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Methane dynamics in continuous permafrost thaw ponds: using clumped isotopes for process characterization

BONNIE J DE BAETS[1,2], PETER M J DOUGLAS[3], JASON M E AHAD[2], PAUL R GAMMON[4], MOMOTAJ BEGUM[1] AND JOSUÉ J JAUTZY[2]

[1]Department of Earth & Planetary Sciences, McGill University
[2]Geological Survey of Canada, Natural Resources Canada, Québec
[3]McGill University
[4]Geological Survey of Canada, Natural Resources Canada, Ottawa

While methane emissions from Arctic ecosystems are predicted to increase due to climate warming, the mechanisms controlling these emissions are not well understood. In addition to standard bulk carbon and hydrogen isotope measurements, clumped isotope measurements of methane can provide further insights into the processes driving both microbial methane production and oxidation, and, how these contribute to Arctic methane fluxes. There are currently very few clumped isotope datasets available from Arctic ecosystems, and even fewer studies that report measurements of both $^{12}CH_2D_2$ and $^{13}CH_3D$, limiting our ability to comprehensively interpret this new tracer for methane cycling in this region.

This study presents a large dataset (32 measurements) of methane clumped isotopes from a wide range of thaw ponds from continuous permafrost landscapes. These include both natural ebullition fluxes and gas forced from sediments from polygonal ponds and ice-wedge trough ponds in Nunavut, Canada, and gas forced from culvert pond sediments in Yukon, Canada. Clumped isotopologue data indicate clear and highly variable microbial disequilibrium, with $\Delta^{13}CH_3D$ varying from –1.496 to 4.127‰, and a much larger variation in $\Delta^{12}CH_2D_2$ from –46.452 to –7.765‰, a range comparable to the entire variability previously reported for microbial methane in experiments and natural environments. Data from Nunavut and the Yukon both span this range, with no clear differences by pond type. This broad spread contrasts with the limited range of methane $\delta^{13}C$ values (–73.6 to –51.3‰), highlighting the utility of methane clumped isotopes to distinguish methane production processes within an ecosystem. While the clumped isotope dataset shows no evidence of significant aerobic or anaerobic methane oxidation effects, substantial variation is observed in $\Delta^{12}CH_2D_2$ (–37.594 to –7.765‰) in gas bubbles collected from discrete periods of bubbling within the same pond, associated with much less variation in $\Delta^{13}CH_3D$ and $\delta^{13}C$. This may reflect methanogens accessing different pools of water with differing $\delta^{2}H$, leading to distinct combinatorial effects. Finally, clumped isotope data is compared with CH_4 concentrations and fluxes to discern how processes revealed by these new measurements could help elucidate Arctic methane cycling.