Methane isotopologue analysis for mud gas logging samples to trace microbial and thermogenic methanogenesis in deep marine sediments

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Methane, and its isotopic composition, maintains enduring interest due to its important implications for understanding the global carbon cycle, atmospheric greenhouse gas dynamics, and the limits of life in deep and extreme environments. Previously, carbon ($\delta^{13}C$) and hydrogen (δD) stable isotope ratios, and methane/ethane ratios have been used to identify the origin of methane in deep marine environments; however, the interpretation of these geochemical proxies becomes challenging where mixing of differently sourced methane occurs. For example, mixing between deep microbial and thermogenic methane may obscure the initial isotope signals.

We analyzed the $^{13}CH_3D$ abundance, as well as the methane/ethane ratios, $\delta^{13}C$, and δD of mud gas logging samples in order to investigate the transition from microbial to thermogenic methanogensis in deep marine sediments. The mud gas logging samples come from the northern Gulf of Mexico and represent the gases associated with return fluids from drilling down to 4,880m below seafloor; they provide a unique opportunity to sample methane and associated light hydrocarbons from deep marine sediments.

The methane/ethane ratios and $\delta^{13}C$ of methane suggest transition from dominantly microbial to thermogenic methane at about 3,200 m below seafloor. Relative abundance of four methane isotopologues ($^{13}CH_4$, $^{12}CH_4$, $^{12}CH_3D$, and $^{13}CH_3D$) yielded apparent temperature of equilibrium ranging from ca. 70 to 150°C, consistent with the transiton. Our results suggest that methane isotopologue abundance can be used to gauge the temperature of microbial and thermogenic methanogenesis in deep sediments, and that mud gas logging provides an excellent opportuntity to survey deep methanogenesis.