

# Using Clumped Isotopes to Constrain Factors Influencing Trends in Atmospheric Methane

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A critical issue facing humankind today is global warming and climate change. A significant part of this change is driven by the rise of atmospheric greenhouse gases such as methane and carbon dioxide. Methane is more efficient on a per molecule basis than carbon dioxide and its ~10 yr lifetime is considerably shorter than carbon dioxide, both of which make it a potential target for mitigation because its concentration is more sensitive to changes in source and sink fluxes. However, considerable uncertainty remains about the strength and variation of these source and sink fluxes. The isotopic composition and proportions of isotopic molecules (isotopologues) has the potential to add information that will be useful for constraining the factors that contribute to the rise of methane in the past, today, and in the future.

Here, we explore how atmospheric methane isotopologues enriched with two heavy isotopes  $^{13}\text{CH}_3\text{D}$  and  $^{12}\text{CH}_2\text{D}_2$  may have varied in the past 40 years using inverse box models. Our approach and findings are similar to that of Chung and Arnold (2021: GBC 35(10), p.e2020GB006883). Our models complement the forward models of Hagnegahdar et al. (2017: GBC 31(9), 1387-1407) and Whitehill et al. (2017: GCA 196, 307-325.). We anchor our model 40-year using measurements of methane in air today and fit to NOAA/GML records of atmospheric methane concentration (Dlugokencky: [gml.noaa.gov/ccgg/trends\\_ch4/](http://gml.noaa.gov/ccgg/trends_ch4/)) and published isotope composition data. We explore variations in 40 year model records for  $\Delta^{13}\text{CH}_3\text{D}$  and  $\Delta^{12}\text{CH}_2\text{D}_2$  to strength of sink reactions, kinetic isotope effects, and the isotopic composition and strength of individual sources. Like Chung and Arnold (2021), we find that the 40 year model records for  $\Delta^{13}\text{CH}_3\text{D}$  and  $\Delta^{12}\text{CH}_2\text{D}_2$  are relatively insensitive to lifetime but can yield significant differences in fossil fuel vs biogenic sources for methane emissions scenarios such as those provided in The Emissions Database for Global Atmospheric Research (EDGAR). These differences are large enough that they should be possible to resolve at the present measurement uncertainties for  $\Delta^{12}\text{CH}_2\text{D}_2$  in samples of archived air. We will discuss these results and our progress with the model calculations.