Appendix 1 - Literature Review – methane measurement - academic papers

(2022) Continuous CH_4 and $\delta^{13}CH_4$ measurements in London demonstrate under-reported natural gas leakage

https://acp.copernicus.org/articles/22/3595/2022/acp-22-3595-2022-discussion.html

Long-term methane stable isotope analysis coupled with mole fraction measurement has been used to link isotopic signature to gas leaks in London, UK.

Location: Imperial College London, Huxley roof

Method: δ13CH4 values were made using a Picarro G2201-i isotopic analyser beginning in early 2018; 2 years continuous measurements 2018 - 2020. Simulations completed with UK Met Office Lagrangian dispersion model NAME.

Results: Isotopic source values revealed a predominance of natural gas CH4 with source values higher than -45 ‰ in ~74-80 % of the afternoon data. In contrast, simulated sectoral contributions using UK NAEI and EDGAR inventories showed the largest fractions from waste sectors, leading to a simulated underestimation of observed δ 13CH4.

These results suggest that natural gas leaks in London are under-reported in both inventories. The underestimation of mole fractions in the NAEI-25km and NAEI-2km might be accounted for by missing natural gas emissions in the NAEI inventory for London.

UK NAEI emissions are approximately 2.5 times smaller than the EDGAR emissions for the London area, but 8 % smaller than the EDGAR emissions across the UK.

Subtracting the 25 km NAEI emissions from the 25 km EDGAR emissions shows largest differences between inventories were in: London, Birmingham and the Leeds-Sheffield area, which have higher emissions in the EDGAR inventory.

(2022) Street-level methane emissions of Bucharest, Romania and the dominance of urban wastewater

https://doi.org/10.1016/j.aeaoa.2022.100153

An example of a city-level study that explores the source apportionment of methane emissions. (A paper similar to this one for London is soon to be published.)

Abstract: "*Mobile surveys were conducted in the urban areas of Bucharest while continuously measuring CH4 and C2H6 for locating enhanced CH4 mole fractions above local atmospheric background, which are referred to as a leak indication (LI). The flux rates were determined for identified clusters of LIs. An annual city-wide total emission estimate was calculated by scaling up the flux rates. Multiple locations, where CH4 exceeded the daily atmospheric background mole fractions, were measured for δ13CCH4, δ2HCH4, and C2:C1 ratios for tracing contributing CH4 sources. As Europe seeks to cut urban emissions, studies like this will be useful for identifying targets for mitigating emissions and for assessing future governmental regulation of greenhouse gas (GHG) emissions*."

(2019) Environmental baseline monitoring for shale gas development in the UK: Identification and geochemical characterisation of local source emissions of methane to atmosphere

https://doi.org/10.1016/j.scitotenv.2019.134600

Abstract: "*Baseline mobile surveys of methane sources using vehicle-mounted instruments have been performed in the Fylde and Ryedale regions of Northern England over the 2016–19 period around proposed unconventional (shale) gas extraction sites. The aim was to identify and characterise methane sources ahead of hydraulically fractured shale gas extraction in the area around drilling sites. This allows a potential additional source of emissions to atmosphere to be readily distinguished from adjacent sources, should gas production take place.*

The surveys have used ethane : methane (C2:C1) ratios to separate combustion, thermogenic gas and biogenic sources. Sample collection of source plumes followed by high precision δ13C analysis of methane, to separate and isotopically characterise sources, adds additional biogenic source distinction between active and closed landfills, and ruminant eructations from manure.

The surveys show that both drill sites and adjacent fixed monitoring sites have cow barns and gas network pipeline leaks as sources of methane within a 1 km range. These two sources are readily separated by isotopes (δ13C of −67 to −58‰ for barns, compared to −43 to −39‰ for gas leaks), and ethane : methane ratios (<0.001 for barns, compared to >0.05 for gas leaks). Under a well-mixed daytime atmospheric boundary layer these sources are generally detectable as above baseline elevations up to 100 m downwind for gas leaks and up to 500 m downwind for populated cow barns. It is considered that careful analysis of these proxies for unconventional production gas, if and when available, will allow any fugitive emissions from operations to be distinguished from surrounding sources."

(2019) Assessing London CO2, CH4 and CO emissions using aircraft measurements and dispersion modelling

https://doi.org/10.5194/acp-19-8931-2019

Pitt, J. R., Allen, G., Bauguitte, S. J.-B., Gallagher, M. W., Lee, J. D., Drysdale, W., Nelson, B., Manning, A. J., and Palmer, P. I.: Atmos. Chem. Phys., 19, 8931–8945.

Abstract: "*We present a new modelling approach for assessing atmospheric emissions from a city, using an aircraft measurement sampling strategy similar to that employed by previous mass balance studies. Unlike conventional mass balance methods, our approach does not assume that city-scale emissions are confined to a well-defined urban area and that peri-urban emissions are negligible. We apply our new approach to a case study conducted in March 2016, investigating CO, CH4 and CO2 emissions from a region focussed around Greater London using aircraft sampling of the downwind plume. For each species, we simulate the flux per unit area that would be observed at the aircraft sampling locations based on emissions from the UK national inventory, transported using a Lagrangian dispersion model. To reconcile this simulation with the measured flux per unit area, assuming the transport model is not biased, we require that inventory values of CO, CH4 and CO2 are scaled by 1.03, 0.71 and 1.61, respectively. However, our result for CO2 should not be considered a direct comparison with the inventory which only includes anthropogenic fluxes*."

(2017) Characterization of interferences to in situ observations of δ13CH4 and C2H6 when using a cavity ring-down spectrometer at industrial sites

https://doi.org/10.5194/amt-10-2077-2017

"An example study conducted on a UK compressor station, carried out by the LSCE Paris group a few years ago now and linked with an NPL survey of the site. This was from the early days of the laser-based instruments and there were difficulties in extracting good ethane data." (Dave Lowry, RHUL)

(2017) Evaluating methane inventories by isotopic analysis in the London region

https://www.nature.com/articles/s41598-017-04802-6

Abstract: "*A thorough understanding of methane sources is necessary to accomplish methane reduction targets. Urban environments, where a large variety of methane sources coexist, are one of the most complex areas to investigate. Methane sources are characterised by specific δ13C-CH4 signatures, so high precision stable isotope analysis of atmospheric methane can be used to give a better understanding of urban sources and their partition in a source mix. Diurnal measurements of methane and carbon dioxide mole fraction, and isotopic values at King's College London, enabled assessment of the isotopic signal of the source mix in central London. Surveys with a mobile measurement system in the London region were also carried out for detection of methane plumes at near ground level, in order to evaluate the spatial allocation of sources suggested by the inventories. The measured isotopic signal in central London (−45.7 ±0.5‰) was more than 2‰ higher than the isotopic value calculated using emission inventories and updated δ13C-CH4 signatures. Besides, during the mobile surveys, many gas leaks were identified that are not included in the inventories. This suggests that a revision of the source distribution given by the emission inventories is needed*."

(2017) Origins and trends in ethane and propane in the United Kingdom from 1993 to 2012

https://www.sciencedirect.com/science/article/pii/S1352231017301103

Abstract: "*Continuous, high frequency in situ observations of ethane and propane began in the United Kingdom in 1993 and have continued through to the present day at a range of kerbside, urban background and rural locations. Whilst other monitored C2 – C8 hydrocarbons have shown dramatic declines in concentrations by close to or over an order of magnitude, ethane and propane levels have remained at or close to their 1993 values. Urban ethane sources appear to be dominated by natural gas leakage. Background levels of ethane associated with long range transport are rising. However, natural gas leakage is not the sole source of urban propane. Oil and gas operations lead to elevated propane levels in urban centres when important refinery operations are located nearby. Weekend versus weekday average diurnal curves for ethane and propane at an urban background site in London show the importance of natural gas leakage for both ethane and propane, and road traffic sources for propane. The road traffic source of propane was tentatively identified as arising from petrol-engine motor vehicle refuelling and showed a strong downwards trend at the long-running urban background and rural sites.* ***The natural gas leakage source of ethane and propane in the observations exhibits an upwards trend whereas that in the UK emission inventory trends downwards****. Also,* ***inventory emissions for natural gas leakage appeared to be significantly underestimated compared with the observations. In addition, the observed ethane to propane ratio found here for natural gas leakage strongly disagreed with the inventory ratio***."

(2016) Spatial and temporal variability of urban fluxes of methane, carbon monoxide and carbon dioxide above London, UK

https://acp.copernicus.org/articles/16/10543/2016/acp-16-10543-2016-discussion.html

Long-term (3-year) measurements on methane fluxes in London.

Results: The wintertime increase above background in CH_4 concentrations and the accompanying enrichment in $\delta\ ^{13}C$ were consistent with North Sea natural gas and attributed to losses of CH_4 from over-pressurized pipelines in response to (or anticipation of) an increase in demand and to incomplete combustion upon boiler ignition.

Annual CH_4 emissions (72 tons km^{-2}) were more than double the LAEI value suggesting that sources are not as well characterized by the inventory. A shortfall in inventoried CH_4 emissions can be explained by the existence of temperature-dependent sources related to natural gas usage and perhaps also of biogenic origin (e.g. sewage).

(2017) Natural gas and climate change

https://pure.manchester.ac.uk/ws/portalfiles/portal/60994617/Natural_Gas_and_Climate_Change_Anderson_Broderick_FOR_DISTRIBUTION.pdf

Policy support paper that covers many aspects of natural gas and climate change, concluding that: *"By 2035 the substantial use of fossil fuels, including natural gas, within the EU's energy system will be incompatible with the temperature commitments enshrined in the Paris Agreement."*

The paper cites uncertainty in methane leakage (*from across the full supply chain, not specific to downstream distribution networks*) as critical in understanding the role that natural gas may play as a viable bridging fuel in the transition to a low carbon economy:

"In order to quantify the maximum level of EU natural gas consumption compatible with existing EU targets and the Paris Agreement, the relative lifecycle carbon intensity of a range of potential natural gas sources must be more fully understood, ***particularly methane leakage****."*

The research alludes to the challenge of super-emitter sources and the difficulty in identifying and assessing them:

"*The production and distribution of natural gas releases methane both deliberately and inadvertently. The exact amount varies widely across locations and production technologies, and through time at a given location.* ***Close monitoring shows that in most supply chains a small number of sites, or pieces of equipment, are responsible for a large proportion of methane emissions, however, they are difficult to identify a priori.*** *Leakage rates affect the relative contribution of methane to the climate change impact of natural gas supply chains*."

(2015) Plume mapping and isotopic characterisation of anthropogenic methane sources

https://www.sciencedirect.com/science/article/abs/pii/S1352231015002538

Methane stable isotope analysis coupled with mole fraction measurement has been used to link isotopic signature to methane emissions from landfill sites, coal mines and gas leaks in UK.

Location: London and SE England

Method: Targeted areas, when the methane plume was intercepted, air samples were collected and analysed using δ13C–CH4 isotopic analysis by CF-GC-IRMS (high precision method).

Results: Methane from landfill isotope signatures −60.2 ± 1.4 to −55.2 ± 0.6‰. Methane of constant origin for the methane of −36.3 ± 0.3‰ consistent with a dominantly thermogenic North Sea gas source. It suggests that there are other leaks in the gas distribution system along with the storage tank ("gasometer") that is located in the middle of the transect. 24 ppm north of Bacton where pipelines bring all southern and much northern North Sea Norwegian gas onshore, as well as gas from the interconnector pipeline to Belgium. Samples collected had 13C signatures of 36.3 ± 0.3‰ and −35.7 ± 1.2‰.