

Carbon and hydrogen isotopic reversals in highly mature coal-derived gases: a case study from Paleozoic gases in the southern Ordos Basin, China

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The positive compositional carbon isotopic series, $\delta^{13}\text{C-CH}_4 < \delta^{13}\text{C-C}_2\text{H}_6 < \delta^{13}\text{C-C}_3\text{H}_8 < \delta^{13}\text{C-C}_4\text{H}_{10}$, is common in thermogenic gases. With the exploration of deeper strata, however, isotopic reversals ($\delta^{13}\text{C-CH}_4 > \delta^{13}\text{C-C}_2\text{H}_6 > \delta^{13}\text{C-C}_3\text{H}_8$) in over-mature unconventional shale gases and conventional (coal-derived) gases have been identified. Paleozoic gases in the southern Ordos Basin, China, with partial or complete isotopic reversals, were studied as examples of isotopic fractionation in over-mature coal-derived gases. Isotopic compositions of gases of different maturities from the Ordos Basin and shale gases from around the world were compared. Results indicate that carbon isotopic series are related to maturity. Complete isotopic reversal mostly occurs in regions with vitrinite reflectance (R_o) $> 2.4\%$. Where $2.4\% > R_o > 2.0\%$, almost all gases display partial isotopic reversal, with $\delta^{13}\text{C-CH}_4 > \delta^{13}\text{C-C}_2\text{H}_6$ or $\delta^{13}\text{C-C}_2\text{H}_6 > \delta^{13}\text{C-C}_3\text{H}_8$. Reversal carbon isotope in coal-derived gases is not caused by an abiogenic origin, mixing of gases from different types of source rock, abiogenic polymerization, wet gas cracking, or other mechanisms that contribute to reversal in shale gases. Based on the unique structure of coaly source rock and the geology of the Ordos Basin, closed-system aromatization–polycondensation reactions are considered the most likely explanation for carbon isotopic reversal. During the reactions isotopic light gases were generated by recombination of previously formed hydrocarbons and residual kerogen–coal. Hydrogen isotopic reversal in southern Ordos Basin may also be caused by aromatization–polycondensation reactions.