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Continental Shelf Research 22(2002) 2429–2442

CONTINENTAL SHELF RESEARCH

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Contribution to atmospheric methane by natural seepages on the Bulgarian continental shelf

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Received 1 January 1999; received in revised form 1 October 1999; accepted 1 January 2000

Abstract

A regional estimation of the contribution to atmospheric methane by natural gas seepages on the UK continental shelf was undertaken by (Mar. Geol. 137(1/2) (1997) 165). This paper is the second in the series, and provides an estimation of the atmospheric methane flux from Bulgarian Black Sea continental shelf.

Potential gas source rocks include Holocene gas-charged sediments, Quaternary peats and sapropels, and deep-lying Palaeocene and Neogene clays, Cretaceous coals, and other sediments of late Jurassic to early Cretaceous age. These cover almost the whole continental shelf and slope and, together with irregularly developed seal rocks and widespread active and conducting faults, provide good conditions for upward gas migration.

A total of 5100 line kilometers of shallow seismic (boomer) and echo-sounder records acquired during the Institute of Oceanology's regional surveys, and several detailed side-scan sonar lines, have been reviewed for water column targets. Four hundred and eighty-two targets were assigned as gas seepage plumes. It is estimated that a total of 19,735 individual seeps exists on the open shelf. The number of seeps in coastal waters was estimated to be 6020; this is based on available public-domain data, specific research, and results of a specially made questionnaire which was distributed to a range of "seamen".

More than 150 measurements of the seabed flux rates were made in the "Golden sands" and "Zelenka" seepage areas between 1976 and 1991. Indirect estimations of flux rates from video and photo materials, and a review of published data have also been undertaken. Based on these data, three types of seepages were identified as the most representative of Bulgarian coastal waters. These have flux rates of 0.4, 1.8, and 3.5 l/min.

The contribution to atmospheric methane is calculated by multiplying the flux rates with the number of seepages, and entering corrections for methane concentration and the survival of gas bubbles as they ascend through seawater of the corresponding water depth. The estimation indicates that between 45,100,000 (0.03 Tg) and 210,650,000 m^3 (0.15 Tg) methane yr^{-1} come from an area of 12,100 km^2.

Keywords: Atmospheric methane; Gas seepage; Shallow gas; Bulgarian shelf; Black Sea

1. Introduction

It is only recently that natural seabed gas seepages have been recognised as a significant source of atmospheric methane (Hovland et al.,

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PII: S0278-4343(02)00055-9

1993; Judd et al., 1993; Cranston, 1994). The few existing estimations of global methane flux from this "minor" source vary greatly: from 0.5–1.9 $Tg\,yr^{-1}$ (Cranston, 1994; Trotsyuk and Avilov, 1986) to 65–146 $Tg\,yr^{-1}$ (Hovland et al., 1993). This is because they all are based on local observations, i.e. the initial data for global extrapolations of the distribution, frequency and flux rates of seeps are different, and they are diverse in character. For this reason it would be preferable if more precise local and regional estimations were made first, and then these estimations were generalized to produce a realistic value of the global contribution to atmospheric methane by natural seabed gas seepages.

The first "brick in the wall" is already in place—the regional atmospheric methane flux from UK continental shelf (Judd et al., 1997). A good working methodology was developed for this estimation. It included the following steps: identification of likely sources of methane; review of evidence of gas seepage; determination of likely seabed flux rates and the survival of methane bubbles as they ascend through seawater. The first step addressed the question: is there enough gas within the sediments which has had time to migrate to the seafloor? In the second step available shallow seismic (pinger, boomer), echo-sounder and side-scan records were reviewed to estimate the total number of the gas seeps, based on their distribution and frequency. Knowledge of seabed flux rates and the percentage of the initial mass reaching the sea surface was established in the third and fourth steps, respectively. Then it was a matter of simple calculation to estimate the annual gas flux to the atmosphere.

The above methodology was used in the study described here to estimate the methane flux from gas seepages on the Bulgarian continental shelf, and also for a comparison of the results. Its general continuity was kept the same, as were some of the theoretical considerations concerning the causes of water column targets (WCT) and the method of estimating the survival of methane bubbles. However, a different approach was applied for determination of the distribution of gas seepages in shallow coastal waters, and significantly changed limits of seabed flux rates were used.

For the purposes of this study the Bulgarian Black Sea shelf was divided into a coastal zone (water depths from 0 to 20 m, an area of 702 km^2) and the open shelf. The shelf, which has a total area of 11,383 km^2, was subdivided into 30 cells, each bounded by 1/4° of latitude and longitude.

2. Sources of methane

The western coast of the Black Sea cuts off heterogeneous structural elements including: the Eastern part of the Moesian Platform (the Varna monocline), the Kemchian foredeep (the Lower Kemchian depression), the Eastern prolongation of the Alpine folded region (the Balkanides), and the Burgas synclinorium (which is represented by the Burgas Palaeocene–Neogene depression), as shown in Fig. 1. These all contain source rocks with a significant oil and gas potential. They include a wide range of sediments lying from the first few meters below the seabed to depths of a few kilometers, and ages from Holocene to Cretaceous.

Holocene gas-charged sediments (described as "rockfor" by Khrischev, 1981) are developed south of Cape Kaliakra (Fig. 1) in water depths of 25–30 to 90–100 m. They cover all central parts of the shelf at depths of 0.5–1.0 to 3.5–5.0 m below the seabed, and have a thickness of 2–3 to 10–12 m. Their extent exactly coincides with the area of unusually high sedimentation associated with the South Black Sea current. This current transports and deposits huge amounts of fine material, mainly from the River Danube, which is highly enriched in fresh organic matter. The quantity of gas provided ongoing methane production within these sediments is estimated to be more than $10 \times 10^9\,m^3$ (Chochov, 1987).

Quaternary peats and organic rich sediments are associated with buried deltaic complexes and lagoons developed during several low stands of the sea level on the periphery of the shelf. These have local importance, but may contain significant amounts of gas when separate bodies are superimposed; this occurs in the southern peripheral

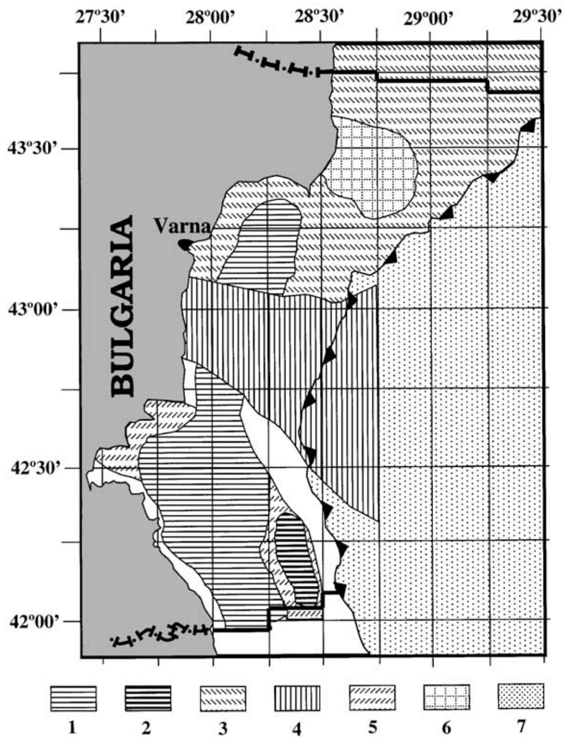

Fig. 1. Distribution of potential methane source sediments: (1) Holocene gas-charged sediments—"rokfor" type; (2) Quaternary peats and organic-rich sediments; (3) Paleozoic source sequence in the Moesian platform; Paleocene and Neogene clays in the Kamchian Foredeep (4) and Burgas Trough (5); (6) Cretaceous coals; and (7) source sediments on the continental slope and apron.

depression (Fig. 1) and the delta of the palaeo Sredetska River (Krustev et al., 1988).

The Moesian platform has been a stable province since at least the Early Mesozoic. Its sediment cover mainly comprises 6–7 km thick Phanerozoic carbonate rocks. The sediment strata lie in monoclinic structures inclined toward the south-east, providing good conditions for upward hydrocarbon migration even from the apron of the continental slope. The highly organically enriched Devonian and Lower Carboniferous sediments which are situated within the lower thermogenic gas window, Upper Triassic and Paleocene clays which lie in the main oil and gas window, together with Cretaceous coals, provide from 1 to 3 million tonne of hydrocarbons/km^2 (Geodekyan et al., 1984; Monakhov et al., 1990).

The process of oil and gas genesis in the deep-lying strata of the Lower Kamchian depression took place with increasing intensity, starting in the Late Cretaceous. Considerable potential for hydrocarbon generation has been achieved within the Eocene and Oligocene sediments; these have a thickness of up to 3–4 km. Between 8 and 15 million tonne of hydrocarbons/km^2 have formed along the axial area of the depression (Geodekyan et al., 1984).

The main source rocks in the Burgas depression are Palaeocene and Neogene sediments. It is estimated that between 0.1 and 0.3 million tonne of hydrocarbons/km^2 have formed in the deepest part of the depression where their thickness reaches 3.5–4 km (Vasilev, 1998).

The continental slope and apron is of secondary importance for hydrocarbon potential after the Kamchian foredeep, but it is much larger (Fig. 1). It provides optimum condition for hydrocarbon generation and migration upward to the shelf areas with its thick (up to 10 km) Cenozoic sediment sequence and monoclinic structures rising toward the continental shelf. The main source rocks here are Upper Eocene and Oligocene sediments with a total production of 12–15 million tonne of hydrocarbons/km^2 (Trotsyuk et al., 1990; Vasilev, 1998).

As can be seen from the above description, source rocks cover almost the whole continental shelf, slope and apron, and part of the adjacent land (Fig. 1). At the same time seal rocks are irregularly developed and there are widespread active and conductive faults which provide good conditions for upward gas migration. Thus, for example, shallow gas (as indicated by acoustic turbidity, bright spots, enhanced reflectors and pockmarks) is widespread on the whole Bulgarian continental shelf and uppermost slope (Dimitrov, 1989, 1990a, b; Dimitrov and Doncheva, 1994). Moreover, Trotsyuk and Avilov (1986) estimated that from 2.5 to 20 million m^3 methane yr^{-1} diffused through the seabed to the water column from this part of the Black Sea. In addition, several shallow (350–500 m below sea level) small gas fields and one oil and gas field are known both inland and offshore (Chembersky, 1995); these serve as a source for many permanent seepages (Dimitrov et al., 1979).

3. Distribution and frequency of gas seeps

3.1. The distribution of gas seeps on the open shelf

It is now widely known that gas bubbles, fish and other "targets" in the water column are detectable by a range of acoustic systems, from high frequency echo-sounders and side-scan sonars to lower frequency pingers and even boomers. Judd et al. (1997) discussed in detail, both theoretically and practically, the problem of distinguishing water column target produced by ascending gas bubbles from those with other possible causes. The targets from shoals of fish may have the most similar acoustic image to gas plumes, and confusion between these two is the most common misinterpretation. The shoaling habits of Black Sea fishes were studied in order to reduce this error to a minimum. When interpreting the data every doubtful target was rejected even though this may have resulted in a very conservative estimation of the number of gas seeps on the open shelf.

Fig. 2 shows selected acoustic images of gas seepages on different sections (side-scan sonar, echo-sounder and boomer) compared to that of shoals of the most common Black Sea fish, a small (about 8–10 cm long) sprat, *Sprattus phalericus*. However, it can be seen that the acoustic images of natural seeps (Fig. 2b–e) are more similar in shape to the 'artificial seep' produced by a SCUBA diver's gas bottle (Fig. 2a) than those of the shoals of fish (Fig. 2f–h). This fish likes cool waters and migrates daily, going deeper at midday and near the surface during the night (Zusser, 1971). There are many factors influencing the shape and size of the shoals but some tendencies can be identified; the sizes of shoals vary in width from 50–60 to 600–700 m, and up to 1500 m; and in depth from a few meters to 20–25 m. Shoals always tend to spread horizontally and to have a higher density at the seafloor (Fig. 2f). They are less common in mid-water (Fig. 2g), and disintegrate to many smaller shoals at the surface (Fig. 2h). However,

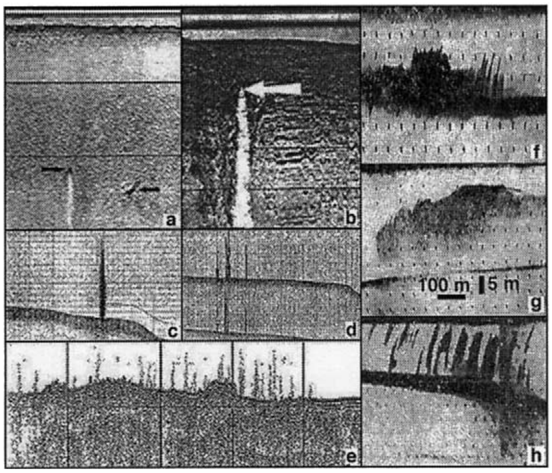

Fig. 2. Examples of some prominent water column targets. Hundred kilo-Hertz side-scan sonar sections showing: (a) a scuba diver laying on flat muddy seafloor (right arrow) and air bottles with 1/4 opened vent imitating a strong gas seep (flux rate more than 10 l/min; (b) a natural seep; (c and d) 32 kHz echo-sounder sections at the shelf edge; (e) 1.2 kHz boomer section in the zone of seepage abundance in the "Golden Sands" seepage area; shoaling fish; (f) at the seafloor; (g) in the middle of the water column; and (h) at the sea surface.

despite significant differences between fish shoals and gas plumes, discrimination between them is somewhat subjective and should be made by an experienced interpreters.

A total of 5100 line kilometers of shallow seismic (boomer) and echo-sounder records acquired during the Institute of Oceanology's regional surveys and several detailed side-scan sonar lines have been reviewed. Water column targets were identified in 26 cells and 482 targets were interpreted as gas seepage plumes.

To determine the total number of gas seepages in each cell two different approaches were used: the first is based on simple extrapolation of the established number of seeps in the footprint of the survey lines (a seabed area of $0.05\,km^2$ was insonified for every survey line kilometer) to the total area of the cell. The second method is based on a calculation of the density of seeps (the number per line kilometer of reviewed profiles), the distribution of these densities was then subdivided into classes with the following assumed densities: 0.19, 0.46, 1.06, 2.55, and $4.15\,km^2$

(although there were indications of up to 35 seeps/ km^2).

The total number of gas seeps for each cell was then estimated assuming that this seepage density applied to the whole area of the cell. For the cells where there are no survey lines or no seeps were identified, the lowest class was applied. The difference between the estimates obtained by these two methods is insignificant.

The total number of gas seeps on the open Bulgarian Black Sea shelf is estimated to be at least 19,735 individuals.

3.2. The distribution of gas seepages in the coastal zone

The distribution and frequency of seeps in the coastal waters has been determined based on a review of public-domain data, specific research and a specially made questionnaire that was distributed to fishermen, scuba divers, "rapana" hunters, etc.

Four seepage areas are reported in the literature: two small areas, "Lozenez" and "Michurin", along the southern coast (Mandev et al., 1978) and larger ones at "Golden Sands" and "Zelenka". These last two areas were reported for the first time in 1951 (Palii et al., 1951), but were known to local fishermen for a long time before. They have been studied in more detail by the specialists of the Institute of Oceanology since 1976 (Dimitrov et al., 1979).

3.3. "Golden Sands" and "Zelenka" case studies

The "Golden Sands" gas seepages area is a long (about 1200–1300 m), narrow (about 250–300 m at its widest) zone parallel to the coast line. It is situated about 1 km offshore of resort of Golden Sands. The water depth where the gas seeps occur varies from 7 to 12 m, and single seeps are found in depths of up to 22 m. A smaller (approximately 250×50 m) gas seepage area occurs north-east of the main area.

The "Zelenka" gas seepage area is situated about 2.5 km to north-west from Cape Kaliakra and 200–250 m from the shore line. It is oval in plane view, with dimensions of $400 \times 250\,m^2$, and occurs in water depths of 5–9 m, and up to 15 m. A chain of single seeps is present to the east of the main area.

The seabed is covered by fine grained sands (about 2–2.5 m thick) which grades into silty sands and clayey sands in water depths greater than 12–15 m. These sediments are underlain by single blocks, boulders and rock banks of Sarmatian limestone and marlstone (Dimitrov et al., 1979) which are exposed on the seafloor in many places within the area. These rocks are covered by a dense layer of black shells of the bivalve *Mytilus Galloprovincialis* (Fig. 4). Most of the gas seeps are concentrated in and around these rock exposures. A grey–white mantle which look like "the foam of boiled milk" is often seen within a few first decimeters of the gas seeps. This seems to be a bacterial mat.

Most probably the seeping gas has gas migrated along tectonic faults from shallow (550–700 mbsl) gas fields (Fig. 3). This contention is supported by the almost complete similarity of the composition of the gas from the gas fields and from the seepages (Table 4), and by the longevity and stable flux rates of the seeps.

Individual bubble streams are scattered irregularly within the seepage areas and active seepage zones are surrounded by zones of less, or no, seep activity. Activity varies between weak seeps with very small gas bubbles (the smallest gas bubbles observed both immediately at the gas vent and at the sea surface have a radius of about 2–3 mm), strong continuous chains of bubbles, and seeps which make the surface of even a calm sea "boil". The average density of the gas seepages in active seeping zones is about 3–5 seeps/m^2 but may reach up to 24 seeps/m^2. The exact total number of the seeps has not been established, but it is estimated to be at least 1200 individuals in the "Golden Sands" area, about 200 seeps in the northern area, and 800 seeps in the "Zelenka" area.

3.4. The number of seepages in the rest of the coastal waters

The main purpose of the questionnaire was to obtain new data about gas seeps along the Bulgarian coast. It was hoped that the study

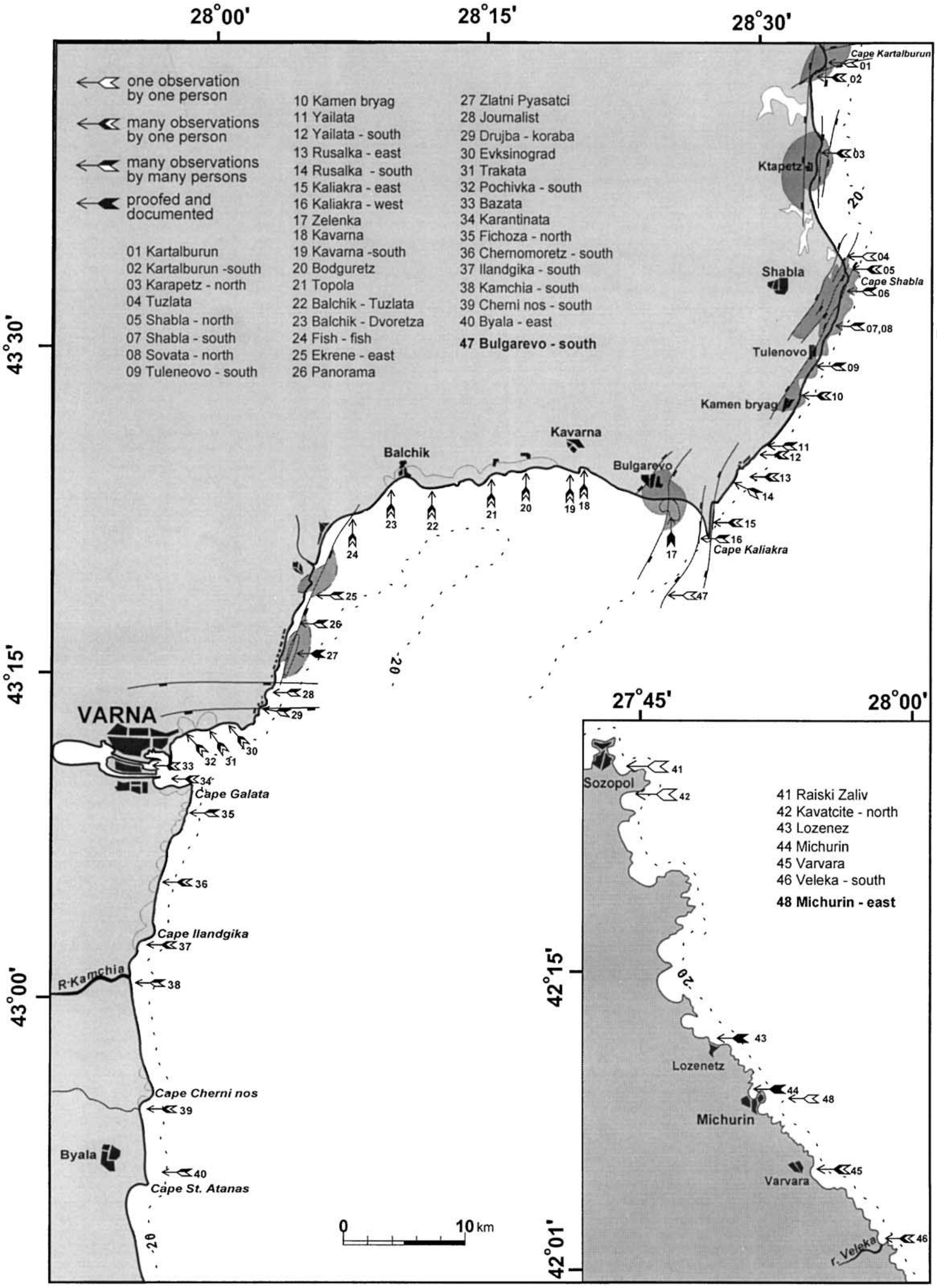

Fig. 3. Location map of known and possible gas seepage areas in the Bulgarian coastal waters showing shallow gas fields and some of the major active coastal landslides.

would identify previously unreported seepage areas, and provide information such as: the number of seeps observed; the number per square meter; the area covered, etc. This information helped with the estimation of the total number of seeps in each seepage area.

A total of 47 possible new gas seepage areas have been identified (Fig. 3). The new areas were grouped into the following categories: one observation by one person; many observations by one person; and many observations by many persons. The first category was rejected as unreliable, but 41 areas was included in the estimation of the total number of seepages (although only four of them have been checked and confirmed).

All seepage areas are situated in water depths between 2–3 m and 15–18 m, with a maximum of 22 m. Most of the seepage areas are situated above known small shallow gas fields or along faults (especially those with an expression on the seafloor, i.e. faults active in the recent near past or at present). Others are situated in front of active coastal landslides (most probably marking the outcrop of the shear surface on the seafloor), in estuaries and in semi-enclosed bays where there are limited or no water circulation. It should be noted that seeps are restricted to specific sites along preferred lineations—faults, fissures, the outcrops of porous rocks, etc. For this reason the combined total area of the individual seepage areas is about 6 km^2, i.e. together the seepage areas occupy less than 1% of the coastal waters. Moreover, the northern half of the coastal zone, which was the most exploited by those who responded to the questionnaire, has a seepage density of about 14 seeps/km^2—ten times greater than the southern coastal zone (Fig. 3).

The number of seepages within the separate areas varies from 15–20 to 500, and the total number of individual gas seeps in the Bulgarian coastal waters is estimated to be at least 6020.

4. Determination of the seepage flux rates

The correct determination of the flux rates is critical if errors of under- or over-estimation are to be avoided. A range of measurements have been carried out in order to produce a realistic figure for the flux rate of a single gas seepage.

More than 150 independent direct flux rate measurements have been made in the zones where seeps are most abundant in the “Golden Sands” and “Zelenka” seepage areas. The quantity of released gas was measured by trapping the gas with an inverted funnel placed over the seeps and timing the collection of the gas in inverted water-filled graduated tubes. The measurements were made at the seabed by scuba divers in 1976, 1982, 1987, 1989, 1990 and 1991 (Table 1).

Individual flux rates varied from 0.28–0.65 to 3.5–5.0 l/min; only two measurements, made in the “Golden Sands” area, showed rates greater than 5.0 l/min. The smallest rate measured was 0.12 l/min, and the strongest seeps vent 6.2 and 9 l/min. In two places the flux was measured using a special funnel with an area of 1 m^2; the flux rates were:

Table 1
Seep flux rate measurements, Bulgarian coastal waters

Flux rates (l/min)	Number of measurements in						Total number	Cumulative (%)
	1976	1982	1987	1989	1990	1991		
<0.36	5	18	15	9	11	7	64	99.9
<0.90	4	11	8	5	4	5	37	59.0
<1.80	7	5	6	3	5	3	29	35.3
<2.70	4	3	5	—	2	—	14	16.7
<5.00	3	3	2	—	2	—	10	7.7
>5.00	—	—	2	—	—	—	2	1.3

- 9 l/min (two "average" seeps with rates of approximately 2.5 l/min and one "strong" seep of about 4 l/min) and
- 10.8 l/min (24 "small" seeps with average rate of 0.45 l/min).

Weighted average flux rates for the selected flux rate classes were found to be:

- 0.4 l/min for the range up to 0.9 l/min;
- 1.68 l/min for the range 0.9–2.7 l/min; and
- 10.8 l/min for the class with an upper limit of 32 l/min.

Indirect estimates of flux rates were also made by including specific questions in the questionnaire: bubble size at the observation site (seafloor, seawater or sea surface); number of bubbles per a time period or the distance between successive bubbles; the type of the seepage (permanent or intermittent), etc. These parameters were also estimated from available video and photo materials.

Bubble radii of about 5 mm have been considered as the most common, and, by assuming a bubble rise rate of 25 cm/s, the flux rates were calculated for a range of possible number of bubbles per second. The data suggest that there are three types of seepages which are the most representative of Bulgarian coastal waters (Fig. 4). These have flux rates as follows:

- slow flux: 0.26–0.62 l/min;
- medium flux: 1.0–2.0 l/min; and
- fast flux: 2.5–3.8 l/min.

The data from Table 1 were plotted on a cumulative probability plot (dashed line in Fig. 5). This indicates a mean flux rate of 1.8 l/min at the 43% probability level. The lower bound (taken as lower quartile) is 0.4 l/min at the 5% probability level, and the upper limit (taken as mean plus standard deviation) is 3.5 l/min at the 89% probability level. To derive a generally applicable estimate of the flux rates, published data have been reviewed (Table 2). These have

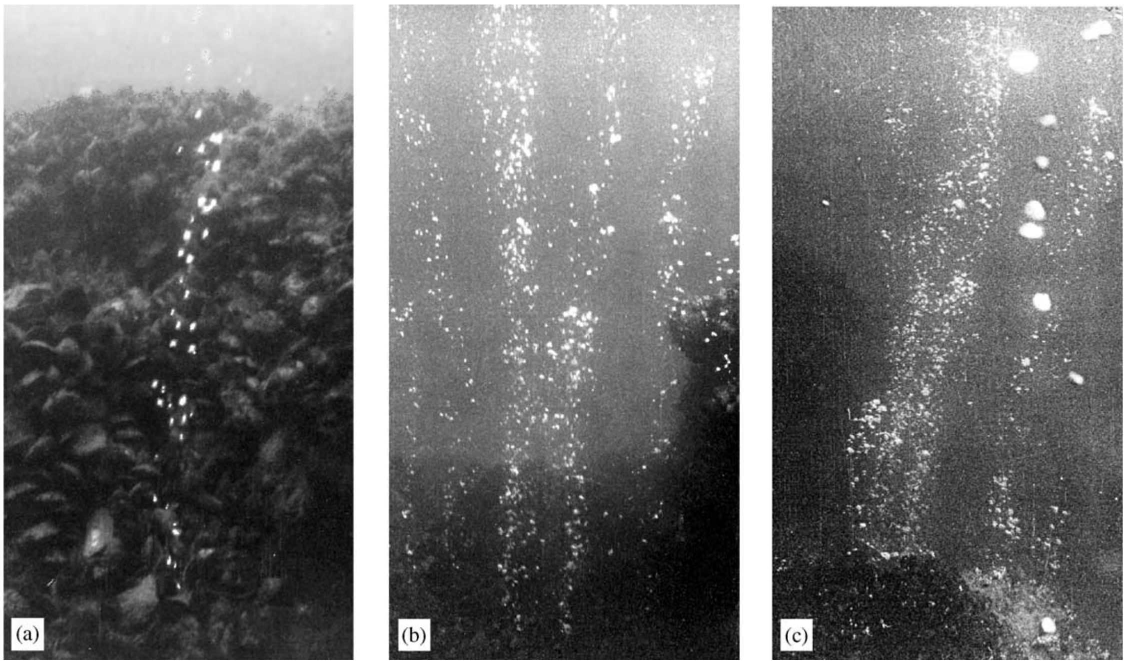

Fig. 4. Photographs of gas seepages in the "Golden Sands" area showing: left, a small seep producing up to 20 bubbles/s (bps) with a flux rate of 0.26–0.62 l/min; centre, two medium strength seeps, 40–60 bps, 1.0–2.0 l/min; and right, a strong seep with more than 80 bps, 2.5–3.8 l/min.

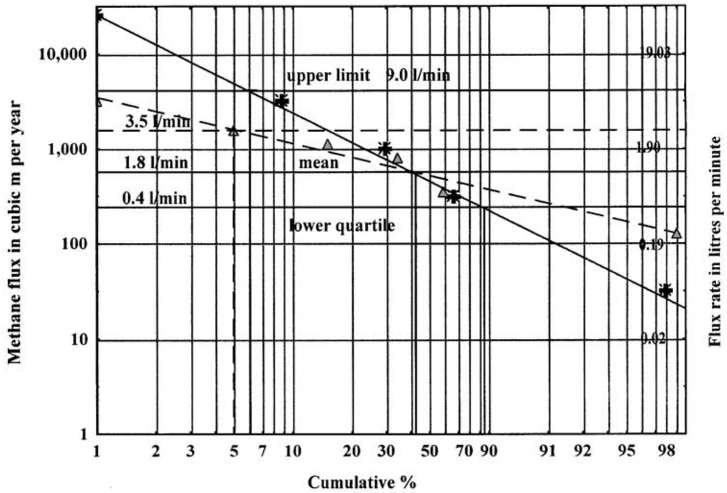

Fig. 5. Single seepage flux rates at the seabed. Cumulative log probability plots from case studies in Bulgarian coastal waters (dashed line) and known seepages worldwide (see Table 3). The values used for calculations are shown.

been plotted on the same graph (Fig. 5, Table 3) as the data from the case studies presented above. The difference between the two plotted lines reflects the local characteristics of the source and suggests that every single seepage area may have its own probability distribution.

It should be noted that the highest measured flux rate reported in 50 l/min (26,280 $m^3\,yr^{-1}$) (Bernard, 1980; quoted in Hovland et al., 1993). *The discovery of the strong seepages is of the first order* [I don't understand what you mean by this!—I mean that the strongest seeps are discovered firstly and then those with less flux rates] and if there are no reports of other such seeps it may concluded that there is a very small number world wide, or even that those in the Gulf of Mexico are the only seepages of such strength. Furthermore, it seems improbable that natural seeps venting several liters of gas per second would have a long life span. If such seeps do exist they should be classified as blow outs.

Generalizing the data, the following seepage flux rate classes were assigned for coastal waters:

- slow flux: 0.4 l/min (210 $m^3\,yr^{-1}$);
- medium flux: 1.8 l/min (945 $m^3\,yr^{-1}$); and
- fast flux: 3.5 l/min (1840 $m^3\,yr^{-1}$).

Based on physical considerations the medium class was assigned for all seep on open shelf. A flux rate of 9.0 l/min (4700 $m^3\,yr^{-1}$, taken at 6% level on the general probability plot) is assumed to be the upper bound.

It has been assumed that all seeps are permanent with relatively constant flux rates, i.e. the contribution to atmospheric methane by natural gas seepages is a long-term, continuous process. This assumption is supported by the visual observations of Palii (one of the discoverers of the "Golden Sands" seepage area) and Trayanov (a scuba diver who participated in the first investigations of the area in 1975–1976 and most of the subsequent flux rate measurements) made both at the seabed and the sea surface in 1990–1991. It seems that the intensity of gas release was as intensive as 40 yr previously. As the sources of these seeps have not been affected by anthropogenic activities, and no geological events have occurred over this time scale, there is no reason to suppose that flux rates have changed, or will change, in the foreseeable future.

5. Gas composition

The composition of the gas from several parts of the "Golden Sands" and "Zelenka" seepage areas,

Table 2
Flux rate at the seabed of a single gas seepage[a]

Area and description	Data source	1/min	$m^3\,yr^{-1}$	Observations
Toney River site, New Foundland, Canada—bubble rise through the 1–3 m water column… have an average diameter of 1 cm and are released at a rate $1\,s^{-1}$	Cranston (1994)	0.04	21	> 1000
Torry Bay, Firth of Forth, North Sea—an average flux of $1270\,m^3\,yr^{-1}$ is estimated for the whole area (50–200 individual bubble streams)	Judd et al. (1999)	0.05	26	50
Stockholm Archipelago—about 60 sites of gas seepages… the rates was estimated at 3–4 l/h	Soderberg and Floden (1992)	0.06	32	60
UK Block 15/25, North Sea—submersible measurements of seepage rates on individual gas vents range from 0.14 to 0.6 l/h at ambient conditions, equivalent to a mean flow rate of 5.74 l/h at STP	Clayton and Dando (1994)	0.10	52	50
Michurin Bay, Black Sea—central group of about 30 seepages… vents from about 0.1–0.2 $m^3\,day^{-1}$	Mandev et al., (1978)	0.11	58	30
Kattegat, North Sea—there are an average of 65 active gas seeps, … over 25% of the gas flux was observed to come from two seeps, one with a rate of approximately 20 l/h and 10 l/h the other… the remaining seeps had measured flows in the range 0.15–5 l/h… mean value of 1.22 l/h	Dando et al. (1994)	0.02	10	63
		−0.17	−87	1
		−0.33	−175	1
UK Block 15/25—total volume of gas escaping as bubbles from these three seeps… was estimated as 200 l/h	Hovland and Judd (1988)	1.11	583	3
North Sea Pockmark—10–15 small vents, each issuing a gentle stream of single bubbles at flow rates of between 0.14 and 0.6 l/h (at 160 m water depth)	Dando and Hovland (1992)	0.04	21	10
		−1.17	−615	5
Tommeliten field, North Sea—the total gas production from main seepage area (120 individuals within an area of 6500 m^2) approximated to 24 $m^3\,day^{-1}$ (at ambient pressure, 75 m water depth)	Hovland and Judd (1988)	1.18	620	120
Milos island, Mediterranean Sea—fifty-six flow rates from 39 individual seeps were measured and these ranged from 0.2 to 18.5 l/h at the depth of collection	Dando et al. (1995)	0.22	115	20
		−1.80	−946	10
		−3.70	−1.945	9
Golden Sands and Zelenka seepage areas, Bulgarian Black Sea Coastal waters—see description in the text	This study	0.40	236	1200
		−1.80	−1840	600
		−3.50	−1.02	300
Sea of Okhotsk—for a water depth of 700 m and temperatures of 4°C, … a single bubble vent releases on the order of $10^7\,g\,C\,a^{-1}$ as methane	Cranston et al. (1994)	0.35	184	10
Gulf of Mexico	Bernard (1980)	50.0	26,280	1
			Total	3543

[a] The flux rates are corrected for water depth.

and from the "Lozenetz" and "Michurin" areas has been studied. The analyses (Table 4) show that it is dry methane gas, with a methane concentration of between 92% and 97% and very small concentrations or no higher hydrocarbons. The gasses are most probably of biogenic origin (Mandev et al., 1978). The similarity of the gases from these seepage areas with the gases from shallow gas fields along the coast, as well as the geographical relationship, suggests that many of the seeps arise from these gas fields.

For the purpose of calculation it is assumed that the average methane concentration of all the seep gases is 95%.

6. The contribution to atmospheric methane

With an estimate of the total number of seeps and the flux rate per seep it is a matter of simple multiplication to estimate the gas flux at the seabed from each seepage area in the coastal zone and for each cell of the open shelf. The calculation indicates that at least 4,046,000 m^3 gas yr^{-1} seeps through the seabed from the coastal zone and approximately 147,000,000–737,000,000 m^3 seeps from the seabed of the open shelf.

The quantity escaping to the atmosphere has been estimated from the percentage of the initial mass of gas reaching the sea surface (Fig. 6—assuming a 10 mm initial bubble diameter) for the average water depth determined for each cell and seepage area. Then, by entering corrections for methane concentration 95%), an estimate of the sea surface methane flux was calculated. This suggests that approximately 3,700,000 m^3 methane yr^{-1} (0.003 Tg yr^{-1}) enters the atmosphere from the coastal zone, and approximately 41,390,000–210,650,000 m^3 methane yr^{-1} (0.03–0.15 Tg yr^{-1}) comes from the continental shelf area (see Table 5).

Table 3
Previously published seep flux rates

Rate ($m^3 yr^{-1}$)	Total number	Cumulative (%)
<50	1183	99.98
<500	1311	66.60
<1000	738	29.61
<5000	309	8.78
<10,000	2	0.06

7. A speculative extrapolation

A speculative extrapolation can be made for whole Black Sea continental shelf taking into account the similarity in geology and its proven oil and gas potential.

For example, the majority of the north-western shelf is underlain by the Skytian platform whose geology is very similar to that of the Moesian

Table 4
Composition of natural gases along the Bulgarian Black Sea coast

	Location	Age of source	CH_4 (%)	C_{2+} (%)	CO_2 (%)	N_2 (%)	H_2S (%)
Shallow gas fields (see Fig. 2 for location)	Blatniza	Oligocene	93–96.5	—	0–1.5	2.0–7.1	Traces
	Krapez	Oligocene	94–98.5	—	0–1.0	1.5–5.2	Traces
	Tulenovo	Oligocene	90–95.5	0.1	0–0.2	5.5–9.0	—
	Bulgarevo	Oligocene	96–98.2	0.3	0–0.4	1.3–3.5	—
	Kranevo	Oligocene	95–98.3	0.2	0–1.6	3.4–6.8	—
	Kamchia	Up. Eocene	92–98.8	0.4	0–2.3	0.6–7.1	Traces
	Babaeski	Oligocene	95–96.8	0.1	0–0.3	0.5–3.1	—
Seepage areas	Zelenka	?	93–95.2	0.1	0–0.4	3.8–5.3	—
	Golden Sands	?	92–96.3	0.2	0–0.9	0.8–4.4	—
	Lozenetz	?	93–94.5	0.2	0–0.5	0.0–2.5	Smell
	Michurin	?	94–96.8	0.2	0–6.2	0.0–0.5	Smell

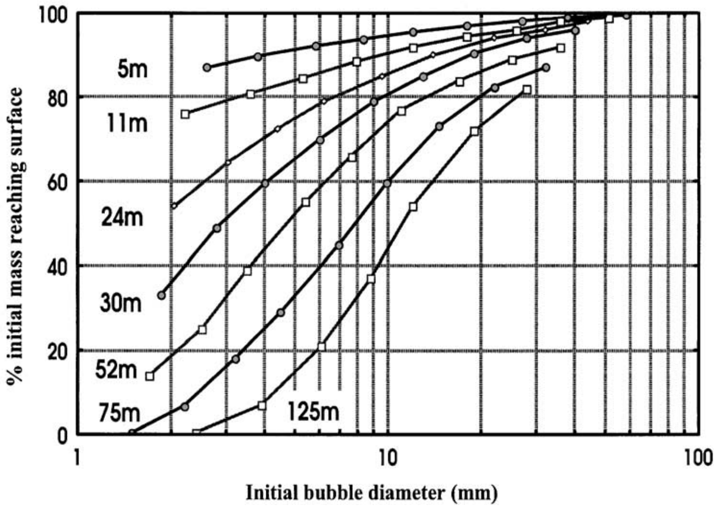

Fig. 6. Percentage of initial mass of gas bubbles reaching the sea surface at corresponding water depth (after Judd et al., 1997).

Table 5
Atmospheric methane from natural gas seepage in the Black Sea

		BG coastal zone	BG shelf	Whole BGCS	Rest B.Sea coastal zone	Rest B.Sea shelf	Whole B.SeaCS
Area (km^2)		702	11,400	12,100	10,100	109,500	131,700
Number of seeps		6020	19,735	25,755	86,860	189,560	302,175
Atmospheric methane	$m^3\,yr^{-1}$	3,700,347	41,390,472	45,090,819	53,390,726	397,566,375	496,047,921
	$Tg\,yr^{-1}$	0.003	0.030	0.033	0.038	0.285	0.356
	$m^3\,yr^{-1}$	3,700,347	206,952,360	210,652,707	53,390,726	1,987,831,877	2,251,875,311
	$Tg\,yr^{-1}$	0.003	0.148	0.151	0.038	1.425	1.614

platform where most of the Bulgarian seeps occur. Also several oil and gas fields are being exploited in Romanian and Ukrainian waters, and several areas of active gas venting are known (Egorov et al., 1989; Gevorkyan et al., 1991). Moreover, the north-eastern (Kerch–Tamin) and eastern shelves may be the most prolific gas venting areas in the Black Sea (even though they are very narrow). There are more than 50 mud volcanoes in the Kerch–Tamin shelf emitting huge amount of gas (Glebov and Shelting, 1998). Many shallow gas features, pockmarks, bright spots, BSRs and gas plumes, and also several mud volcanoes, have been found in the Caucasus area (Korsakov et al., 1989; Glebov and Shelting, 1998). It is estimated that between 0.5 and 15 billion m^3 methane yr^{-1} are vented from the Georgian shelf (Tkelashvili et al., 1998). Several oil and gas seeps (Iztan, 1996) and mud volcanoes (Glebov and Shelting, 1998) are known along the Turkish coast line, etc.

This brief review suggests that a simple extrapolation from the Bulgarian continental shelf to the whole Black Sea continental shelf is justified (Table 5). Therefore, it is suggested that from 0.36 to 1.6 Tg CH_4 enters the atmosphere every year from gas seeps on the Black Sea continental shelf—an area not exceeding 0.5% of the world's continental shelves. This does not even consider onshore gas seeps.

Acknowledgements

Dr. K. Prodanov is acknowledged for helpful discussions about the behaviour of shoaling fish in the Black Sea. T. Trayanov provided the results of flux rate measurements and Dr. A. Judd edited the manuscript.

Part of this work was financially supported by the Bulgarian Ministry of Education, Science and Technology, Project III-2568/95.

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