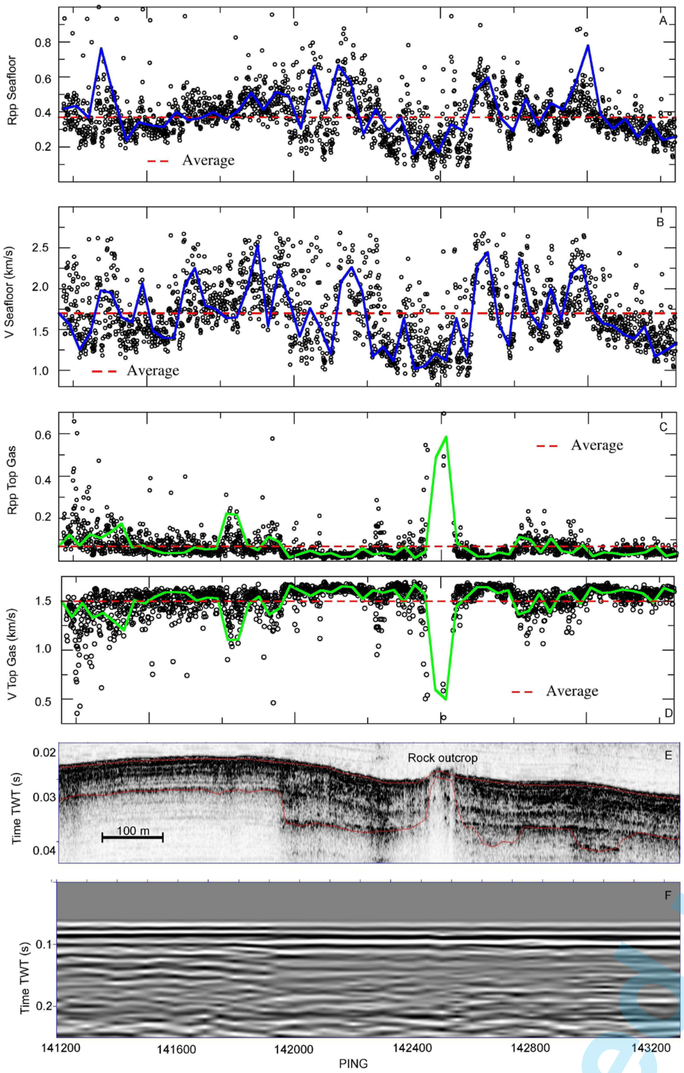


Fig. 8 - CHIRP data analysis performed on the GA 06 CHIRP profile (see Figure 2 for location). Black circles: picked events; blue line: envelope; red dotted line: average of picked events. **A**. Reflection coefficient of the seafloor; **B**. P-wave velocity of the seafloor extracted from the reflection coefficients; **C**. Absolute reflection coefficients of the gas front; green line: envelope; **D**. P-wave velocity of the gas front extracted from the reflection coefficients; **E**. portion of the CHIRP profile with the picked reflections, i.e. seafloor and gas front (red lines); **F**. coincident portion of the near offset multichannel seismic profile.

in addition to the observed stable isotopes and the age of the organic source material for the methane collectively support a relatively shallow primary microbial methane source (Schoell, 1980; Whiticar *et alii*, 1986).

ORIGIN OF THE GAS SEEPS

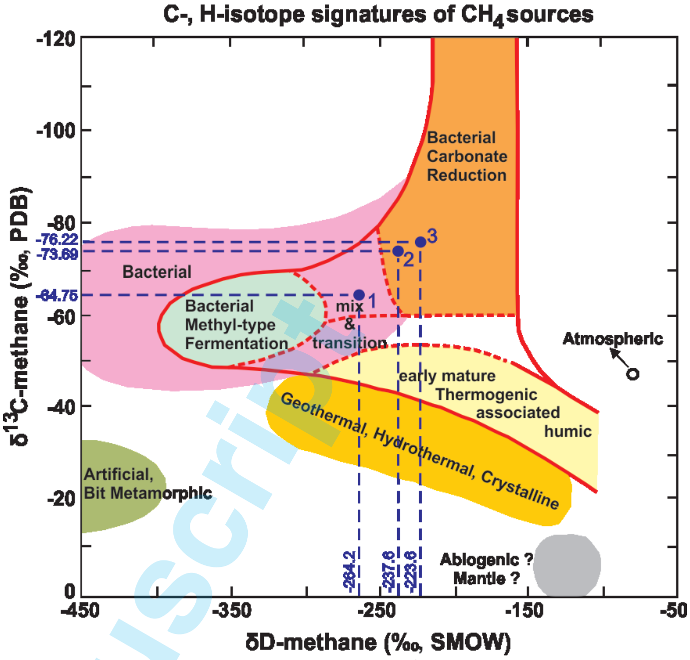


Fig. 9 - $\delta^{13}CH_4$ vs. δD-CH_4 plot showing the major methane types (after Whiticar, 1999, modified). The plotted stable carbon and hydrogen isotope compositions of the gases sampled in the northern Adriatic Sea (indicated as "1", "2" and "3") reveal the microbial origin of the methane.

Different types of gas-related features are recognizable within the whole Plio-Quaternary succession in the northern Adriatic Sea (Fig. 10), and we hypothesize two main sources feeding the shallow gas accumulations and the gas seeps (Fig. 11). The deeper one is evidenced on the multichannel seismic data by seismic features such as bright spots and gas chimneys rooting at depths of ca 1 s (TWT), and by direct evidence of gas occurrence provided by the available composite logs of boreholes drilled in the study area. No information about the depth of the gas reservoirs are presently available, but the bright spots identified in correspondence of these reservoir, lying at a depth of about 1 s (TWT), would represent their geophysical expression.

Available data regarding the gas composition of the gas reservoirs reveal the predominance of CH_4 (ca 99%), with the occurrence of N_2 and CO_2 in the order of 1% and 0.02% in vol, respectively (e.g. Panieri, 2006). We suggest that these gases contribute to the shallow gas accumulations, hypothesizing an upward migration of gases through high-permeability layers hosted within the Plio-Quaternary succession and, locally, to sub-vertical faults. Although tectonic stresses appear to be the most efficient triggering mechanism for seeps (Talukder, 2012), gases may migrate upward via permeable pathways or they could be driven by buoyancy forces that exceed the threshold pressure of the overburden sediments, according to García-García *et alii* (2007).

Only part of the deeper gas would reach the surface, since the characteristics of the uppermost (i.e., 50 m) stratigraphic succession, which is dominated by fine-grained sediments according to data from the Venezia 1 and Triglia Mare 1 boreholes (www.videpi.com), indicate that it could have acted as a seal for upward-migrating gases; this is evidenced by the high amplitude reflections at the top of gas chimneys, which record localized, shallow gas accumulations. The acoustic characteristics of the uppermost sedimentary sequence reveal that gas is widely distributed throughout the study area. Isotopes of our gas samples revealed an apparent organic matter age ranging from approximately 32,000 to 34,000 years BP. These ages are consistent with the Denekamp Interstadial, during which time the improved

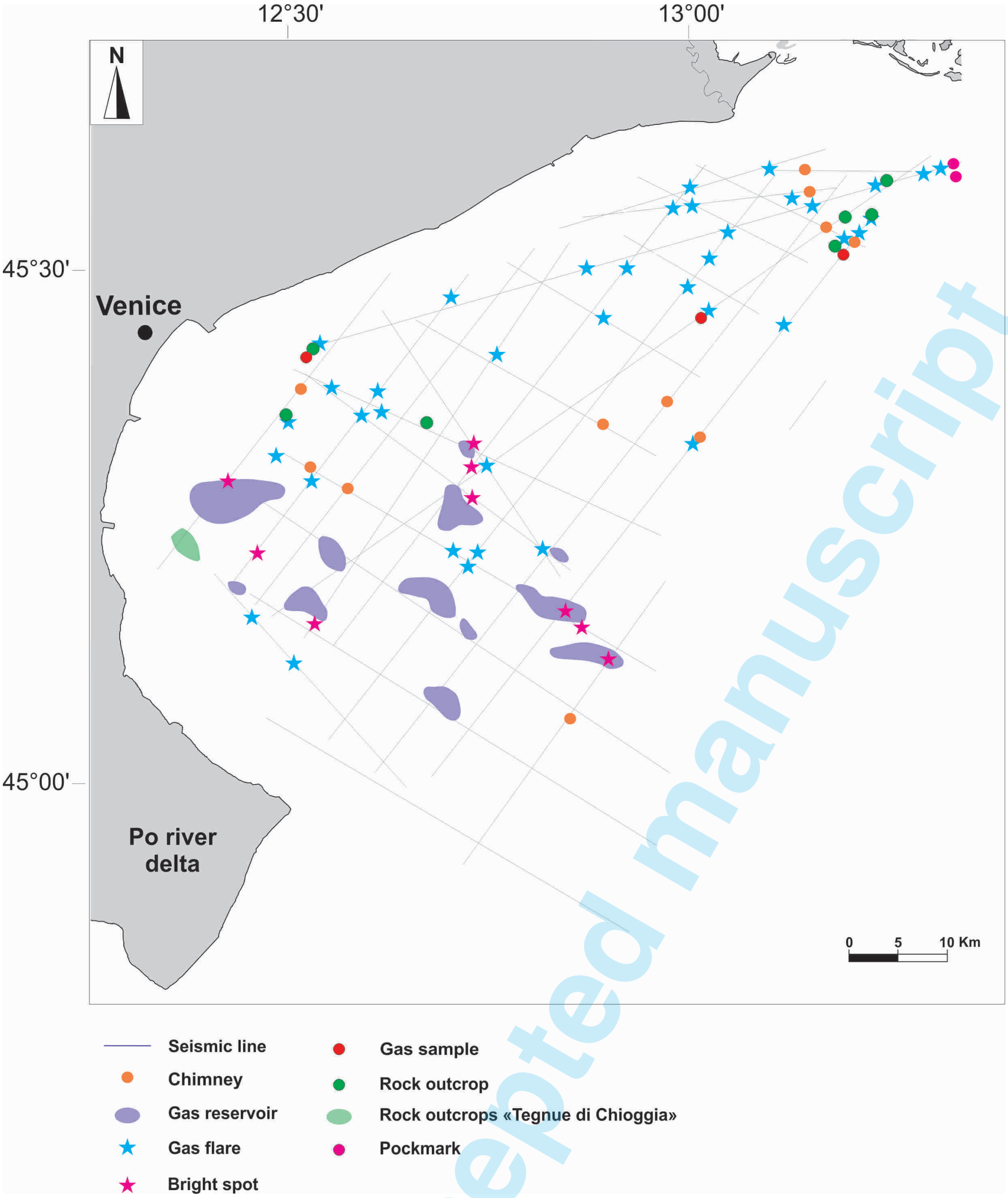


Fig. 10 - Distribution of the gas-related features identified in the study area. The “Tegnue di Chioggia” and the gas reservoirs have been mapped following Tosi *et alii*, (2017) and Casero (2004), respectively.

climate conditions would have favored the deposition of peat layers (Canali *et alii*, 2007). We then suggest that the widespread occurrence of shallow gas, identified through both the seismostragraphic and quantitative analyses of the CHIRP data, mostly derives from the Late Pleistocene peat layers, possibly mixed with some gas of deeper origin (Fig. 11).

CONCLUSIONS

Geological, geophysical and geochemical analyses allowed to identify and characterize the gas-charged fluids occurring throughout the Plio-Quaternary succession, and to date the shallow gas seeping at three leakage sites. In particular:

- Different types of gas-related features have been identified within the whole Plio-Quaternary succession up to the seafloor and to the water column;
- Quantitative analyses of CHIRP data led to the determination of the presence of gas within the uppermost stratigraphic succession, and a definition of its mean volumetric concentration, which is on the order of a few percent.
- Geochemical analyses performed at three leakage areas revealed that seep gases are microbial, and ^{14}C isotopes indicate that they mostly originate from the degradation of organic material with an apparent age of ca. 32,000-34,000 yrs BP. These isotopic signatures suggest the formation of shallow methane within relatively laterally persistent, Late Pleistocene peat layers, which are widely distributed throughout the northern Adriatic Sea.

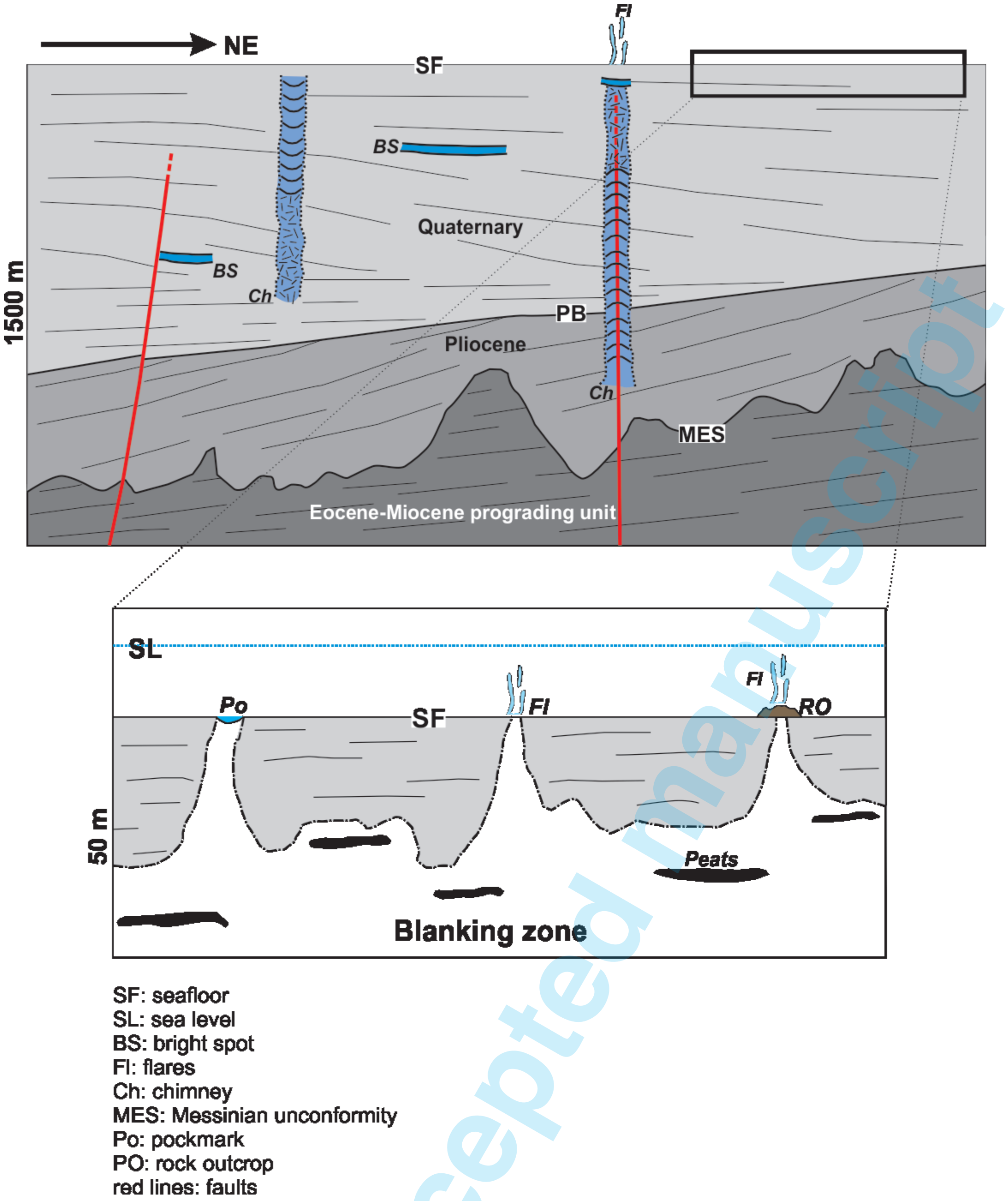


Fig. 11 - The plumbing system model for the northern Adriatic Sea. In the upper panel, two reflector configurations, i.e. pull up and pull down, are identifiable within the chimneys. The up-tilted reflections could be due to sediment deformation induced by upward migration of gas-enriched fluids (e.g. Donda *et alii*, 2015) or to localized higher seismic velocities, possibly indicating carbonate cementation within otherwise poorly cemented sediments (e.g. Cowley & O'Brien, 2000). Pull down reflector configurations would testify to a local decrease of the seismic velocity due to gas occurrence. In the lower panel, the blanking zone reflects the occurrence of gas-charged sediments.

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