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Gasgeochemical features in the western part of the East Vietnam Sea (Bien Dong Sea)

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ABSTRACT

During the joint Russian-Vietnamese studies in the East Vietnam Sea (known as Bien Dong or the South China Sea), manifestations of new hydrocarbon-accumulating zones were discovered; a series of evidence of the presence of mineral indicators for solid minerals in shelf deposits were obtained; a unique zone, where accumulations of ferromanganese crusts and nodules have been formed, was revealed for the first time; evidence of unique properties of deep-water fine-dispersed carbonate sediments (which makes it possible to attribute them to an independent type of mineral resources of the East Vietnam Sea) were obtained; the *ikaite* mineral (that serves as an indicator of methane migration zones and cold marine paleo-conditions) was discovered in the Phu Khanh basin; heavy concentrates with indicators of rare forms of ore mineralization were collected; features of methane and mercury fluxes into the atmosphere, cultures of methane-oxidizing, oil-oxidizing and sulfate-reducing bacteria were registered for the first time within the study areas. New data on gravity and magnetic field anomalies along the shelf and slope of Vietnam have been obtained. The article presents the results of coastal geophysical and gasgeochemical research obtained in 2010–2020 within the Joint Russian-Vietnamese Laboratory for Marine Geosciences (POI FEB RAS - IMGG VAST). For the first time, anomalous methane fields (comparable to anomalies on the oil- and gas-bearing shelf and the gas hydrate-bearing slope of Sakhalin island) were registered in the water column. The gravimagnetic survey was carried out on the Central and Southern shelf and the continental slope of Vietnam; the executed works supplemented the geophysical data, which had been obtained in the 80s–90s of the preceding century. The paper contributes to the Russia-Vietnamese program within the UN Decade of Ocean Sciences for Sustainable Development framework.

Keywords: Methane, hydrogen, helium, Bien Dong Sea, gasgeochemical fields, seismotectonic.

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INTRODUCTION

Gasgeochemical fields (along with the data from geophysical, stratigraphic, mineralogical, magmatic, lithochemical, and other studies) are the essential characteristics of the geostructures related to continent-ocean transitional zones. The study's relevance to gasgeochemical fields of hydrocarbon gases, helium, hydrogen, carbon dioxide, nitrogen, oxygen, radon, and others is associated with their possible use as geological, oceanological, and geoecological indicators and processes. The submarine gas manifestations influence the upper part of the lithosphere, the hydrosphere, and the lower part of the atmosphere. At the same time, underwater degassing areas occupy, in aggregate, more than 10% of the World's Oceans, but at present, they are insufficiently studied [1]. Since the areas of dispersion of natural gases from their lithospheric sources can extend for hundreds of kilometers, penetrating the water column and the atmosphere [2], this phenomenon is becoming global. The gasgeochemical research is also significant in applying geoecological issues, for example, studies of alternative sources of hydrocarbons (gas hydrates, coal methane, mud volcanic and geothermal gas, etc.), natural greenhouse gas emissions, explosive and toxic gases (CH_4, CO_2, CO, H_2, etc.), and mapping of gas-saturated sediments which is vital for the design of engineering structures [3].

The study of natural gas fluxes contributes to solving the problem of the origin of hydrocarbon minerals, which becomes more and more closely related to the issues of oil and gas potential of crystalline rocks [4, 5] and deep fluid [6, 7]. The cost-effective gasgeochemical methods make investigating large territories and water areas possible. In active transitional continent-ocean zones, within relatively small areas, various hydrocarbons and lithosphere–degassing accumulations can be described: methane fluxes, mud volcanoes, gas hydrates, coal-and-gas deposits, oil-and-gas deposits, geothermal and hydrothermal systems. In this regard, the East Vietnam Sea (Bien Dong Sea), a typical representative of the near-continental marginal seas of East Asia, is currently an essential focus of geochemical research.

Issues of distribution, genesis, and other scientific aspects related to natural gas fluxes within the World Ocean and its continental framing, including low-temperature seeps, gas hydrotherms, gas fluxes from coal-bearing strata, mud volcanoes and, especially, gas hydrates (future energy sources) are reflected in hundreds of publications, reports, and patents [1, 8–19]. At the same time, the amount of knowledge decreases sharply with distance from the coast toward deep-water regions.

The distribution and variability of gasgeochemical fields of natural gases in marginal seas and surrounding geostructures of the continent-ocean transitional zone are the most complex and relevant.

The study of the gasgeochemical regime of continental margins is impossible without studying the relationship between marine and surrounding terrestrial geological structures in terms of stratigraphy, tectonics, deep structure, volcanism, and the forecast of mineral and hydrocarbon resources [20–26].

Studies of gasgeochemical fields should consider the migration of gases not only from oil-and-gas and coal-containing deposits but also from active local gas-discharge systems (mud volcanoes, geothermal systems, water mineral springs, etc.). These types of natural gas manifestations are widely developed in the East Asian Seas and adjacent areas but have not been studied enough.

Interest in gasgeochemical fields is caused by fundamental and energy problems, and gas fluxes from the continental and sea basin lithosphere are involved in forming climatic and environmental conditions [27–36]. Over the past decades, the concentration of CO_2 and CH_4 in the atmosphere has increased by 0.4 and 0.3–1.2% per year, respectively [37, 38], which also requires complete knowledge about the distribution and sources of anomalous gasgeochemical fields in the geological structures of continental margins.

There are necessary prerequisites and conditions for studying gasgeochemical fields in bottom sediments and surface gas manifestations within a single methodological complex. On the other hand, there is a lack of expeditionary and analytical data for the Bien Dong (Sea) to illuminate the relevant fundamental and applied issues of natural gas dispersion. In this regard, the article's purpose is to research the gasgeochemical features of the Bien Dong Sea. These modern studies are especially relevant and aim at the distribution patterns of natural gases in near-surface, near-bottom conditions and their relationship with geological structures, oil-and-gas deposits, gas hydrates, and seismotectonics in the continent-ocean transitional zone of East Asia.

The research on gasgeochemical fields of the World Ocean

Lithochemical and gasgeochemical studies made it possible to discover the universal phenomenon of geochemical zoning to establish the most critical role of landscape-geochemical and structural-tectonic conditions in the manifestations and structural features of primary and secondary dispersion aureoles of matter [39]. Using the example of the Western Pacific Seas, the role of a geodynamic factor in the mineral distribution and chemical composition of bottom sediments was studied [40]. Gasgeochemical fields of hydrocarbon accumulations are well studied on the continent; as a result, domestic scientists created fundamental works and gas-genetic classifications [8, 11, 41–44]. In recent years, research abroad also came to the forefront [5] (due to deep-water drilling programs).

One of the undoubted successes in the study of gasgeochemical fields in the near-bottom layer of marginal seas was the searching concept for oil and gas deposits by express methods of gasgeochemistry, developed in the works of A. I. Obzhirov et al., (1992; 1993; 2004) [11, 45, 46]. However, despite these and other works [8], gasgeochemical fields in sedimentary strata in the Sea of Okhotsk, Sea of Japan, Bien Dong Sea, and East Siberian Seas have been poorly studied (especially at the horizons below the bottom surface), the study of gasgeochemical fields is very significant from the point of view of fundamental and prospecting aspects.

An increase in concentrations of various gases in bottom sediments is usually caused by diffusion and filtration processes during gas migration [47–52]. These processes are associated with the active transport of matter, especially along boundaries of lithospheric plates, intraplate rift zones, and post-magma chambers, in junction zones of different types of earth crust (oceanic-suboceanic-continental), in areas of asthenosphere uplifts and other zones, including active neotectonic structures, as well as points and belts of increased seismicity.

The genetic relationship of underwater seeps of methane, its isomers, helium, hydrogen, and carbon dioxide, their distribution areas with oil and gas deposits, accumulations of gas hydrates, deep chambers, and fault zones have been proven for many Western Pacific Seas and adjacent insular and continental structures [53–58].

The geochemical volcanic processes within Kamchatka and the Japanese islands are being studied deeply [59–63]. Methods (which are very important for monitoring the activity of gas emissions and for integrated estimates of gas fluxes) for remote measurements of volcanic gas fluxes (Global Volcanism Program, DOAS, FLYSPEC, etc.) and methane fluxes are widely used [64].

The primary sources and manifestations of natural gases and hydrocarbon fluids are in the Pacific oil-and-gas-bearing region [8, 19, 52]. Russian and foreign researchers have discovered thousands of terrestrial and underwater centers of hydrocarbon discharge (mainly of methane-bearing fluids) along the continental margins (Fig. 1). More than 300 areas with seeps of hydrocarbon gases, oil, and groundwater have been studied [1, 8].

In the Eastern Pacific, gas manifestations on the Oregon continental slope are among the most studied [65]. There are other well-known areas; for example, in the Northern part of the Santa Barbara Channel (California, USA), more than 1,500 concentrated seeps release hydrocarbon gases and oil. About 4,000 pockmarked structures (pockmarks) within the bottom of the underwater Californian margin [66] are formed by local fluxes of thermogenic natural gas. More than 30 similar regions with local seeps of natural gases, including fluxes of abiogenic methane in the hydrothermal systems of the MAR (Mid-Atlantic Ridge) spreading zone, have been found in the Atlantic Ocean [67].

The most numerous seeps of hydrocarbon gases of thermogenic (oil-and-gas) origin in the Atlantic Ocean are detected in the Gulf of Mexico; seeps of natural gases are registered near the Azores, in the Gulf of Cadiz (the Northwestern slope of Spain) [68], and other areas. Underwater seeps of natural gases, not associated with hydrothermal activity, are common within continental margins on shelves and slopes [8, 69–71]. Areas with natural gas seeps occur within thick (over 2 km) sedimentary strata containing various accumulations of hydrocarbons: oil and gas deposits, gas hydrates, and methane-saturated sediments. A necessary condition for hydrocarbon degassing in such areas is, as a rule, discontinuities [58]; folded dislocations and increased seismicity are additional conditions.

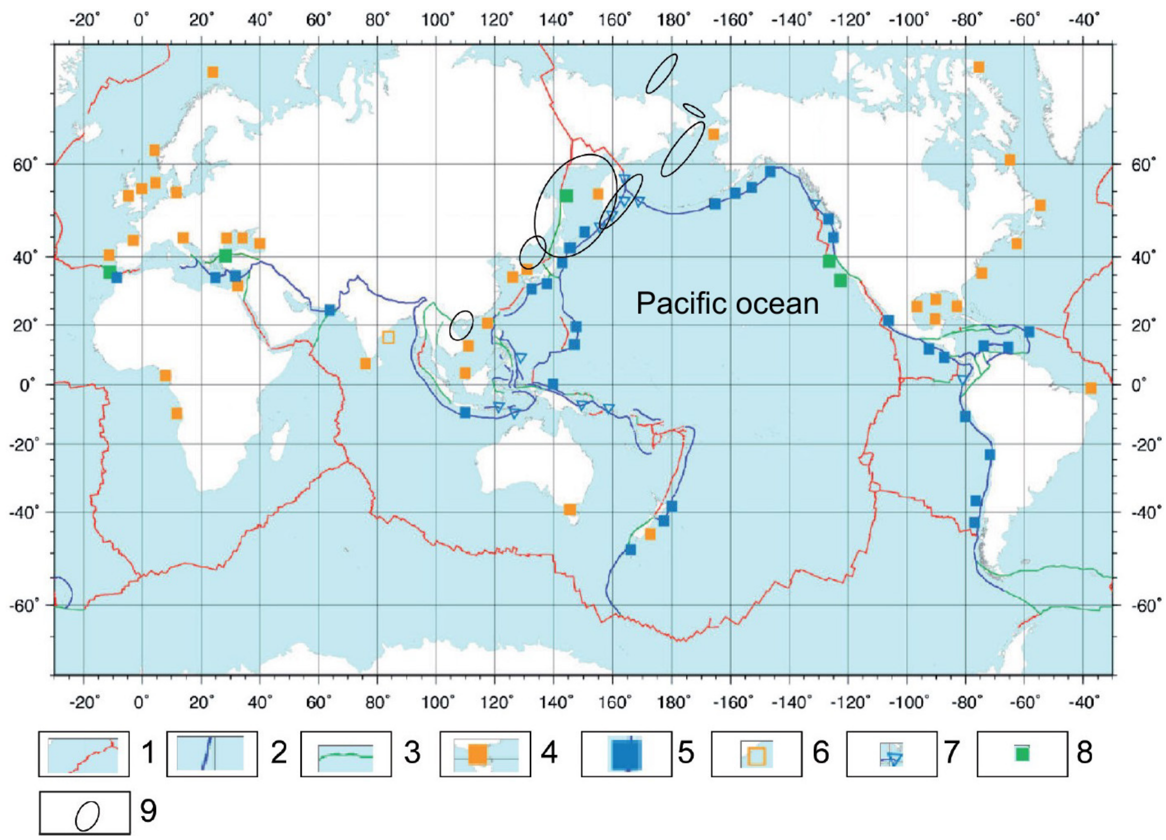

Figure 1. Global seep distribution [19]. 1- boundaries of lithospheric plates and planetary fault zones; 2- active continental margins; 3- transform plate boundaries; 4- groundwater and hydrocarbon fluid manifestations; 5- active seeps of natural gas; 6- sites of methane paleoseeps near the West of India; 7- gas manifestations, recorded by benthic fauna; 8- areas of the most intense methane fluxes, associated with gas hydrates; 9- study areas of the team of the Gasgeochemistry Laboratory, POI FEB RAS

The research on gasgeochemical fields in the Bien Dong

The Bien Dong (East Vietnam Sea, or the South China Sea) is characterized by significant oil and gas potential. Integrated geological and geophysical expeditions to the Bien Dong were organized by POI FEB RAS (on board the R/V Alexander Nesmeyanov (1983), Morskoy Geofisik (1989), Professor Bogorov (1995), etc.). These geological and geophysical expeditions laid the foundation for studying the geological structure of the Vietnamese part of the Bien Dong and contributed to understanding the bottom tectonic structure and the discovery of hydrocarbon deposits. The beginning of gasgeochemical research is closely related to the oil-prospecting works of the USSR in Vietnam. The Gasgeochemistry Laboratory, headed by A. I. Obzhirov, studied the distribution of natural gases (methane, heavy hydrocarbons, and other gases) in the near-bottom water layer on the shelf and slope to forecast hydrocarbon deposits. During the expeditions, anomalous fields of methane and HCGs were discovered, which, combined with geophysical studies, positively characterized the prospects for oil and gas in the East Vietnam Sea (later confirmed).

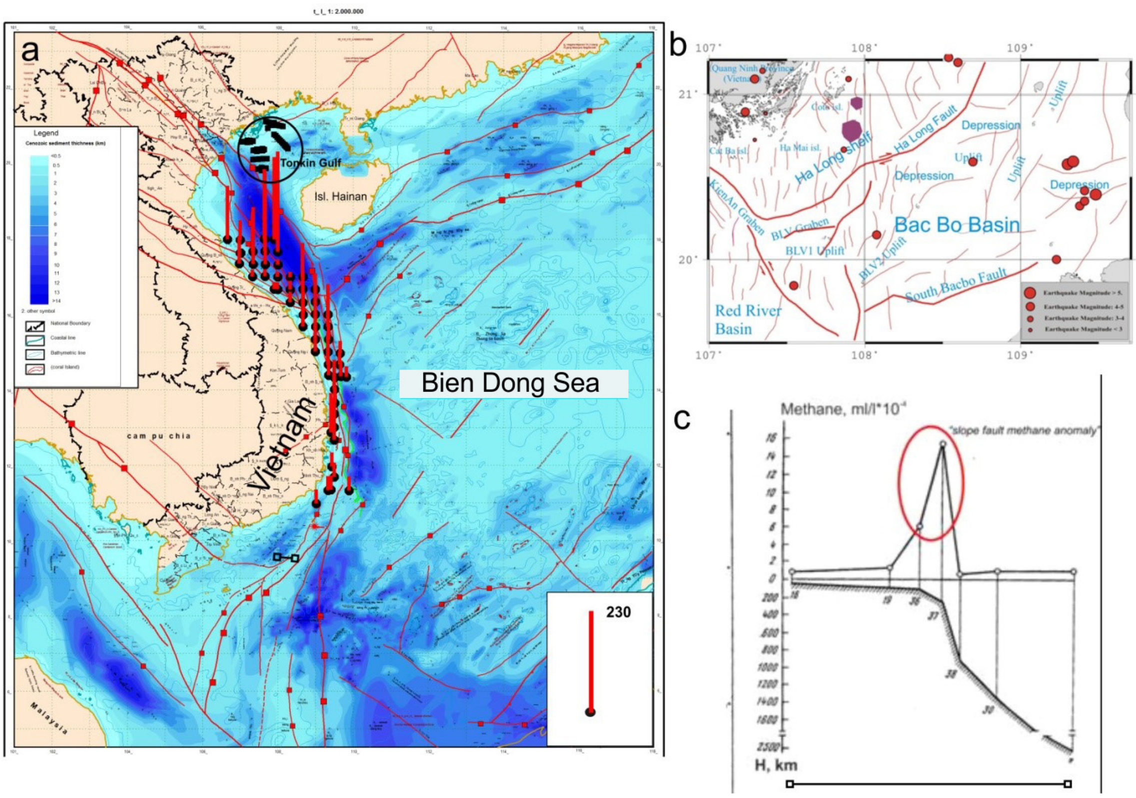

Figure 2. The scheme of gasgeochemical research in the Bien Dong. *Legend:* a- map of faults and sediment thickness (km). Red charts- methane concentrations in the near-bottom water layer (1994, R/V Professor Bogorov); b- tectonic map of the Gulf of Tonkin (Beybuwan basin), earthquakes pointed for the all observations historically; c- profile of methane anomaly above the slope break in the Nam Con Son basin (1989) [11]. Black circle (a) corresponds to the present research (Beybuwan basin) 2013–2014

Geophysical and drilling operations were carried out at promising structures with the participation of the Dalmorgeofizrazvedka trust (Yuzhno-Sakhalinsk). As a result, oil and gas fields were discovered on the shelf in the Southern part of the East Vietnam Sea (Cuu Long sedimentary basin and other structures). About 15–17 million tons of oil annually are currently produced there.

In the 1980s, the researchers of the Gasgeochemistry Laboratory detected a sharp variability of methane concentrations in the near-bottom waters in the western part of the Bien Dong Sea. Anomalous methane concentrations were registered in the Northern and Southern parts of the Vietnamese shelf. The maximum methane concentration of 1,540 nL/L was recorded in the South of the shelf at station M33-37 (9°7’1”N and 108°8’5”E) at a depth of 230 m (Fig. 2). Around this station, at depths of 110–280 m, an anomalous methane field with concentrations of 500–1,000 nL/L, which exceeds its background contents by 10–50 times, was recorded in the near-bottom waters. Small concentrations of hydrocarbon gases (ethane and propane) - 0.5–1.0 nL/L - were observed in this anomalous methane field.

In the last decade, the Bien Dong and the territory of Vietnam have been actively studied by gasgeochemical methods [72–79]. Many works were devoted to methane distribution in seawater [74, 77], methane emissions on the water-atmosphere interface [75, 76], and methane distribution in sediments in the Northern and Southwestern parts of the Bien Dong [78]. As a result, mainly due to the efforts of the Gasgeochemistry Laboratory of POI FEB RAS and the Joint Russian-Vietnamese Laboratory for Marine Geosciences (POI FEB RAS - IMGG VAST), gasgeochemical research has been developed in Vietnam along with other geological and geophysical studies. However, a detailed analysis of the distribution of hydrocarbons and other gases in the Bien Dong Sea sediments is still ahead. This review aims to describe the main features of the distribution of HGCs and other gases and to discuss the possibility of issues of origin and indicatory aspects for the Vietnamese shelf and slope and some geostructures of terrestrial framing.

Currently, much attention is paid to studying the Western Pacific areas, which had been previously considered unpromising in terms of oil and gas potential. Also, modern research is aimed at studying geological structures and deep faults, which can be channels for the migration of hydrocarbon gases. (Fig. 3). Methane, hydrogen, and helium dissolved in water might be indicators of fault zones for forecasts of seismic activity, ecological assessment, and search for hydrocarbon deposits. Combined methane and helium anomalies may indicate the presence of a deep ascending fluid.

The basins of the Red and Mekong rivers, aquifers, lakes, and shallow bays of the Bien Dong Sea are active areas regarding hydrocarbon, hydrogen, and carbon dioxide degassing [11, 75, 80].

The possibility of penetration of significant amounts of deep gases from the interior into the surface horizons of sedimentary basins and tectonic depressions of Vietnam is one of the critical issues for elucidating the role of this region in the gasgeochemical regime of the Northwestern Pacific. The presence of numerous oil- and gas-bearing structures on the shelf of Vietnam creates preconditions for the sub-vertical migration of magmatogenic, metamorphogenic and thermogenic gases through permeable zones. However, the share of migrating components is unclear in the gas emission on the shallow shelf of Vietnam and adjacent land.

According to R. B. Shakirov et al., (2017) [81], the Bien Dong Sea is a part of the East Asian Gas Hydrate Bearing Belt stretching from the coast of New Zealand to the Bering Sea. We believe that the Vietnamese continental shelf can be divided into three geochemical provinces regarding the distribution of methane and other gaseous components. The Northern part is roughly between 18°N and 15°N, the central part is bounded by 15°N and 11°N, and the Southern part extends between 11°N and 6°N. The first methane anomalies were observed mainly in the Northern and Southern parts of the Vietnamese shelf. These anomalies indicate a long-term methane diffusion from sediments [11]. Luong et al., (2019) [77] found contrasting variability in dissolved methane concentrations in the near-bottom waters of the western Bien Dong Sea. Low methane concentrations were detected in the deep-water area (500–3,800 m), while higher methane concentrations were observed within the shallower shelf and slope areas (30–500 m). This aspect may coincide with the methane distribution in the bottom sediments of the sea.

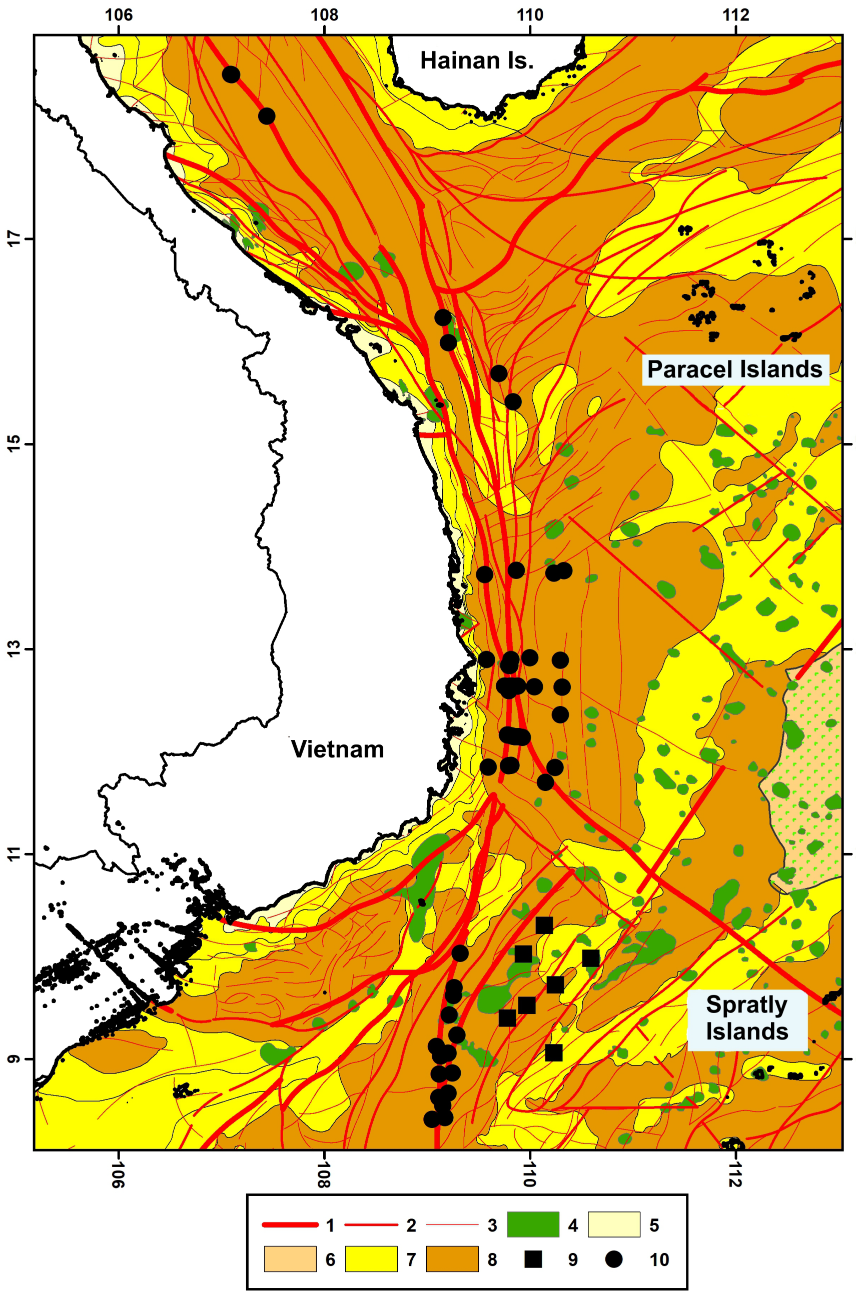

Figure 3. Simplified geological map of the Bien Dong and sampling sites. 1- first order faults; 2- second order faults; 3- third order faults; 4- βN_2-Qp: olivine basalt, tholeiitic basalt, andesite-basalt. Sedimentary columns of different ages: 5- Q; 6- N_1; 7- N_2-Q; 7- N_2-Q; 8- E_2. 8- E_2. Sampling points: 9- sampling during the expedition of DK-105; 10- sampling onboard the R/V Akademik M. A. Lavrentyev (LV88, 2019)

Material and Methodology

Gasgeochemical studies during the 88th cruise of the R/V Akademik M. A. Lavrentyev were conducted in three areas of the western part of the Bien Dong [82, 83]: the Nam Con Son sedimentary basin (the Southwestern part of the Bien Dong Sea); the Phu Khanh sedimentary basin (the central Vietnamese shelf and slope); the Red river sedimentary basin. During the cruise, a large amount of geological, geophysical, and oceanographic data was obtained, making it possible to reveal the new geochemical and mineralogical features of the Vietnamese continental shelf and adjacent deep-water basins [82, 83]. In addition, data on gases in sediments in the Southwestern sub-basin were also obtained in the DK 105 cruise (August–September 2019) [78].

Onshore research in North and South Vietnam was carried out within the joint Russian-Vietnamese Laboratory for Marine Science and Technology framework, within a series of local grants, and within the framework of Vietnamese national projects. During the works together with colleagues from the Institute of Marine Geology and Geophysics of VAST within the framework of the joint Russian-Vietnamese Laboratory for Marine Science and Technology (Pacific Oceanological Institute named after V. I. Il'yichev, FEB RAS, and the Institute of Marine Geology and Geophysics, VAST) on the Vietnamese shelf of the Bien Dong, a representative set of samples was obtained for gasgeochemical studies. During the marine expeditions, gasgeochemical studies on the contents of methane, hydrocarbon gases, carbon dioxide, helium, and hydrogen were carried out in the deep, surface, and subsurface layers of the water columns, and in bottom sediments of the sedimentary basins of Nam Con Son, Phu Khanh and Red river (including the Gulf of Tonkin).

Water sampling. Onboard water sampling used a 6-position Rosette (USA) combined with a CTD probe. The sounding complex was equipped with a cassette of NISKIN bottles (6 bottles). Water sampling was carried out at different horizons, considering the vertical distribution of temperature, salinity, and other hydrological parameters during CTD sounding. The study examined horizons characterized by significant gradients of oceanographic parameters, such as temperature, salinity, turbidity, etc., to detail the distribution of methane concentrations in the water column. Sampling was conducted at six horizons; the standard ones were 0 (surface), 20, 50, 200, and 600 meters. The "HeadSpace" method of equilibrium concentrations was used for the gas chromatographic water analysis. For a more accurate and detailed analysis of methane of concentrations and the number of its homologs (ethane, propane, butane) in seawater, sampling to obtain the gas phase using the vacuum degassing method was also conducted.

Sediment sampling. Sampling for gasgeochemical analysis was carried out using 12 mL syringes with cut-off nozzles into 43 mL and 68 mL flasks filled with a saturated NaCl solution with a preservative (0.5 mL of chlorhexidine digluconate, 0.05%). The sediment sampling was carried out using a gravity sampler 6 meters long (in the Gulf of Tonkin, the sampler was 3 meters long) and up to 120 mm in internal diameter. During the sampling, the sedimentary cores' lithological features -were considered.

Methods for analyzing the stable carbon isotopes. The analysis of stable carbon isotopes of methane and carbon dioxide was performed on a Finnigan MAT - 252 mass spectrometer using the CF-IRMS system at Nagoya University, Japan [63]. The mass spectrometric method was used to study the genesis of gasgeochemical fields in these areas. The values (δ) were calculated as $R_{sample}/R_{standard}$ ratio, where R is the $^{13}C/^{12}C$ ratio for both the testing sample and the standard (VPDB).

Methods of analysis of water and sediment samples. The analysis of hydrocarbon gases, nitrogen, oxygen, and carbon dioxide was carried out using a two-channel gas chromatograph CristalLux 4000M equipped with ionization flow and thermal conductivity sensors (sensitivity is 10^{-5}%). For helium and hydrogen analysis, we used the gas chromatograph "Chromatek-Gazochrom 2000" (JSC "Chromatek", Yoshkar-Ola) with high-sensitivity thermal conductivity sensors (1–2 ppm for helium and hydrogen), as well as a vacuum degassing unit. The analysis duration for hydrocarbon gases was 20 min; for helium and hydrogen - 5 min.

The Gas Geochemistry Laboratory of the Pacific Oceanological Institute of FEB RAS has State Standard Certificate No. 58 for the passport of the laboratory PS 1.051-21.

Molecular biological studies of sediments were conducted on the surface layer of the core and along the core length in increments of 50 cm, considering layers with different colors, odors, and consistency. Microbiological communities were considered: anaerobic, microaerophilic, and anaerobic bacteria. The intensity of aerobic and anaerobic destruction, the balance of methanogenesis/methanotrophy, sulfate reduction, nitrogen fixation, and nitro-denitrification were evaluated.

Gasgeochemical provinces of the Vietnamese shelf

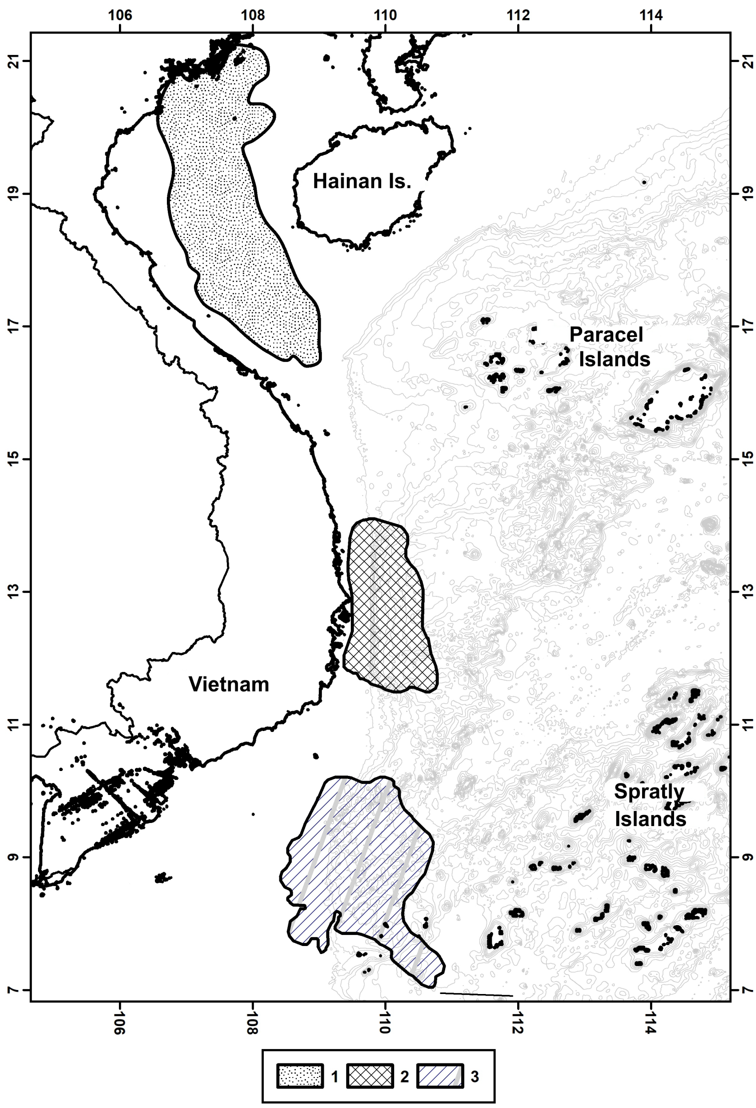

Figure 4. Scheme of the main gasgeochemical provinces, identified based on the analytical results of gasgeochemical studies. 1- the Northern gasgeochemical province; 2- the Central gasgeochemical province; 3- the Southern gasgeochemical province

The results of the gasgeochemical analysis allow us to distinguish three gasgeochemical provinces. The Northern is represented by the sedimentary basin of the Red river and Beybuwan, the Central - by the Phu Khanh sedimentary basin, and the Southern - by a part of the Nam Con Son sedimentary basin and adjacent areas of the Southwestern sub-basin of the Bien Dong. The provinces are distinguished according to their gasgeochemical, geochemical, geostructural, and other characteristics. The Northern area lies between 18°N and 15°N, the central area locates between 15°N and 11°N, and the Southern area extends from 11°N to 7°N. Morphologically, these parts are also different. The Northern and Southern provinces occupy a gentle and wide shelf; the central area is characterized by a narrow shelf with a steep slope. The outline of the selected areas is shown in Fig. 4. The averaged indicators of the gasgeochemical characteristics of the provinces are presented in Table 1.

Table 1. Averaged gas concentrations in sedimentary basins of the western part of the Bien Dong

Province	C mean, ppm	C mean, nL/L	AMC, nL/L	BMC, ppm	BMC, nL/L	W, %	$C_1 \backslash (C_2+C_3)$	He mean, ppm	H_2 mean, ppm	Mean $\delta^{13}C$-CH_4, ‰
Northern	4.4	3,146	9,460	1.15	786.5	14.5	21.1	11	5	-52.6
Central	12.7	12,486	58,000	12.9	12,543	14.5	26.2	15	12	-48.9
Southern	19	18,587	56,700	18.7	18,228	27.07	7.62	3	5	-36.9

Note: BMC- background methane concentrations; AMC- anomalous methane concentrations; W- coefficient of the moisture content.

The Northern gasgeochemical province

The province was identified based on the encircling fault zones of the Red, Ma, and Lo rivers. Faults run through the center of the Gulf of Tonkin and control the Song Hong Trough.

According to microbiological surveys at sampling stations, methanotrophic organisms and a small number of sulfate-reducing bacteria were detected [84]. Methane concentrations in sediments vary from 2,860–9,460 nL/L. There is a pattern in the spatial distribution of the anomalous methane concentration (AMC) fields - a considerable increase in concentrations towards the fault zones (relative to the background field of methane concentrations, indicating methane emission caused by a network of active near-surface faults. The microbial component in the background methane field is characterized by $\delta^{13}C$-CH_4 values of -70.5–93.6‰ PDB. River waters are marked by the “lightest” composition of methane carbon. Mixed thermogenic-microbial gases in the Gulf of Tonkin sediments are represented by $\delta^{13}C$-CH_4 values of -52.2–58‰. An increased background for thermogenic and metamorphogenic methane and its homologs and hydrogen anomalies, indicates an ascending emission of natural gases from depths, probably corresponding to meso- and apocatagenesis zones. In this setting, elevated helium concentrations indicate the presence of components from depth.

In the Northern province of the Vietnamese shelf, an extensive field of anomalous methane concentrations in the near-bottom waters was found in the sedimentary basin of the Red River Rift (Fig. 5). At depths of 70 m and 88 m, the maximum methane concentrations of 250 nL/L and 230 nL/L, respectively, were observed in this area. In addition, elevated methane concentrations are found along the coastal line of the shelf within this area. The methane distribution characterizes the presence of an anomalous gasgeochemical field. Since the intensity of the abnormal gasgeochemical field is weak and there are almost no heavy hydrocarbon gases, effec productive oil and gas traps are assumed occur at a depth of more than 3,000 m and are controlled by a fault zone. The presence of a fault is confirmed by high concentrations of carbon dioxide (0.40–0.50 mL/L). Separate fields of elevated methane concentrations in the coastal part of the shelf characterize the prospects for oil and gas exploration here (under favorable geological conditions) positively.

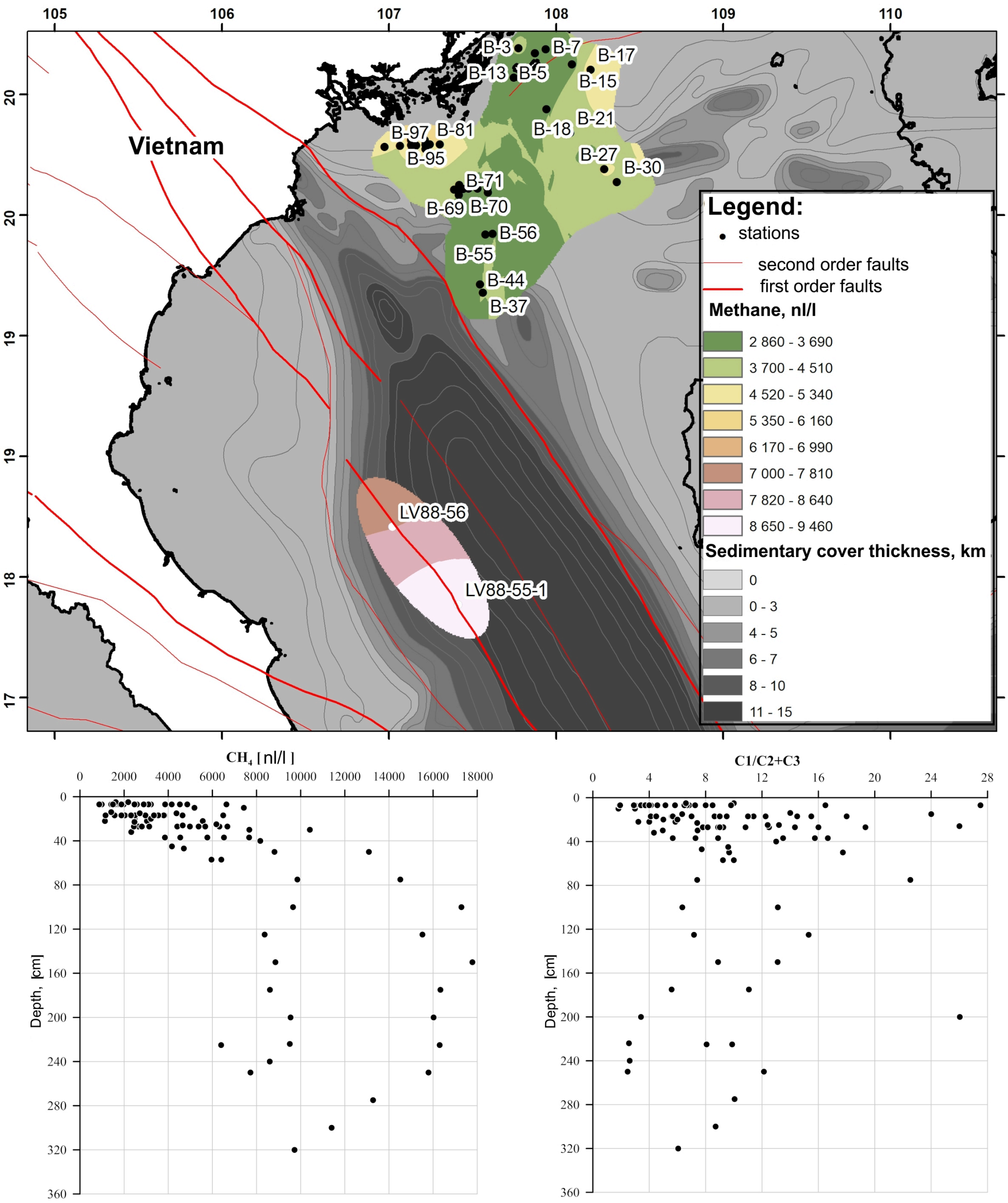

Figure 5. The main gasgeochemical characteristics of the province. 1- spatial distribution of methane; 2- methane distribution in depth; 3- $C_1/(C_2 + C_3)$ ratio by depth

The paper [85] established the transforming character of fractures, directed Northeasterly, that cross all the structures in the Gulf of Tonkin; this is caused by their penetration into the sedimentary complexes of the Triassic, Cretaceous, and Cenozoic rifts from older buried complexes. Thus, the identified system of fluid-conducting zones is determined by a network of discontinuous structures, along which the conditions are most favorable for the sub-vertical gas and fluid transport towards the upper part of the Earth's crust (in our case, towards the sedimentary cover of the Song Hong Trough, including marine sandy-clayey deposits). Fracture systems in geodynamical active rift structures of the Triassic, Cretaceous, and Cenozoic are favorable for the migration of deep fluid. Thus, the relationship of gasgeochemical anomalies with tectonics features reflects an increased gas and fluid conductivity in Northern Vietnam. According to a previous gasgeochemical survey [81, 86], the microbial composition in the background methane field is characterized by $\delta^{13}C\text{-}CH_4$ values of -70.5–93.6‰ PDB. Mixed thermogenic-microbial gases in the Gulf of Tonkin sediments are represented by $\delta^{13}C\text{-}CH_4$ values of -52.2–58‰ [75]. Based on the character of the isotopic composition, the researchers assumed that gas was generated in the zone of meto-apocatagenesis, and ascending methane emission took place along the fault zones. Isotopy data obtained in 2019: $\delta^{13}C\text{-}CO_2$ -17.4–23.8‰ and $\delta^{13}C\text{-}CH_4$ -40.4–60.0‰. According to the isotopic composition values, methane is of thermogenic origin; it was probably generated at great depths, most likely related to the top of the crystalline basement. We assume that towards the center of the Song Hong Trough, methane carbon will become "heavier" following analogies for gas fields in Siberia [87]. The main gas geochemical characteristics are shown in Fig. 5.

The central gasgeochemical province

The province is distinguished within the basement of the Phu Khanh sedimentary basin. The Red river fault crosses the basin in the North and the Tuy Hoa fault zone (shift). Also, according to the results of microbiological surveys at sampling stations along the axial fault, three groups of microorganisms were identified immediately: methanotrophic, hydrocarbon-oxidizing, and sulfate-reducing [84]. At the same time, the growth of some methanotrophic organisms was noted only at low temperatures. The main gasgeochemical characteristics are shown in Fig. 6. Methane concentrations in the basin ranged from 2,110–58,000 nL/L. All anomalous methane concentrations are confined to fault zones. At high methane concentrations in the province, the $C_1/(C_2 + C_3)$ ratio also indicates significant microbial activity. The results of a series of isotopic analyses show that the values for the stable isotope of methane carbon lie within the range of -27.7‰ and -66.6‰ (of carbon dioxide carbon lie within the range of -15.4‰ and -25.9‰), which indicates a predominantly thermogenic gas genesis (depths of more than 2 km) with an admixture of gases of microbial origin. In an intense upward natural gas flux, favorable conditions for developing microbial processes are created. The $^{13}C/^{12}C$ isotopic ratio of thermogenic methane can be masked by the admixture of a significant proportion of microbial gas from the upper sedimentary horizons. The analysis found high methane concentrations (up to 666.4 nM/dm^3), ethane, and propane in sediments at the considered stations in the Phu Khanh Basin. In the nearest stations' deposits, methane content in sediments is even higher, up to 3,422 nM/dm^3. The highest hydrogen and helium concentrations were found in the deep-water part of the Phu Khanh basin; further to the South, there is a trend towards a gradual decrease in concentrations. In the central region, evidence of "local" fluid discharge zones was observed in its Northern and Southern parts.

In the central gasgeochemical province of the Vietnamese shelf, background methane concentrations (30–40 nL/L, depths of 100–400 m) were recorded. High concentrations were observed (0.74–0.79 mL/L) in the depth interval of 200–400 m. In general, background fields of methane and elevated carbon dioxide concentrations were discovered here in the 1980s, which indicated the passivity of geological processes, the occurrence of ancient volcanic structures close to the surface, the presence of tectonic boundaries between them, and the absence of oil- and gas-bearing strata. At present, evidence of gas "flares" and the prospects for gas manifestations at volcanogenic structures and in zones of decompacted ledges have appeared in this area. The occurrence of mud volcanism is also known on the shelf of Central Vietnam [88].

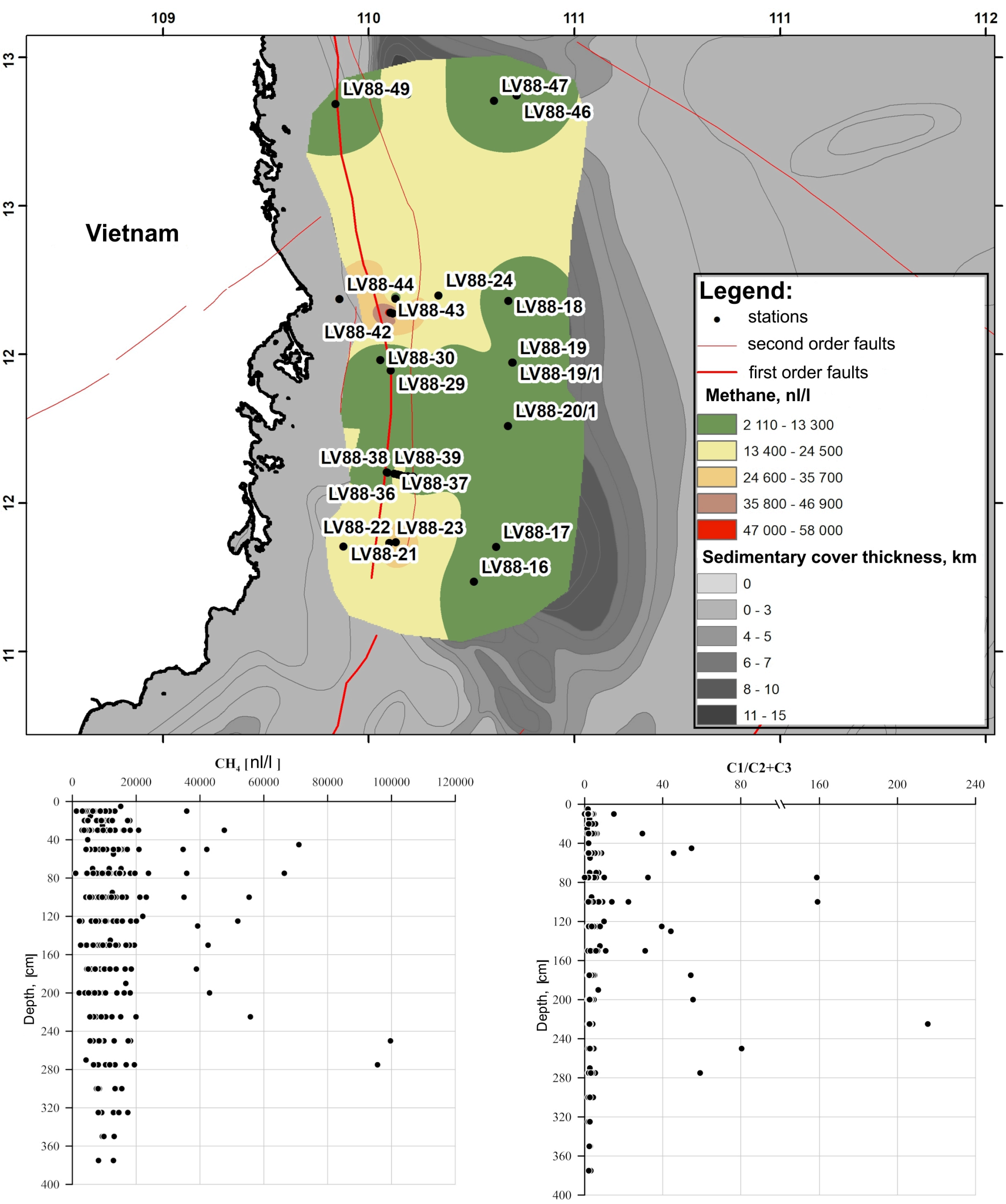

Figure 6. Some gasgeochemical characteristics of the Central province. 1- spatial distribution of methane; 2- methane distribution in depth; 3- $C_1/(C_2 + C_3)$ ratio by depth

The Southern gasgeochemical province

The Northern part of the Southern province is represented by a ridge connecting the Cuu Long and Nam Con Son sedimentary basins. A deep meridional fault zone (with manifestations of degassing) passes through the ridge. Anomalies of hydrocarbon gases, hydrogen sulfide (an unusual phenomenon for sediments of the Vietnamese shelf), and ferromanganese formations with a rubidium content of up to 2% were detected in sediments by X-ray fluorescence analysis [79]. Both methanotrophic microorganisms at the sea depth of 400 m and the sulfate-reducing bacteria were observed in all three provinces.

Methane concentrations ranged from 386–56,700 nL/L. Anomalous methane concentrations are mainly confined to faults.

"Heavy" composition of methane carbon and carbon dioxide carbon (-25‰ and -17.6‰ VPDB, respectively) was observed in the sediments of the deep-water part of the Phu Khanh basin and the Northern part of the Nam Con Son basin; anomalous concentrations of helium and hydrogen in sediments were recorded in the same areas, indicating metamorphogenic and thermogenic sources. Following the results obtained, a deep hydrocarbon-degassing zone is assumed to exist in this area. The ratios of hydrocarbon gases, as well as increased gas moisture values, confirm this assumption.

The main gasgeochemical characteristics are shown in Fig. 7. According to the data obtained, the isotopic composition of methane carbon varies from -25‰ to -64‰, and of carbon dioxide carbon from -17.5‰ to -24.8‰ VPDB.

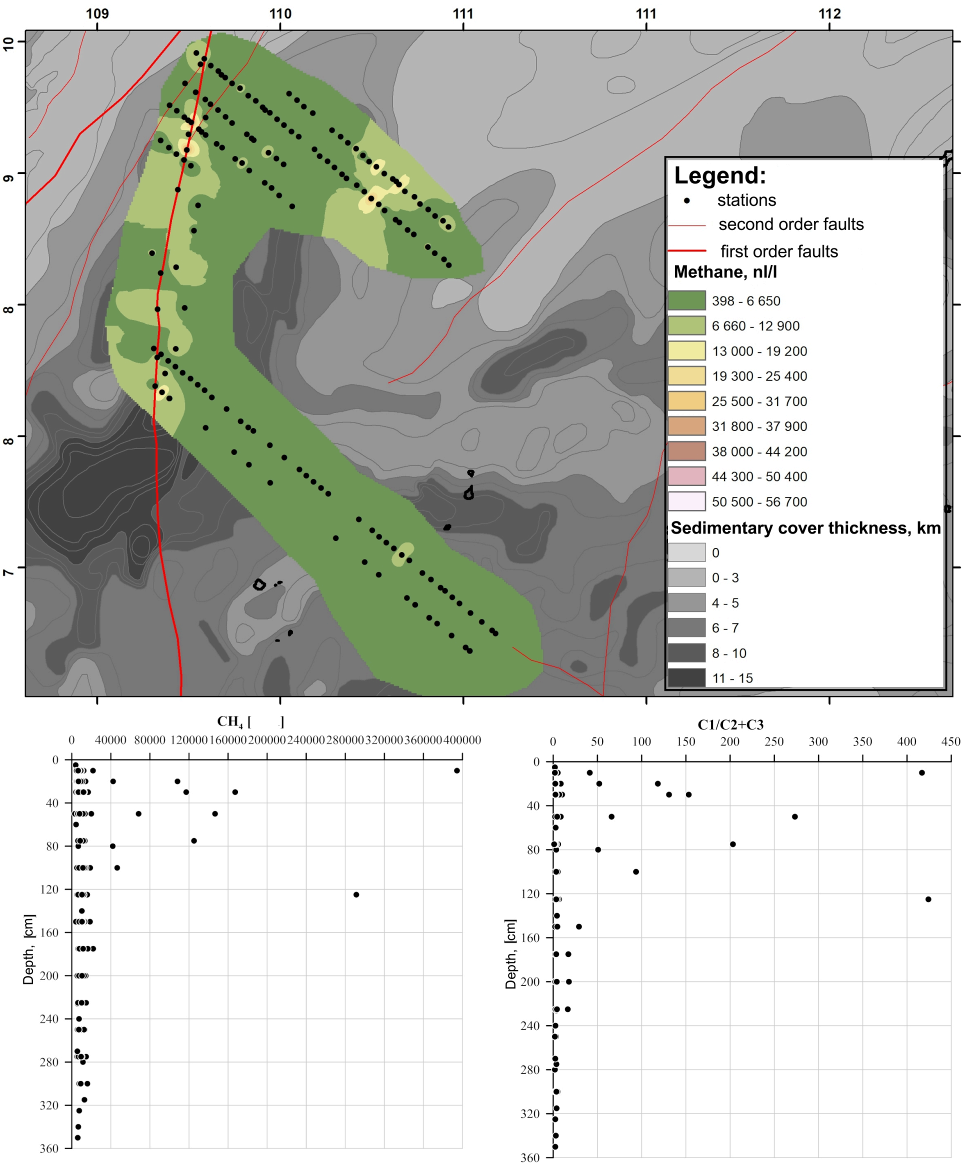

Figure 7. Main gasgeochemical characteristics of the Southern Province. 1- spatial distribution of methane; 2- methane distribution in depth; 3- $C_1/(C_2 + C_3)$ ratio by depth

In the Southwestern part of the Vietnamese shelf, anomalous fields of methane, hydrogen, and carbon dioxide were detected in the near-bottom waters within the meridional fault zone, which extends along the boundary of the shelf and slope of the western part of the Bien Dong Sea at the sea depths of 200–300 m. Background gasgeochemical fields of methane (10–20 nL/L) were observed in the sea's deep water (500–3,800 m). On the shelf (depths of 30–200 m) and slopes (depths of 200–500 m), methane concentrations increased up to 30–40 nL/L. The presence of anomalies of methane and its homologs characterizes the hydrocarbon potential positively; hydrogen anomalies point to this zone's depth and seismo-tectonic activity; carbon dioxide anomalies indicate the presence of intrusive complexes [55]. At the same time, the variability of anomalous methane fields is associated with the oil-and-gas content in the basement rocks and sedimentary cover of the continental shelf, not with earthquakes, as in some areas of the Sea of Okhotsk and the Sea of Japan.

Medium-intensity high methane gasgeochemical fields (up to 1,540 nL/L) and trace amounts of heavy hydrocarbon gases were registered at the Mekong, White Tiger, Yuzhno-Konshonskaya, Zapadno-Nutovskaya structures (currently, their oil and gas potential is proven), The finding suggests that anomalous gasgeochemical fields of weak and medium-intensity are indicators of oil and gas deposits in low-seismic regions. These structures must be located at the Southern end of the progradation wedge of the oceanic crust [89].

The Sea of Japan has a similar picture: low-intensity anomalous gasgeochemical methane fields were found in the Genzan trough (the Southern edge of the oceanic crust block) [90]. Thus, in the practice of regional oil and gas prospecting, attention should be paid to the features of the deep structure, namely, the interaction zones of different types of the earth's crust.

The region is an area of an extension where the lower sedimentary strata and the dislocated basement can form reservoirs due to dilatancy: there are conditions for the migration of hydrocarbons from the surrounding parent strata. Under such conditions, strong upward methane fluxes, and other structures (such as chimneys), are not formed. There are very few active faults, and with the low seismicity of the area, the probability of detecting them is small. However, diffusive seepage is the cause of anomalies of methane and other hydrocarbon gases along the sides of folded structures, which is similar to inland deposits. This geological phenomenon can explain large oil reserves in the crystalline basement of the Cuu Long and Nam Con Son basins. Under such conditions, a gasgeochemical survey can be especially effective in searching for traditional oil and gas deposits.

Gasgeochemical criteria indicate that in the marginal seas of East Asia, the prospects for oil and gas deposits and gas hydrates are also high within those numerous structures where exploration work has not been carried out yet.

Thus, the results make it possible to identify the features and patterns of the spatial distribution and genesis of natural gases in sediments in the Red river sedimentary basin, the Phu Khanh sedimentary basin, and the Nam Con Son sedimentary basin.

Gasgeochemical provinces differ by a set of gasgeochemical indicators, geological structure, oil-and-gas potential, generation mechanisms, accumulation, and migration of gases and fluids.

Relationship between gasgeochemical fields and seismotectonics in the Bien Dong

The gasgeochemical regime of the continent-ocean transitional zone is determined by geological processes in the junction zone of geostructures (continent-marginal sea). An essential aspect of the distribution, composition, and intensity of gasgeochemical fields is their relationship with the seismotectonic setting. The study of seismotectonic control over the distribution of heterogeneous gas fluxes is closely associated with the forecast, search for hydrocarbon minerals, and monitoring of greenhouse gas emissions into the atmosphere.

The Bien Dong is distinguished by its unique geological history, the development of uncompensated tectonic troughs, special deposits of oil and gas in the crystalline basement, and few gas manifestations against the background of the "calm" gasgeochemical regime. The geodynamics of the Bien Dong Sea is determined by the interaction of three lithospheric plates: Pacific, Indo-Australian, and Eurasian. The Bien Dong is known for the commercial oil and gas potential of the South Vietnam shelf basement [91, 92], and the prospects for oil potential of the Central and Northern shelves are being discussed [93].

The Bien Dong Sea is a low-seismic basin in the western and central regions; the basin is sandwiched between four lithospheric plates and is formed by a subduction zone in the East [94], which, however, is not a determining factor in the formation of this sea [89]. Oil and gas deposits in the granitoid basement have been discovered in the Southern Vietnam shelf, and gas condensate deposits have been detected in the Neogene sediments of the Hanoi Basin in the North of the country. Both areas are located in vast and poorly studied subsidence zones. In terms of hydrocarbons, the Vietnamese shelf, slope, and even deep-water areas are highly promising.

Gasgeochemical studies in the Northern part of Vietnam, carried out by the Joint Russian-Vietnamese laboratories of POI FEB RAS - IMGG VAST and FEGI FEB RAS - POI FEB RAS - IGS VAST, annually bring new facts about the wide distribution of active manifestations of methane and its homologs, carbon dioxide, carbon monoxide, hydrogen, and helium. For example, on the slope of Mount Phan Xi Pang (Northwestern Vietnam), anomalous methane concentrations were first detected in thermal springs in 2016 [95]. In this area, using the combinatorial recognition algorithm KORA-3, seismogenic nodes were identified, and the possibility of earthquakes with $M > 5–7$ was described [95]. In Bien Dong, methane release into the atmosphere was established [96]; methane values were generally small (compared to the Sea of Okhotsk and the Sea of Japan) [76]. The methane release is probably caused by the low modern gas activity of the bottom of the Bien Dong, which is associated with its low seismicity. Here, the pre-seismic and long-term post-seismic relationship between gasgeochemical fields and earthquakes probably prevails.

Vietnam's Southern and Northern shelves have high oil and gas potential [91]. The Bien Dong is a promising basin searching for methane-hydrate deposits [97]. The shallow water, up to 50 m deep, the Northwestern shelf of the Bien Dong is still poorly studied, but there is direct evidence of its oil and gas potential. For example, in the Southwest of the shallow shelf of Hainan island, there are about 120 seeps of hydrocarbon gases with "thermogenic" carbon isotopic composition of methane (> 71%) -33.91–(-38.24)‰ [98]. Moreover, a large gas reservoir, consisting mainly of carbon dioxide, was discovered near Hainan island, possibly due to thick carbonate strata and intrusive formations. The main migration channels for gas-liquid fluids within the continental slope are faults below the shelf edge [99]. In its Northern part (the Shenhu Basin), Chinese researchers, while drilling with special sealing devices, discovered gas hydrates in the sedimentary interval of 153–225 m below the bottom surface in the area with coordinates 19.9°N, 115.2°E [100].

In this area, at least eight paleo-outbursts of methane (associated with sea level decrease, dissociation of gas hydrates, hydrological parameters, and a source of hydrocarbon (methane) fluid) have been identified [101].

Our primary studies were carried out in the Beibuvan sedimentary basin, which occupies a shallow water area with depths from 20–70 m along the continental and insular coasts [72, 73].

The Gulf of Tonkin (Beybuwan sedimentary basin), one of the least studied and associated with the most extensive rift system of the Red river regions, was chosen as a research area. Most earthquakes are recorded in Northern Vietnam and on its coast, compared to the Central and Southern regions (Fig. 8) [102].

An extended linear region of earthquake epicenters in the Red River Rift zone, consisting of deep-seated faults, is distinguished here; displacements along them are recorded for the entire tectonic development of Northern Vietnam [102]. The faults in the Red river lineament, stretching from the continent towards the Gulf of Tonkin, are currently active: right-sided displacements, which are responsible for numerous strong crustal earthquakes, occur along with them ($M < 7$; [102]). At least one hundred manifestations of thermal waters have been identified along the Red river shear zone, and our studies [82, 86] have revealed anomalous gasgeochemical fields of thermogenic hydrocarbon gases, helium, hydrogen, and carbon dioxide there. These data were confirmed by analyzing the chemical composition of sediments and biomarkers (for example, Pr/Ph 0.8–1.1) and by other means [86]. The peculiarities of the gas distribution and the biomarker occurrence of biomarkers are most likely caused by the migration of gases and hydrocarbon fluids along the fault system of the Red river shear zone. The presence of C_2–C_4 hydrocarbon gases in sediments (in concentrations close to methane) indicates the signs of the epigenetic thermogenic genesis of hydrocarbon gases.

In 2012–2014 it was shown for the first time [73] that there are anomalous gasgeochemical fields of methane (mainly of low and medium intensity), hydrocarbon gases, carbon dioxide, helium, and hydrogen in sediments, near-bottom and surface waters in the Beibuvan basin, within the islands and coastal framing. Not more than 20 earthquakes have been registered within the marine area of the basin during historical times. The low number of earthquakes recorded is caused by the low seismic activity of the Northwestern part of the Bien Dong Sea [103] but is also, apparently, due to the lack of instrumental observations.

Profiles (6), selected according to preliminary geophysical studies of IMGG VAST, were executed. The main criteria for the profiles were the geophysical signs of gas migration channels.

Propane and butane were detected in 80% of samples. Methane concentrations in the study area sediments range from 1,010–8,000 nL/dm^3. Such methane concentrations in surface sediments, by analogy with the known oil and gas fields of the Sea of Okhotsk, usually indicate the presence of a lithological layer, which probably, delays the migration of hydrocarbon gases, provided that their source is in the sedimentary column or basement. Background methane concentrations in bottom sediments of the Gulf of Tonkin reach 3,490 nL/dm^3, a reasonably high background, indicates a long-term diffusive natural gas infiltration through low-permeability strata.

Gasgeochemical data, shown on time sections of seismic profiling, made it possible to reveal the confinement of anomalous gasgeochemical fields to tectonic faults. The presence of thermogenic hydrocarbon gases, biomarkers of pristane and phytane, together with geological data, determine the positive oil and gas potential of the Beybuwan sedimentary basin, which is controlled by the Red River Rift in the West, where hurricane concentrations of methane (13 mL/L) were detected in freshwater sources. Based on the low ratios of methane to hydrocarbon gases of the C_2–C_4 series and the distribution of hydrogen, carbon dioxide, and helium anomalies on fault maps, we concluded that the gas anomalies are predominantly migratory in origin.

In 2014, the polygon around Cat Ba island was detailed; while surveying the island itself since 2013, the maximum methane and carbon dioxide concentrations were registered there. Methane was detected in all sedimentary samples; ethane was recorded in 95% of all samples. The ethane concentrations in the samples were 10 times lower than the methane concentrations but sometimes reached equal values, for example 1,050 nL/dm^3 [104].

Notably that the methane concentrations dissolved in the fresh waters of the study area decreased from west to east within the study area. This low value can be explained by the decrease in the influence of gas-fluid fluxes at a distance from the Red River Rift fault system. The difference between very high methane concentrations in wells and water sources of islands and low methane concentrations in sediments and seawater is notable. On the one hand, judging by the low content of organic carbon and other signs, intensive methane oxidation and organic matter processes may occur in marine sediments and seawater within the Gulf of Tonkin. Large-scale methane oxidation is also expressed in the carbonate accumulation North of the Bien Dong, as in the geological past [101]. Another, more likely factor is that the solid carbonate rocks (observed at most stations) are good seals that unify the gas flux towards the seabed.

Due to the presence of shale and clayey layers and low seismotectonic activity in the study area, the high-intensity, brightly contrasting anomalous gasgeochemical fields are not formed.

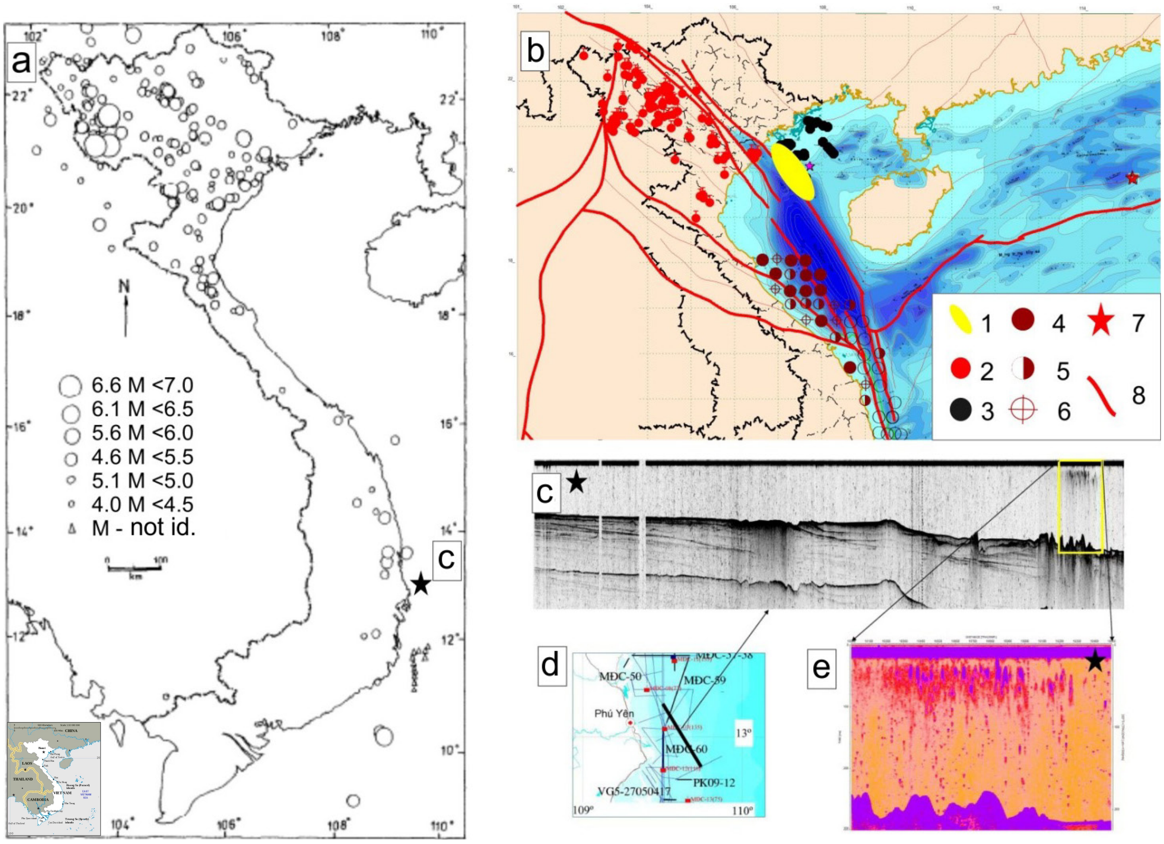

Figure 8. Map of earthquakes (a); geothermal manifestations (b), wells 30–40 m deep: 1- area of increasing intensity of the anomalous gasgeochemical field and concentrations of chemical elements (Fe, As, Al/Si, and others), the occurrence of anoxic biomarkers; 2- geothermal manifestations; 3- stations (2013–2014); 4- anomalous methane fields of medium intensity; 5- anomalous methane fields of low-intensity; 6- background methane field; 7- gas hydrates; 8- fault zones; (c)- section of CSP (the gas seep is indicated by the yellow rectangle); (d)- location of the profile of CSP (see also Fig. (a)); (e)- gas bubbles in the water column

In the Northwestern part of the Bien Dong, anomalous gasgeochemical fields controlled by the geological structure of the Beybuwan sedimentary basin are distinguished. The faults in the region are low-active seismically, expressed in low methane concentrations. Methane was registered in all sedimentary samples (at concentrations up to 9,000 nL/L, against a background of 4,000 nL/L). Ethane was detected in 95% of all samples.

The data are consistent with the studies of the Gasgeochemistry Laboratory of POI FEB RAS in the East Siberian Sea. It has been established that the areas of predicted gas-oil, oil-and-gas, and oil deposits on the East Siberian Sea shelf are characterized by the minimum methane and hydrocarbon content in bottom sediments (less than 0.05 cm^3/kg and 0.001 cm^3/kg, respectively) and by the maximum thickness of the sedimentary cover within the North Chukchi (more than 10 km), South Chukchi, and Ayon basins (more than 3 km) [105]. Thus, the formation of low-intensity anomalies of hydrocarbon gases is typical for promising oil- and gas-containing areas of the East Asian Seas in sedimentary basins with low seismic activity and more than 10 km thick in depocenters.

Notably, the features of the distribution of natural gases, the low seismic activity of the shelf, the distribution of thermal springs and coal-bearing areas on adjacent land, the linear nature of extended deep permeable zones, and the geochemical features of hydrocarbons make it possible (in our opinion) to compare regional permeable systems, located at a distance from each other. For example, we can compare the Red river fault system with the Mesozoic-Cenozoic graben-rift system of the Chukchi Sea, where anomalous low- and medium-intensity methane fields have been identified [106, 107].

In hydrological wells (as well as in boreholes and water sources) of Cat Ba, Co To, and other islands within the Gulf of Tonkin, methane concentrations reach up to 11 mL/L (against a background of 0.3 mL/L), which is probably associated with the presence of deep permeable zones and the sources of hydrocarbon gases, drained by these zones.

The distribution of hydrocarbon gases is unusual compared to the Sea of Okhotsk and the Sea of Japan: in bottom sediments and seawater, high concentrations of hydrocarbon gases (up to pentane) are recorded at relatively low concentrations of methane.

The low concentrations of methane in the presence of the overall promising hydrocarbon potential of the basin are explained by the low seismotectonic activity of this part of the Bien Dong Sea. Hydrocarbon potential manifests through the hydrological horizons in wells of islands of the Gulf of Tonkin.

Tectonic depressions are assumed to be the most promising. It is noteworthy that the Northwestern part of Vietnam is an area of modern seismic activity, and, probably, an increase in the number and intensity of anomalous gasgeochemical fields, which is caused by the rise in seismic activity, can also be registered within the shelf area of Northern Vietnam.

CONCLUSIONS

The processes of interrelation between near-surface gasgeochemical fields and sub-surface gas-bearing sources are taking place in the marginal seas of East Asia. Sub-surface gas-bearing sources include gas-bearing, coal-and-gas-bearing, oil-and-gas-bearing sediments, basement gases, crustal, and mantle.

The ascending emission of natural gases causes the formation of anomalous polygenetic gasgeochemical fields of low, medium, and high intensity. Lower crustal and upper mantle sources along the active margin contribute significantly to gas emissions.

These areas are characterized by various geological types of lithosphere degassing and processes that form and destroy the hydrocarbon deposits and gas hydrates.

Based on the existing data and our research, we identified gasgeochemical fields of natural gases in the bottom sedimentary strata along the Vietnamese continental shelf, slope, and deep-water part of the Bien Dong Sea.

The obtained results make it possible to distinguish three gasgeochemical provinces in the study region: the Northern (Red river basin), the Central (Phu Khanh basin), and the Southern (Nam Con Son basin). The provinces differ by gasgeochemical indicators, geological structure, oil-and-gas potential, oil-and-gas generation mechanisms, and gas and fluid accumulation, and migration features.

In the Phu Khanh and Nam Con Son sedimentary basins, the evident indicators of anomalous gasgeochemical fields created by previously unknown sources (which are producing hydrocarbon gases) have been identified.

The relationship of gasgeochemical fields with the seismotectonics of the Bien Dong Sea can be characterized as preseismic.

Underwater geological sources play a dominant role in near-bottom gasgeochemical methane anomalies. Data on the distribution of gasgeochemical fields in the Bien Dong Sea water column generally correlate with anomalies in bottom sediments. For the shelf stations, of course, the influence of complex hydrological and hydrometeorological factors that control the activity of gas exchange on the water-atmosphere interface and gas migration between water masses of different origins is also important.

Based on the gas distribution features in the sediment cores, it can be assumed that there is a large extended zone where hydrocarbons are produced, and hydrocarbon-degassing is taking place along the Vietnamese continental slope of the Bien Dong Sea. Focused areas of intense vertical migration of natural gases can be distinguished within this zone.

Research on thermal springs along the rift zone of the Red river is of great importance in terms of oil and gas exploration and fundamental research into the genesis of natural gases. The deep faults that control Northern Vietnam (the Red River Rift) are likely to be fluid-dynamically related to the permeable zones of the Northern Vietnamese shelf. Analogies can be traced to the Sea of Japan and Okhotsk [108]. One of the most important conclusions is the existence of the relationship between the gasgeochemical regime of thermal springs, the deep gas component, and the geological structure of Northern Vietnam.

The paper makes an important contribution to the United Nations Decade of Ocean Science for Sustainable Development, as well as to the research program of the WESTPAC working group on “Gas Hydrates and Methane Flows”.

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