Abundances of paired methane isotopologues as evidence for the origin of methane from mud volcanoes in Taiwan.

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The abundances of methane isotopologues with two rarely substituted isotopes have been theoretically calculated and experimentally tested to estimate the temperature at which methane is formed or thermally equilibrated. Such a thermometer overcomes the inherent disadvantages of the conventional approach that uses the bulk isotopic compositions of two coexisting, equilibrated phases.

This study aims to determine the source of methane from mud volcanoes or seeps in Taiwan using the abundances of two rare isotopologues of methane ($\Delta^{13}CH_3D$, $\Delta^{12}CH_2D_2$), bulk isotopic compositions of methane ($\delta^{13}C_{CH4}$ and δD_{CH4}), carbon dioxide ($\delta^{13}C_{CO2}$), dissolved inorganic carbon, and fluid (δD_{H20} and $\delta^{18}O_{H20}$). Our analyses yielded $\Delta^{13}CH_3D$ and $\Delta^{12}CH_2D_2$ values that ranged between 1.9 and 7.1‰, and 0.5 and 19.9‰, respectively. A portion of the samples were characterized by values falling on the range delineated by the equilibrium line, and predict a temperature range between 100 and 280°C. These temperature estimates together with the bulk isotopic compositions suggest that methane is formed by thermal maturation at great depths. Other samples were characterized by values deviating from the equilibrium line at various degrees. Considering geological backgrounds and methanogens detected at some sites, the non-equilibrated gases most likely represent mixtures between microbial and thermogenic sources. Overall, these results suggest the extraction of methane and other gases from multiple depth sources from the strata fragmented by fault displacement in an active orogenic belt.