Methylation of aromatic nuclei and marine petroleum evolution in the Tarim Basin, China

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The Tarim Basin occupies an area of ≈ 560 × 10^3 km^2. It is one of the most important marine oil-gas bearing basins in China. Marine petroleum is mainly buried below 5 km. It has received much attention due to its unique features:

① The methylated aromatic hydrocarbons are abundant in marine oil and source rock and the carbon isotopic compositions of aromatics have a ^{12}C enrichment with the increasement of methyl substitutes [1]; ② The average $\delta^{13}C$ values of the fractions of oil and bitumen "A" show an abnormal order [2]; ③ Most of the oil-cracking gases at the depth of above 6 km are characterized by the ^{13}C-depleted methane (-54.9‰), which are globally rare; with the increasing depth, methane has a ^{12}C enrichment and the $\delta^{13}C_2$-$\delta^{13}C_1$ values increase [3].

However, the mechanism of the special isotopic fractionation is still difficult to explain. The thermal simulation experiments of model compounds indicate that: because of methylation reactions, aromatic hydrocarbons have a ^{12}C enrichment as the increasement of the amount of methyls and methane has a ^{12}C enrichment as the temperature increases.

Then, we can find that the above surprising phenomena in the Tarim are consistent with those in the simuleted experiments, indicating that the methylation of aromatic nuclei is responsible for the anomaly. The isotopic compositions of individual aromatics are mainly affected by the methylation during the maturation process. Then, the fractions containing aromatic nuclei (aromatics, resins and asphaltenes) have a ^{12}C enrichment compared with saturate hydrocarbons, resulting in an abnormal order among the fractions of organic matter. In addition, the carbon isotopically lighter methyls are "stored" in the aromatic nuclei due to methylation, resulting in the isotopically heavier methyls to form gases; the gases become lighter with increasing maturity.

The study suggests a widely occurrence of methylation in the Tarim Basin and the methylation of aromatic nuclei significately affects the deep marine petroleum evolution.

[1] Zhang et al. (2014) Sci. China Earth Sci. **44,** 1723-1730.
[2] Zhou et al. (2019) J. Petrol. Sci. Eng. **175,** 83-96.
[3] Wang et al. (2018) Mar. Petrol. Geol. **91**, 532-549.