Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.37268

Resolving the information content of bulk and clumped methane isotopologues in atmospheric inversions: sensitivity, uncertainty, and source–sink attribution

BIBHASVATA DASGUPTA[1], MS. MALAVIKA SIVAN[2] AND THOMAS RÖCKMANN[1]

[1]Institute for Marine and Atmospheric Research Utrecht
[2]Royal Netherlands Meteorological Institute (KNMI), De Bilt

Reducing uncertainty in atmospheric methane source attribution is essential for evaluating mitigation strategies, and stable isotope measurements are among the few tools that can distinguish emission types without direct flux measurements. The conventional approach uses bulk isotope ratios ($\delta^{13}C\text{-}CH_4$ and $\delta D\text{-}CH_4$) as source fingerprints, exploiting the fact that biogenic and thermogenic methane exhibit distinct carbon and hydrogen isotope signatures. More recently, multiply-substituted isotopologues $\delta(^{13}CH_3D)$ and $\delta(^{12}CH_2D_2)$ have become measurable in the atmosphere, and their strong sensitivity to the kinetic isotope effect during OH oxidation has raised hopes that they could independently constrain sink strength and break degeneracies that bulk isotopes cannot resolve. Here we employ a two-hemisphere atmospheric inversion for all combinations of the four isotopologue tracers, optimising emissions from five source categories, per-source isotopic signatures, OH sink strength, and isotopic fractionation during loss over 1980–2024. Rather than comparing overall fit quality directly, we decompose the information each tracer contributes using an averaging-kernel analysis—a method that quantifies how many independent pieces of information each measurement type adds to the inversion and whether that information reduces uncertainty in source signatures, emission magnitudes, or sink parameters. We find that $\delta^{13}C\text{-}CH_4$, with its high measurement-to-prior-uncertainty ratio, consistently reduces the normalised per-observation cost and constrains emission budgets. $\delta(^{13}CH_3D)$, despite an apparent improvement in posterior fit, directs less than 3% of its information content toward source-signature discrimination; the bulk of its contribution constrains emission magnitudes through its temporal trend, not source identity. $\delta D\text{-}CH_4$ primarily constrains OH sink strength rather than sources. $\delta(^{12}CH_2D_2)$ systematically degrades posterior fits across all configurations: its current measurement precision of ~2‰ is a factor of three to ten below the threshold at which it would provide net-positive source information, given the combined uncertainty in source signatures and isotopic fractionation during loss. These results show that apparent improvements in inversion performance attributed to clumped isotopologues can reflect analytical precision rather than genuine gains in source attribution, and that tightening source-signature priors and isotopic fractionation constants is as important as improving atmospheric measurement precision before clumped isotopologues can meaningfully distinguish methane emission sources.