Formation processe(s) of methane in submarine mud-volcanoes from the Marianna trench (IODP Exp. 366)

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Active serpentine mud volcanoes located on the overlying plate in the mariannas subduction fore-arc are unique geological features. Hydrogen and methane (among other volatile species) are locally produced within the serpentinite, the latter being a potential byproduct of deep microbial activity. Alternatively, these species could have been produced abiotically, for instance during Fischer-Tropsch type (FTT) reactions.

IODP expedition 366 (dec. 2016-feb. 2017) on the Mariannas convergent margin recovered a suite of core drills and associated pore fluids from three serpentine mud volcanoes. In order to determine the contribution of biogenic and abiotic gases, we measured the abundances of doubly-substituted methane isotologues, $^{13}CH_3D$ and $^{12}CH_2D_2$ in 10 gas samples from the Asùt Tesoru seamount. The samples are from three different drill cores and have been collected at depth ranging between 1 and 100 meters below the sea-floor. Measurements were performed using the Panorama (Nu-Instruments) mass spectrometer at UCLA, using a mass resolving power of ~40,000.

The $\delta^{13}C$ and δD values range between -5 and -23.6‰ (±0.1‰, vs. PDB) and between -95 and -112‰ (±2‰, vs. SMOW) respectively. The abundances of clumped isotopologues show small but significant variations: $\Delta^{13}CH_3D$ values range between 2.3 and 4.3‰ (±0.2‰) and $\Delta^{12}CH_2D_2$ range between 9.7 and 11.7‰ (±0.8‰).

If formed under thermodynamic equilibrium, the temperatures derived from the abundance of $\Delta^{13}CH_3D$ would range between +120 and 220°C. However, all samples display small but significant $\Delta^{12}CH_2D_2$enrichments or depletions at a given $\Delta^{13}CH_3D$ value, relative to values predicted for equilibrium. These disquilibriums range between -2 and +5‰, indicating that mixing or mass fractionation had occurred during or after the formation of methane in the Mariannas mud volcanoes. However, no simple combination of methane formed at high- (250-300°C) and low-temperature (< 90C) can account for the observed gas compositions. As an alternative, preliminary interpretations involve isotope mass fractionation of methane within the serpentinite volcanoes.