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Determining UK regional isotopic source signatures of methane using long-term records

Ceres A. Woolley Maisch[1], Rebecca E. Fisher[1], James L. France[1], David Lowry[1], Grant Forster[2], Mathias Lanoisellé[1], and Euan G. Nisbet[1]

[1]Department of Earth Sciences, Royal Holloway, University of London, Egham, UK
[2]Centre for Ocean and Atmospheric Sciences (COAS), School of Environmental Sciences, University of East Anglia, Norwich, UK

Long term records are vital for understanding the way in which our environment is changing.

A significant rise in atmospheric methane began in 2007 and has accelerated thereafter, particularly since 2014. This trend was observed globally and was coupled with a sustained isotopic shift to values more depleted in ^{13}C (more negative $\delta^{13}C\text{-}CH_4$). Currently, there is no consensus as to why these observations have occurred. However, long term methane isotopic measurements can provide information about changes in the source mix of this important greenhouse gas. Here, long term records of both methane mole fraction and $\delta^{13}C\text{-}CH_4$ from 5 sites across the UK are presented, showing an increase in CH_4 and a decrease in ^{13}C from 2007, similar to those recorded globally, but at the regional-local scale. The approximately weekly in-situ measurements offer an insight into both suburban and background regions of the UK.

A method for isotopic discrimination from longer term atmospheric measurements of CO_2 and $\delta^{13}C\text{-}CH_4$ as outlined by Miller and Tans (2003) is utilised in this work. Miller-Tans analysis allows for the explicit specification of background values, vital when dealing with long term records due to both seasonal, local, regional and global background variations in atmospheric CH_4 and $\delta^{13}C\text{-}CH_4$.

When applying the Miller-Tans method to the long-term data from UK sites, as expected, the heaviest $\delta^{13}C\text{-}CH_4$ source signatures, which are associated with industrial sources such as gas leaks, are observed for the suburban sites, and biogenic, lighter, sources for the background sites. The methane source distribution is compared to results from mobile measurements carried out at Royal Holloway, University of London and to the UK National Atmospheric Emissions Inventory (NAEI).

From the initial results, it seems that there is a larger proportion of thermogenic/pyrogenic emissions in this data compared to the NAEI inventory. At all sites, there has been a post 2006 decline in $\delta^{13}C\text{-}CH_4$. Using the Miller Tans method, it is possible to calculate bulk regional source signatures which highlight a distinction between rural and suburban emissions, in general agreement with the NAEI.