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Differentiating thermogenic from microbial methanogenesis: insights from multiply substituted isotopologues of methane

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Microbial methanogenesis – the biological production of methane by microorganisms – generates the vast majority of methane on Earth [1]. An increasing number of studies report the presence of methane in the atmospheres of extraterrestrial bodies such as Mars and Enceladus, leading to speculations about the potential for life on other planets. However, a number of non-microbial processes, such as thermogenesis or water-rock reactions can also produce methane, and may result in the production of a false positive biosignatures.

Classical 'bulk' isotopic measurements of methane $\delta^{13}C$ and δD, together with associated $C_1/(C_2+C_3)$ ratio, and isotopic characterization of source C and H, are often sufficient to disentangle the sources and processes that resulted in the production of methane in terrestrial environments. Yet, these types of environmental information will not be available in the context of samples collected from extraterrestrial bodies.

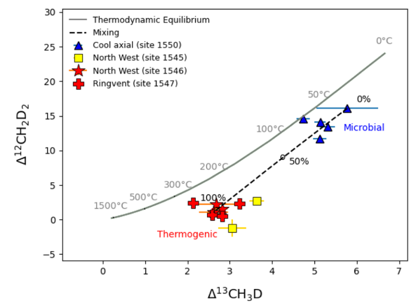
Several studies demonstrated that multiply substituted isotopologues ('clumped isotopes') of methane ($\Delta^{13}CH_3D$ and $\Delta^{12}CH_2D_2$) are mainly sensitive to the genetic pathway of methane, making this type of measurements particularly promising for assessing the biogenicity of environmental methane samples.

In this work we studied the methane produced within organic-rich marine sediments in Guaymas Basin, a young rift basin characterized by seafloor spreading. There, the methane has been suggested to be produced by both microbial methanogenesis in surficial cool sediments and thermal cracking of organic carbon molecules in deeper, hydrothermally heated sediments. These two processes have the potential to produce low $\Delta^{12}CH_2D_2$ values relative to thermodynamic equilibrium (combinatorial effect) that occur when methane is produced with hydrogens atoms having distinct isotopic compositions.

By performing clumped isotopic analyses of methane from shallow to deep sediments cored during IODP Expedition 385 [2] we explore the range of isotopic compositions expressed by these two main pathways of methane production in natural settings. This approach allows us to untangle the relative contribution of high and low maturity thermogenic methane versus microbial methanogenesis and confirms the utility of clumped isotopes in methane as a promising tool for the research for life elsewhere.

[1] Saunois, M. et al., "The Global Methane Budget 2000-2017" 12, no. 3 (2020),

[2] http://publications.iodp.org/proceedings/385



Multiply substituted ($\Delta^{13}CH_3D$ vs. $\Delta^{12}CH_2D_2$) isotopic measurements and associated errors for methane from IODP Exp.385 subsurface sediments.

The dashed black line tracks mixing between putative thermogenic and microbial gases.
The gray curve follows the thermodynamic equilibrium among methane isotopologues as a function of temperature.