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# Using Methane Isotopologues to Illuminate Earth's $O_2$-$CH_4$ History: Implications for Biosignatures on Habitable Worlds

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Methane is an important biogenic trace gas on Earth since the emergence of life and is thought to be a potential biosignature on other planets. Its presence has also been reported on other planetary bodies, such as Mars and Enceladus, and, recently, tentative hints of its presence were reported in a rocky exoplanet's atmosphere. Methane can be produced biotically and abiotically in varying Earth environments. Discerning its sources on Earth can advance our interpretation of methane observed elsewhere. Traditionally, bulk isotopic compositions of methane (D/H and $^{13}C/^{12}C$) have been central to constraining its origin. However, they are controlled by the isotopic composition of the source C and H used to build the molecule. Recent advances in measuring clumped methane ($^{13}CH_3D$ and $^{12}CH_2D_2$) provide a powerful approach for constraining formation processes and alteration pathways of methane, independent of bulk isotope ratios. As false positive detections of methane remain a major concern in methane's role as a biosignature, we investigate this promising proxy at a ferruginous lake—Brownie Lake (Minnesota), an important but unexplored analog site on modern Earth with relevance to early Earth (Archean and Proterozoic) and beyond due to its low oxygen and iron-rich features. We sampled dissolved methane from the water column and sediments, as well as ebullitive methane. $^{13}CH_3D$ and $^{12}CH_2D_2$ were analyzed to determine how the isotopic signal is transferred and possibly altered along its pathway to the surface. We present preliminary results in Figure 1. In opposition to previous sites [1] that display $\Delta^{12}CH_2D_2$-$\Delta^{13}CH_3D$ co-variations, methane from Brownie exhibits limited to no variations in $CH_2D_2$ with depth, while $^{13}CH_3D$ tends to show higher values when the condition changes to more oxic. This relationship could be due to a mixed pathway for methane oxidation in Brownie Lake—primarily by aerobic oxidation of methane (AeOM) but with possible contributions from iron-based anaerobic oxidation of methane (AOM). Our study will validate the application of this clumped approach to early Earth and gain insights into the role of methane as a potential biosignature on other worlds.

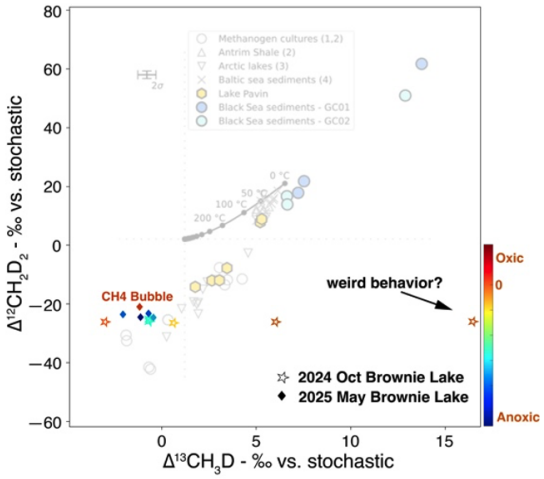



Figure 1. Clumped methane measured from Brownie Lake on top of previous data reported in Giunta et al., 2022. Previous data are shown transparently. The colors in our data represent the oxygen level.

Reference:

[1] Giunta, T., et al. (2022). *Geochimica et Cosmochimica Acta*, *338*, 34–53.