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Stable carbon isotopes of coal-derived gases sourced from the Mesozoic coal measures in China

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ABSTRACT

Coal-derived, large scale gas fields derived from the Mesozoic coal measures in China are mainly distributed in the Middle–Lower Jurassic coal measures in the Tarim, Junggar and Turpan–Hami basins in northwest China, and the Upper Triassic Xujiahe Formation coal measure in Sichuan Basin, central China. In 2011, the annual production was $21.6 \times 10^9 \text{ m}^3$ and the proved geological reserves were $2485 \times 10^9 \text{ m}^3$, accounting for 21% and 30% of the total in China, respectively. Based on analyses of gas composition and stable carbon isotopes ratios of 203 samples and stable carbon isotopes of 102 CO_2 samples, the following conclusions were made. (a) Based on diagnostic plots using the stable carbon isotopic and molecular composition of gas samples, alkane gas from the Mesozoic coal measures in China is shown to be coal-derived. (b) According to the $\delta^{13}\text{C}_2$ vs. C_2H_6 plot of a great number of oil-derived and coal-derived gases in China, it is concluded that gases with $\delta^{13}\text{C}_2 > -28.5\text{‰}$ are coal-derived and those with $\delta^{13}\text{C}_2 < -28.5\text{‰}$ are oil-derived in most cases. (c) Among the natural gases from the Mesozoic coal measures in China, primary coal-derived gases with normal carbon isotopic distribution pattern among the C_1 – C_4 alkanes (i.e. $\delta^{13}\text{C}_1 < \delta^{13}\text{C}_2 < \delta^{13}\text{C}_3 < \delta^{13}\text{C}_4$) are dominant. (d) Carbon isotopic pattern reversal mainly results from the mixing of coal-derived gases having different maturities but the same source and secondly from microbial oxidation of propane (e.g. Mu 3 and Mu 4 wells in the Gumudi gas field, Junggar Basin). (e) CO_2 in the coal-derived gases from the Mesozoic coal measures in China has both biogenic and abiogenic origins. The biogenic origin is dominant and the abiogenic CO_2 is mainly found in the Kuqa Depression in the Tarim Basin and western Sichuan Basin. (f) Isotopic differences between heavy hydrocarbon gases and methane become less with increasing maturity.

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1. Introduction

Mesozoic coal measures are well developed in China and form a number of large scale coal-derived gas fields in some areas such as the Tarim, Junggar and Turpan–Hami basins in northwest China where coal-derived gases are sourced from the Early–Middle Jurassic coal measures, and the Sichuan Basin in central China where gases are derived from the Late Triassic Xujiahe Formation (Fig. 1). Natural gases sourced from the Mesozoic coal measures have played a major role in the recent rapid development of the natural gas industry in China. The annual production in 2011 of gases sourced from the Mesozoic coal measures was $21.6 \times 10^9 \text{ m}^3$, which accounts for 21% of the total gas produced. The proven reserves of these gases are $2485 \times 10^9 \text{ m}^3$, which accounts for 30% of the total. Therefore, studies on their carbon iso-

topes will have great significance not only for the natural gas industry, but also on gas-source correlation.

2. Study area

The Early–Middle Jurassic coal measures are widely developed in the Tarim, Junggar and Turpan–Hami basins in northwest China. Their coal resources account for 60% of the total in China and they are considered to be good gas source rocks (Dai, 1986).

2.1. Tarim Basin

The Tarim Basin has an area of $560 \times 10^3 \text{ km}^2$ and half of it has Early–Middle Jurassic coal measures. The related gas fields are mainly found in the Kuqa and southwest Tarim Depressions (Fig. 2).

The Kuqa Depression, which has an area of 28,500 km^2 , is dominated by Mesozoic sediments with thicknesses up to 10,000 m (Liang et al., 2003). The distribution area of Early–Middle Jurassic

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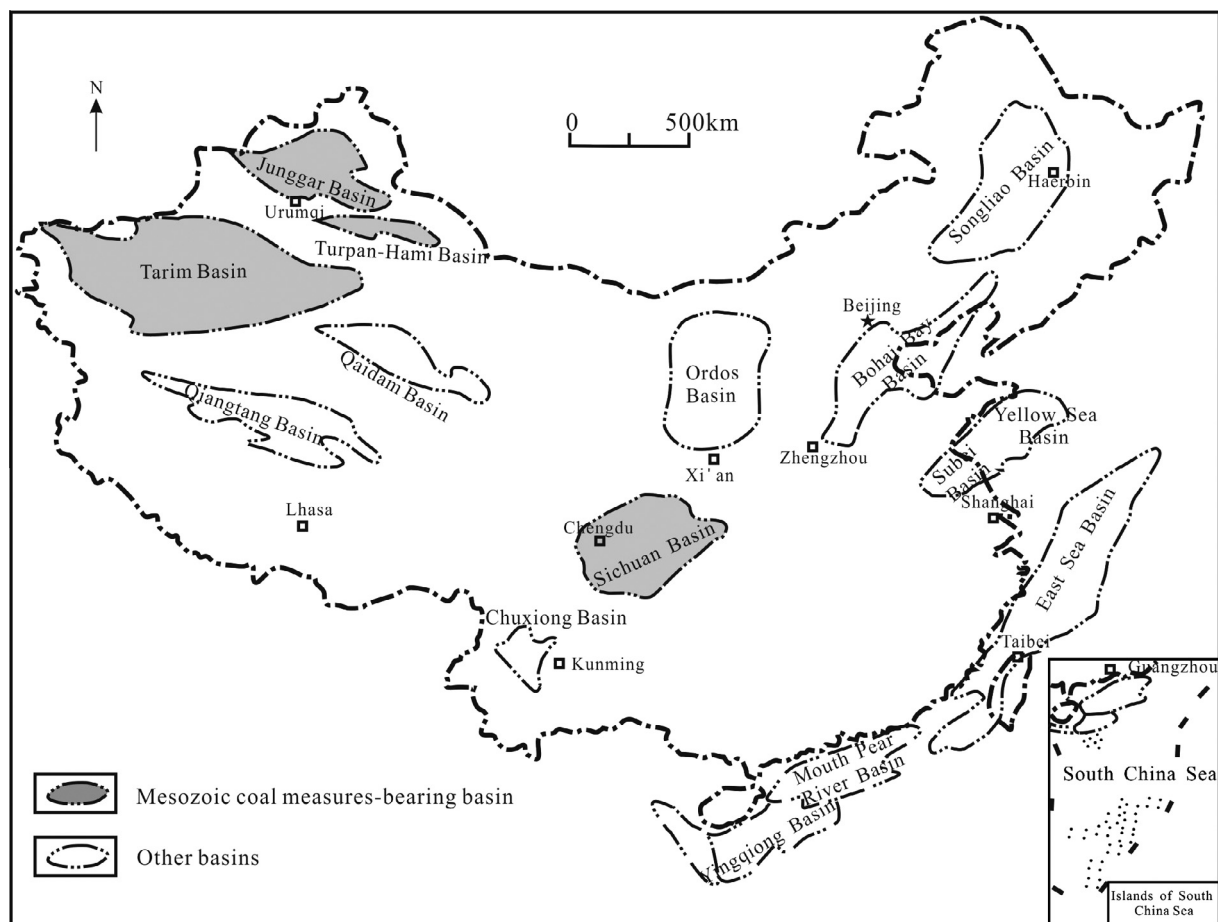


Fig. 1. Location map of basins with coal-derived gas fields sourced from Mesozoic coal measures in China.

(including some Late Triassic) coal measures is $> 10,000 \text{ km}^2$, and the thickness can be up to 1000 m. The organic carbon content (TOC) of dark shale, carbonaceous mudstone and coal is ca. 6%, 40% and 93%, respectively. The organic matter is dominantly humic (Jia et al., 2002). Based on the hydrocarbon generation index as shown in Table 1 (Dai et al., 2009a), the Middle–Late Jurassic has considerable gas generation potential. The Kuqa Depression has three sets of reservoir caps from the bottom to top: (a) combination of coal measures, sandstones and pebbly sandstones of the Ahe and Yangxia formations; (b) combination of Lower Tertiary gypsum–salt rock and gypsum mudstone, Lower Tertiary sandy conglomerate till of the Cretaceous Bashijiqike Formation (most gas fields were found in this combination) e.g., Kela2 gas field which has the greatest gas reserves in China; (c) Combination of gypsum mudstone, siltstone and pebbly sandstone of the Upper Tertiary Jidike Formation. Gas fields such as Tuziluoke gas field have been found in this combination. The Tarim Basin is the most gas enriched basin in China and the Kuqa Depression is its main gas producing area with the Kela 2 gas field with annual gas production more than $10 \times 10^9 \text{ m}^3$ ($11 \times 10^9 \text{ m}^3$ in 2007) being the first discovery in China. Table 2 shows geochemical data for gases from the main gas fields in the Kuqa Depression (Fig. 2).

The southwest Tarim Depression has an area of $150 \times 10^3 \text{ km}^2$. The terrigenous Jurassic strata were mainly developed along the southwest margin (Fig. 2) and in angle unconformity contact with other different Mesozoic strata; most of the rest are uplift areas. The Upper Jurassic Kuzigongsu Formation consists of varicolored sandy conglomerate and mudstone which is absent from some local areas. The Middle Jurassic Yangye and Taerga formations

consist of fluvial swamp and lacustrine mudstone interbedded with coal beds or black mudstones. These are considered to be good gas source rocks based on the average TOC value of 1.79% (181 samples) (Dong and Xiao, 1998). The Lower Jurassic Kangsu Formation consists of fine-coarse sandstone, pebbly sandstone, mudstone, shale and coal beds. It is the second most important gas source rock. The Lower Jurassic Shalitashi Formation consists mainly of alluvial fan sandy conglomerate. The Jurassic strata have thickness from 200–2000 m. At present, only three gas fields with generation at depth accumulation in upper reservoirs have been found (Fig. 2). Table 3 shows the geochemical data for these natural gases.

2.2. Junggar Basin

The Early–Middle Jurassic coal-bearing strata are widely distributed in the Junggar Basin, which has an area of $130 \times 10^3 \text{ km}^2$. Both the depression and deposition centers are located in the piedmont area of northern Tianshan Mountain in the southern basin (Fig. 2). It is also an area where the Early–Middle Jurassic coal measures are most developed in northwest China. The thickness of the coal beds ranges from 30–167 m, accounting for 5–10% of total coal measures, i.e., coals are well developed in the Lower Jurassic Badaowan (J₁b) and Middle Jurassic Xishanyao (J₂x) formations (Fig. 3) (Dai et al., 1997). The coal measures have dark mudstones varying in thickness from 100–500 m and the organic matter is characterized by type III (Humic)–II₂ kerogen (i.e. dominated by humic organic matter with some contribution from sapropelic organic matter). As shown in Table 1, the

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