Geochemical characteristics and genesis of natural gas in the Yan'an gas field, Ordos
Basin, China

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ABSTRACT

The Yan'an gas field in the Ordos Basin, China, was discovered in 2014 and contains reserves of up to 100 billion m³. Its occurrence in the basin contradicts the distribution model of "gas in the north, oil in the south". Upper Paleozoic natural gas in the Yan'an area is dominated by methane, with an average dryness coefficient (C_1/C_{1-4} , %) of 99.5% ($\delta^{13}C_1 = -30.8$ to -27.5‰), with minor amounts of $CO_2 = 3.8\%$, $N_2 = 0.5\%$, and ethane ($\delta^{13}C_2 = -37.2$ to -30.5‰) and traces of higher hydrocarbons. The gas is predominantly coal-derived originating from Carboniferous–Permian humic hydrocarbon source rocks and minor oil-type gas from sapropelic source rocks. The carbon isotope series includes reversals, with $\delta^{13}C_2 < \delta^{13}C_1$ and rare $\delta^{13}C_3 < \delta^{13}C_2 < \delta^{13}C_1$. The enriched $\delta^{13}C_1$ value is a result of the extremely high thermal maturity, while the depleted $\delta^{13}C_2$ value cannot be explained solely by the simple addition of oil-type gas. The complex carbon isotope reversals result from the combined effects of several secondary reactions under extremely high thermal maturity.

Keywords: Carbon isotope values, Light hydrocarbon, Coal-derived gas, Carboniferous-Permian, Yan'an gas field, Ordos Basin

1. Introduction

The Ordos Basin is a petroliferous basin with Paleozoic strata occurring over an area of more than 250,000 km². The basin has the largest proven reserves of natural gas, the largest annual output, and the most stable tectonics in China (Liu et al., 2009, 2015; Dai et al., 2014). The Upper Paleozoic stratum is a tight-sand gas pool, where the widely distributed coal-bearing rocks are superimposed with sandstone reservoirs. Tight sand gas accumulations were formed over a large area as the coal-derived natural gas migrated only short distances for efficient trapping and accumulation (Yang and Liu, 2014).

Large-scale gas exploration in the Ordos Basin started in the 1980s when the exploration strategy evolved from targeting structural to lithological traps. Ordovician paleo-weathered crust gas fields, such as the Jingbian gas field, were discovered. The exploration focus moved from Lower Paleozoic carbonate traps to Upper Paleozoic clastic lithological traps in the middle of the 1990s and some clastic lithological gas reservoirs (such as the Yulin gas field) were discovered. Since 2000, exploration has moved to the Upper Paleozoic tight sand gas reservoirs and several (such as the Sulige gas field) were discovered (Yang et al., 2012). The annual average growth of natural gas reserves has exceeded $150 \times 10^9 \text{ m}^3$ in the last 10 years. Since 2007, newly confirmed reserves exceeded $500 \times 10^9 \text{ m}^3$ per year (Yang and Liu, 2014). Large gas fields such as Jingbian, Sulige, Yulin, Daniudi, Shenmu, and Zizhou, occur in the northern parts of the basin and define a regional paradigm for the hydrocarbon distribution of “Paleozoic gas in the north, Mesozoic oil in the south” (Dai et al., 2005;

Li et al., 2005; Zhao et al., 2014). With the exception of the Jingbian gas field, for which the reservoir comprises Ordovician carbonate rocks, the large gas field reservoirs consist of Carboniferous–Permian sandstone that produce coal-derived gas (Dai et al., 2005, 2014; Li et al., 2005) and minor oil-type gas. Since 2014, exploration has discovered economic gas flows in several Upper Paleozoic successions in the southeastern Ordos Basin that do not conform to the regional hydrocarbon distribution. Proven reserves had been discovered in the Yan'an gas field, including the YQ2-Y128, Y113-Y133 and Y353-Y145 well areas, with an initial cumulative proven gas reserve of $337.4 \times 10^9 \text{ m}^3$.

The genesis of Ordovician gas in the Jingbian field in the southern Ordos Basin is disputed. The carbon isotope composition of ethane can be used as an indicator of parent material (Dai et al., 2016). Oil-derived gas is typically characterized by $\delta^{13}\text{C}_2 = < -28$ to -29‰ , while the $\delta^{13}\text{C}_2$ value of coal-derived gas is $> -28\text{‰}$ (Dai et al., 1992, 2014). Ethane in the Jingbian gas field is unusually depleted and $\delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$. Some studies have suggested that it is a mixture of Upper Paleozoic coal-derived gas and Ordovician oil-type gas, while others have proposed that it resulted from the injection of an oil-type gas generated from Carboniferous–Permian marine limestones (Dai et al., 1992, 2014; Zou et al., 2007; Hu et al., 2008).

This study presents the first analysis and discussion of the gas composition, carbon and hydrogen isotope characteristics, light hydrocarbon composition and distribution, genetic type, and carbon isotopic reversal phenomena of the Yan'an gas field. These new data provide a set of reference characteristics and details of unusual aspects of

natural gas under over-mature conditions in the Ordos Basin. Since the Lower Paleozoic strata of the Yan'an gas field have not been drilled extensively, the hydrocarbon potential of the source rocks cannot be studied directly sufficiently and can only be inferred from the gas geochemistry.

2. Geological setting

The Yan'an gas field is located in the southern-central part of the Yishan Slope (a first-order tectonic unit) of the Ordos Basin, in eastern Yan'an, Yanchang, and southern Yanchuan counties (Fig. 1). It is bounded by the Yulin–Mizhi gas fields in the north (Zone II), and overlaps the lower Paleozoic gas zone (O_{1m5}) in the south of the Jingbian gas field (Zone III). The gas-producing units of this gas field comprise the Carboniferous Benxi Formation (C_{2b}), middle Permian Xiashihezi Formation (P_{2h}), and Shanxi Formation sandstones (P_{1s}; Zhao et al., 2014; Li et al., 2015). The Benxi Formation is composed of tidal flat–barrier island deposits; the reservoir rocks comprise a single barrier island sand bar and sand flat, with a thickness of 2–10 m, which occurs in the southern portion of the field. The Shanxi and Xiashihezi (Member 8) formations represent a lake–delta sedimentary system; the reservoir rocks are composed of 4–20 m thick delta-front, underwater distributary channel sands, with a wide lateral distribution and vertical superposition. Porosity (4.4–5.6%) and permeability (0.6–7.1 mD) of the reservoir rocks are low (Li et al., 2015).

3. Analytical methods

3.1. Sampling

Twenty-seven gas samples were taken from the YQ2–Y128 gas region of the Yan'an gas field (Fig. 1). These were collected in high-pressure cylinders (pressure resistant up to 15 MPa) at pressures of ~5 MPa after the vessels had been repeatedly flushed and refilled.

3.2. Geochemical and stable carbon and hydrogen isotopic analyses

Measurements of gas composition and carbon and hydrogen isotopic ratios were conducted at the PetroChina Research Institute of Petroleum Exploration & Development (RIPED) in Beijing, China. A HP7890A gas chromatograph with a capillary column was used to separate single gas hydrocarbon compositions (PLOT Al₂O₃, 50 m × 0.53 mm). The gas chromatograph temperature was first set at 30 °C, where it was held for 10 min, and then raised to 180 °C at a rate of 10 °C/min.

A mass spectrometer (MS; Thermo Delta V Advantage) was used to analyze carbon isotopes. The initial temperature was set at 33 °C using a GC–C-irm-MS, then raised from 35 °C to 80 °C at a rate of 8 °C/min, and to 250 °C at a rate of 5 °C/min, where it was held for 10 min. Each sample was analyzed three times, yielding an analytical precision of ± 0.3‰. Results are reported relative to the Vienna Pee Dee Belemnite (VPDB). Hydrogen isotopes were analyzed using a Finnigan MAT 253 mass spectrometer using the GC–TC–IRMS method. The gas was separated through a chromatographic column (HP-PLOTQ column, 30 m × 0.32 mm × 20 mm).

Analytical precision was $\pm 3\%$, and H-isotope results are reported relative to Vienna Standard Mean Ocean Water.

3.3. Analysis of Light hydrocarbons

Light hydrocarbons were analyzed at the Langfang branch of RIPED using a HP5890A gas chromatograph, and a PONA capillary column ($50\text{ m} \times 0.25\text{ mm} \times 0.5\text{ }\mu\text{m}$) with helium as the carrier gas. The front of the capillary column was immersed into a liquid nitrogen trap to enrich the low content light hydrocarbon. After injecting a large volume of gas (about 15–20 ml) over a 20 min period, the cold trap was removed. The eluted light hydrocarbons were analyzed using a FID at 320 °C. The initial temperature was set at 30 °C, where it was held for 10 min, then raised to 70 °C at a rate of 1 °C/min and then to 160 °C at 3 °C/min, and finally the temperature increased from 160 °C to 270 °C at a rate of 5 °C/min, where it was held for 20 min.

Light hydrocarbon was qualitatively analyzed using an Agilent PONA gas chromatograph. The standard test mixture contained 53 compounds (from isobutane to *n*-octane). The compounds were quantified from peak areas of individual compounds.

The geochemical parameters of the Upper Paleozoic gas (II) of the Yulin–Mizhi gas fields in the northern Yan'an gas field, and the Ordovician Majiagou Formation gas (III) of the Jingbian gas field in the northwest, were selected for comparison of genetic types. Data and literature sources are listed in Tables 1 and 2.

4. Results

4.1. Natural gas components

The composition of natural gas in the Yan'an gas field is dominated by hydrocarbons (95.7%), of which methane CH₄ accounts for 89.9–97.5% (mean CH₄ = 95.2%), while ethane (C₂H₆) (0.5%) and propane contents are low (Fig. 2). The dryness coefficient (C₁/C_{1–4}, %) averages 99.5% (Table 1), typical of dry gas. Non-hydrocarbon gases include a small amount of CO₂ (1.4–9.1%) and N₂ (0.5%).

The Jingbian gas field is similar to the Yan'an gas field with respect to its dry coefficient values, and CH₄ and CO₂ contents. In the north, the Upper Paleozoic gas of the Yulin–Mizhi gas fields has a low dryness coefficient and CO₂ content (Fig. 2).

4.2. C-isotope compositions

The carbon isotope composition of methane in the Yan'an gas field has $\delta^{13}\text{C}_1$ values ranging from –30.8 to –27.5‰, more enriched than that of the Upper Paleozoic gas in the north. The $\delta^{13}\text{C}_2$ values range from –37.2 to –30.5‰ (mean $\delta^{13}\text{C}_2$ = –34.8‰), which is more depleted than that of the Yulin–Mizhi gas fields (Hu et al., 2008; Zhao et al., 2014). The propane content of the gas is very low, averaging 0.04%, and it has a $\delta^{13}\text{C}_3$ value of –37.3 to –30.4‰, which is slightly more depleted than that of the gas from the Jingbian gas field (mean $\delta^{13}\text{C}_3$ = –29.3‰; Table 1).

Alkane gases from a single source show a linear relationship between carbon number (1/n) and $\delta^{13}\text{C}_n$ and the slope change of the regression line can be used to identify whether mixing between different types of gas at different evolutionary stages

and biogas occurred, and to monitor the effects of methane seepage (Rooney et al., 1995). Upper Paleozoic gases in the eastern Ordos Basin and gas in the north are typically mature-stage and coal-derived and exhibit a linear relationship between $\delta^{13}\text{C}_n$ and $1/n$ suggesting a homogeneous accumulation of gas in this area (Fig. 3). In contrast, the carbon number and isotopic composition of gas in the Yan'an gas field show similar characteristics to those of gas from the Jingbian gas field (Fig. 3a), such as $\delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$ and a negative carbon isotope series ($\delta^{13}\text{C}_3 < \delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$; Fig. 3b–c).

A plot of $\delta^{13}\text{C}_1 - \text{C}_1 / (\text{C}_2 + \text{C}_3)$ is widely used to identify kerogen types for gas generation (Bernard et al., 1978; Whiticar, 1999). Gases in the eastern Ordos Basin show the typical characteristics of thermogenic gas (Fig. 4). The Yan'an gases, as well as Permian gases from Yulin and Mizhi gas fields, plot most closely to a more mature, Type III kerogen, suggesting that gases are derived predominantly from Permian humic hydrocarbon source rocks. The Ordovician gases of the Jingbian gas field are more variable and span the area between Type II and Type III kerogens suggesting that they are mixtures of coal-derived and oil-type gases.

Based on the model of CO_2 genesis in natural gas proposed by Dai et al. (1996), gas that forms by organic genesis contains less than 15% CO_2 and $\delta^{13}\text{C}_{\text{CO}_2} < -10\text{‰}$, while gas of inorganic origin contains $\text{CO}_2 > 60\%$ and $\delta^{13}\text{C}_{\text{CO}_2} = -8$ to -3‰ . The $\delta^{13}\text{C}_{\text{CO}_2}$ values range from -12.98 to 2.4‰ ; the $\delta^{13}\text{C}_{\text{CO}_2}$ values of 11 samples are higher than -8‰ (Fig. 5). A relatively thick limestone unit is developed in the Benxi (C_2b) and Taiyuan ($\text{P}_{1\text{t}}$) formations in the area, and it is likely that CO_2 was the product of

thermal cracking of these inorganic carbonates.

4.3. H-isotope compositions

The hydrogen isotopic compositions of natural gas are controlled by the types of organic matter, thermal maturity and the environmental conditions of the aqueous medium (Dai et al., 1992; Liu et al., 2008; Wang et al., 2015). δD_{CH_4} values of the Yan'an gas field gas range from -170 to -163‰ (mean $\delta D_{CH_4} = -166.6\text{‰}$), similar to that of the Ordovician Jingbian gas (Table 1). δD_{CH_4} values in the south of the basin are generally enriched and shows hydrogen isotope reversal ($\delta D_{CH_4} \geq \delta D_{C_2H_6}$; Fig. 6) consistent with an over-mature stage. The δD_{CH_4} values of the Yan'an gas field samples yield a relatively large variation in δD_{CH_4} and consistent with a transitional facies environment within the study area.

The Upper Paleozoic gases in the Yan'an gas field display the characteristics of gases from coal-series source rocks in the correlation diagram between δD_{CH_4} and $\delta^{13}C_1$ values (Fig. 7a). The Lower Paleozoic gases in the Jingbian gas field display similar characteristics, indicating that the methane is mainly coal-derived.

Furthermore, the Upper Paleozoic gases of the Yan'an gas field also display the trend of mixing by coal-derived and oil-type gases as a result of the wide range of $\delta^{13}C_2$ values, which is similar to the gases from the western Jianbian areas (Fig. 7b).

Hydrogen isotopic compositions of Yan'an gas field do not display a positive correlation between δD_{CH_4} and carbon isotope values or follow the maturity trend.

Rather they exhibit δD_{CH_4} and $\delta^{13}C_1$ values spanning the region for coal-derived and

oil-type gases suggesting a mixed origin.

4.4. Light hydrocarbon composition

Light hydrocarbon distribution and carbon isotope composition reflect natural gas genesis (Hu et al., 2007). The maturity and dryness coefficient of Yan'an gases are extremely high (Table 1) and the light hydrocarbon components in the gases have been subjected to extensive cracking. Their low abundance make their detection difficult. Table 2 list the light hydrocarbon data of 27 gas samples that were measured after concentration using liquid nitrogen.

C₆–C₇ hydrocarbons of the Yan'an gas field are rich in aromatic hydrocarbons (12.6%) and branched alkanes (55.4%). Isoalkanes (56.2%) and cycloalkanes (29.4%), are the major constituents of the C₅–C₇ aliphatic compounds, a characteristic of humic organic matter (Leythaeuser et al., 1979; Snowdon et al., 1982; Hu et al., 2007). Since aromatic hydrocarbons are highly soluble and vulnerable to absorption and water action during migration, a ternary chart of C₅–C₇ aliphatic compounds can be used to distinguish oil-type from coal-derived gas (Dai et al., 1992; Odden et al., 1998; Hu et al., 2007). Fig. 8 shows that the light hydrocarbons in the Yan'an gas field are characteristic of coal-derived gas and plot close to the field of Upper Paleozoic coal-derived gas in the basin.

The C₇ light hydrocarbons include *n*-heptane (*n*-C₇), methylcyclohexane (MCH, also labeled MCC₆), and dimethylcyclopentane isomers (Σ DMCP, also labeled Σ DMCC₅). *n*-Heptane is predominantly derived from algae and bacteria and is

sensitive to variations in maturity, while dimethylcyclopentanes form from the lipids of aquatic organisms and their predominance is a characteristic of light hydrocarbons in oil-type gas. Methylcyclohexane can be derived from lignin, cellulose, and components of higher plants and is a major constituent of the light hydrocarbons in coal-derived gas (Dai et al., 1992; Odden et al., 1998; Whiticar, 1999; Hu et al., 2007). Coal- and oil-type gas can be distinguished from each other by examining the relationships between $n\text{-C}_7$, MCH, and ΣDMCP in ternary space (Fig. 9). The C_7 light hydrocarbon compound of Yan'an gas is high in methylcyclohexane (48.7–88.1%), while n -heptane (5.9–44.0%) and dimethylcyclopentanes (0–23.1%) are low. The parent material has humic characteristics (Table 2), and all samples plot in the field of coal-derived gas (Fig. 9). Many C_7 components of gases are not present in the Yan'an gas field; however, they have been identified in the relatively low-maturity Sulige and Shenmu gas fields in the northern parts of the basin. The composition of light hydrocarbons in this area is significantly influenced by thermal maturity, and Yan'an gases are predominantly over-mature and plot near the aromatic curve (Fig. 10).

5. Discussion

5.1. Genesis of alkane gas

The carbon isotope composition of ethane and other hydrocarbons can be used to distinguish oil-type and coal-derived gas from each other. While this method is effective for gases derived from a single source, identifying the sources of mixed

gases is more complicated (Dai et al., 2005; Zou et al., 2007; Hu et al., 2008). Most Upper Paleozoic gases in the southern Ordos Basin, including the Yan'an gas field, have depleted $\delta^{13}\text{C}_2$ values. If genetic types of gas are defined simply by a $\delta^{13}\text{C}_2 \geq -28\text{‰}$ threshold, contradictory conclusions arise regarding the origin of gases in the Yan'an area (Chen, 2002; Zou et al., 2007; Hu et al., 2008; Yang et al., 2012). Yan'an gas shows carbon isotope reversals ($\delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$, $\delta^{13}\text{C}_3 < \delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$), indicating that it has been affected by secondary processes. In addition, the natural gas in the southern Ordos Basin (dryness coefficient = 95%) is over-mature, and methane is the dominant alkane gas (mean $\text{CH}_4 = 95\%$). The genesis of the gas cannot be modeled with confidence using ethane, propane, and other minor components; therefore, in over-mature gas the carbon isotopes of ethane do not provide reliable data for identifying the gas source (Zou et al., 2007; Yang et al., 2012).

The hydrocarbon source rocks in the Upper Paleozoic Shanxi, Taiyuan, and Benxi formations in the eastern Ordos Basin include extensive coal seams and dark mudstones, which are considered to be the gas source rocks of the Upper Paleozoic gas fields (Dai et al., 2005, 2014). In the Yan'an gas field, the coal seam is 6–18 m thick, with an organic carbon content of 60.2–86.9%. The dark mudstone unit in the Shanxi Formation is 30–120 m thick, contains Type II₂–III organic matter, has an organic carbon content of 0.1–9.4%, and a mean thermal maturity of 2.4 %Ro (Li et al., 2015), which increases towards Fuxian County in the southwest (Fig. 1). These properties indicate that the mudstone unit had great hydrocarbon-generating potential. In addition, the underlying marine–transitional facies Benxi and Taiyuan formations

source rocks also were able to generate natural gas. The dark mudstone has an organic carbon content of 0.1–9.0%, with %Ro = 2.5, and the argillaceous limestone is over-mature. The kerogen contains sapropel-type components, has an average organic carbon content of 0.9%, and its abundance of organic matter and gas-generating ability are higher than those of the Ordovician carbonate rocks (Dai et al., 2014).

Yan'an gas has extremely low $\delta^{13}\text{C}_2$ values, but the $\delta^{13}\text{C}_1$ values are closer to those of coal-derived gas in the coal seam. Natural gas maturity is related closely to gas-source correlation. The maturity of gas source rocks could be estimated with the methane carbon isotope values, based on $\delta^{13}\text{C}_1$ —%Ro empirically derived equations for different types natural gas (Dai and Qi, 1989). Based on equation for coal-derived gas (Dai and Qi, 1989), the %Ro value is 1.8–3.1 and with an average of 2.5, indicating that most samples are highly over-mature gases and conform to the actual characteristics of geochemical parameters of the Upper Paleozoic coaly source rock. However, according to the oil-type gas equations the %Ro values range from 4.6–7.4 and with an average of 6.1, which obviously do not conform to the actual parameters. Furthermore, the light hydrocarbon compositions show characteristics typical of coal-derived gas (Figs. 4, 8 and 9). On the other hand, the Carboniferous source rocks of gas field have a certain ability to generate natural gases, such as samples from wells Sh12, Sh37, Sh48 and Y175. Additionally, the hydrogen isotopic composition of Yan'an gas field shows different characteristics from the typically coal-derived or oil-type gas in the correlation diagram between $\delta\text{D}_{\text{CH}_4}$ and carbon isotope values, suggesting mixed origin (Fig. 7). Therefore, we conclude that the Yan'an gas field is

dominated by coal-derived gas sourced from Carboniferous–Permian humic rocks with minor oil-type gases from sapropelic source rocks.

5.2. Analysis of the carbon isotope reversal

Dai et al. (1992, 2014) proposed five mechanisms for carbon isotope reversal: (1) mixing of organic and inorganic gases; (2) mixing of oil-type and coal-derived gases; (3) mixing of gases from the same source during different periods, or mixing of gases from different sources during the same period; (4) oxidization of gas components by bacteria (Li et al., 2012), and (5) thermochemical sulfate reduction (TSR). Geological structures in the eastern Ordos Basin are stable and occur at depth, resulting in unsuitable conditions for inorganic gas injection and bacterial oxidation. TSR reactions would not affect the carbon isotope composition of methane, but may cause the carbon isotope of ethane to be enriched when the H₂S content is high (10%); these characteristics are not observed in the gases analyzed during the present study (Hu et al., 2007; Dai et al., 2014).

5.2.1. Enriched $\delta^{13}\text{C}_1$ values

The carbon isotope composition of methane is influenced by the degree of thermal evolution (Rooney et al., 1995; Dai et al., 2014). $\delta^{13}\text{C}_1$ values become enriched within increasing thermal evolution from the north-northeast to the south of the Ordos Basin (Hu et al., 2007; Yang et al., 2012; Zhao et al., 2014; Dai et al., 2016). The %Ro value of the Upper Paleozoic coal-rich source rocks indicates that the Yan'an gas field is in

the over-mature stage of evolution, and enriched $\delta^{13}\text{C}_1$ values are the result of thermal maturity, consistent with the fact that the maturity and $\delta^{13}\text{C}_1$ values are higher than those of the Upper Paleozoic gas fields in the north.

5.2.2. Depleted $\delta^{13}\text{C}_2$ values

Mixing of gases that came from the same source but vary in evolution degrees should not be considered as the main cause of relatively depleted ethane carbon composition. Previous studies found that $\delta^{13}\text{C}_2$ of shale gas trapped in the Shanxi Formation coal measure strata in the Ordos Basin is generally less than -29‰ (Wu et al., 2015). The $\delta^{13}\text{C}_2$ value of gases in the coal seam should be more enriched than -28‰ even in the early stages of evolution. In addition, the internal gas of source rocks should not have interacted with migrated gas (Wu et al., 2015).

The Carboniferous source rocks of Yan'an gas field had the potential to generate natural gases, and it has been proposed that the unusually depleted $\delta^{13}\text{C}_2$ value of gas in the southern part of the Ordos Basin was caused by the addition of oil-type gas formed in Ordovician or Carboniferous–Permian argillaceous limestones (Hu et al., 2008; Yang et al., 2012; Dai et al., 2014; Zhao et al., 2014). However, the significance of the mixing of coal-derived and oil-type gas can be explored further. Once mixing occurred, the $\delta^{13}\text{C}_2$ and $\delta^{13}\text{C}_3$ values of the Permian ($\text{P}_{1\text{s}}$ and $\text{P}_{2\text{h}}$) gas in the Yan'an gas field would be more enriched than those of the other sources of natural gases.

In the Yan'an gas field, the $\delta^{13}\text{C}_2$ value of the Permian natural gas (mean $\delta^{13}\text{C}_2 = -34.6\text{‰}$) is more enriched than that of gas in the Benxi Formation ($\text{C}_{2\text{b}}$), thereby

meeting the criteria of mixing, and is equivalent to that of Ordovician natural gas in the southern Jingbian gas field (mean $\delta^{13}\text{C}_2 = -34.8\text{‰}$; Dai et al., 2014; Kong et al., 2016). However, the $\delta^{13}\text{C}_3$ value of the Permian gas (mean $\delta^{13}\text{C}_3 = -33.2\text{‰}$) is more enriched than that of gas in the Benxi Formation, but more depleted than that of gas in the Jingbian gas field (mean $\delta^{13}\text{C}_3 = -30.2\text{‰}$; Kong et al., 2016).

Widespread reversals ($\delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$, $\delta^{13}\text{C}_3 < \delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$) exist in Yan'an gas field, which are rarely seen in natural gases (Fuex, 1977). According to the results of mixed sources calculations from previous research (Dai et al., 2004), the reversal of $\delta^{13}\text{C}_1$ and $\delta^{13}\text{C}_2$ cannot happen until the volumetric ratio of coal-derived and oil-type gases in the mixture reach the 20:1. Moreover, if the obvious mixing of oil-type gas mentioned above occurred, the light hydrocarbons would show significant variation. However, the characteristics of coal-derived gas was clearly shown by the light hydrocarbon composition in the Yan'an gas field and in a major portion of the Jingbian gas field (Figs. 4, 8 and 9). Thus, if the addition of oil-type gas occurred in the Yan'an gas field, the interfused oil-type gas is more likely to have been generated from the Upper Paleozoic argillaceous limestone. In addition, the unusually depleted $\delta^{13}\text{C}_2$ value cannot be explained solely by the simple addition of oil-type gas, but by a combination of complex factors.

5.3. Secondary negative carbon isotope series

Dai et al. (2016) proposed that a secondary negative carbon isotopic mechanism is responsible for the large-scale carbon isotope anomalies observed in recently

discovered over-mature thermogenic gas deposits, and defined some of the dominant controlling factors, including secondary cracking, the participation of transition metals in reactions at high temperatures, diffusive phase migration leading to fractionation, and high temperature ($> 200\text{ }^{\circ}\text{C}$). The degree of thermal evolution in the Ordos Basin gradually increases from north-northeast to south, and the sediments enter the over-mature stage near the Yan'an, Wuqi, and Qingyang areas (Hu et al., 2008; Yang et al., 2012), thereby meeting the conditions for the occurrence of secondary negative carbon isotope phenomena.

Secondary cracking refers to the over-mature stage process where shale gas results from the mixing of cracking gases from kerogen, retained oil and wet gas. Until the dry gas stage, the addition of depleted carbon isotope ethane from early stage generation, or small amounts of condensate cracking, can lead to carbon isotope reversal (Tilley et al., 2013; Xia et al., 2013). Previous studies (Zhao et al., 2014; Wu et al., 2015; Kong et al., 2016; Dai et al., 2016) have suggested that secondary cracking might also be an important contributing factor in the generation of the unusually depleted $\delta^{13}\text{C}_2$ values seen in the Shanxi Formation shale gas in the Yan'an area and found that the Upper Paleozoic gas in the basin that also show a phenomenon similar to the rollover of shale gas $\delta^{13}\text{C}_2$ with decreasing wetness. The Yan'an region was a depositional center in the late Paleozoic. It contains a thin sandstone reservoir (Li et al., 2015), has a high shale content that is of high maturity. The close association between source and reservoir rocks suggests that gases produced by the secondary cracking occurring in the shale gas system may have diffused into the

conventional reservoir (Wu et al., 2015).

Burruss and Laughrey (2010) proposed that carbon isotopic reversal in highly mature, deep basin gas in the Appalachian Basin was caused by Rayleigh fractionation and a redox reaction resulting from the occurrence of a transition metal in the presence of water at temperatures of 250–300 °C (Tilley et al., 2011). Rayleigh fractionation would have occurred during the secondary reversal stage in the shale gas, when the $\delta^{13}\text{C}_2$ value changed from depleted to enriched, becoming more enriched than $\delta^{13}\text{C}_3$ (Wu et al., 2015). The natural gas in the Yan'an gas field plots near the Rayleigh fractionation curve, but there is no clear change in the carbon isotope systematics of ethane (Fig. 11).

The diffusion and migration of natural gas occurring when a gas is over-mature can lead to the occurrence of carbon isotope fractionation and reversal (Dai et al., 2016). Natural gas in the Ordos Basin shows trends of aromatic hydrocarbon index (toluene/heptane) increase and a decrease in the saturated hydrocarbon index (heptane/methylcyclohexane). This process is consistent with the “evaporation fractionation” of crude oil, indicating the occurrence of an oil–gas separation process (Wu et al., 2015; Thomson, 1987). These processes possibly occurred in the Yan'an gas field (Fig. 12) as a result of uplift in the southern Ordos Basin and maximum burial in the Early Cretaceous. The high degree of thermal evolution meant that natural gas diffusion resulted in dissipation and the removal of large amounts of saturated hydrocarbons and small amounts of aromatic hydrocarbons. As a result, “evaporation fractionation” of the residual natural gas occurred (Wu et al., 2015).

Over-maturity or high temperatures ($> 200\text{ }^{\circ}\text{C}$) can facilitate the occurrence of secondary negative carbon isotope mechanisms (Dai et al., 2016). Some sedimentary basins, not fully entered the over-mature stage, can be affected by high temperature and hydrothermal fluids (Burruss and Laughrey, 2010; Zumberge, 2012). These basins also show the reversal phenomenon of $\delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$, and under conditions of gas formation result in a change in carbon isotopic exchange equilibrium. When the temperature exceeds $150\text{ }^{\circ}\text{C}$, $\delta^{13}\text{C}_1$ would be greater than $\delta^{13}\text{C}_2$, and when it exceeds $200\text{ }^{\circ}\text{C}$ a negative carbon isotope series ($\delta^{13}\text{C}_3 < \delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$) occurs (Vinogradov et al., 1970). The carbon isotope systematics in the Yan'an gas field show reversals of $\delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$ and rare $\delta^{13}\text{C}_3 < \delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$, which are associated with conditions of over-maturity as well as other dominant controlling factors.

6. Conclusions

Over-mature natural gas in the Yan'an gas field, southeastern Ordos Basin, contains methane (95.2%), ethane (0.5%), and negligible amounts of propane and butane. The dryness coefficient of the gas is 99.5%. Non-hydrocarbon gases include minor amounts of CO_2 of inorganic origin, and N_2 . Branched alkanes and aromatic hydrocarbons are the predominant $\text{C}_6\text{--C}_7$ light hydrocarbons. $\text{C}_5\text{--C}_7$ saturated compounds are dominated by isoalkanes and cycloalkanes. The organic matter type, carbon isotopic compositions of methane, and characteristics of light hydrocarbons are typical of coal-derived gas. The $\delta\text{D}_{\text{CH}_4}$ value of the Yan'an gas field gas ranges from -170 to -163‰ , and shows characteristics of the mixing of coal-derived and

oil-type gas. Thus, the Yan'an gas field is dominated by coal-derived gas generated from Carboniferous–Permian humic source rocks with minor oil-type gases from sapropelic source rocks

The $\delta^{13}\text{C}_1$ value of alkane gas ranges from -30.8 to -27.5% , and $\delta^{13}\text{C}_2 = -37.2$ to -30.5% , with $\delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$ and $\delta^{13}\text{C}_3 < \delta^{13}\text{C}_2 < \delta^{13}\text{C}_1$, where the enriched $\delta^{13}\text{C}_1$ value resulted from thermal maturity. The unusually depleted $\delta^{13}\text{C}_2$ value cannot be explained solely by the simple addition of oil-type gas, and complex carbon isotope reversals resulted from the combined effects of several secondary reactions under extremely high thermal maturity.

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Stratigraphy		Lithologic section	Thickness (m)	Facies	Petroleum system		
System	Formation				S	R	C
Upper Permian	Shiqianfeng (P ₁ q)		250~270	Shallow lacustrine			
Middle Permian	Shangshihezi (P ₂ sh)	(P ₂ h ₁)	120~150	Shallow lacustrine			
		(P ₂ h ₂)					
		(P ₂ h ₃)					
		(P ₂ h ₄)					
	Xiaoshihezi (P ₂ xh)	(P ₂ h ₅)	130~160	Fluvial and delta			
		(P ₂ h ₆)					
		(P ₂ h ₇)					
Lower Permian	Shanxi (P ₃ s)	(P ₃ s ₁)	90~140	Fluvial delta and swamp			
		(P ₃ s ₂)					
	Taiyuan (P ₃ t)		60~80	Restricted platform			
Upper Carboniferous	Benxi (C ₂ b)		20~60	Restricted platform			
Lower Ordovician	Majiagou (O ₂ m)		120~600	Platform evaporites			

Siltstone Sandstone Sandy mudstone Mudstone Coal bed
 Gypsum Aluminum mudstone Gas-bearing Dolomite Limestone

Fig. 1. Geological map, sampling sites of Yan'an gas field, and generalized Paleozoic stratigraphic column for the Ordos Basin (modified after Dai et al., 2014, 2016).

Fig. 2. Relative proportions of gas components of natural gas in the southeastern Ordos Basin (data from Hu et al., 2008; Dai et al., 2014; Zhao et al., 2014).

Fig. 3. Relationship between carbon number $1/n$ and $\delta^{13}\text{C}_n$ of natural gas in the eastern Ordos Basin, for: (a) Yan'an gas field, (b) Yulin–Mizhi gas fields, and (c) Jingbian gas field (data from Hu et al., 2008; Dai et al., 2014; Zhao et al., 2014).

Fig. 4. Relationship between C_1/C_{2+3} and $\delta^{13}C_1$ of natural gas in the Yan'an gas field (modified after Bernard et al., 1978; Whiticar, 1999; data from Hu et al., 2008; Dai et al., 2014; Zhao et al., 2014).

Fig. 5. Relationship between $\delta^{13}\text{C}_{\text{CO}_2}$ and CO_2 content (%) of gases in the Yan'an gas field (data for Jingbian gas field sources from Dai et al., 2014).

Fig. 6. Hydrogen isotopic series of alkanes in natural gas from the eastern Ordos Basin, for: (a) Yan'an gas field, and (b) Jingbian gas field (data for Jingbian gas field sources from Dai et al., 2014).

Fig. 7. Cross-plots of: (a) δD_{CH_4} vs $\delta^{13}C_1$ and (b) δD_{CH_4} vs $\delta^{13}C_2$ of natural gas from the Daniudi gas field (modified after Wang et al., 2015; data for the Jingbian gas field from Dai et al., 2014).

Fig. 8. Ternary diagram of C_{5-7} light hydrocarbons in natural gas from the Ordos Basin (modified after Dai et al., 1992; Hu et al., 2007; light hydrocarbon data for the Jingbian gas field and upper Paleozoic samples are from Hu et al., 2007, 2010; Dai et al., 2014).

Fig. 9. Ternary diagram showing C_7 light hydrocarbons in natural gas in the eastern Ordos Basin (modified after Dai et al., 1992; Hu et al., 2007; light hydrocarbon data for the Jingbian gas field and upper Paleozoic samples are from Hu et al., 2007, 2010; Dai et al., 2014).

Fig. 10. Heptane and isoheptane distribution in natural gas of the eastern Ordos Basin (modified after Thompson, 1983; data for upper Paleozoic gas are from Hu et al., 2007, 2010; Li et al., 2012).

Fig. 11. Relationship between $\delta^{13}\text{C}_2$ and C_2H_6 showing mixing and Rayleigh fractionation (modified after Thompson, 1987; data are from Burruss and Laughrey, 2010; Dai et al., 2014).

Fig. 12. Relationship between saturated hydrocarbon content and aromatic hydrocarbon content for identification of the evaporative fraction (modified after Thompson, 1987; data for upper Paleozoic gas are from Hu et al., 2007, 2010; Li et al., 2012; this study).

Table 1. Component and isotope properties of natural gas in the southeast of Ordos Basin

Gas field	Well	Strata	Main component, (%)					$\delta^{13}\text{C}(\text{‰})$, VPDB					$\delta\text{D}(\text{‰})$, VSMOW					Reference
			C	C	C	i-	n-	C	C	C	C	C	C	C	C	C	C	
			H ₄	H ₆	H ₈	C ₄ H ₁₀	C ₄ H ₁₀	O ₂	C ₂	H ₄	H ₆	H ₈	C ₄ H ₁₀	O ₂	H ₄	H ₆	H ₈	
Yan'an gas field	Sh 2	P _{2h}	9.68	0.73	0.09	0.02	0.06	1.41	1.07	—	—	—	—	12.9	—	—	190	This paper
	Sh 217	P _{2h}	9.63	0.62	0.05	—	—	1.27	1.76	—	—	—	—	0.2	—	—	183	
	Sh 6	P _{1s}	9.63	0.76	0.07	—	—	1.97	1.86	—	—	—	—	9.1	—	—	187	
	Sh 25	P _{1s}	9.84	0.43	0.02	—	—	1.15	1.48	—	—	—	—	—	—	—	—	
	Sh 26	P _{1s}	9.71	0.37	0.02	—	—	1.14	1.24	—	—	—	—	—	—	—	—	
	Sh 28-1	P _{1s}	9.73	0.41	0.02	—	—	1.52	1.39	—	—	—	—	—	—	—	—	
	Sh 36	P _{1s}	9.39	0.43	0.02	—	—	1.93	1.72	—	—	—	—	3.6	—	—	182	
	Sh 38	P _{1s}	9.59	0.42	0.03	—	—	1.11	1.53	—	—	—	—	1.1	—	—	185	
	Sh 56	P _{1s}	9.96	0.48	0.03	—	—	1.00	1.47	—	—	—	—	—	—	—	—	
	Sh 127	P _{1s}	9.45	0.43	0.03	—	—	1.72	1.37	—	—	—	—	0.2	—	—	184	
	Sh 207	P _{1s}	9.94	0.28	0.02	—	—	1.09	1.56	—	—	—	—	—	—	—	—	
	Sh 209	P _{1s}	9.90	0.42	0.02	—	—	1.08	1.57	—	—	—	—	—	—	—	190	
	Sh 210	P _{1s}	9.39	0.43	0.03	—	—	1.86	1.30	—	—	—	—	0.1	—	—	187	
	Sh	P _{1s}	9	0	0	—	—	1.4	1	—	—	—	—	—	—	—	—	

	212		3.24	.41	.02			.63	.69	29.7	35.1	34.5		0.7	167	184
	Sh	P	9	0	0			4	(	-	.					
	214	1s	4.86	.43	.03			.14	.43	28.3	35.4					
	Sh	P _{1s}	9	0	0			4	(	-	.			2	-	-
	217-1		4.45	.30	.02			.79	.43	29.3	34.0			.4	167	178
	Sh	P _{1s}	9	0	0			4	(	-	.			-	-	-
	225		3.87	.42	.03			.01	.67	28.8	34.1			0.9	163	167
	Sh	P	9	0	0			4	(	-	.	-				
	228	1s	4.68	.41	.03			.30	.46	28.5	35.2	34.7				
	Sh	P	9	0	0			4	(	-	.	-				
	229	1s	4.74	.47	.03			.16	.47	27.8	34.5	33.5				
	Sh	P _{1s}	9	0	0			4	(	-	.	-		(	-	-
	231		3.14	.40	.02			.96	.47	29.4	34.4	34.0		.2	168	197
	Y	P	9	0	0			2	(	-	.					
	161	1s	5.37	.47	.03			.53	.47	28.2	34.5					
	Y	P	9	0	0			2	(	-	.	-				
	186	1s	5.98	.42	.02			.97	.40	28.1	36.4	35.5				
	Y	P	9	0	0			1	(	-	.	-				
	201	1s	7.48	.39	.04			.37	.65	28.9	34.9	32.3				
	Sh	C	9	0	0			2	(	-	.	-		-	-	-
	12	2b	5.31	.53	.04			.51	.59	30.6	37.2	35.8		0.6	165	183
	Sh	C	9	0	0			2	(	-	.	-		-	-	-
	37	2b	6.60	.42	.03			.74	.22	30.8	37.1	37.3		2.1	170	173
	Sh	C	9	0	0			4	(	-	.			1	-	-
	48	2b	4.89	.52	.04			.29	.25	29.9	36.5			.7	163	186
	Y	C	9	0	0			2	(	-	.	-				
	175	2b	6.49	.62	.05			.16	.59	27.5	33.4	33.3				
	Y	P _{1s}	9	3	0	0.	0.	1	(	-	.	-	-			
	u28-12		4.25	.25	.47	06	07	.08	.65	32.4	27.0	24.8	23.0			
Yu	Y	P _{1s}	9	2	0	0.	0.	1	(	-	.	-	-			
lin,	u35-8		5.32	.67	.34	07	06	.45	.03	32.6	24.9	23.7	22.6			
Mizhi	Y	P _{1s}	9	2	0	0.	0.	1	(	-	.	-	-			
gas	u43-10		4.39	.73	.41	08	08	.80	.52	31.9	26.3	23.0	22.0			
field	Y	P _{1s}	9	2	0	0.	0.	(	(	-	.	-	-			
	u44-7		5.65	.65	.32	04	04	.57	.69	32.8	25.5	23.8	24.8			

Hu
et al.,
2008

		M	P	9	4	0	0.	0.	(	(	-	.	-	-					
	i17	₂ h		2.75	.39	.86	18	15	.43	.93	34.1	23.8	22.4	21.2					
		M	P	9	5	0	0.	0.			-	.							
	i18	₂ h		3.32	.09	.19	04	04			34.1	23.5							Zha
		M	P	9	3	0	0.	0.			-	.							o et al.,
	i21	₂ h		5.18	.38	.50	09	07			35.1	22.7							2014
		Y	P	8	6	0		0.			-	.	-	-					
	u17	₂ h		8.79	.68	.36		14			34.2	25.5	23.1	21.1					
		G	O	9	0	0	0.	0.	£	(	-	.	-	-	-	-	-		
	19-15	₁ m ₅		3.18	.60	.06	01	00	.80	.31	29.1	33.8	30.6		0.4	169	184	186	
		G	O	9	0	0	0.	0.	£	(	-	.	-	-	-	-	-		
	19-13	₁ m ₅		3.32	.70	.07	01	01	.55	.31	27.0	32.7	30.1	27.7	0.5	167	184	189	
		Sh	O	9	1	0	0.	0.	£	(	-	.	-	-	-	-	-		
	an58	₁ m ₅		4.13	.09	.14	02	02	.43	.12	33.9	26.9	27.3	23.0	2.8				
		Sh	O	9	0	0	0.	0.	£	(	-	.	-	-	-	-	-		
	Jing an133	₁ m ₅		3.22	.49	.05	00	00	.88	.30	29.6	35.0	29.7			168	190	183	Dai
	bian	Sh	O	9	0	0	0.	0.	£	(	-	.	-	-	-	-	-		et al.,
	gas	an13	₁ m ₅	4.20	.56	.07	01	01	.82	.27	31.0	32.5	29.4			167	178	181	2014a
	field	Sh	O	9	1	0	0.	0.	£	(	-	.	-	-	-	-	-		
	an95	₁ m ₅		5.25	.02	.11	01	01	.00	.52	27.4	28.1	27.1	27.6	2.8	168	176	187	
		Sh	O	9	0	0	0.	0.			-	.	-	-					
	an62	₁ m ₅		6.55	.55	.07	01	01			32.7	33.1	30.0						
		Sh	O	9	0	0	0.	0.	£	1	-	.	-	-					
	an377	₁ m ₅		5.86	.27	.03	00	00	.80	.05	32.9	36.5	29.6						
		Sh	O	9	0	0	0.	0.	£	£	-	.	-	-					
	an323	₁ m ₅		1.14	.40	.06	01	01	.93	.43	33.4	35.9	30.1						This
																			paper

Table 2. Main light hydrocarbon parameters of the natural gases in the Yan'an gas field

Well		C ₆₋₇ , (%)			C ₇ , (%)			*C ₅₋₇ ^d , (%)			n-Heptane			Aromaticity
		Aromatics	Alkanes	Cycl	n-C ₇ ^a	MCC6 ^b	ΣD	n-alkanes	iso-alkanes	Cycloalkanes	Heptane value ^e	Heptane value ^f		
S _{h2}		4.6	84	11.3	30	69.	0.0	14.19	74.74	11.07		0.	0.4	
S _{h217}		8.2	51	40.3	11.	72.	15.	12.74	54.67	32.59	2.46	10	0.1	
S _{h6}		10.	50	39.1	10	74.	15.	7.47	50.57	41.95	3.56	9.	0.1	
S _{h25}		15.	53	30.5				21.73	64.68	13.59				
S _{h26}								22.64	67.80	9.56				
S _{h28-1}			73	23.3				27.74	54.86	13.40				
S _{h36}								24.46	75.54	0.00				
S _{h38}		19.	50	30.3				19.52	64.34	16.13				
S _{h56}		8.9	68	22.3				16.05	64.90	19.05				
S _{h127}		27.	29	43.0				19.37	58.34	22.28		0.		
S _{h207}		20.	79	0.00				6.24	93.06	0.69		0.		
S		29.	0.	70.7				0.00	0.00	100.0		0.		

h210	₁ S	22	00	8					0		00			
	S	18.	20	61.6	9.	85.	4.6					14	0.1	
h212	₁ S	23	.13	4	71	67	1	6.38	18.23	75.39		.59	1	
	S	19.	31	49.0										
h214	₁ S	35	.59	6				13.06	53.20	33.73				
	S													
h217-1	₁ S							0.00	59.20	40.80				
	S	12.	37	19.1	13	71.	15.					11	0.1	1.
h225	₁ S	93	.96	1	.45	19	36	16.26	41.32	42.42	2.23	.79	9	85
	S	20.	63	15.4										
h228	₁ S	70	.89	0				8.86	65.94	25.20				
	S	0.0	94											
h229	₁ S	0	.10	5.90				19.53	73.84	6.63				
	Y	4.0	79	16.4										
161	₁ S	8	.51	1				19.75	68.20	12.05				
	Y	15.	71	12.9										
186	₁ S	51	.56	3				25.04	67.14	7.82				
	Y	5.6	72	21.5	11.	88.	0.0					10	0.1	
201	₁ S	3	.84	3	86	14	0	6.23	63.45	30.32		.10	3	
	S	6.0	68	25.5										
h12	₂ b	6	.42	2				16.64	53.31	30.55				
	S	12.	30	56.9										
h37	₂ b	68	.41	1				8.90	23.96	67.13				
	S	12.	30	57.0	5.	71.	23.					5.	0.0	2.
h48	₂ b	59	.34	7	87	07	06	6.69	28.02	65.29	3.49	.07	8	68
	Y	5.7	75	18.8	43	48.	7.3					41	0.9	0.
175	₂ b	3	.41	6	.98	70	2	17.51	66.03	16.46		.86	0	15

$$^a n\text{-}C_7\% = n\text{-}C_7 \times 100 / (n\text{-}C_7 + MCC_6 + \Sigma MCC_5);$$

$$^b MCC_6\% = MCC_6 \times 100 / (n\text{-}C_7 + MCC_6 + \Sigma MCC_5);$$

$$^c \Sigma \text{MCC}_5\% = \Sigma \text{MCC}_5 \times 100 / (n\text{-C}_7 + \text{MCC}_6 + \Sigma \text{MCC}_5);$$

^dThe distribution of ^{*}C₅₋₇ light hydrocarbon includes only the *n*-alkanes, isoparaffins and cycloalkanes, but excludes the aromatic hydrocarbons;

$$^e \text{iso-Heptane value} = (2\text{MH} + 3\text{MH}) / (\text{t1,2-DMCP} + \text{c1,3-DMCP} + \text{t1,3-DMCP}); \text{P3} = 2,2\text{-DMP} + 2,4\text{-DMP} + 3,3\text{-DMP} + 2,3\text{-DMP} + 3\text{-EP};$$

$$^f \text{n-Heptane value} = (100 \times n\text{-C}_7) / (\text{CH} + 2\text{MH} + 2,3\text{-DMP} + 1,1\text{-DMCP} + 3\text{MH} + \text{c1,3-DMCP} + \text{t1,3-DMCP} + \text{t1,2-DMCP} + 3\text{EP} + 2,2,4\text{-TMP} + n\text{-C}_7 + \text{MCH})$$