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Multiple origins of the Paleogene natural gases and effects of secondary alteration in Liaohe Basin, northeast China: Insights from the molecular and stable isotopic compositions

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ABSTRACT

The molecular and isotopic (C and H) compositions of Paleogene [Shahejie Formation ($E_{2-3}s$) and Dongying Formation (E_3d)] natural gases from 11 new gas samples as well as the data of 71 gases published from previous studies in the Liaohe Basin were analyzed, and the results were considered in combination with geochemical and distribution characteristics of the source rocks. The genetic type, gas–source correlation and secondary alteration of the natural gases were studied in detail, and the source rock maturities of the gases were estimated. The following conclusions were reached: (1) with the exception of dry gases in the major part of the third member of the Shahejie Fm. (E_2s^3) reservoirs, the gases in other reservoirs are mainly wet; (2) variable carbon and hydrogen isotopes of the alkane gases are observed in different reservoirs of the basin with $\delta^{13}C_1$ values ranging from −60.7‰ to −34.2‰, $\delta^{13}C_2$ from −47.3‰ to −22.3‰, $\delta^2H_{CH_4}$ from −261‰ to −188‰, and $\delta^2H_{C_2H_6}$ from −269‰ to −148‰; (3) the natural gases of the basin are of multiple origins, and mixing of thermogenic and microbial is observed — the mixing ratio in the first member of the Shahejie Fm. (E_3s^1) is the highest among all the reservoirs; (4) gases of different reservoirs suffered varying extents of biodegradation, which caused partial reversal of the carbon isotope series of methane and its homologues ($\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3 > \delta^{13}C_4$); the reversal of the gases in the shallow reservoirs (E_3s^1 and E_3d) is greater, while those in the deep reservoir (E_2s^3) is less; (5) humic natural gases of the ED were mainly generated by source rocks at early maturity, while the sapropelic ones of the ED and WD were generated during the mature and high mature stages; (6) both the source rocks of E_2s^3 and E_2s^4 strata could contribute to the gas reservoirs of E_2s^3 and E_2s^1 in the west depression. Sapropelic gases in east depression were generated by E_2s^3 source rocks, while the other gases reservoired in the E_2s^3, E_3s^1, and E_3d strata are probably sourced from the source rock strata of E_2s^3 and E_3s^{1-2}.

1. Introduction

Natural gases have two genetic types: microbial or thermogenic. Microbial gas, which is mainly methane, is a product of biogeochemical processes associated with the remineralisation of labile organic matter at temperatures <80 °C by communities of microbes (Whiticar et al., 1986; Schoell, 1988; Whiticar, 1999; Huang and Larter, 2014). Microbial gas is characterized by a high dryness coefficient with $C_1/C_{1-5} > 0.95$ and is enriched in ^{12}C with $\delta^{13}C_1 < -55$‰, and it accounts for >20% of the world's gas (Rice and Claypool, 1981; Rice, 1993) and is considered to be one of the most important gas sources of gas hydrate (Collett, 2002). Thermogenic gas is generated by the degradation or cracking of organic matter (kerogen and oil) under high thermal stress (Stahl, 1977; Schoell, 1983; Dai et al., 1992). Thermogenic gas can be divided into two types: humic gas (generated by humic organic matter with kerogen type III) and sapropelic gas (generated by sapropelic organic matter with kerogen type I–II). The $\delta^{13}C_1$ value of thermogenic gas is often larger than −50‰. When generated by a low maturity source rock, thermogenic gas is typically rich in heavy alkane gases (C_{2-5}); when generated by high-maturity source rocks, it has a high dryness coefficient. The carbon isotope values of the alkane gases of humic gas indicate enrichment in ^{13}C compared with those of sapropelic gases of similar maturity (Stahl, 1977; Dai et al., 1992; Galimov, 2006). The molecular and isotopic values of the gases can be affected by secondary alteration, including mixing, migration and biodegradation (Schoell, 1980, 1983; James, 1990; Dai et al., 1992; Whiticar, 1999; Martini et al., 2008; Hao et al., 2008; Osborn and McIntosh, 2010; Liu et al., 2013). Thus, isotopic values must be used to reliably identify the origins, sources and maturities of the gases.

Many previous works have studied the genetic types of the natural gases in the Liaohe Basin, and various genetic types (e.g. microbial gas,

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sapropelic gas, humic gas and mixed gas) have been identified (Xu et al., 1993; Liu et al., 1993; Zhang et al., 2002; Li et al., 2011; Huang and Larter, 2014; Li et al., 2015). Many of these previous studies focused on one oil and gas field or a local study area in the Liaohe Basin. However, the geochemical characteristics of the gases throughout the entire basin have not been deeply studied, and little has been reported on the effects of biodegradation, migration and mixing on the gases produced in different reservoirs of different depressions. In this study, the molecular and stable isotopic compositions of the gases of the entire basin were studied comprehensively in combination with the Paleogene geological characteristics to determine the origins and sources of the gases in different reservoirs and depressions along with the effects of secondary alteration (e.g. biodegradation, migration and mixing). The findings of this work can significantly improve the understanding of the petroleum system and are beneficial to petroleum exploration in the Paleogene strata of the Liaohe Basin.

2. Geological setting

The geological setting of the petroleum systems of the Liaohe Basin has been summarized by numerous authors (e.g., Koopmans et al., 2002; Tuo et al., 2003; Fuhrmann et al., 2004; Huang et al., 2004; Hu et al., 2005; Li et al., 2013; Gu, 2016). The Liaohe Basin, located in northeast China, is one of the secondary tectonic elements of the Bohai Bay Basin groups (Fig. 1a). The Liaohe Basin is bounded by several structural elements: the Yanliao fold belt to the west, the Inner Mongolia axis to the north, the Liaodong anticline to the east, and the Liaodong Bay and Bohai Sea to the south (Fig. 1b). The basin is a Cenozoic rift basin developed after Mesozoic rifting and spans approximately 65 km in width and 470 km in length (Hu et al., 2005). The basin is subdivided into six structural units: the western uplift, central uplift, western depression (WD), eastern depression (ED), and the da'mintun depression. The WD and ED are separated by the central uplift (Fig. 1b). The three depressions were mostly isolated from each other during their development, and the tectonic and sedimentary differences among them caused the differences in their petroleum systems (Xu et al., 1993).

The thickness of the Cenozoic sedimentary layer is primarily 3000–4000 m, although some deep parts contain up to 6000 m of sediments. The Paleogene sediments mainly consist of clastic rocks with a few carbonates and volcanic horizons (Fig. 2). The Paleogene contains three

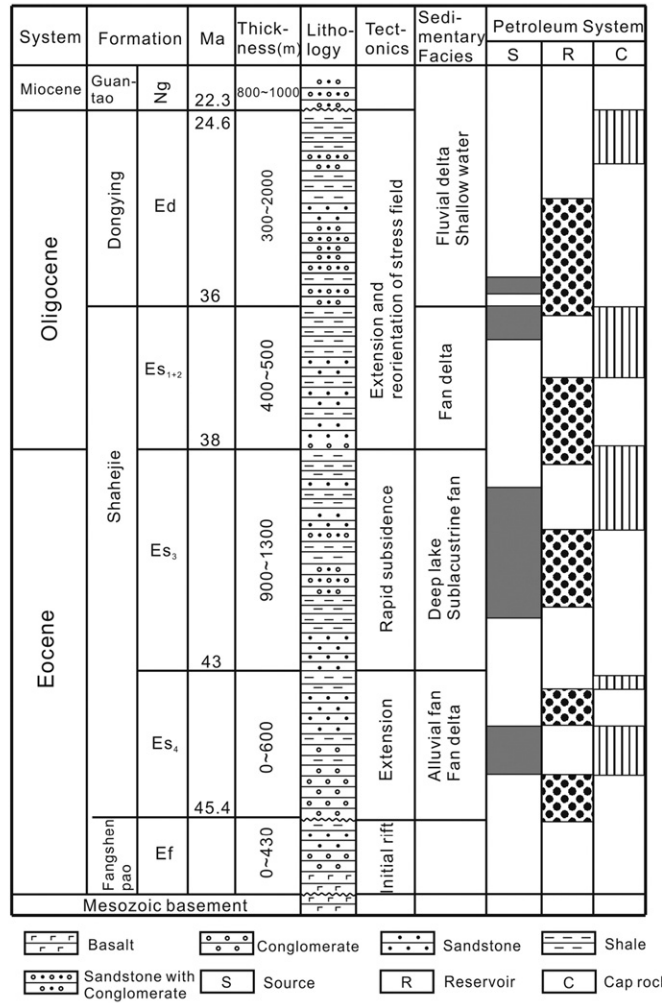

Fig. 2. Stratigraphic column and the petroleum system for the Liaohe basin. (Modified from Hu et al., 2005).

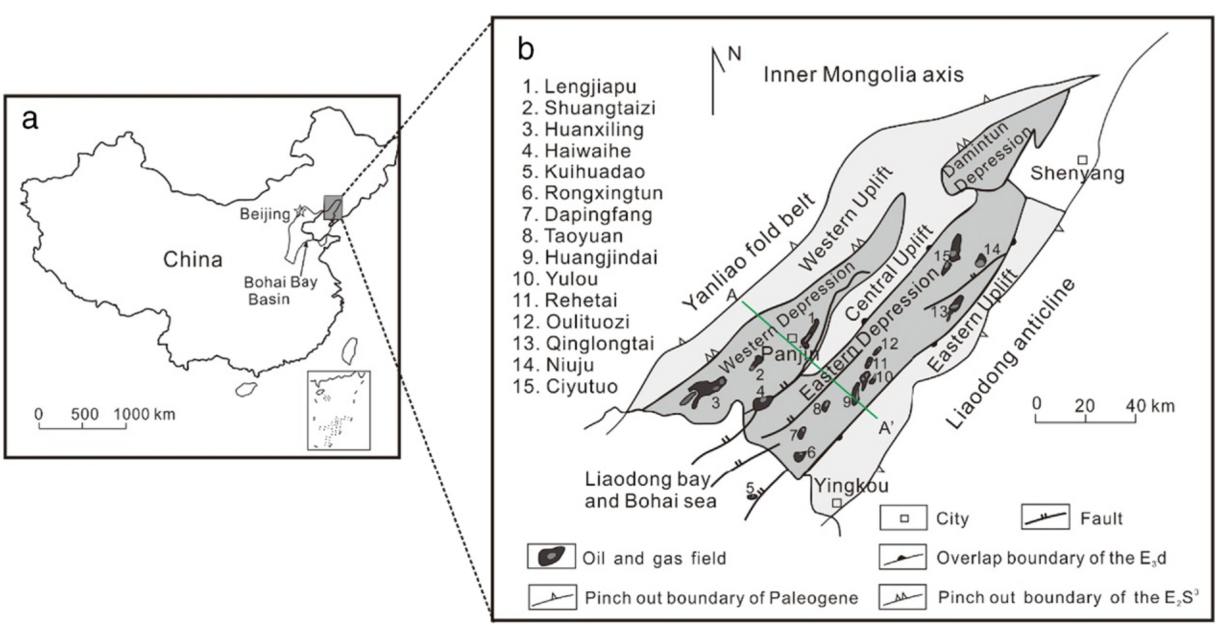

Fig. 1. Location map (a), structural subdivision and the oil-gas field distribution (b) of the Liaohe Basin. (After Zhai, 1991).