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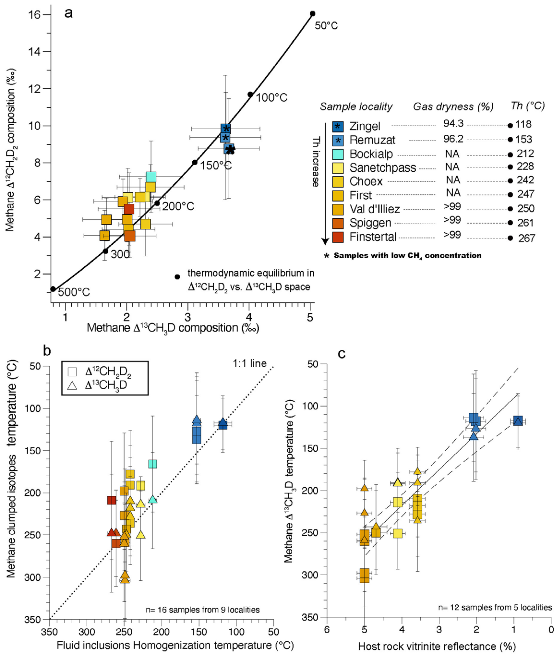

Geochemistry of CH_4 isotopologues ($^{13}CH_3D$ and $^{12}CH_2D_2$) within fluid inclusions in Alpine tectonic quartz fissures

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Petroleum-bearing fluid inclusions are small encapsulations of oil and gas that persist in the rock record long after their parent fluids have moved on.The chemical and isotopic compositions of inclusions offer a unique way to investigate hydrocarbon formation pathways in deep Earth environments and in ancient geological times. Here we use stable isotope compositions of methane, including clumped isotopologues $^{13}CH_3D$ and $^{12}CH_2D_2$, to evaluate the formation conditions of methane within fluid inclusions in Alpine tectonic quartz fissures. We observe that this CH_4 is relatively ^{13}C and ^{2}H enriched ($\delta^{13}C$ = -26 to -39‰; $\delta^{2}H$ = -126 to -146‰), consistent with the formation by advanced catagenesis of organic matter and has clumped isotope compositions indicating intramolecular isotopic equilibrium at catagenetic to low-grade (sub-greenschist) metamorphic temperatures ($T\Delta^{13}CH_3D$: 120-300°C; $T\Delta^{12}CH_2D_2$=120-260°C). The methane clumped-isotope temperatures of Alpine quartz-hosted inclusions agree well with temperatures reconstructed from fluid inclusion thermometry (*T*h) and re-enforces the interpretation that thermogenic methane reaches equilibrium with respect to $\Delta^{13}CH_3D$ and $\Delta^{12}CH_2D_2$ values under natural geological conditions between the wet-gas and dry-gas temperature windows. These findings, combined with an analysis of the geographic distributions of inclusion-bearing fissures and fluid inclusion *PVT* behaviours, also provide direct evidence of reservoirs of thermogenic methane trapped beneath the Alps during Mid-Miocene tectonic nappe emplacement (c.a. 25 to 15 Ma).