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Bulk and intramolecular carbon isotopic reversal of the Paleozoic natural gas in the Ordos Basin: Insights into the aromatization of hydrocarbons at the early maturation

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This study focuses on the Ordos Basin and for the first time reports the bulk and intramolecular carbon isotopic reversals in conventional gas reservoirs at the high-evolution stage. The intramolecular isotopic compositions of propane determined by an improved GC-Pyrolysis-GC-IRMS method, which was calibrated by the quantitative isotopic ^{13}C NMR method. Through analysis of chemical compositions, compound-specific isotope compositions, and the position-specific (PS) carbon isotope compositions of propane in 12 natural gas samples from the northern part of the basin and the Longdong area in southwest basin, we found that the natural gas in the Longdong area is oil-type gas or mixed gas, while the natural gas in the Taiyuan Formation of the northern basin is typical coal-type gas. Analysis of the propane generation pathway reveals that propane in the coal-type gas of the northern basin mainly originates from the n-propyl reaction pathway. In the Longdong area, although the source rocks of the natural gas have a high maturity (Ro~3.4%), the $\Delta_{C\text{-}T}$ values of propane are negative, indicating characteristics of the early low-maturity stage. Majority (25 - 75%) of propane in this area is derived from the isopropyl reaction pathway. This phenomenon is attributed to the preservation of branched-chain hydrocarbon molecules during the early maturation stage. Specifically, the thermal stabilization process preserves the structures formed in the early low-maturity stage, allowing these gases to retain low-maturity characteristics even at the high-maturity stage. Then, during the high-maturity (or over-maturity) stage, propane is preferentially formed via the isopropyl pathway. The PS isotope distribution of propane confirms early hydrocarbon generation from ancient source rocks in the southeastern Ordos Basin in the early stages of thermal evolution.