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Alkanes and alkenes in Mediterranean volcanic-hydrothermal systems: origins and geothermometry

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It is still a matter of debate if nature provides conditions for abiogenic production of hydrocarbons. Methane (C1) and the C2+ alkanes emanating from ultramafic hydrothermal systems such as Lost City have been considered to be abiogenic in origin, mainly because of the occurrence of an isotopic reversal between methane and the C2+hydrocarbons and C1/C2+ ratios >1000 [1]. Abiogenic production of methane has been postulated to occur under the relatively oxidizing redox conditions of continental-hydrothermal systems, too. It was observed that temperatures received from the H_2-H_2O-CO-CO_2-CH_4 geoindicator were coincident with temperatures derived from carbon isotope partitioning between CO_2 and CH_4in gases released from the Mediterranean volcanic-hydrothermal systems of Nisyros (Greece), Vesuvio and Ischia (both Italy) [2]. Such equilibrium pattern, if not fortuitous, can only be obtained if mantle- and marine limestone-derived CO_2 is reduced to CH_4. At Nisyros, observed C1/C2+ ratios from 300-4000 are in agreement with an abiogenic origin of the methane. Ethane and propane, however, were shown to be non-genetic with CO_2 and methane. C1/C2 and C2/C3 distribution ratios may point to the admixture of small amounts of hydrocarbons deriving from the thermal decomposition of organic matter along with abiogenically equilibrated methane essentially devoid of the higher hydrocarbons [3].

Here, we provide new isotopic and hydrocarbon concentration data on several Mediterranean volcanic-hydrothermal systems, including Nisyros, Vesuvio, Ischia, Vulcano, Solfatara and Pantelleria. Wherever possible, we have extended our data set for the hydrogen isotope composition of CH_4 and H_2, n-alkane- and alkene/alkane-distribution ratios. At Nisyros, measured alkene/alkane- and H_2/H_2O concentration ratios confirm the attainment of equilibrium between CO_2 and CH_4. CO_2 and CH_4 appear to have equilibrated in the liquid phase at temperatures of $\sim$360°C and redox conditions closely corresponding to the metastable mineral paragenesis of fayalite-hematite-quartz. At Pantelleria and Solfatara, apparent isotopic CH_4-CO_2 equilibration temperatures of $\sim$560°C and $\sim$450°C, respectively, are in agreement with measured alkene/alkane- and H_2/H_2O ratios, too. However, at Baia di Levante (Vulcano), these concentration ratios do not correspond to the apparent carbon isotopic temperature. These findings imply that carbon isotopic analysis of discharged CO_2 and CH_4 might be a powerful tool to determine temperatures of volcanic aquifers. Alkene/alkane and H_2/H_2O concentration ratios should be measured along with the carbon isotopic composition of CO_2 and CH_4 to be able to check independently if isotopic equilibrium between CO_2 and CH_4 has been attained.

[1] Proskurowski et al. (2008) Science 319, 604-607; [2] Fiebig, J. et al. (2007) Geochim. Cosmochim. Acta **71**, 3028-3039. [3] Fiebig, J. et al. (2009) Geology 37, 495-498.