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Characterisation of the regional source mix of methane at different locations in Europe using continuous isotope ratio measurements of δ^2H and $\delta^{13}C$.

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Methane (CH4) is a potent greenhouse gas with a global warming potential of about 84 over a 20-year timescale, and an atmospheric lifetime of about 9 years. The increase in CH4 emissions has contributed about 0.6°C to the observed global warming since pre-industrial times. The ongoing increase in atmospheric CH4 undermines efforts to mitigate climate change. To effectively mitigate CH4, it is essential to understand the location, strength and temporal variability of its most important sources, which vary in different regions. A widely used method to distinguish emissions from different source categories is the measurement of CH4 isotopic composition. Such measurements provide additional insight because different CH4 production processes emit CH4 with different isotopic compositions.

Traditionally, CH4 isotope measurements have been carried out on atmospheric air samples under controlled laboratory conditions, but since a few years, instruments measuring isotopic composition continuously at monitoring stations have become available. An important application of continuous isotopic CH4 measurements is the evaluation of regional-scale emissions with respect to the existing emission inventories. In model simulations using emissions from these inventories, the relative contributions of different source categories to observed enhancements can be calculated. This information can be used to simulate time series of the isotopic composition. By comparing these simulations with observed isotopic data, we can not only assess whether total emissions in a model are over- or underestimated, but also identify which source categories are responsible for any discrepancies.

The mobile isotope ratio mass spectrometry system developed at Utrecht University has been deployed at more than 10 different locations in Europe over the past decade, in most cases for approximately 7 months. The recorded 20-min resolution and high-precision isotope data of both d13C and d2H provide empirical constraints to the CH4 source mix at the different locations. The combination with high-resolution model simulations has provided many new insights into regional-scale emissions. We present an overview of key findings and discuss the value of high-resolution isotope measurements for improving our understanding of the regional budgets of this important greenhouse gas.



