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An Insight into Atmospheric Methane Sources and Sinks Using $^{13}CH_3D$ & $^{12}CH_2D_2$ Clumped Isotopes

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Methane is an important greenhouse gas with a significant impact on the chemistry of the troposphere and stratosphere. However, there are still large uncertainties in the atmospheric methane budget and its components.

Clumped isotopes of methane have shown a good potential as a complimentary tool to bulk isotope measurement in fingerprinting atmospheric methane sources. Here we evaluate the contribution of some important microbial methane sources in atmospheric methane during different hours of the day using isotopologue measurements ($^{13}CH_4$, $^{12}CH_3D$, $^{13}CH_3D$, and $^{12}CH_2D_2$) of methane from air collected at sites located on the suburban campus of the University of Maryland (near campus buildings, a restored creek, and at an open-sided dairy barn) and from a semi forested freshwater wetland site near Goldsboro, Maryland. To fingerprint the role of methane emission from these biogenic sources in atmospheric methane, we also measured methane samples collected with chambers, bubble collection, during soil incubations, and from the stomach of a fistulated cow. Air samples in the wetland and barn sites were collected in different time of the day including both early in the morning after methane accumulated in the nocturnal boundary layer, and late in the afternoon when convection mixed air to a cloud layer. Collections were also made at different heights, from within centimeters to a few meters above the surface. Our measured $\delta^{12}CH_2D_2$ values of air samples reveal mixing of microbial methane with low D/H and air methane. The $\delta^{12}CH_2D_2$ of the near surface air collected at the RSC wetland during the early time of the day ranges from ~23‰ to ~44‰ contrasts with background air (with $\delta^{12}CH_2D_2$ ~50‰) collected later in the day. Methane collected directly from wetlands, from cattle, and from incubation experiments has negative $\delta^{12}CH_2D_2$. The effect of mixing with biogenic sources on $\delta^{13}CH_3D$ is much smaller. The mixing behavior can be used to provide a way to fingerprint local contributions to this greenhouse gas.