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# In Plume Miller-Tans Time Series Analyses for Improved Isotopic Source Signature Characterisation

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To characterise the isotopic chemistry of a source it is common to collect three or more grab bag samples of air from the plume downwind of the source of interest. Each air sample is then transported to a laboratory for precise analysis of the concentration and isotopic chemistry for the molecule of interest. The isotopic signature of the source is then determined by analysing the composition data in a Keeling or Miller-Tans plot (Keeling, 1961; Miller and Tans, 2003), which provides only a single estimate of the isotopic source signature. However, there is often considerable variability within the same source category, both at a single sample site and more commonly between sites. Ideally, the variability in the source category isotopic signature needs to be characterised for subsequent use in modelling blended regional-scale isotopic signatures. Another positive aspect of the in-field time series data sets is the real-time feedback, which may assist with refining sampling strategies.

Laser spectrometers enable the collection of in situ time series data for both the concentration and isotopic chemistry of specific molecules. These data can be analysed using either the moving Keeling or moving Miller-Tans methods (Vardag et al. 2016). In this presentation we show that continuous measurements taken within an emission plume for at least an hour generate enough data to make better estimates of the population statistics that summarise the variability in the source category isotopic signature compared to a small set of grab bag samples, despite the analytic technique having lower precision than analysis of grab bag samples by isotope ratio mass spectrometry.

We present examples of moving Miller-Tans analyses from urban gas leaks in Sydney, Australia, and analyses of the primary methane sources in the Surat Basin, Australia, including cattle feedlots, abandoned leaky exploration wells, and coal seam gas infrastructure. For each methane source we compare the moving Miller-Tans results with grab bag analyses and global databases of isotopic source signatures.

We conclude that collecting concentration and isotopic chemistry time series data in plumes downwind of a source improves characterisation of isotopic source signature variability. Such data sets provide useful input for modelling blended regional methane concentration and isotope data measured at either tall-tower air monitoring sites or as part of airborne atmospheric chemistry surveys.

References

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