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Potential for long-term, high-frequency, high-precision methane isotope measurements to improve UK emissions estimates

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On the global scale methane (CH_4) concentrations have more than doubled over the last 150 years, and the contribution to the enhanced greenhouse effect is almost half of that due to the increase in carbon dioxide (CO_2) over the same period. Microbial, fossil fuel, biomass burning and landfill are dominant methane sources with differing annual variabilities; however, in the UK for example, mixing ratio measurements from a tall tower network and regional scale inversion modelling have thus far been unable to disaggregate emissions from specific source categories with any significant certainty. Measurement of the methane isotopologue ratios will provide the additional information needed for more robust sector attribution, which will be important for directing policy action

Here we explore the potential for isotope ratio measurements to improve the interpretation of atmospheric mixing ratios beyond calculation of total UK emissions, and describe current analytical work at the National Physical Laboratory that will realise deployment of such measurements. We simulate isotopic variations at the four UK greenhouse gas tall tower network sites to understand where deployment of the first isotope analyser would be best situated. We calculate the levels of precision needed in both δ-^{13}C and δ-D in order to detect particular scenarios of emissions.

Spectroscopic measurement in the infrared by quantum cascade laser (QCL) absorption is a well-established technique to quantify the mixing ratios of trace species in atmospheric samples and, as has been demonstrated in 2016, if coupled to a suitable preconcentrator then high-precision measurements are possible. The current preconcentration system under development at NPL is designed to make the highest precision measurements yet of the standard isotope ratios via a new large-volume cryogenic trap design and controlled thermal desorption into a QCL spectrometer.

Finally we explore the potential for the measurement of clumped isotopes at high frequency and precision. The doubly-substituted $^{13}CH_3D$ isotopologue is a tracer for methane formed at geological temperatures, and will provide additional information for identification of these sources.