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Spatial distribution and geochemistry of the nearshore gas seepages and their implications to natural gas migration in the Yinggehai Basin, offshore South China Sea

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ABSTRACT

About 120 gas seepage vents were documented along the west and southwest coast of the Hainan Island, South China Sea, in water depths usually less than 50 m. The principal seepage areas include the Lingtou Promontory, the Yinggehai Rivulet Mouth, Yazhou Bay, the Nanshan Promontory and the Tianya Promontory. They occur along three major zones, reflecting the control by faults and lateral conduits within the basement. It is estimated that the total gas emission from these seepage vents is $294-956 \text{ m}^3/\text{year}$. The seepage gases are characterized by a high CH₄ content (76%), heavy δ^{13} C₁ values (-38 to -33‰) and high C_1/C_{1-5} ratios (0.95–1.0), resembling the thermogenic gases from the diapiric gas fields of the Yinggehai Basin. Hydrocarbon-source correlation shows that the hydrocarbons in the sediments from seepage areas can be correlated with the deeply buried Miocene source rocks and sandstone reservoirs in the central depression. The 2D basin modeling results based on a section from the source rock center to the gas seepage sites indicate that the gas-bearing fluids migrated from the source rocks upward through faults or weak zones encompassed by shale diapirism or in up-dip direction along the sandstone-rich strata of Huangliu Formation to arrive to seabed and form the nearshore gas seepages. It is suggested that the seepage gases are sourced from the Miocene source rocks in the central depression of the Yinggehai Basin. This migration model implies that the eastern slope zone between the gas source area of the central depression and the seepage zone is also favorable place for gas accumulation.

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1. Introduction

Natural hydrocarbon seepages from sea bottom have been reported in quite a few offshore petroleum-bearing basins, such as North Sea of Netherlands (Hovland and Judd, 1988; Schroot and Schuttenhelm, 2003), Gulf of Mexico (Behrens, 1988), offshore California (Scott et al., 1999), Great Britain (Selley, 1992), the Torry Bay of Scotland (Judd et al., 2002), and southeast Asia (Macgregor, 1993). Because hydrocarbon seepages are proxies to migration pathways and/or deeper hydrocarbon reservoirs (Abrams, 2005), they are very important clues for petroleum exploration. The hydrocarbon seepages along the eastern margin of the Yinggehai Basin, South China Sea, have been known and recorded for more than 100 years (Zhang, 1965). However, little information has been reported on their geochemistry and origin. With increasing petroleum exploration activities in the Yinggehai Basin, it has been realized that the gas seepages may provide new information to trace the hydrocarbon sources and thus reduce the exploration risk in the basin. In this paper, we present the spatial distribution, gas flux rates and geochemical characteristics of the hydrocarbon seepages, and discuss their origins and migration models in the basin.

2. Geological setting

The Yinggehai Basin is one of the most gas-rich Cenozoic rift basins in China (Gong, 1997; Huang et al., 2003). It trends northwest–southeast, and is separated from the Hainan Island to the east by the No.1 fault (Fig. 1). The tectonic evolution of the basin can be divided into two stages: a Paleogene extensional rifting event and a Neogene post-rift thermal subsidence (Gong, 1997). The basin was filled with thick deposits of Cenozoic clastics on Paleozoic and Mesozoic basement rocks (Fig. 2). It was estimated that the maximum thickness of the Cenozoic deposits reaches 16 km at the basin center (Huang and Xiao, 2004).





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Fig. 1. (a) General map showing the location of the Yinggehai Basin and (b) the distribution of the Yinggehai nearshore gas seeps.

The basin is characterized by a rapid subsidence rate, a high geothermal gradient and overpressure (Zhang et al., 1996; Huang et al., 2003; Huang and Xiao, 2004). It is evaluated that the maximum sedimentation rate was up to 1.2 mm/year. The geothermal gradient was as high as 4.25–4.56 °C/100 m during the Tertiary period (Huang et al., 2002, 2003). The maximum pressure coefficient reaches 2.0–2.3 (Zhang et al., 1996; Huang et al., 2003) with the major part of the basin being an overpressure system. The combination of overpressure and high palaeo-geothermal gradient had an important influence on the generation, migration and accumulation of natural gases in the basin. Available geological and geochemical data indicate that the main hydrocarbon–source rocks in the basin reside in the Meishan and Sanya Formations (Huang et al., 2003). These source rocks contain gas prone higher plant-derived organic matter (Huang et al., 2003).

Another important geological character of the basin is the widely developed diapiric structures in the basin center area. The diapiric faults act as preferential pathways for upward migration of natural gases from the deep sources into the Pliocene sandstone bodies and Quaternary strata, forming the major



Fig. 2. Schematic stratigraphic column of the Yinggehai Basin.

exploration targets in this basin. Several gas fields related to the diapiric structures have been discovered in the Yinngehai Basin in recent years (Dong and Huang, 1999; Huang et al., 2002, 2003).

The discovered natural gas pools principally occur in the Pliocene–Ouaternary marine sandstones from the central diapir zone. with a burial depth of 390-2000 m. Most of them are of diapiric origin. The gases are believed to be derived from a set of Type II₂-III sources rocks in the Meishan and Sanya Formations occurring in the central depression of the Yinggehai Basin (Huang et al., 2003; Huang and Xiao, 2004). The gas seepages occur in the west and southwest coastal area of the Hainan Island, and cover the southeast part of the hanging wall of the No.1 fault in the Yinggehai Basin (Fig. 1), with water depths usually ranging from 10 to 50 m. Most of the seafloor topography appears to be relict and related to the erosion of older sedimentary strata. A thin layer of surfacial sediments consisting of silts, clays, and sands is deposited in this area. The basement rocks comprise inter-bedded deltaic sandstones, siltstones and shales of Pliocene and late Miocene age. Some of the gases derived from the deep buried source rock in the central basin migrate to the seabed through tortuous pathways and exit from the seafloor surfacial sediments, leading to the formation of the nearshore gas seepages in the Yinggehai Basin, which will be discussed in following sections.



Fig. 3. Sonar records of 3.5 kHz through the Yinggehai Rivulet Mouth seep site. Seep bubbles in water column are shown as maroon curtains or bubbling plumes.

3. Methods and experiments

The data contained in this paper are mainly from a joint CNOON–BP research project and a CNOOC survey project on the seepage investigation in the Yinggehai Sea, carried out in 1991 and 2001, respectively (Huang and Zhang, 1992; Xie et al., 2001).

The surveys were navigated by Global Positioning System (GPS). The cruising speeds for the surveys were about 3–5.5 knots. The gas seepage investigation was made using two side-scan sonars equipped on the surveying ships with frequencies of 3.5 kHz and 12 kHz, respectively. The analog signals were recorded by a paper chart recorder. The acoustic intensity returned from gas bubbles in water is proportional to their cross-sectional area.

Most of the seepage gas samples were collected from the water about 5–25 m below sea level using a funnel. Some gases in the Yazhou Bay were sampled directly from the seafloor using a remotely operated vehicle (ROV). Seafloor sediment samples were collected by a few methods, for example, the shallow core samples were collected using a lead bomb tube with a maximum sampling depth of 30 cm, and mudsnapper samples were also collected at or near the surface of the seafloor.

The sediments were extracted in dichloromethane and were analyzed by GC and GC/MS. The seepage gases were analyzed for their composition using a Hewlett Packard 5890 II gas chromatograph, equipped with a thermal conductivity detector. Methane and ethane gaseous hydrocarbons and CO₂ were separated for δ^{13} C measurements following procedures similar to those described by Schoell (1980) using a Finnigan-MAT251 mass spectrometer. The δ^{13} C values are reported in reference to the PDB standard, with an analytical precision of $\pm 0.02\%$. Stable deuterium–hydrogen isotope ratios were determined for some of the samples. The δD_{C1} values are reported in reference to SMOW, with an analytical precision of $\pm 3\%$.

4. Results and discussion

4.1. Gas seepage distribution

The two investigations have found that there are more than 120 gas seepage vents along the west and southwest coast of the Hainan Island, in water depths usually less than 50 m (Fig. 1). They are distributed mainly in the Lingtou Promontory, the Yinggehai

Rivulet Mouth, the Yazhou Bay, the Nanshan Promontory and the Tianya Promontory to form three major seepage zones (Fig. 4). Gas bubbles cover these areas to form nearshore bubble zones. For instance, within an area measuring about 100 m by 50 m in the Yinggehai Rivulet Mouth, there are numerous (>40) individual vents, with each producing visible bubbles (1–2 cm in diameter for a single bubble) continuously, displaying spectacular seeps (Fig. 3). The gases escape from the seafloor sediments through fractures in



Fig. 4. Sparker profiler records showing pockmarks at the Yinggehai gas seepage site. The locations of cross sections are shown in Fig. 1.

Table 1

Gas flow rate measurements from the Yinggehai nearshore seep vents.

Location	No. of measurements	Flux for one vent (ml/h)			
Yinngehai Promontory	3	420-910			
Yazhou Bay	4	310-760			
Nanshan Promontory	5	350-800			
Lingtou Promontory	2	280-430			

the Pliocene Yinggehai Formation strata or the seafloor outcrop of the pre-Tertiary basement rocks.

Since the leaking hydrocarbons have affected shallow sediments (Chen et al., 2005) and seafloor (Hovland and Judd, 1988; Taylor, 1992; Schroot and Schuttenhelm, 2003), the seabed morphology features also provide evidence of near-surface fluid expulsions. The sparker seismic profiles (Fig. 4) show seabed pockmarks in the Yinggehai seep site. These pockmarks have sizes of 0.5–2 m wide and 0.5–1 m high, and are believed to be related to gas emissions. A similar scenario was reported in the Norwegian Trench where pockmarks occur in weak zones in the soft sediments within the seepage areas (Hovland and Judd, 1988).

Based on the gas seepage occurrences, it is believed that the gases are originated from deeper burial rocks, and not from seafloor soft sediments.

4.2. Gas flux rates

Gas seepage flux rates were estimated using the displacement of water by an inverted funnel or ROV. The methods have been shown to be capable of accurate measuring flux over short time periods (Scott et al., 1999; Judd et al., 2002). Twelve samples were collected at the gas seepage areas of the Yinggehai Basin using an inverted 25 cm diameter plastic funnel, and transferred to the measuring cylinder through a short plastic tube. The gas was stored in a 200-cm³ glass cylinder. Two samples were taken by ROV at the Yazhou Bay and the gas bubbles were collected in a 200-cm³ glass cylinder which was sealed in water with a silicone septa and aluminum cap.

The results are summarized in Table 1. The flux rates vary widely in the different gas seepage areas. The highest flux rate of individual seep vent occurs at the Yazhou Bay, and the lowest at Lingtou Promontory, with flux rates of 280 ml/h and 910 ml/h, respectively. Assuming that the flux rate measurements undertaken are representative, a crude approximation of the total gas flux from the Yinggehai Basin can be estimated. By applying the total 120 seepage vents, the total flux rates range from 294 to 956 m³/year.

It should be pointed out that the flux rate is possibly underrated, because some continuous seepage may be occurring in water depths up to 50 m, but no measurements have been made. The flux rate and volume of hydrocarbon seepage to the surface provide direct and critical information to the petroleum system analyst regarding source and gas potentials (Hunt, 1996; Abrams, 2005). The Yinggehai Basin is typical of a basin containing rich gas resources which will be further elaborated in the following sections.

4.3. Geochemistry and origin of gas seepages

The geochemistry of samples from the gas seeps shows that the composition is dominated by methane, with minor N₂, CO₂ and O₂ (Table 2). The gas seepage samples collected by ROV contain high methane, nitrogen and carbon dioxide. The difference between samples collected from the seafloor by ROV and from the water surface by a funnel (SWF) suggests that the SWF samples were contaminated by a small amount of air during sampling. The seepage gases have a high dry index (0.95–1.00) with relatively heavy $\delta^{13}C_1$ values (–33.91 to –38.24‰) and light deuterium–hydrogen isotope ratios (–131 to –162‰), indicating a thermogenic origin without biogenic gas presence (Fig. 5a).

In order to trace the seepage gas sources, a comparison was made between the seepage gases and the subsurface trapped gases in the central diapiric zone of the Yinggehai Basin with regard to their compositions and compound-specific isotopic ratios. Huang et al. (2003) reported the geochemical characteristics of the reservoired gases from the Yinggehai Basin. The thermogenically

Table 2

Composition and stable isotopes of gases sampled at the Yinggehai nearshore gas seepage sites.

Sample no.	Site ref.	Composition (%)							C_1/C_{1-5}	$\delta^{13}C_1$			δD_{C1}
		C ₁	C ₂	C ₃	C ₄	CO ₂	N ₂	02		C ₁	C ₂	CO ₂	
YS1	Lingtou Promontory	46.04	0.21	0.03	0.02	0.89	43.28	9.50	0.99	-35.51			-143.6
YS2	Yinggehai Rivulet Mouth	39.21	0.60	0.14	0.03	0.09	50.50	9.87	0.98	-36.06	-22.99	-17.20	
YS3		42.30	0.74	0.16	0.04		45.73	12.01	0.98				
YS4		36.33	0.65	0.13	0.03		50.31	12.51	0.98	-35.90			-131.0
YS5	Yazhou bay	65.22					27.43	7.29	1.00	-33.91			-149.6
YS6*		76.24				11.02	12.17	0.57	1.00	-34.38			-162.2
YS7		59.55					32.17	8.28	1.00				
YS8		70.01					23.06	5.93	1.00				
YS9	Nanshan Promontory	66.37					27.27	6.36	1.00	-38.24			-158.7
YS10		59.34				0.24	31.23	9.19	1.00				
YS11		63.48					28.14	8.38	1.00				
YS12		62.91				0.32	29.11	7.66	1.00				
YS13		53.84				0.49	35.31	10.36	1.00				
After deductin	g air in the samples:												
YS1	Lingtou Promontory	81.33	0.37	0.05	0.04	1.57	16.64		0.99				
YS2	Yinggehai Rivulet Mouth	70.79	1.08	0.25	0.05	0.16	27.66		0.98				
YS3		91.63	1.60	0.35	0.09	0	6.34		0.98				
YS4		84.76	1.52	0.30	0.07	0	13.35		0.98				
YS5	Yazhou Bay	97.83	0	0	0	0	2.17		1.00				
YS6*		78.28	0	0	0	11.31	10.41		1.00				
YS7		95.72	0	0	0	0	4.28		1.00				
YS8		97.32	0	0	0	0	2.68		1.00				
YS9	Nanshan Promontory	93.51	0	0	0	0	6.49		1.00				
YS10	-	99.60	0	0	0	0.40	0.00		1.00				
YS11		100	0	0	0	0.00	0.00		1.00				
YS12		96.73	0	0	0	0.49	2.78		1.00				
YS13		99.10	0	0	0	0.90	0		1.00				



Fig. 5. (a) The Yinggehai seep gas genetic classification diagram by $\delta^{13}C_1$ versus δD_{C1} (Scheoll diagram, Scheoll, 1980; Whiticar et al., 1986). (b) Cross plot of $\delta^{13}C_1$ values versus C_1/C_{1-5} for the seep gas and the reservoired gas samples collected from the Yinggehai Basin, showing that both seep gas and reservoired gas are thermogenic in origin, and may have same sources. The boundary lines were taken from Tissot and Welte (1984).

derived gases have dry indexes ranging from 0.93 to 0.96, and $\delta^{13}C_1$ values from -47 to -30%, and a maturity corresponding to a source maturity of 1.0–2.2% Ro. As shown in Fig. 5b, the geochemical data from the seepages fall within the reservoired gas boundaries, indicating that they may have derived from the same source.

A source rock rich in humic organic matter may have generated mainly gases with a small amount of oil (Hunt, 1996) and required much greater temperature to expel the remaining hydrocarbons as gas. Dark mudstones with a significant thickness in the Meishan and Sanya Formations occur mainly in the central area of the Yinggehai Basin. They have a TOC of 0.4–2.97%, with a typical Type II₂–III kerogens (Huang et al., 2003), and are believed to be the main source rocks for the reservoired gases in the Yinggehai Basin (Huang et al., 2002, 2003). The burial depth in Meishan and Sanya Formation source rocks varies greatly, from 4000 to 7500 m in the basin, which led to the great discrepancy in the maturity levels of the source rocks from VRo of 1.0 to 2.5% (Huang et al., 2003). Therefore, it is reasonable to consider the deeply buried source

rocks in the Meishan and Sanya Formations in the central area of the Yinggehai Basin as the seepage gas source.

The total soluble extract yields for all of the surfacial sediment samples from the Yinggehai seep sites were low. Only two extracts (91-SR6 and 91-SR7) yielded sufficient quantity to perform conventional GC and GC/MS analyses. Compared with the extracts from the reservoir sandstone samples from DF1-1 gas field, the seepage sample extracts seem to contain less lower-molecular weight *n*alkanes due to slight biodegradation, but their GC spectra are still quite similar (Fig. 6). The Pr/Ph (pristane/phytane) ratios are moderate (1.5-1.8), together with an observed C₂₉aaa sterane predominance (Fig. 7), which may imply a terrestrial source input in a marine depositional environment. The hopane and sterane molecular maturity parameters are fully isomerized, indicating that they are derived from matured source rocks. Based on the hopane and sterane bimarkers, the characteristics of extracts from the surfacial sediments in the seepage areas are similar to those of the source rocks from the Meishan and Sanya Formations and oils from the gas-reservoir in the basin. This suggests that the extracted hydrocarbons from the surfacial sediments in the Yinggehai seepage sites are mainly originated from mature source rocks in the Yinggehai Basin.

4.4. Migration of gas seepages

Based on the above geochemical data and discussion, the seepage gases were likely sourced from the matured source rocks in



Fig. 6. Gas chromatogram of saturated hydrocarbons for two sand extracts from the Yinggehai seepage site and a gas-reservoir sandstone extract from DF1-1 gas field, showing the light biodegradation of the seepage hydrocarbons and their correlation with the reservoired hydrocarbon.



Fig. 7. Representative *m*/*z* 191 and 217 mass fragmentograms showing the correlation among migrated hydrocarbons from the sand of the Yinggehai seepage site, the oil–sandstone in DF gas field and potential source rocks in the central basin.

the Meishan and Sanya Formations in the Yinggehai Basin. However, the Yinggehai nearshore gas seepages are primarily in the areas with no gas fields or discoveries, and there are no Meishan and Sanya Formations or any other possible source rocks around the seepage areas. Therefore, the widely developed matured source rocks in the Meishan and Sanya Formations in the central area of the Yinggehai Basin is the most likely source for the seepage gases. For a further explanation, the migration pathway analysis should be made using both geological–seismic data and fluid flow modeling (Selley, 1992). This is also critical in understanding the surface seepage in terms of petroleum system dynamics (Abrams, 2005).

As pointed out in the above section, the Yinggehai seepages are not randomly distributed, but confined in three zones (Yinggehai Rivulet Mouth, Yazhou Bay, Nanshan Promontory, see Fig. 1b), implying the possible control by faults and lateral conduits in the basement. In fact, all the nearshore gas seepages investigated are directly or indirectly related to the shale diapirism, or to the lateral gas migration induced by pressure difference. Seismic records show that widely developed high angled faults related to the diapirs extend from a few meters to 2 km, and cut through seal rocks, even reaching to seafloor in the Yinggehai Basin (Fig. 8, Xie et al., 2001; Huang et al., 2005), leading to deeply sourced gases to be expelled from overpressured compartments into the overlying strata to form gas-reservoirs or migrate laterally into higher permeability units. The DST data from drilled wells in the Yinggehai Basin show that the burial depth of the overpressured zone is from 1500 m to 2480 m in the diapiric structure zone of the basin center, and becomes deeper toward the east slope (to about 4300 m). The overpressure is believed to be the main driving force for the formation of the diapir structures and the migration of overpressured fluids (Huang et al., 2002).

Under above geological and dynamic framework, a 30 km long section from the source rock center to the gas seepage sites is selected to model the fluid flow using the IES' *PetroFlow* software. In this model, the sedimentary sequences and lithologies were based on seismic interpretations and well data, the paleogeothermal gradients and heat flow data were taken from Gong (1997), relative permeability curves were determined experimentally on reservoir rocks from the basin, and the modeled maturities and overpressure were calibrated using measured vitrinite reflectances and DST



Fig. 8. A seismic profile across L8-1diapir and the eastern part of the Yinggehai Basin, showing the diapir piercing zone and associated faults. The location of seismic section is shown in Fig. 1.

pressure data, respectively. It was assumed that source rocks in the Meishan and Sanya Formations distributed in the central area of the Yinggehai Basin, with an average TOC of 1.05% and Type II₂–III kerogens. The modeling result (Fig. 9) provides an overall indication of the major gas flow directions in the section. The fluids migrate predominantly along faults, fractures and stratigraphic surfaces. The mud diapiric fault system would act as an effective pathway for fluids releasing from the overpressured source rocks of the Meishan and Sanya Formations in the basin center. The fluids migrate upward and were trapped over the diapir structures or migrate laterally through the east slope along the pathways below the sealing surface in the sandstone-rich strata of the Huangliu Formation. The gases may seep in the outcrops of the Neogene strata or accumulate to form gas pools in suitable structures along the pathway.

Fig. 10 shows the lateral–vertical–lateral migration model of gases along a west to east cross-section across the eastern part of the Yinggehai Basin. The migration directions are based on the integration of the modeling results with the regional stratigraphic data. Gases generated from the overpressured source rocks in the basin center may migrate laterally through the framework sandbodies of the Huangliu Formation to the major basin boundary (No.1 fault) by the drive of abnormal differential pressure. The gases would then migrate upward into the overlying sand-rich strata mainly along the No.1 fault, and then migrate laterally into higher permeability up-dipping units. Consequently, the gases migrate laterally to the margin of the basin through coarse-grained sediments, which unconformably overlie the impermeable basement, and eventually escape from the seafloor basement outcrop unconformity and/or the seafloor sediments through fractures.

In this model, the No.1 fault acts as an important pathway for fluid migrating upward. This fault extends 250 km to the northwest and appears to have connected to the Red River fault system (Fig. 1). The fault can be divided into three sections: the northwest section, the middle section and the southeast section based on its activity which increases gradually from northwest to southeast. New data collected in recent years suggest that the middle and the southeast sections of the fault were active from pre-Tertiary to Pliocene or Quaternary, with relatively small fault displacement from approximately 10.5 to 2.7 Ma B.P, during the deposition of the deltaic sand and mudstone sequence. The results of lithology contact analysis of the fault's two sides indicate that sand/sand juxtapositions in the upper Miocene or Pliocene have occurred in different parts of the southeast and middle sections (Fig. 9), which would clearly favor gas migration cross the fault and along the permeable up-dip layers of the hanging wall (Knipe, 1997). This may account for the



Fig. 9. 2D gas migration modeling results at present day along a transect across the eastern part of the Yinggehai Basin, showing possible sources and migration directions of seeping petroleum.



Fig. 10. Migration pathway and formation model of the Yinggehai seeps along the west-southwest coast of Hainan Island. The location of section is shown in Fig. 1 (C–C').

concentrated distribution of gas seeps in the shallow water area along the southeast coast of the Hainan Island.

4.5. Implications for favorable sites of gas accumulation

The constructed gas migration model for the Yinggehai nearshore seeps has a significant implication for hydrocarbon exploration in the region. Hunt (1996) reported that the largest oil-producing area in the world was discovered directly by visible oil-seepages. In south-east Asia, seep-rich basins are usually rich in subsurface reserves, and eight of nine basins with more than 35 documented seepages contain at least one billion barrels oil equivalent (BBOE) proven reserves (Macgregor, 1993). The Yinggehai Sea seepage gases originating mainly from deeply buried source rock (Miocene) must have migrated through complex pathways to the seafloor surface along the eastern margin of the Yinggehai Basin. Consequently, in addition to the diapiric structures in the central depression of the Yinggehai Basin, the pathways of migrating gases on the east slope zone should be an ideal location for searching for important gas accumulation in the Yinggehai Basin.

5. Conclusions

The Yinggehai gas seeps cover a relatively large nearshore area of the Yinggehai Basin, extending about 100 km along the west to southeast coasts of the Hainan Island, in water depths usually less than 50 m and have been documented for nearly 100 years. The major sources for the seepage gases are the Miocene source rocks in the central depression of the Yinggehai Basin. Our investigation indicates that these gas seeps, which have mainly originated from deeply buried source rocks (Miocene) in the central depression, migrate laterally and vertically to the hanging wall of the No.1 fault through the Huangliu Formation framework sand-bodies on the east slope, and escape from the seafloor sediments through fractures or subcrops of the basement. This suggests that the Yinggehai Basin contains huge gas potential and future exploration should be focused on the east slope zone between the active gas kitchen and the Yinggehai seepages.

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