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Using continuous high-precision isotope measurements over several months to characterise sources of atmospheric methane at various European locations

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Because of the strong greenhouse effect of methane, the increasing mole fraction of this gas in the atmosphere represents an important contribution to radiative forcing, and the associated global temperature increase. The main different sources of methane to the atmosphere are qualitatively identified, but their relative contributions are still poorly quantified. Source attribution of observed methane enhancements can help reducing these uncertainties. Isotope analysis is a widely used technique for source characterisation, but due to analytical challenges it has been difficult to obtain long-term high-resolution time series that could help deciphering sources on hourly to daily timescales. At the same time, isotopic source signatures are not always well characterised and may vary in time and space, which is usually not taken into account in the analysis. Through measurements of both the ^{13}C and deuterium isotopic signatures in CH_4 performed over several months, we improve our knowledge about the actual local methane sources, as well as their temporal variations.

We report continuous high-resolution δ^{13}C and δD -CH_4 measurements performed in Lutjewad (North of the Netherlands) and in the city of Kraków, Poland, using a continuous flow isotope ratio mass spectrometry (CF-IRMS) system. The results from Lutjewad clearly illustrate a prevalence of biogenic sources. This matches previous observations made in Cabauw (central Netherlands). Yet, in Kraków, the isotope data indicate that observed methane enhancements are dominated by thermogenic sources. Interpretation of the measurements is supported by Lagrangian particle dispersion simulations, allowing to connect the observations to upstream sources, and to challenge our current understanding of CH_4 isotope source signatures in these regions.

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