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ABSTRACT BOOK

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Multiple origins of methane in fluids circulating in the Othrys ophiolite, central Greece

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The complex geology of Greece includes two important parallel running ophiolitic belts. The Othrys Massif in central Greece belongs to westernmost of them. In and around this wide ophiolite outcrop, some cold hyperalkaline and some hypothermal ($T < 40^{\circ}\text{C}$) alkaline springs are present. Thirty water samples were collected at 17 different sites and both bubbling and dissolved gases were analysed for their chemical (He , Ne , H_2 , O_2 , N_2 , CH_4 , C_2H_6 , CO_2 and H_2S) and isotope (He , $\delta^{13}\text{C}-\text{CO}_2$, $\delta^{13}\text{C}-\text{CH}_4$, $\delta^2\text{H}-\text{CH}_4$) composition. All samples except one have H_2S contents below detection limit ($10\ \mu\text{mol/mol}$), whilst H_2 (from <2 to $2500\ \mu\text{mol/mol}$), CO_2 (up to $26,000$ but generally below $1000\ \mu\text{mol/mol}$) and O_2 (up to $16,000$ but generally below $3000\ \mu\text{mol/mol}$) present low concentrations. Gases in alkaline waters ($\text{pH} < 10$) are generally dominated by CH_4 (from $128,000$ to $915,000\ \mu\text{mol/mol}$), while hyperalkaline ($\text{pH} > 11$) waters are N_2 dominated (from $727,000$ to $977,000\ \mu\text{mol/mol}$). Generally, He isotope composition excludes contributions from a mantle source, showing a mostly pure crustal contribution for the alkaline waters and a prevailing atmospheric contribution for the hyperalkaline ones.

Methane may have different origins, which can be subdivided in biogenic (either directly produced by microbial activity or deriving by decay of organic matter at $T > 150^{\circ}\text{C}$) and abiogenic (from pure inorganic reactions). Among the latter, one of the most debated origins comes from serpentinization processes of ultramafic rocks in ophiolitic sequences at low temperatures ($T < 80^{\circ}\text{C}$). Furthermore, secondary processes (diffusion, inorganic or microbial oxidation, etc.) may mask the original chemical and/or isotope composition. Primary and secondary processes acting on CH_4 can be recognised mainly through its isotope ($\delta^{13}\text{C}$ and $\delta^2\text{H}$) composition and the ratio between CH_4 and C_2+C_3 hydrocarbons (Bernard ratio).

Samples collected in the Othrys Massif display a wide range of both isotope compositions of CH_4 ($\delta^{13}\text{C}-\text{CH}_4$ from -74.5 to $-14.5\ \text{‰}$ and $\delta^2\text{H}-\text{CH}_4$ from -343 to $-62\ \text{‰}$) and Bernard ratio (from 220 to $15,800$). The relatively high values of the ratio seem to exclude great contributions from thermogenic CH_4 . Alkaline waters present the most negative isotope values for CH_4 , evidencing a biogenic (microbial) origin, whereas many of the hyperalkaline waters have CH_4 isotope values compatible with an abiogenic origin through serpentinization processes but also at points, very negative values are present, indicating sometimes a clear biogenic contribution. Finally, few samples both from alkaline and hyperalkaline waters show some evidence of secondary oxidation processes.



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