

# **Methane clumped isotope signatures from continuous permafrost thaw ponds: Implications for tracing methanogenesis and methanotrophy**

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Methane is a potent green house gas, and there are concerns that climate warming-induced permafrost thaw could generate a major increase in methane emissions from Arctic ecosystems. However, the mechanisms controlling Arctic methane emissions are not fully understood. Isotope tracers are critical tools for understanding microbial methane production and oxidation processes, as well as quantifying methane sources to the atmosphere. Conventional isotope measurements ( $\delta^{13}\text{C}$ ,  $\delta\text{D}$ ) cannot fully resolve the different methane production and oxidation pathways, and combining these measurements with novel clumped isotope analyses could provide a clearer understanding of how these pathways contribute to methane fluxes from Arctic environments. Currently there are few clumped isotope data from Arctic ecosystems, and even fewer data that include measurements of both  $^{12}\text{CH}_2\text{D}_2$  and  $^{13}\text{CH}_3\text{D}$ .

Here we present clumped isotope data of methane sampled from permafrost thaw ponds in continuous permafrost environments from the Yukon and Nunavut, Canada, including both natural ebullition gas and gas from sediments. The samples span a wide range of methane concentrations,  $\text{CO}_2:\text{CH}_4$  ratios, and conventional methane isotope values, including samples with low  $\delta\text{D}$  values. The clumped isotope data plot within the range of microbial disequilibrium data from previous experimental and *in situ* studies, and do not indicate a significant influence of anaerobic methane oxidation. Comparisons of the clumped isotope data with concentration, flux, gas composition, and conventional stable isotope data to test current hypotheses for the processes that control clumped isotope values in microbial methane. We also compare our dataset with other clumped isotope datasets of microbial methane to improve the source signature of natural microbial methane, including evaluating the potential for an Arctic-specific signature.