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Using $^{13}CH_3D$ & $^{12}CH_2D_2$ Clumped Isotopes for a Better Understanding of Atmospheric Methane Sources and Sinks

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Although methane (CH_4) is the most abundant organic molecule in the atmosphere and an important greenhouse gas, considerable uncertainty remains in the apportionment of its sources and the strengths of its sinks. Wetlands are the largest natural sources of atmospheric methane, but the difficulty of sampling these widespread systems through the seasons affects understanding of the balance between in-situ microbial production and oxidative consumption of the gas. Likewise, uncertainties in the magnitude and distribution of atmospheric sink reactions leads to corresponding uncertainties in estimates of methane residence time. The emerging ability to analyze clumped isotopes of methane has the potential to improve reconstructions of atmospheric methane sources and sinks [1]. Biogenic methane, for example, is strongly depleted in $^{12}CH_2D_2$ [2], and predictions for atmospheric methane based on rates of protium-rich sink reactions should lead to equally strong $^{12}CH_2D_2$ enrichment in the atmosphere [3]. Here, we provide a progress report from the UMD high mass resolution IRMS (Panorama) Laboratory investigation. Our first measurements of methane from wetland soil incubations, chamber collections, and air, confirm that wetland emissions in our region possess $^{12}CH_2D_2$ deficits and that air has $^{12}CH_2D_2$ enrichments in urban tropospheric air - each of nearly 50 ‰. Our measurements from laboratory experiments and natural systems support a role for re-scrambling of clumped $^{12}CH_2D_2$ isotopologues towards random distributions by methane-consuming processes as argued by [4]. This presentation will discuss how the addition of clumped isotope measurements to more traditional measures of atmospheric methane can be used to understand cycling of this important greenhouse gas in urban and rural environments of the densely populated Mid-Atlantic States, the focus of our ongoing investigation.

[1] Goldschmidt, Douglas et al., (2017), Org. Geochem 113, 262-282

[2] Goldschmidt, Young et al., (2017), GCA 203, 235-264

[3] Goldschmidt,Haghnegahdar et al., (2017), Global Biogeochemical Cycles 31,1387-1407.

[4] Goldschmidt, Ono et al., (2021), GCA 293, 70-85.