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ORIGINAL PAPER

Origin and accumulation of natural gas in the Ningbo Tectonic Zone in the Xihu Sag

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Abstract

In this study, the geochemical characteristics of the oil and gas and the related source rocks in the Ningbo Tectonic Zone in the Xihu Sag are analysed to determine the source of the natural gas. The natural gas in the Ningbo Tectonic Zone is dry gas. It has a high content of methylcyclohexane and low contents of n-heptane and dimethylcyclopentane in the C_7 light hydrocarbons and a heavy ethane-carbon isotope composition, indicating that the natural gas in the study area was derived from humic parent material, making it a coal-type gas. The ratios of the light hydrocarbon parameters of the NB-A and NB-B natural gas are significantly different from those of crude oil and the Pinghu Formation source rocks, indicating that there is no pronounced relationship between the natural gas and the Pinghu Formation source rocks, and the natural gas may be from the source rocks in the deeper horizons of the Pinghu Formation. The homogenization temperatures of the fluid inclusions in the upper member of the Huagang Formation (NB-A1) are mainly 140–150°C, which correspond to 3.5–0 Ma (Late Cenozoic) according to the paleogeothermal history, indicating that in this area, the natural gas in the upper member of the Huagang Formation has charged since the sedimentary period of the Santan Formation.

Keywords Natural gas · Light hydrocarbons · Gas–source correlation · Accumulation time · Xihu Sag

Introduction

It is currently believed that there are three sets of source rocks in the Xihu Sag in the East China Sea Basin, namely dark mudstone, carbonaceous mudstone, and coal; and the main organic matter types are II_2–III (Wei et al. 2019; Yu 2020; Xu et al. 2015; Jiang et al. 2020). The Middle-Upper Eocene Pinghu Formation marine–continental transition facies source rocks are the main source rocks, and the Lower Eocene Baoshi Formation and Oligocene Huagang Formation fluvial–lacustrine facies source rocks are the secondary source rocks (Su et al. 2013; Zhou et al. 2020; Abbas et al. 2018). The Xihu Sag is rich in oil and gas resources. Our current understanding of the source of the crude oil is that it was derived from the Pinghu Formation source rocks (Zhang et al. 2020). However, the characteristics of the natural gas differ by region, indicating that their sources may be different. At present, the natural gas found in the central inversion zone and its surrounding areas is mainly pyrolysis gas, followed by deep source gas, which is believed to have been mainly derived from the source rocks in the Eocene Pinghu Formation and below. However, the sources of the natural gas differ in the different regions. Thus, the source and accumulation processes of the natural gas in the Ningbo Tectonic Zone in the northern–central part of the central inversion zone are still unclear. To address this problem, based on the natural gas composition, carbon isotope composition, and light hydrocarbon fingerprints, in this study, we analyzed the correlations between the geochemical characteristics of the crude oil and the source rocks, investigated the origin and source of the natural gas in this area, and determined the natural gas charging period by studying the fluid inclusions. Our results provide a theoretical basis for studying the mechanism of natural gas accumulation in this area.

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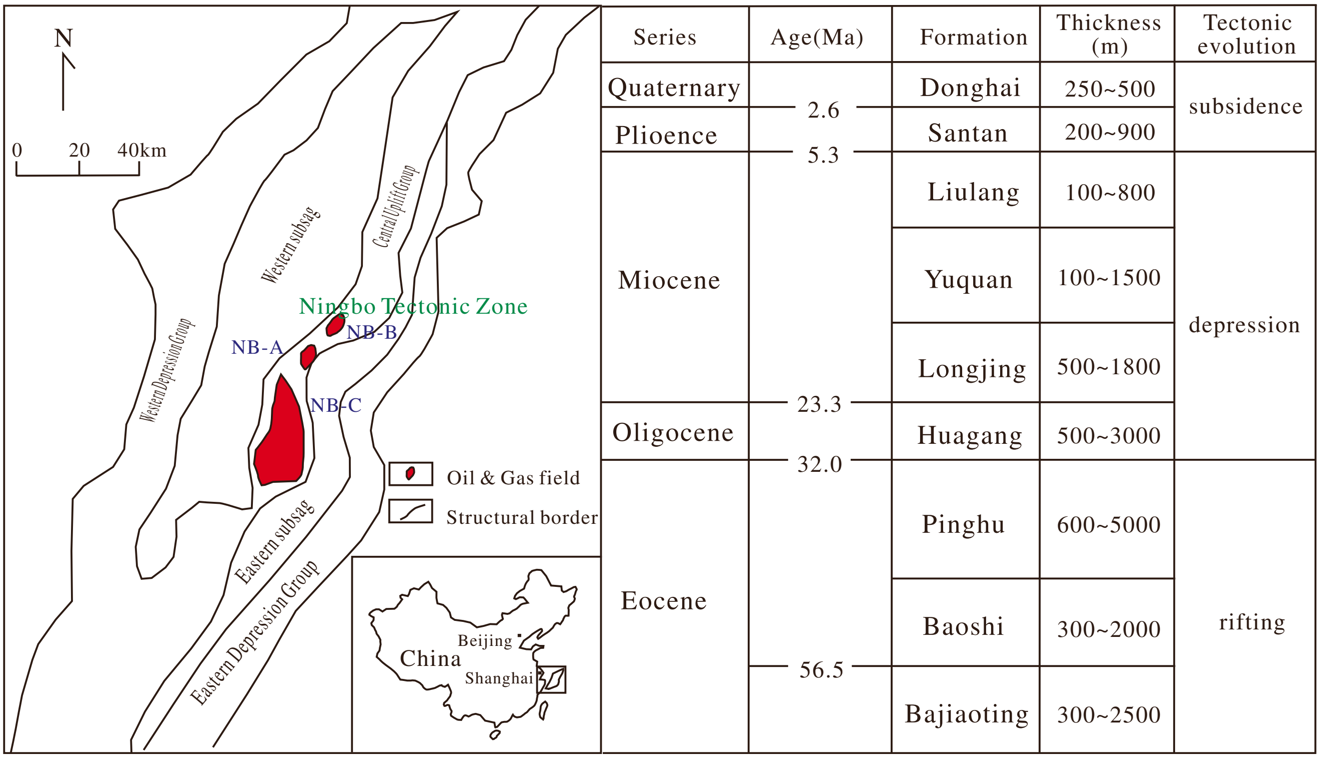


Fig. 1 Tectonic division of the Xihu Sag, locations of the well areas, and stratigraphic Diagram (Modified from Zhu et al. 2020)

Geologic setting and sample collection

The Xihu Sag is located in the northeastern part of the East China Sea Shelf Basin. It is approximately 400 km long from north to south and 100 km wide from east to west, and it has an area of approximately 5×10^4 km^2. From west to east, it can be divided into five secondary tectonic units: the western slope belt, the western subsag, the central inversion tectonic zone, the eastern subsag, and the eastern step-fault zone (Fig. 1). The Oligocene Huagang Formation sandstone reservoir is the main natural gas producing pay zone (Xu et al. 2020; Zhao et al. 2019). Among them, the central inversion zone in the middle of the Xihu Sag is an inversion anticline formed in the Late Eocene by the east to west compression during the Yuquan Movement. From south to north, it can be divided into four tertiary tectonic zones: the Tiantai, Huangyan, Ningbo, and Jiaxing tectonic zones. A series of large anticline structures were developed in an en echelon arrangement in the Ningbo Tectonic Zone, with trap areas of mostly greater than 50 km^2 and a total trap area of approximately 400 km^2. Industrial gas flow has been obtained in multiple wells, with a huge resource potential, mainly including major hydrocarbon-bearing structures such as the NB-A, NB-B, and NB-C.

Table 1 Geochemical parameters of saturated hydrocarbons in source rocks and crude oil from Ningbo structural belt

Well	Strata	Sample types	TOC(%)	nC_{21}^-/nC_{22}^+	Pr/Ph	Pr/nC_{17}	Ph/nC_{18}	OEP
NB-A1	H3	Mudstone	0.38	2.3	0.8	0.79	1.03	1.16
NB-A1	H3	Mudstone	0.34	2.1	0.83	0.54	0.63	0.95
NB-A2	H5	Mudstone	0.13	0.81	0.66	1.13	1.37	0.76
NB-A2	H5	Mudstone	0.49	1.68	0.8	0.78	0.8	0.79
NB-A3	H6	Mudstone	1.16	1.72	1.43	0.48	0.35	1.03
NB-A3	H10	Mudstone	2.05	1.81	0.91	1.18	1.58	1.26
NB-C1	P4	Mudstone	0.4	0.24	8.25	8.63	1	1.39
NB-C1	P4	Mudstone	0.25	0.59	4.3	5.7	1.26	1.21
NB-C1	P4	Mudstone	0.24	0.87	3.28	3.58	1.39	1.26
NB-C1	P4	Mudstone	0.23	0.8	3.64	5.45	1.49	0.99
NB-C1	P5	Oil	—	1.02	6.94	1.90	0.25	0.87

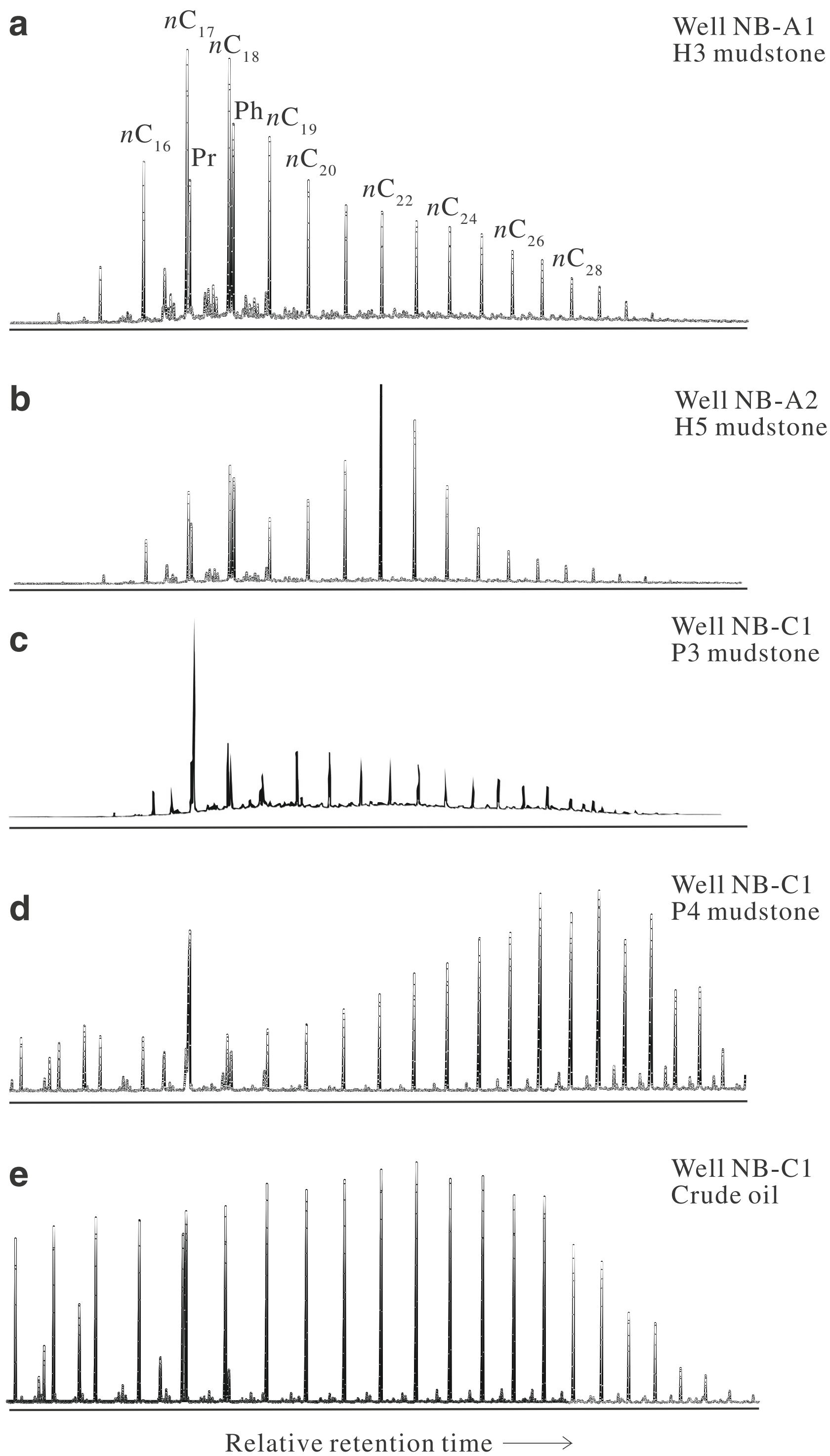


Fig. 2 Mass chromatograms of source rocks from (**a**, **b**) Huagang and (**c**, **d**) Pinghu Formation, and (**e**) crude oil from NB-C1 well (m/z85)

In this study, nine natural gas samples were collected from the NB-A and NB-B well areas in the Ningbo Tectonic Zone in the Xihu Sag. Crude oil and source rock samples were collected from the NB-A and NB-C well areas. The source rocks were analyzed using the absorbed hydrocarbon method, chromatography-mass spectrometry, and crude oil and natural gas composition and isotopic analyses.

Table 2 Geochemical parameters of light hydrocarbons in crude oil and natural gases from Ningbo structural belt

Well area	Well	Sample types	Heptane value	Isoheptane value	Compounds of $C_{5\text{-}7}$ light hydrocarbon (%)					
					nC_7	$MCyC_6$	$DMCYC_5$	$nC_{5\text{-}7}$	$iC_{5\text{-}7}$	$CYC_{5\text{-}7}$
NB-A	NB-A1-1	Gas	9.45	8.04	24.28	53.04	22.68	21.19	62.17	16.64
	NB-A1-2	Gas	8.71	6.85	20.95	55.24	23.81	22.00	61.82	16.18
	NB-A2	Gas	7.87	8.01	21.25	51.25	27.50	26.27	65.78	7.95
	NB-A3	Gas	9.33	4.62	19.71	52.55	27.74	26.77	62.64	10.59
	NB-A4	Gas	15.26	2.90	28.33	33.02	38.63	23.46	53.39	23.15
NB-B	NB-B1-1	Gas	8.56	10.29	24.72	51.69	23.59	35.78	33.03	31.19
	NB-B1-2	Gas	8.07	8.60	23.18	52.79	24.03	43.28	26.70	30.02
	NB-B1-3	Gas	5.27	14.94	27.66	55.17	17.17	37.91	14.60	47.49
	NB-B1-4	Gas	20.64	4.38	39.05	38.69	22.26	50.87	19.98	29.14
NB-C	NB-C1	Oil	38.13	0.58	17.59	64.58	17.83	15.37	8.31	76.32

Results and discussion

Geochemical characteristics of the source rocks, crude oil, and natural gas

The mudstone in the Huagang Formation in the Ningbo Tectonic Zone has a low Pr/Ph ratio, indicating that the sedimentary environment was weakly reducing to reducing, and a low nC_{21}^{-}/nC_{22}^{+} value with a prepeak characteristic, indicating that the source of the organic matter was mainly lower aquatic organisms. The mudstone in the Pinghu Formation has a high Pr/Ph ratio, indicating that the sedimentary environment was oxidizing, and a low nC_{21}^{-}/nC_{22}^{+} value, indicating that the source of the organic matter was mainly higher plants. According to the Pr/nC_{17} and Ph/nC_{18} values, the Huagang Formation source rocks in the study area mainly contain type II kerogen formed in a reducing environment, and the Pinghu Formation source rocks contain type III kerogen formed in an oxidizing environment.

The NB-C1 crude oil *n*-alkanes show a unimodal post-peak distribution with a Pr/Ph value of 6.94, indicating that the sedimentary environment was strongly oxidizing, which is consistent with the sedimentary environment of the Pinghu Formation source rocks. The main carbon peak occurs at nC_{23}, and the sedimentary organic matter was mainly derived from higher plants (Fig. 2, Table 1).

Light hydrocarbon compounds are widely used for hydrocarbon–source correlation (Chai et al. 2020; Hu et al. 2014; Mango 1997; Odden et al. 1998; Hu et al. 2008). Leythaeuser et al. (1979) argued that the light hydrocarbon components of crude oil derived from sapropel-type parent source rocks are rich in n-alkanes, while those derived from terrestrial parent source rocks are rich in naphthenes. Snowdon and Powell (1982) also pointed out that crude oil rich in naphthenes is an important feature of terrestrial parent materials. In terms of the relative compositions of the C_5–C_7 n-alkanes, isoparaffins, and naphthenes in the NB-C1 crude oil samples, naphthenes are dominant, with a relative content of 76.3%, while the n-alkanes and isoparaffins have relative contents of 15.4 and 8.3%, respectively (Fig. 3). The relatively high abundance of methylcyclohexane indicates that the crude oil source rocks in this area are dominated by humic organic matter (Fig. 4). Thompson (1979) proposed that the heptane value and the isoheptane value can be used to determine the maturity of crude oil. The heptane and isoheptane values of the NB-C1 crude oil sample are 38.13 and 0.58, respectively, indicating that it is highly mature crude oil (Table 2).

The methane content of the natural gas is high in the NB-A and NB-B well areas in the Ningbo Tectonic Zone, exhibiting

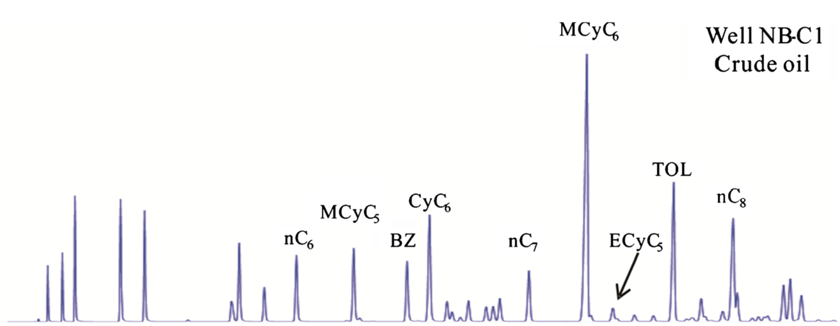


Fig. 3 Gas chromatogram of light hydrocarbon composition of crude oil from NB-C1 well

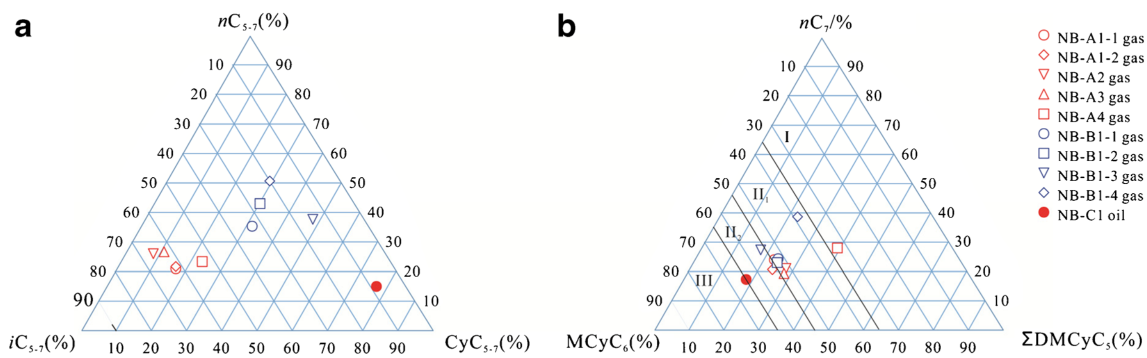


Fig. 4 (**a**) C_5–C_7 compound compositions and (**b**) C_7 light hydrocarbon compositions of the crude oil and natural gas in the Ningbo Tectonic Zone

the characteristics of dry gas. This is also one of the significant differences between the natural gas in this area and that in the other areas of the Xihu Sag. The methane in the natural gas and its homologous isotopes are characterized by a positive distribution pattern of $\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3 < \delta^{13}C_4$, indicating that it is organic gas. The ethane in the natural gas has a heavy carbon isotopic composition of greater than −28.0‰, which is characteristic of coal-type gas (Fig. 5). In addition, it has Ro values ranging from 1.42 to 1.73 (Stahl 1977), indicating that it is highly mature natural gas, similar to the origin of the oil described in the foregoing.

Oil-gas-rock correlation

The carbon isotopes of the ethane in the natural gas, the whole oil carbon isotopes of the crude oil, and the carbon isotopes of the kerogen in the different lithologies in the study area were statistically analyzed and correlated. It was found that the carbon isotopes of the ethane in the natural gas and crude oil in the Ningbo Tectonic Zone are significantly heavier than in other areas, and both exhibit the characteristics of coal-

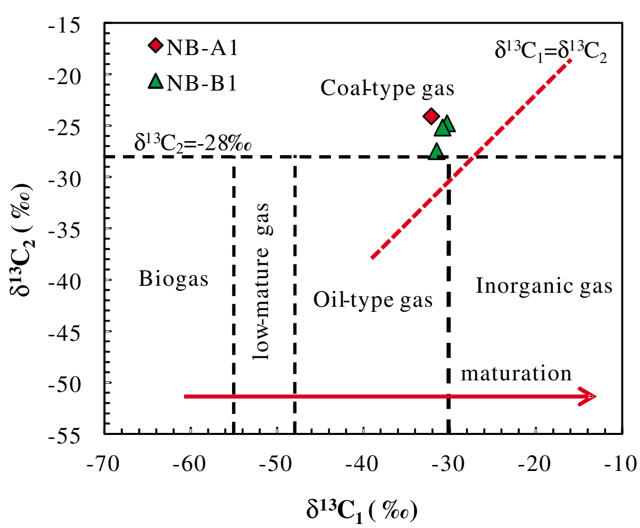


Fig. 5 $\delta^{13}C_1$–$\delta^{13}C_2$ characteristics of the natural gas in the NB-A and NB-B well area

measure strata. Previous studies have shown that there is a good relationship between the crude oil and the Pinghu Formation source rocks (Su et al. 2018; Zhu et al. 2020; Cheng et al. 2020); whereas currently, there are few studies on the source of the natural gas in this area. This study aims to identify the source of the natural gas based on the light hydrocarbon fingerprint correlation of the oil-gas-rock.

Based on the characteristics of the C_5–C_7 alkanes, the contents of the C_5–C_7 naphthenes in the NB-A and NB-B natural gas are lower than those of the NB-C1 crude oil samples, indicating that the oil and gas may have different sources. In addition, the higher abundance of C_5–C_7 naphthenes in the NB-B gases (Fig. 4(a)) indicates that the NB-B gases had a larger terrestrial organic matter input than the NB-A gases. Among the light hydrocarbon C_7 compounds, n-heptane (nC_7), methylcyclohexane ($MCyC_6$), and dimethylcyclopentane ($MCyC_6$) have different organic matter sources, which are closely related to the type of crude oil parent material. N-heptane is mainly derived from algae and bacteria. Methylcyclohexane is mainly derived from higher plant lignin, cellulose, and sugar, and it has stable thermodynamic properties and is a good indicator of the biogenic origin. Dimethylcyclopentane is mainly derived from the lipid compounds of aquatic organisms. As can be seen from Fig. 6, the methylcyclohexane content of the crude oil is relatively high, indicating that it was mainly derived from higher plants; while the methylcyclohexane content of the NB-B natural gas is relatively low, indicating that its source may be different from that of the NB-C crude oil.

The light hydrocarbon components of the natural gas in the study area do not contain benzene or toluene. The source rocks and natural gas light hydrocarbons were correlated using the relevant gas source correlation parameters, including iC_5/nC_5, methylcyclohexane/n-heptane, methylhexane/n-heptane, the methylcyclohexane index, the isoheptane index, and the n-heptane index. The results show that in the study area, the characteristics of the crude oil are similar to those of the Pinghu Formation source rocks. Based on the source rock light hydrocarbon parameter diagram and the analysis of the light hydrocarbon parameters of the natural gas in the NB-A

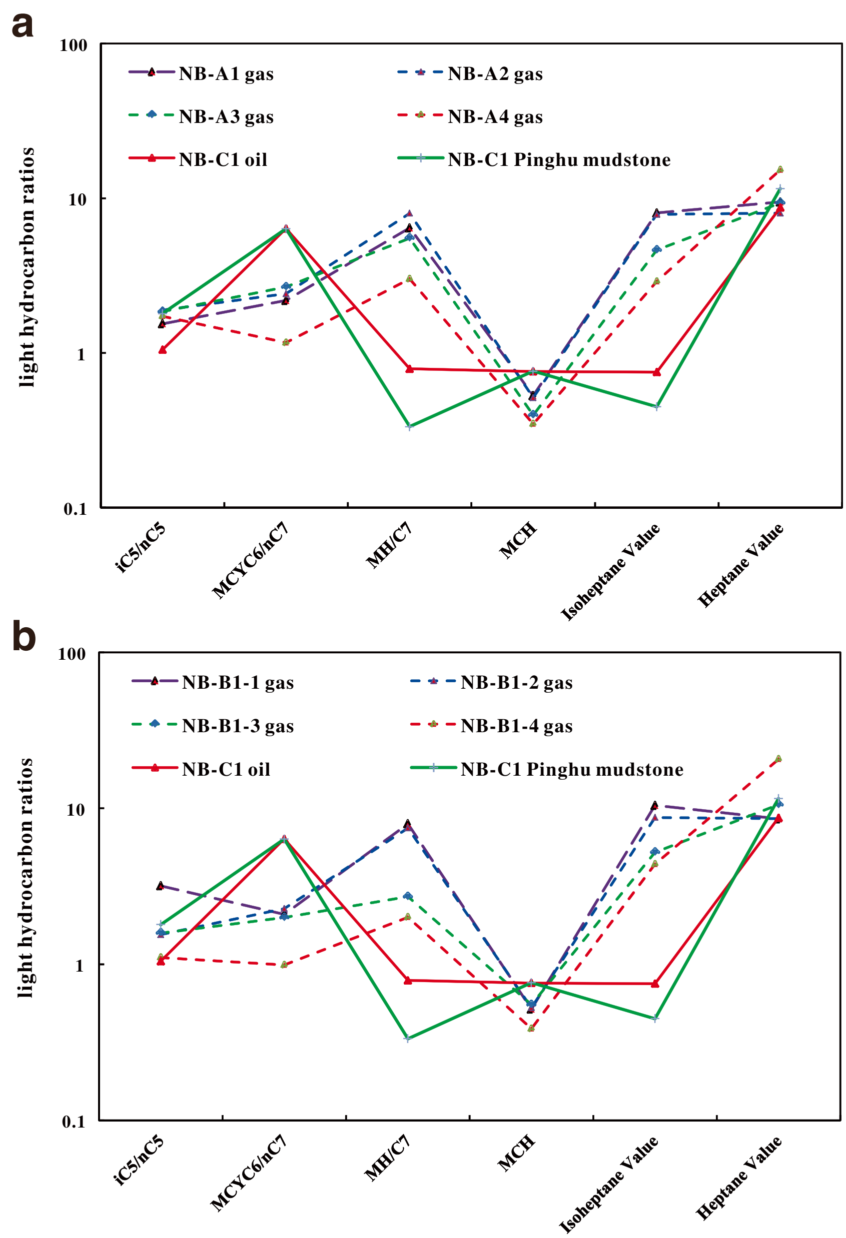


Fig. 6 Correlation between the light hydrocarbon parameters of the natural gas and those of the crude oil and source rocks of (**a**) NB-A and (**b**) NB-B

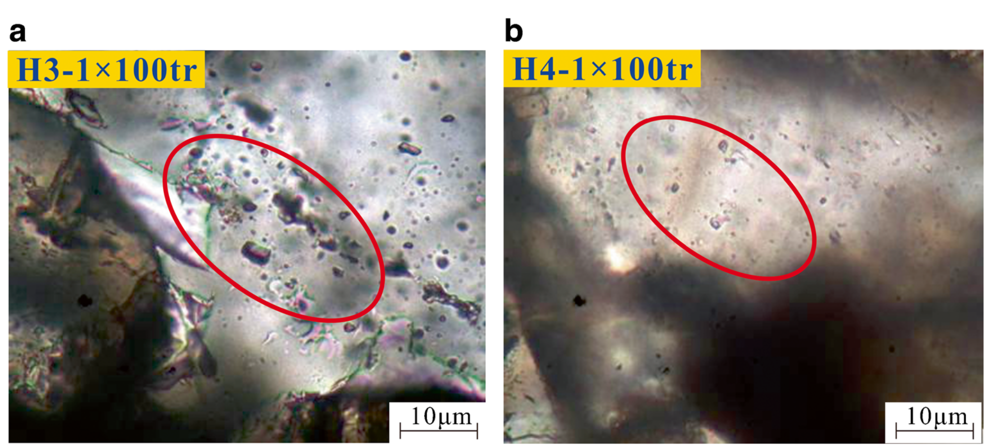
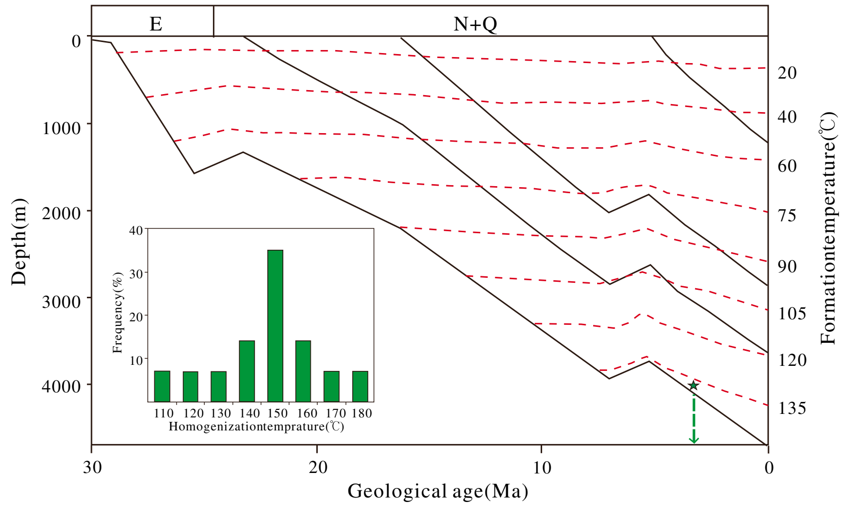


Fig. 7 Gaseous inclusions in (**a**) H3 sandstones and oil inclusions in (**b**) H4 sandstones from well NB-A1

Fig. 8 Determination of the accumulation period of well NB-A1 based on the homogenization temperature of the fluid inclusions and considering the burial history and thermal history of a single well



and NB-B well areas, it was found that the ratios of the light hydrocarbon parameters of the natural gas are not similar to those of the Pinghu Formation source rocks. In particular, the methylcyclohexane/n-heptane, methylhexane/n-heptane, and the isoheptane index are significantly different, indicating that the relationship between the natural gas and the source rocks is not significant, and the natural gas may have been derived from the source rocks in the deeper horizons of the Pinghu Formation, perhaps mostly from the Baoshi Formation. Like the Pinghu Formation, the Baoshi Formation is predominantly composed of cyclic intercalations of mudstone, sandstone, and coal seams (Zhang et al. 2020; Ding et al. 2021).

Natural gas accumulation period

In the thin section analysis, the thin sections exhibiting diagenesis were selected for inclusion analysis. Finally, two sandstone samples from well NB-A1 were selected, and the inclusion thin sections were ground to determine the homogenization temperatures of the inclusions in the upper members (H3 and H4) of the Huagang Formation. Raman spectroscopy analysis revealed that the sandstone samples contained small amounts of pure gas phase inclusions (CH_4 component), blue fluorescent oil inclusions, and hydrocarbon-bearing brine inclusions, which were distributed along the microfractures formed after the diagenesis of the quartz grains (Fig. 7). Among them, the hydrocarbon-bearing brine inclusions were mainly elliptical and oblong, and the size of a single inclusion was primarily 3–8 μm. Their gas-liquid ratio was generally less than 5%. Their homogenization temperature was 112.4°C, which corresponds to approximately 11.0 Ma in the paleogeothermal history, indicating that the natural gas charging occurred in the early deposition period of the Liulang Formation. The brine inclusions have a relatively wide homogenization temperature distribution range of 120°C to 180°C, with a main peak between 140°C and 150°C. Considering the stratigraphic burial history and temperature evolution history of this area (Yang et al. 2004; Shen et al. 2020; Wang et al. 2020), the homogenization temperature distribution interval of the fluid inclusions in the reservoir in the upper member of the Huagang Formation in this area correspond to 3.5–0 Ma in the paleogeothermal history, that is, the natural gas in the upper member of the Huagang Formation in well NB-A1 has been charged since the deposition of the Santan Formation (Fig. 8).

Conclusions

The natural gas in the Ningbo Tectonic Zone in the Xihu Sag exhibits the characteristics of dry gas. The C_7 light hydrocarbons are predominantly methylcyclohexane, the n-heptane and dimethylcyclopentane contents are relatively low, and the ethane's carbon isotopic composition is greater than −28.0‰, indicating that the natural gas was derived from humic parent material and is coal-type gas. The natural gas and crude oil in the NB-A and NB-B well areas have reached a high maturity level, but the ratios of the light hydrocarbon parameters of the natural gas are significantly different from those of the crude oil and the Pinghu Formation source rocks. This indicates that the relationship between the natural gas and the Pinghu Formation source rocks is not significant, and the natural gas may have been derived from the source rocks in the deeper horizons of the Pinghu Formation. The homogenization temperature distribution interval of the fluid inclusions in the reservoir in the upper member of the NB-A1 Huagang Formation is mainly 140–150°C, which corresponds to 3.5–0 Ma in the paleogeothermal history. This indicates that the natural gas in the upper member of the Huagang Formation in this area has been charged since the deposition of the Santan Formation.

Author's contribution D. H.: conceptualization, methodology, writing, review, and editing. Y. T.: supervision, project administration, and funding acquisition. All of the authors have read and agreed to the published version of the manuscript.

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Declarations

Conflict of interest The authors declare that there are no conflicts of interest.

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