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Methane Fingerprinting: Isotopic Methane and Ethane-to-Methane Ratio Analysis Using a Cavity Ring-Down Spectrometer

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Emissions of Natural gas, and methane (CH_4) specifically, have come under increased scrutiny by virtue of methane's 28-36x greenhouse warming potential compared to carbon dioxide (CO_2) while accounting for 10% of the total greenhouse gas emissions in the US. Large uncontrolled leaks, such as the recent Aliso Canyon leak, originating from uncapped wells, coal mines and storage facilities have increased the total global contribution of methane missions even further. Determining the specific fingerprint of methane sources, by quantifying δ^{13}C values and C_2:C_1 ratios, provides the means to understand methane producing processes and allows for sources of methane to be mapped and classified through these processes; i.e. biogenic vs. thermogenic, wet vs dry.

In this study we present a fully developed Cavity Ring-Down Spectrometer (CRDS) that precisely measures $^{12}CH_4$ concentration and its $^{13}CH_4$ isotope concentration, yielding δ^{13}C measurements, C_2H_6 concentration, along with CO_2 and H_2O. This provides real-time continuous measurements without an upfront separation requirement or multiple analyses to derive the origin of the gas samples. The highly sensitive analyzer allows for measurements of scarce molecules down to sub-ppb 1-σ precision in 5 minutes of measurement: with CH_4 $<$0.1ppb, δ^{13}C $<$1‰ C_2H_6 $<$1ppb and CO_2 $<$1ppm. To complement this work, we provide the analysis of different methane sources providing a 2-dimensional mapping of methane sources as functions of δ^{13}C and C_2:C_1 ratios, which can be thought of as a modified Bernard Plot. This dual ratio mapping can be used to discriminate between naturally occurring biogenic methane sources, naturally occurring enriched thermogenic sources, and natural gas distribution sources. This also shows future promise in aiding gas and oil exploration, in distinguishing oil vs coal gases, as well as a valuable tool in the development of methane sequestration.