Fugitive natural gas emissions in York, United Kingdom: Updating the parameters of existing algorithms to be based on instrumental limitations.~~Fugitive emissions of natural gas in York, United Kingdom: Adapting existing algorithms parameters to be based on instrument specifications.~~

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Abstract

Reducing methane emissions has become increasingly important in recent years due to its importance for radiative forcing. One area of particular interest is the oil and gas sector, which often results in fugitive emissions of methane from natural gas distribution. ~~Of the many sources of methane, fugitive emissions from a country's domestic natural gas network are one that can have a direct impact on the citizens of a country.~~ Previous studies have shown the ability to detect these emissions by use of mobile surveys measuring methane, some of these use secondary co-emitted compounds as a means of confirming the nature of the emission. This study aims to adapt existing algorithms parameters by investigating the limitations of equipment used within the platform used for mobile surveys. ~~This has led to reduced enhancement parameters as well as reduced time clustering parameters.~~ These changes suggest that previous methods may underpredict the number of Leak Indications (LIs) by 53.5% with number of LIs detected in the old method being 27 and the new method detecting 58. The majority of these LIs were found to be emitting in a leak rate category of 0 - 2 L min^{-1}. Source appointment was included as a core step within the algorithm itself, this stage was shown to reduce the misassignment of LIs, suggesting the previous methodology may include emissions from pyrogenics and biogenics within their LI assignments.~~When source appointment was included as a core step~~

~~within the algorithm itself, the total fugitive natural gas emissions within a city was reduced from 185.10 L min^{-1} to 60.23 L min^{-1}, nearly three times lower.~~

1 Introduction

Following COP26 and the methane pledge *(European Commission and United States of America 2021)*, methane and its emissions have received increased attention. The pledge states that the signatories will attempt to reduce their methane emissions by 30% of their 2020 levels by 2030. This~~and~~ was brought about due to increasing concern over the potency of methane as a greenhouse gas. With a warming potential 28 times greater than CO_2 over a 100-year timescale and 84 times higher over a 20-year timescale *(IPCC, 2021)*.

65 % of all methane emissions are thought to be anthropogenic in nature and atmospheric methane has seen a consistent growth rate of > 5 ppb year^{-1} since 2007, with 2021 and 2022 seeing growth rates of 17.8 ppb year^{-1} and 14 ppb year^{-1} respectively *(Saunois et al., 2025)*, therefore understanding and mitigating anthropogenic methane is a key goal to comply with the global methane pledge.

Of the anthropogenic emissions, the agricultural sector has the largest contribution towards atmospheric emissions *(Saunois et al., 2025)*. Although there are means of reducing the emissions from these areas, including changing cattle, crop or land management as well as changing the feedstock of the cattle, for example grass silage to maize silage (*Bačėninaitė et al., 2022; Nisbet et al., 2025)*, these changes may still require time to implement, so this sector cannot be the sole focus in order to reach the 2030 deadline.~~Although the growth rate of methane concentrations in the atmosphere has slowed since the 1980s, it is still responsible for 20% of the warming caused by long-lived greenhouse gases since pre-industrial times *(Kirschke et al., 2013)*. The difficulty in reducing methane emissions is in part due to the wide range of emission sources, both natural and anthropogenic. 60% of methane emissions are thought to be anthropogenic in nature *(Karakurt et al., 2013, Saunois et al., 2020)* and of those emissions, by far the largest contribution arises from the agricultural sector with the two main areas of this being enteric fermentation from ruminant livestock like cattle and emissions from paddy fields *(Karakurt et al., 2013, Yusuf et al., 2012)*. Although there are means of reducing the emissions from these areas, such as changing the feedstock of the cattle, for example grass silage to maize silage (*Bačėninaitė et al., 2022)*, there is still a requirement for social change and land management change, both of which are difficult to implement for a 2030 deadline.~~

After agriculture the largest contribution to anthropogenic emissions is from the energy sector with oil, natural gas and coal having relatively similar contributions to methane emissions. Natural gas is of particular importance to the UK, with it being the 19th largest country emitter of methane from the natural gas network (*Scarpelli et al., 2022)*.

One of the sources of methane emissions from the natural gas network is fugitive emissions. A fugitive emission is an unexpected or unwanted emission of gas from a pressurised network that is not detected by standard means *(Sotoodeh, 2021)*. Within the natural gas network, they are commonly referred to colloquially as “Gas Leaks”, however the stigma surrounding

this term, both from industrial operators and the public, means the term fugitive emission is preferable to be used where possible.

In the United Kingdom in 2023, 63.5 billion cubic metres of natural gas was consumed *(Energy Institute, 2024)*. This is used in a range of applications including industrial use, electricity generation and domestic use. Of ~~the UKs~~the UKs natural gas consumption, 33.8 % is from the domestic sector *(DESNZ, 2024)*, with 73.8 % of households in England and Wales using mains gas for either heating or cooking purposes. *(Stewart et al., 2024)*, in 2022 it was estimated that 117 kT of methane was emitted as a result of fugitive emissions related to natural gas distribution *(NAEI, UK Emissions Data Selector)*.

Within the UK, after natural gas is either produced or imported, it is first transported through the National Gas' National Transmission System (NTS).~~The natural gas network consists of production or importing of natural gas; transport of high-pressure natural gas through the National Gas' National Transmission System (NTS),~~ a network of over 5,000 miles of high-pressure steel pipes and more than 500 above ground installations. This natural gas is then transported by one of several of the UK's Gas Distribution Networks (GDNs), a GDN first reduces the pressure from the NTS then oversees the pipework for pre-meter distribution of natural gas to homes and businesses.~~; and finally, distribution of natural gas to homes and businesses which occurs through the 4 Gas Distribution Networks (GDNs) in the UK.~~

~~A GDN first reduces the pressure from the NTS before then supplying natural gas to its customers.~~ The GDN responsible for York covers 2.7 million homes and businesses not just across Yorkshire, but also the northeast of England and northern Cumbria, which means the operation of tens of thousands of kilometres of pipework. This all leads to many unknowns in the locations of fugitive emissions. To combat this, many previous studies have implemented mobile measurement approaches centred around the detection of areas with elevated methane.

1.1 Previous Mobile Measurement Methodology

There have been many different approaches to designing an algorithm to detect fugitive emissions of natural gas, all of which utilise detection of an enhancement in CH_4 concentrations, the major component of natural gas. These algorithms define an enhancement based on whether CH_4 mixing ratios are higher than a certain value *(Phillips et al., 2016)*, are above a certain percentile in measured readings *(Hopkins et al., 2016, Chamberlain et al., 2016)* or by using an outlier detection model *(Keyes et al., 2020)*.

The paper upon which our methodology is based, *(von Fischer et al., 2017)*, defines Observed Peaks (OPs) as methane enhancements > 110% of a 2.5-minute rolling background of the mean CH_4 concentrations two minutes before and after each measured point, and that they occur over less than 160 m ~~*(von Fischer et al., 2017)* .~~ Enhancements occurring within 5 seconds of each other are then grouped together. Mobile surveys ~~It is required that drives~~ are repeated multiple times and Leak Indications (LIs) are determined after grouping OPs that occur within 20 m of one another and seeing which of these grouped clusters contain OPs from separate ~~drives~~mobile surveys. The LIs are then quantified into emission rates in L min^{-1}, using an equation derived from the results ~~from~~ of a controlled release experiment, shown in ***Equation 1***.~~.~~

$$(release\ rate\ /\ L\ min^{-1}) = 0.1178 + 0.08267 \times M - 0.005175 \times A + 0.08626 \times K \quad (Eq.1)$$

Where:

- M is the maximum CH_4 reading
- A is the peak area in ppm.metres ~~index~~
- K is the ratio of ppm.metres to maximum CH_4

This methodology was then further developed in *(Weller et al., 2019)*, this changed the baseline such that it was the median CH_4 value over 2.5 minutes, the spatial grouping of OPs to LIs from 20 m to 30 m and the quantification equation to ***Equation 2***.~~:~~

$$ln\,(excess\ CH_4 / ppm) = -0.988 + 0.817 \times ln\,(emission\ rate\ / L\ min^{-1}) \quad \text{(Eq. 2)}$$

Where the excess CH_4 term is the mean of all CH_4 enhancements associated with the resulting LI.

In *(Maazallahi et al., 2020)*, it was proposed that the existing methodology categorised certain burning emissions as fugitive emissions, to counter this an additional stage using CO_2 ratios with CH_4 was introduced to filter burning emissions.

Source attribution was also used in *(Fernandez et al., 2022)*, using isotopic measurements of CH_4 in addition to ethane: methane ratios.

Most recently in *(Tettenborn et al., 2025)*, the approach was changed further, adapting the quantification equation to be based on peak area as opposed to peak height. Resulting in the quantification equation shown in ***Equation 3***.

$$(\boldsymbol{release\ rate\ /\ L\ min^{-1}}) = \boldsymbol{exp(1.292\ ln(peak\ area) - 2.377)} \quad \text{(Eq. 3)}$$

Where *ln(peak area)* is the mean *ln(peak area)* of all OPs within the LI cluster.

-Variations of this algorithm has been used in many major cities across the US and Canada *(Ars et al., 2020; Weller et al., 2022)* as well as European cities *(Defratyka et al., 2021; Fernandez et al., 2022; Wietzel et al., 2023; Vogel et al., 2024)* and Asian cities (Joo et al., 2024, Ueyama et al., 2025, Umezawa et al., 2025). This paper attempts to detect smaller enhancements of methane by adapting detection and clustering ~~the~~ parameters to be specific to the limitations of the instrumentation used. The paper also explores the effect of introducing a source attribution filter at the OP stage of the algorithm and how this affects the number and the magnitude of LIs.

2 Methodology

2.1 Instrumentation

The WACL Air Sampling Platform (WASP) detailed in *(Wagner et al., 2021)* is the base for these measurements. T~~,~~ ~~t~~he inlet for the WASP~~'s sampling~~ is located at the front of the van on the driver's side, meaning that the vehicle will sample the centre of the road regardless of direction of travel. Since publication of *(Wagner et al., 2021)*, t~~T~~he WASP has since been updated to include a Quark-Elec QK-AS07-0183 for GPS readings. For the measurements surrounding natural gas, the WASP was equipped with a Los Gatos Microportable Greenhouse Gas Analyser (MGGA) for measurements of methane and CO_2, Iterative Cavity Enhanced Differential Optical Absorption Spectrometer (ICAD) for measurements of NO_x (NO_2 + NO), and an

Aerodyne Tuneable Infrared Laser Direct Absorption Spectrometer (TILDAS) Laser Trace Gas Analyser for measurements of ethane *(Yacovitch et al., 2014)*. Measurements of ethane are calibrated using a three point calibration of a high standard of ethane (17.5 ppb), medium (2.5 ppb) and a zero, where calibration standard concentrations were confirmed via GC-MS. For each mobile survey~~drive~~ a calibration is performed before and after the mobile survey~~drive~~ itself, a linear regression is performed to find the slope and intercept of the calibration concentrations versus measured concentrations. The average is taken for the two calibrations to account for instrument drift during the sampling ~~drive~~ survey and the resulting equation, ***Equation 4***, is then used to apply a correction to ethane concentrations.

$$C_2H_{6\,corrected} = C_2H_{6\,uncorrected} \cdot m + c \quad (Eq. 4)$$

Where:

- m = Gradient of calibration concentration vs mean response averaged over the two calibrations
- c = Intercept of calibration concentration vs mean response averaged over the two calibrations

2.1.1 Instrument Response Time

Response time of the MGGA is reported as < 0.5 s from the manufacturer's specification. The response rate of the TILDAS however was unknown. The TILDAS is capable of recording measurements at a rate of 10 Hz, however, the flow rate through the instrument needed to be altered to make these measurements true to the 10 Hz values. Originally, the inlet to the TILDAS had two valves in series, Stainless Steel Integral Bonnet Needle Valve, 0.37 Cv, 1/4 in. #SS-1RS4 and an electronic Upstream Flow Control Valve, 10,000 sccm, 0.25 in. Tube, Viton Seal #0248A-10000SV which allows small changes to maintain the internal pressure at 70 Torr. With the two valves in series, the instrument is unable to achieve a high enough flow rate for true 10 Hz measurements. Moving the valves to be parallel, the instrument was able to achieve a flow rate close to 5 Hz, at this point it was found that the pump was the limiting factor for the flow rate of the instrument.

These changes to increase the flow rate of the instrument were made to allow for a response time as close to that of the MGGA as possible. To find the accurate response time of the TILDAS, an experiment was devised whereby a high concentration of ethane (17.630 ± 0.715 ppb, measured via GC-MS) was flowed through the TILDAS and switched to ambient air 10 times on 2 separate valve setups for a total of 20 repeats of low-high-low transitions in the concentration of ethane. The transition times were located by eye and then the transition time to go from 90% of the maximum value to 10% of the maximum value was calculated *(Symonds, 2017)*, an example of the high to low transition with the 90 % and 10 % limits is shown in ***Figure 1***. The transition time on the first valve ranged from 0.7 – 1.1 s with a mean value of 0.9 s, the second valve having responses ranging from 0.7 – 1.4 s also with a mean response of 0.9 s, giving confidence in a sub 1 s response rate from the TILDAS and therefore showing the capability of a sub 1 s response in both instruments. The data however was still averaged to 1 s with a 1 s clustering time due to the data now being limited by the data acquisition rate of the WASP's GPS.

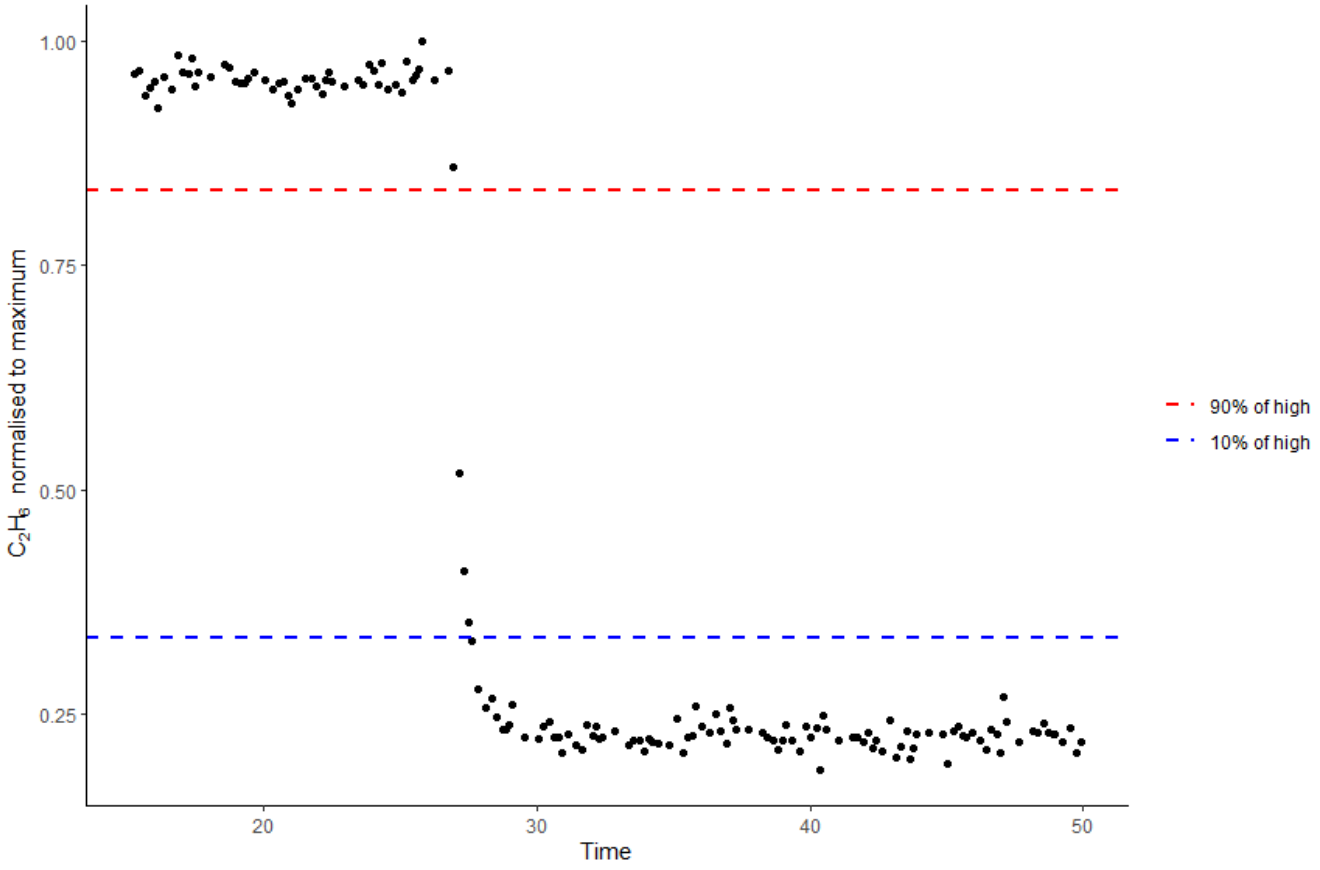

Figure 1: *Example response transition of TILDAS high concentration to low concentration, normalised to maximum recorded response*

2.1.2 Variation in methane measurements

Previous algorithms define an enhancement as being higher than 1.1 times a 2.5 minute rolling median background. This work however seeks to understand if this parameter holds true for the specific instrumentation used in the mobile surveys. To understand what this parameter may be, a variance experiment. The standard deviation of methane measurements over this period was calculated to understand the minimum detectable enhancement for the methane detection algorithm. ~~For better understanding of minimum detectable enhancements of methane possible with the equipment available a variance experiment using compressed air flowing through the instrument for 2 hours. The standard deviation of methane measurements over this period was calculated to inform the enhancement criteria for the methane detection algorithm.~~

For 2 hours compressed air was flown through the Los Gatos MGGA, measured at a median value of 7.2 ppm, the standard deviation of these results was found to be 0.006 ppm. An enhancement criteria was proposed as ~~The enhancement criteria was~~

~~calculated as~~ 5 times this standard deviation divided by the median baseline, resulting in an enhancement criteria of ~~in this case~~ 1.005 times the baseline. However, this assumes a stable baseline that is replicated in the field. In reality, when applying this enhancement criteria it leads to the detection of enhancements that are too small to be reliably quantified.

Instead, the methane mixing ratios measured during each mobile survey, were collated and the standard deviation was calculated for each mobile survey. Again enhancement criteria was calculated as anything larger than 5 times the standard deviation divided by the median of methane mixing ratios, this was done for each mobile survey and resulted in a median enhancement criteria of 1.01 times the baseline. However as this would result in detection of enhancements that would be very small and could be diffuse or from enhancements that occurred far from the roads and therefore harder to quantify as true natural gas emissions, a larger enhancement criteria of 1.05 times the background was selected. This ensured there was still a large difference from the original methodologies criteria, while still remaining within the instrumentation known variation.

~~Instead, the variation in the calculated baseline for drives was found and it was therefore determined that an enhancement of 1.05 times the baseline should be used instead.~~

2.2 Driving Route

York is a city in the north-east of England with a population over 200,000. A driving campaign took place over two separate weeks in May and June of 2024 resulting in 18 ~~drives~~ mobile surveys of a "flower petal" route, shown in ***Figure 2***, staying within the outer ring roads of the A64 and A1237 and mainly sampling residential areas of the city.~~,~~ T~~t~~he majority of the roads sampled on the route were only driven in one direction but due to the position of the sampling inlet this allowed the middle of the road to be ~~the~~ sampled ~~point~~regardless of the direction of travel. The route was driven 18 times as, in order to capture~~this number of times due to findings that to capture~~ > 90 % of emissions, a route should be driven at least 5 – 8 times over separate days *(Luetschwager et al., 2021)*. The route was chosen as it covers multiple different neighbourhoods within York, but was not intended to be used to compare measurements to the cities emissions inventory due to only covering a small fraction of the total miles of road within the York urban area, 27 miles of a total 507 administered by the local authority *(Department for Transport, 2025)*.

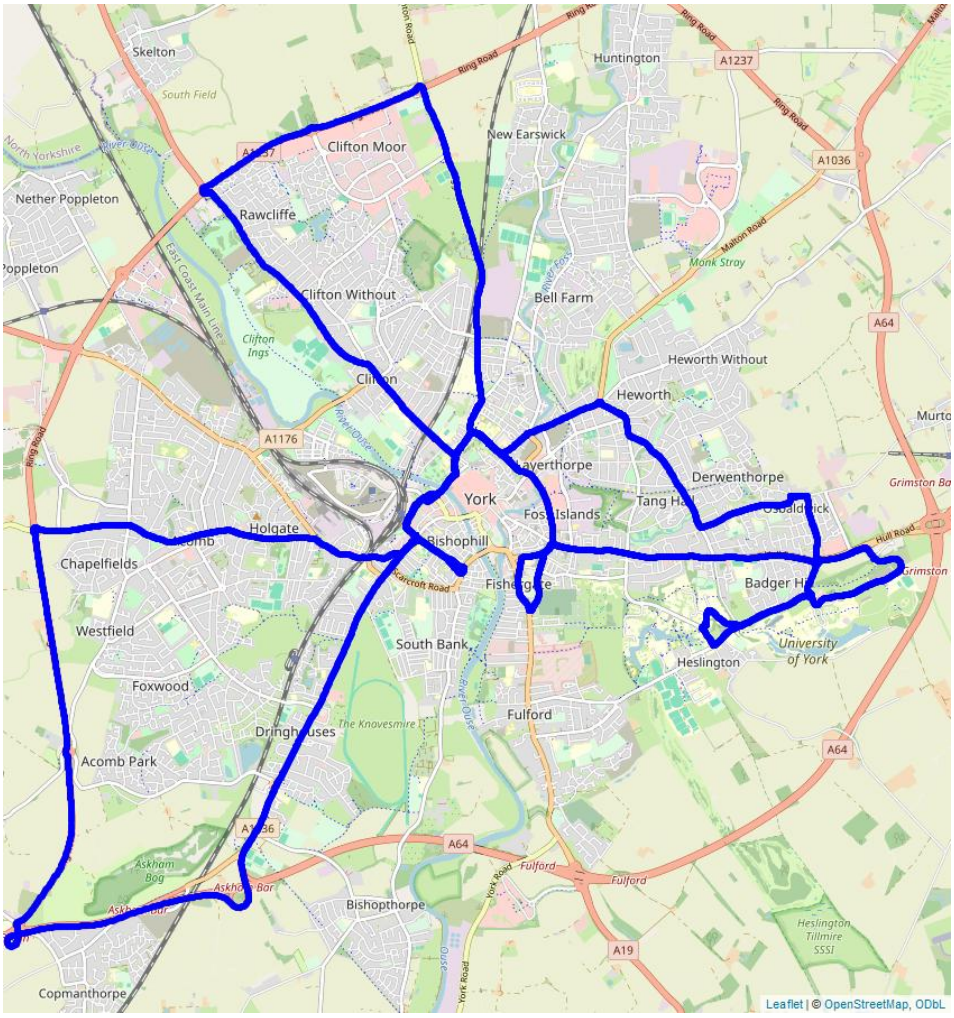

Figure 2: *Map of the route taken in WASP surveys.*

2.3 Enhancement Detection Algorithm

The original algorithm, used in *(Weller et al., 2019)*, has been adapted following the findings in ***2.1.1*** and ***2.1.2***. OPs are clustered within 1 s as opposed to 5 s. With a faster instrument response, it was expected that the measurements would more readily distinguish between two separate enhancements that occurred spatially close to one another. By clustering over a time of 5 s, assuming an average speed of 20 miles per hour (8.9 m s^{-1}), this would mean the potential to cluster together enhancements 44.5m apart, whereas a cluster time of 1 s would at most be clustering enhancements 8.9 m apart, the reason for this change was discussed in ***2.1.1***. Enhancement criteria was also changed, instead of an enhancement needing to be more than 110 % of the baseline, it must be 105 % of the baseline. This allows detection of smaller enhancements, the change was discussed in ***2.1.2***. LI determination occurred after identifying the source type of each OP, ensuring LI analysis occurs only

on OPs of the same source type, to further reduce the chance of comparing long standing thermogenic fugitive emissions with possible nearby single occurrence pyrogenic or biogenic emissions.~~Unlike the previous iterations of the algorithm, OPs are clustered within 1 s as opposed to 5 s. With a faster instrument response, it was expected that the measurements would more readily distinguish between two separate enhancements that occurred spatially close to one another. Additionally, by clustering over a time of 5s, assuming an average speed of 20 miles per hour (8.9 m/s), this would mean the potential to cluster together enhancements 44.5m apart, whereas a cluster of 1s would at most be clustering enhancements 8.9 m apart, the reason for this change was discussed in *2.1.1*. Enhancement criteria was also changed such that instead of an enhancement needing to be more than 110% of the baseline value to instead being an enhancement related to the results of the variance experiment to allow detection of smaller enhancements, this variation is discussed in *2.1.2*. LI determination occurred after identifying the source type of each OP, to allow LI analysis to occur on OPs of the same source type, to further reduce the chance of comparing long standing thermogenic fugitive emissions with possible nearby single occurrence pyrogenic and biogenic emissions.~~

2.4 Controlled Release Experiment

To attain a quantification equation specific to the equipment used at York, a controlled release experiment was conducted at the Bedford Aerodrome. The controlled release took place at the Bedford Aerodrome over 5 days in June of 2024. A MiniCRF was deployed to manage releases of methane and ammonia, while a MidiCRF was deployed for releases of ethane. In total there were 41 releases lasting an average of 30 minutes each. Releases consisted of varying amounts of methane (0.2 – 70.48 L min^{-1}), ethane (0 – 7.01 L min^{-1}) and ammonia (0 – 7.87 L min^{-1}) to reflect a range of methane sources, including natural gas and farm emissions. Releases were from a mixture of linear vertical releases, a muti emission point ring, multi point source emissions and single point releases, occurring at heights ranging from ground level to 3 m elevation. Over the course of the experiment wind speeds were measured using four Gill Met Pak Pro instruments deployed at 3 m, 6 m, 9 m and 12 m~~tres~~ elevation, winds were recorded as 1 minute vector averages. Average wind speed over the 5 days was 3.87 m s^{-1} with wind speeds ranging from 0 - 9.75 m s^{-1}. During each release, an initial period was spent locating the plume before sampling the plume at set distances for 10 repeats~~-~~, the platform then stepped ~~before stepping~~ further away in distance for another set of 10 repeats, , this continued ~~repeated~~ until the plume was either lost, or a lack of driveable ground was left available.~~,~~ I~~i~~t was noted that larger releases were detectable further away.~~,~~ h~~H~~owever, as the data from the controlled release was intended to be used in quantifying sub-road and near-road fugitive emissions of natural gas, a maximum distance of 30 m from the controlled release point was applied due to this reflecting the maximum road widths typically found within a city like York *(Essex Planning Officers Association, 2018).*

Of the 41 releases conducted in the controlled release, only 26 releases were able to be used for processing data, this is due to several reasons, including some releases not having detectable enhancements. Within these 26 releases, 3525 methane enhancements were detected over distances between 5.8 m and 382.1 m from the release point;~~,~~ the majority of releases detected further away from the release point were from higher emission rate releases. When this was filtered to include

enhancements from only 30 m from the release point this resulted in including 1226 enhancements from 23 ~~drives~~mobile surveys. Density plots of number of detected enhancements against distance from source are shown in ***Figure 3*** for all detected enhancements and ***Figure 4*** for enhancements detected within 30 m from the source.

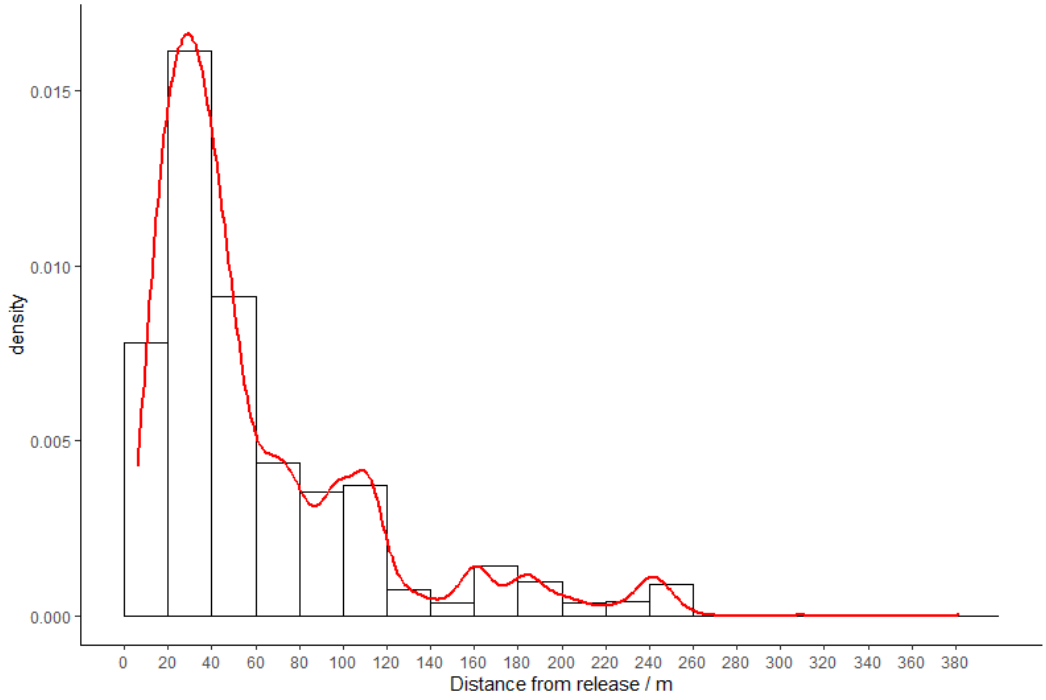

Figure 3: *Density plot of number of detected enhancements against distance from release point*

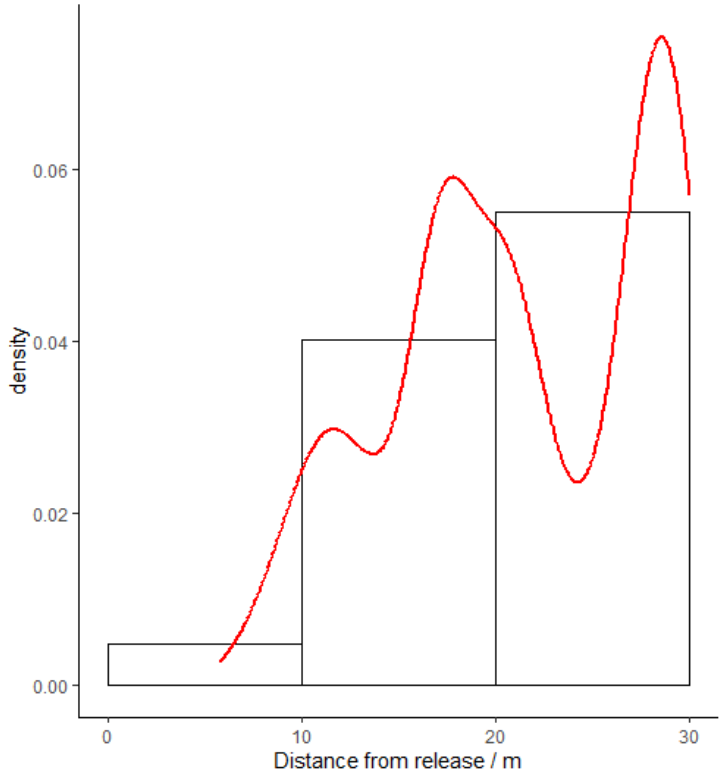

Figure 4: *Density plot of number of detected enhancements against distance from release point (Limited to 0 – 30 m)*

2.4.1 Quantification equation

There has been much development and advancement in the last few years on the use and application of "Advanced Mobile Leak Detection" systems for natural gas emissions detection and reporting. The original methodologies, where algorithms were developed to convert peak height maxima of measured methane plumes to estimated emission rates *(Weller et al., 2019)* have been superseded with plume area algorithms *(Tettenborn et al. 2025)* which are instrument and vehicle speed agnostic. However, this is still not a precise conversion, and can only be treated as a generalised guide to emissions estimation as factors such as wind, instrument inlet location and local variability due to buildings and unknown source locations.

In order to reduce the uncertainty for the WASP as much as possible, we present the results of a 1-week controlled release experiment conducted under variable wind conditions in a simple open field environment. Whilst this does not replicate the complex conditions of an urban setting, previous work in *(Tettenborn et al. 2025)* shows that combined results from both urban and open field settings can be combined to give a generalised trend to create their plume area emission algorithm. For the WASP, the setup is slightly different to the work in *(Tettenborn et al. 2025)*, with the WASP's inlet located on the driver's side at low elevation. This may influence the impact of vehicle turbulence on the measurements and the difference in elevation

will lead to a different vertical section of the plume being sampled. A comparison between the results of the York controlled release, and the *(Tettenborn et al. 2025)* methodology averages are shown below in ***Figure 5***. All data shown is for downwind transects where the plume was intercepted at a maximum of 30 m from the controlled release location. The plume area is calculated as a function of distance travelled (as opposed to time) to correct for vehicle speed differences as done in the original *(Tettenborn et al. 2025)* work.

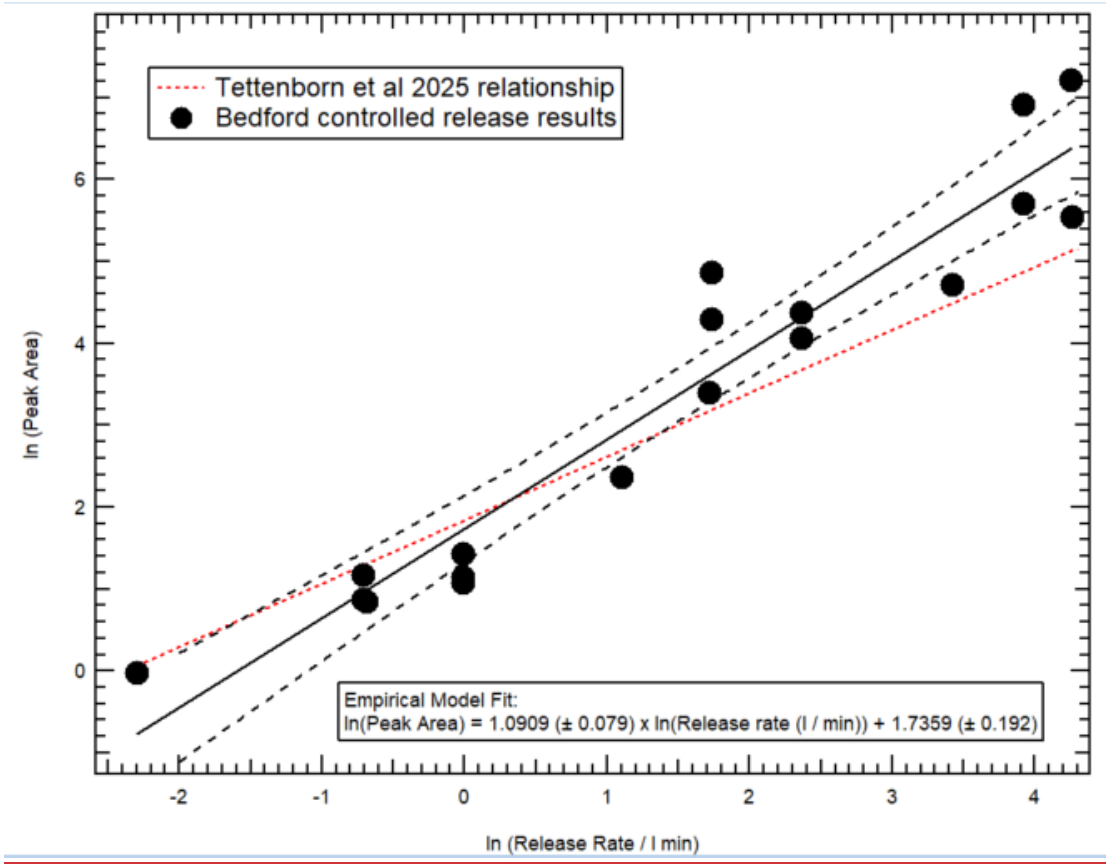

Figure 5: *Peak area vs actual release rate for plume transects within 30m of release. Data shown is an average of multiple transects (at least 10) for each release.*

As it can be seen in ***Figure 5***, whilst the general trend of increasing plume area with release rate is adhered to, the gradient of the trend is steeper – implying that a near-ground based inlet is potentially more capable of ascribing differences in emission rates.

One of the expected limitations of these algorithmic methods is the assumption that the effect of windspeed is ignored, which given the importance of windspeed in more distal emissions modelling such as Gaussian plume modelling from vehicles *(Dowd et al. 2024)* may be an oversight. Using the 3 m mast, 1 Hz wind data (averaged to 1 minute data) located on site at the

controlled release, the wind data is incorporated into the algorithm to give a volumetric correlation with the release rate according to ***Equation 5***.

$$wind\ speed\ \times \int_{plume\ start}^{plume\ end} [CH_4] \quad \text{(Eq. 5)}$$

The results of the integration of the wind speed into the algorithm are shown below in ***Figure 6***. Possibly somewhat surprisingly, there is a slight decrease in the goodness of fit to the relationship – potentially due to plume dynamics close to source not being immediately controlled by the atmospheric conditions, but the dynamics of the emission. This may also provide evidence as to the reasons why the results of previous studies have ended up with metrics that would at first hand seem unlikely to be able to produce reliable results from atmospheric dispersion principles. Given this result, that it seems to be as robust to consider wind as to not, it may be prudent for future controlled release experiments to focus on recreating the conditions of gas migration prior to emission to the atmosphere to see if this result still holds true.

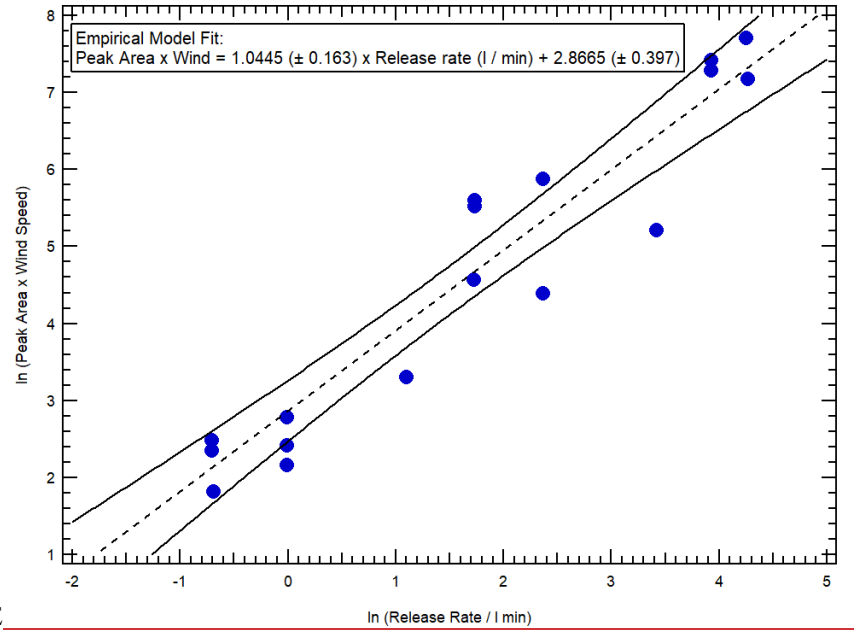

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Figure 6: *Peak area x Wind speed vs actual release rate for plume transects within 30m of release. As with Figure X, data shown is an average of multiple transects (at least 10) for each release.*

Due to these findings, the quantification equation used within York mobile surveys is shown in ***Equation 6***.

$$ln(release\ rate\ /\ L\ min^{-1}) = 0.9167 \times ln(Peak\ Area) - 1.7359 \quad \text{(Eq. 6)}$$

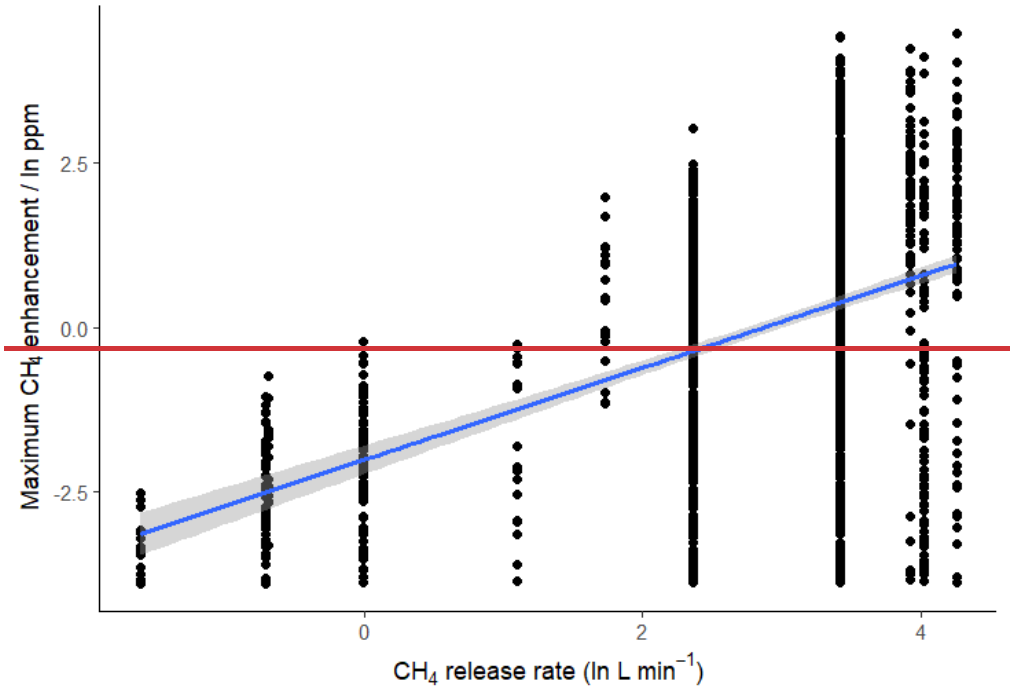
Additionally, leak rates were then reported within bins, similar to that within *(Tettenborn et al., 2025)*, where three possible bins were assigned; high (> 40 L min^{-1}), medium (6 - 40 L min^{-1}) and small (< 6 L min^{-1}). This was adapted for the York surveys, the small category was changed to 2 - 6 L min^{-1} and a new category, very small, was introduced which contained leak

rates of 0 - 2 L min^{-1}. This change was introduced due to the lower enhancement criteria within the York methodology which allows for detection of much smaller fugitive emissions.

It is important to note that these results are only suitable for the specific set up utilised here, and should not be more widely applied without corroboration with other instruments or platform packages.

~~A quantification equation to calculate emission rate from CH_4 measurements could be calculated using enhancements that were detected within 30 m of the controlled release emission point, to better reflect the enhancements likely to be detected from under road natural gas emissions. By first using the methane detection algorithm a linear regression model could then be used to find a relationship between the release rate and the maximum enhancement. 1226 enhancements were detected from 23 separate releases that were within 30 m of the emission point. These enhancements were determined from CH_4 releases ranging from 0.49 to 70.48 L min^{-1}. A linear regression was then performed using the equation:~~

~~$$ln\,(maxexcess\,CH_4) = b_0 + b_1 * ln\,(emission\,rate) + \varepsilon, \varepsilon \sim^{iid} N(0, \sigma^2)$$~~



~~***Figure 5:*** *Plot of the ln of the maximum detected CH_4 enhancement against the ln of the known CH_4 release rate.*~~

~~This resulted in the quantification equation;~~

~~$$ln\,(maxexcess\,CH_4) = -1.2668 + 0.6323 * ln\,(emission\,rate)$$~~

2.4.2 Instrument Lag Time

For each of the releases, the lag time between detecting an ethane enhancement and a methane enhancement was calculated. ~~With knowledge of~~ Due to the response times of the instruments, it was expected that the TILDAS would respond to an enhancement before the MGGA, however, this assumes that both instruments receive the same packet of air at the same time, while, in reality, the packet of air will take a different amount of time to flow through manifold to each instrument. To find this more accurate lag time of the instruments, the maximum methane enhancement for each pass was found, following this the maximum ethane enhancement was found (that occurred within 5 seconds of the methane). The ~~5~~ resulting 10 second window was selected as on transects of the controlled release the van travelled at roughly 20 miles hour^{-1} ~~ph~~. Over the course of 10 s (5 s either side of the methane maximum) this would mean 85 m covered in the van, the average length of a transect being 180 m. The time lag between ethane and methane showed that in most cases (88.1 %), maximum ethane concentration preceded maximum methane concentration with a mean lag of 2.7 s before and a median of 3.8 s before. Observing a window of max methane to 5 s before max methane resulted in a mean lag of 3.3 s from ethane to methane and a median lag of 3.9 s. This helped inform the detection algorithm to look for maximum ethane within a window only up to 5s before the maximum methane. Density plots showing the time lag of maximum ethane from maximum methane are shown in ***Figure 7*** for the full 10 second time window and ***Figure 8*** for up to 5 seconds before the time of maximum measured methane.

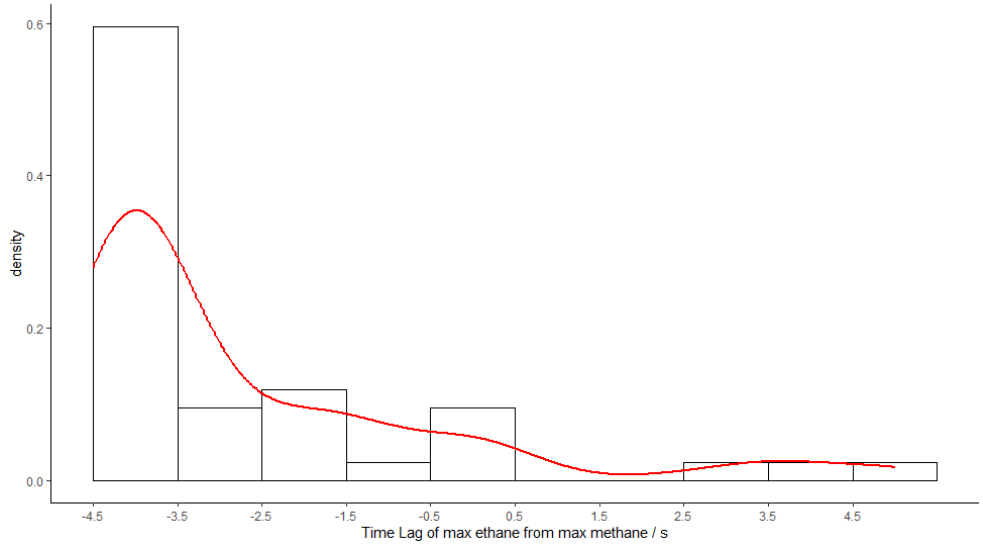


Figure ~~7~~6: *Density plot of time lag of the maximum ethane measurement from the maximum methane measurement.*

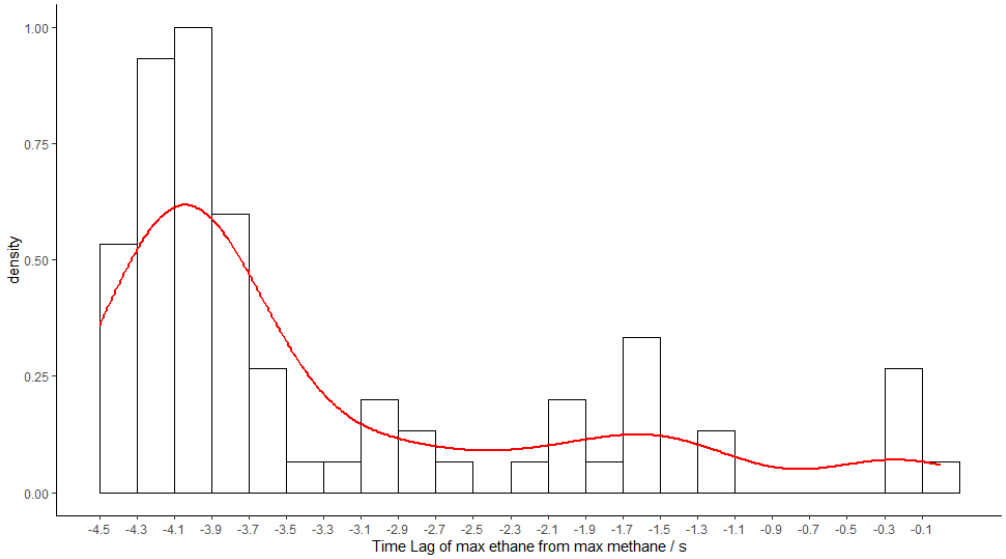


Figure ~~7~~8: *Density plot of time lag of the maximum ethane measurement from the maximum methane measurement (Only including ethane measurements that precede the methane)*

2.5 Source Appointment

Source determination using ethane-methane ratios has been shown to be effective, due in part to the knowledge that ethane is present in measurable quantities in thermogenic gas but not biogenic gas ***(****Fernandez et al., 2022)*~~,~~, ethane:methane (C_2:C_1) ratios can be used in order to determine the source of a methane emission. Demonstrated in *(Fernandez et al., 2022; Defratyka et al., 2021; Lowry et al., 2020; Yacovitch et al., 2014)*, C_2:C_1 < 0.005 may be~~is~~ associated with biogenic sources, > 0.005 to < 0.09 are thermogenic and > 0.1 are considered pyrogenic or combustion. Ideal examples of these relationships are shown in ***Figure 9***. In order to calculate these ratios methane and ethane values must first be aligned in time, ~~To complete this time alignment of methane and ethane values needs to be completed~~ due to them being measured on separate instruments, the criterion for aligning these times is based on the results from ***2.4.2***. Additionally, enhancements are removed where the R^2 of CO_2:CH_4 is greater than 0.9 to ensure no combustion sources are wrongly assigned as thermogenic.

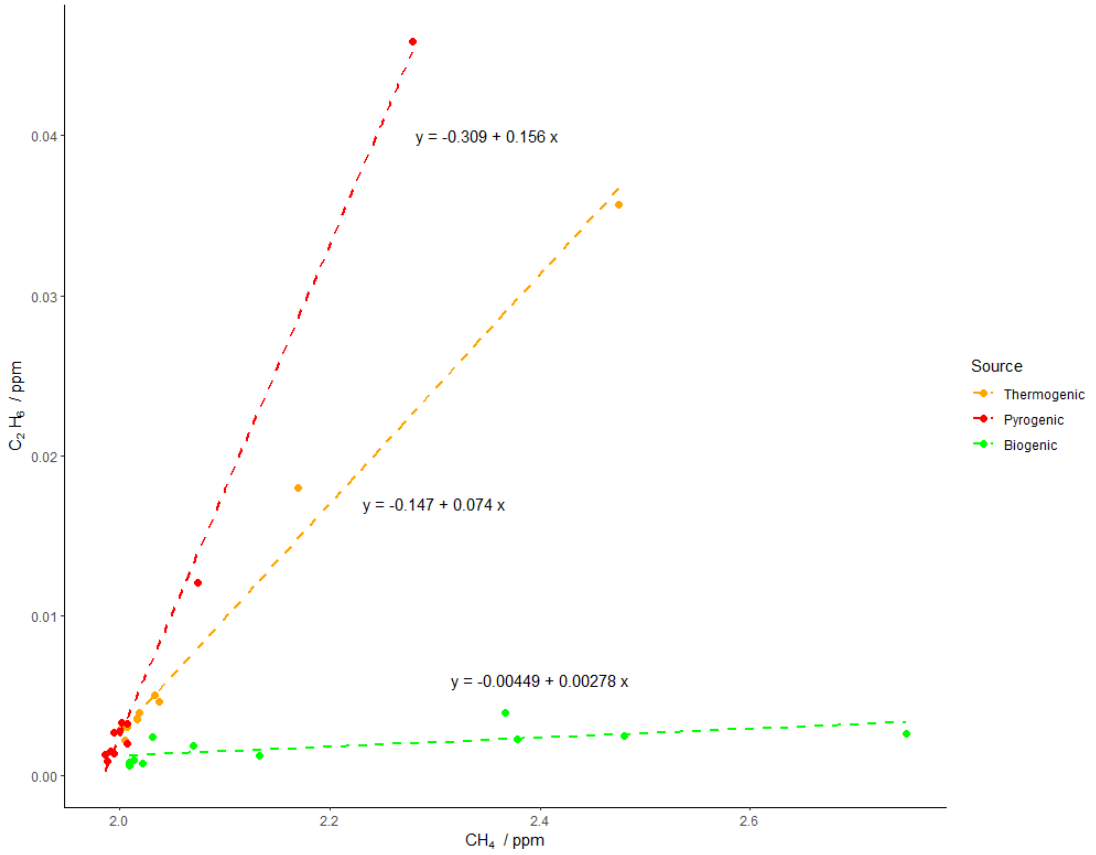


Figure ~~9~~8: *Relationship between CH_4 and C_2H_6 for three OPs of different source types located during the sampling campaign.*

3 Results

3.1 Results of York ~~Drives~~mobile surveys

17 ~~drives~~ mobile surveys were conducted across the route of York, the raw data was taken from 10Hz files for methane (MGGA) and ethane (TILDAS) and time averaged to 1 Hz data to be of the same response time as the WASPs other internal components (e.g. GPS) , a colour map of the resulting measured methane concentration is shown in ***Figure 10***.. The data was then processed to remove data taken when speeds were 0 or > 40 miles hour^{-1}~~ph~~ as well as removing data within the area of Wolfson Atmospheric Chemistry Laboratories due to this also being the location within which calibrations and other instrument tests were conducted. A rolling 2.5-minute median background of CH_4 was then ~~taken~~ applied and enhancements were determined as any CH_4 measurement taken that was greater than 1.05 times the calculated background.~~,~~ T~~t~~he enhanced readings were then clustered such that any elevated reading within one second of another were assumed to correspond to the

same enhancement. These enhancements were then spatially averaged such that 4687 OPs were detected over the course of the 18 ~~drives~~mobile surveys.

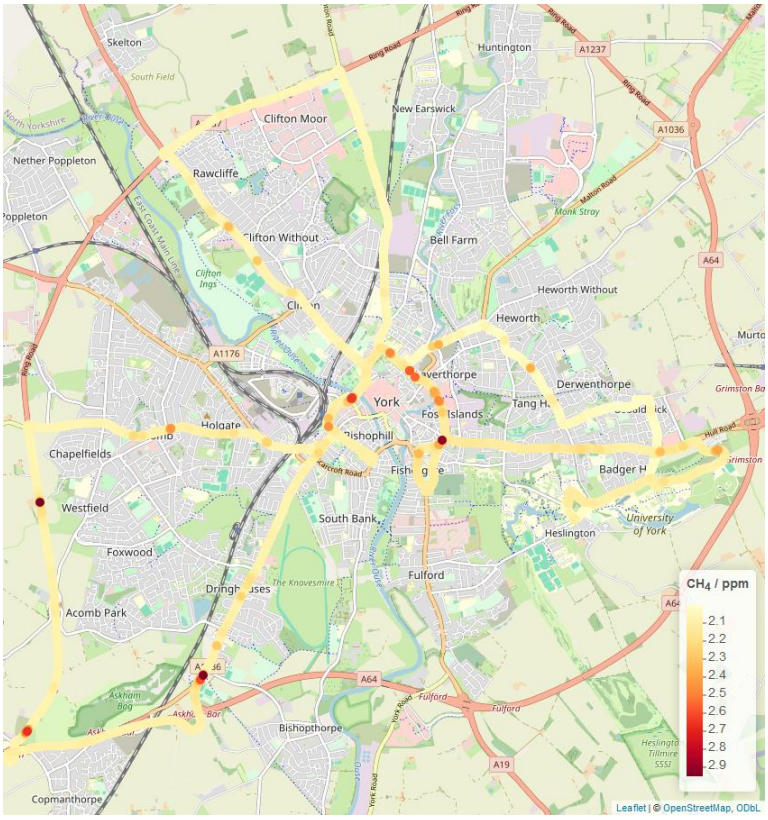


Figure ~~9~~10: *Colour map of CH_4 concentration from one of the York ~~drives~~mobile surveys.*

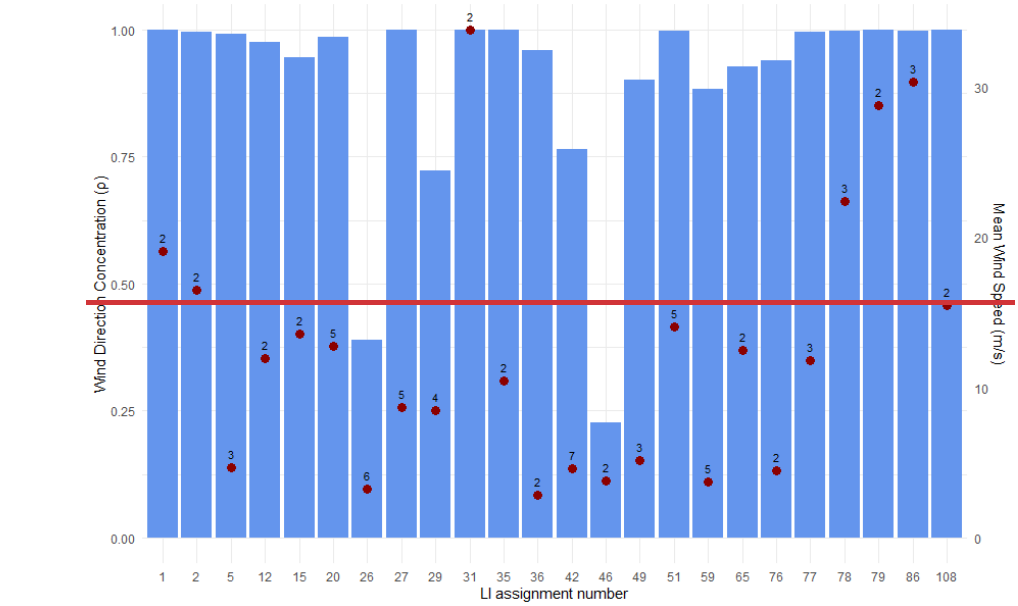
For each of these OPs the maximum ethane value was found from the time of maximum methane to 5 seconds prior. The two instruments' data were then aligned for each OP such that time of maximum methane was equal to time of maximum ethane measurement. A linear regression was then taken of values from 5 seconds prior to the maximum methane to 5 seconds after and a source type was assigned such that C_2:C_1 < 0.005 is associated with biogenic sources, > 0.005 to < 0.09 are thermogenic and > 0.1 are considered pyrogenic or combustion.

This meant that of the 4687 OPs, 1778 (~~38.1~~37.8 %) were found to be thermogenic in origin. Each Thermogenic OP was then clustered to find others within 30 m of one another and filtered to ensure that clusters contained OPs occurring on at least two separate ~~drives~~mobile surveys, to remove any OPs occurring from an event happening on only one ~~drive~~mobile survey. The remaining clusters were then averaged into LIs such that the ~~CH_4 enhancement was the maximum from the OPs and the~~ latitude and longitude were~~as~~ a weighted spatial average, resulting in 24 t~~T~~hermogenic LIs from the 1778 thermogenic OPs. Leak rate

was determined using the equation present in ***2.4.1*** using the mean *ln(peak area)* of all OPs within the LI cluster. The smallest leak rate was determined to be 0.0126 L min^{-1} and the largest being ~~16.04~~ 4.13 L min^{-1}, when assigned to bins 2 were small (2 - 6 L min^{-1}) and 22 were very small (0 - 2 L min^{-1}).~~.~~ When the source type filter is omitted, this results in 58 LIs with leak rates ranging from 0.0124 to ~~23.10~~4.70 L min^{-1}, when assigned to bins 9 were small (2 - 6 L min^{-1}) and 49 were very small (0 - 2 L min^{-1}).~~ In terms of cumulative leak rates, without source attribution cumulative leak rate would be 185.1 L min^{-1} whereas when source attribution was factored in a cumulative leak rate of 60.2 L min^{-1} was found.~~

3.1.1 Industry applicability

As many gas distribution companies have signed up to voluntary emission reporting programmes, such as the Oil and Gas Methane Partnership (OGMP) 2.0, they are now obligated to report emissions through measurement based methods. One of the most popular methods for such a reporting programme is through comprehensive, repeated vehicle based measurement surveys of an operator's gas network. Here, we have a repeated route of measurements where thermogenic emissions have been reported at certain locations throughout the campaign. It is therefore interesting from a mitigation perspective to investigate how many times each of those thermogenic emissions were identified over the course of the campaign.



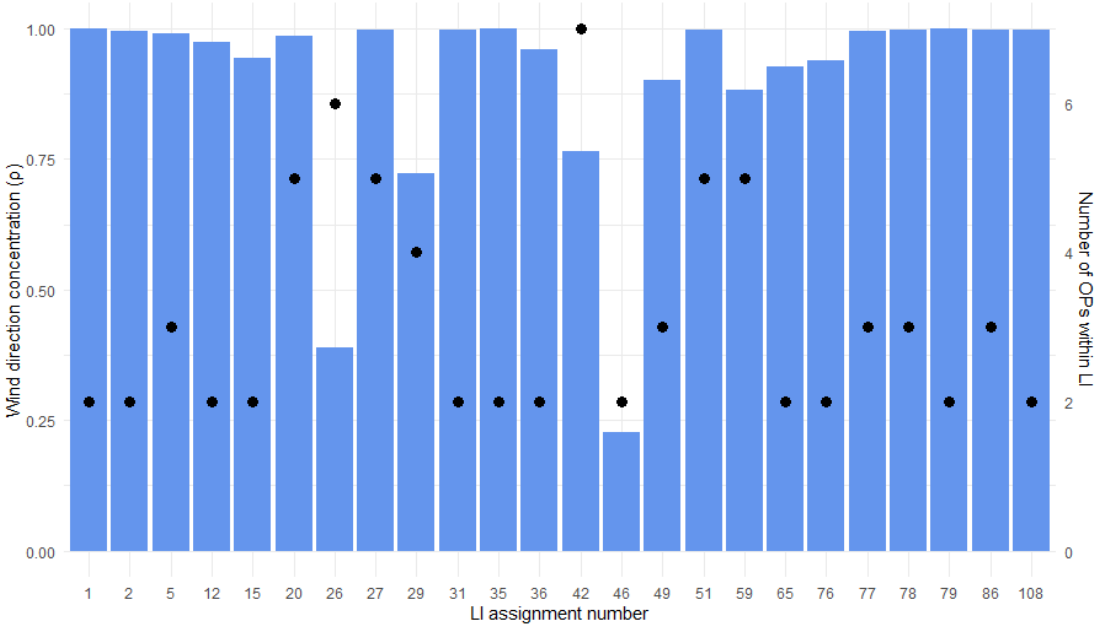


Figure 1~~0~~1: *Wind direction consistency and and number of OPs~~wind speed~~ per Thermogenic leak indication ~~(Labelled with number of enhancements in each LI)~~*

The effect of wind on detection of LIs was initially investigated by calculating the mean resultant length of wind directions when a Thermogenic OP was detected. This was calculated using ***Equation 7***.~~the equation:~~

$$\rho = \frac{1}{n}\sqrt{(\sum_{i=1}^{n} cos\theta_i)^2 + (\sum_{i=1}^{n} sin\theta_i)^2} \quad (Eq.7)$$

Where:

- ρ is mean resultant length
- n is number of data points
- θ_i is the angle in radians

For this analysis ρ is close to 1 when the wind directions are concentrated (similar) and close to 0 when more dispersed. ***Figure 10*** shows that for the majority of LIs detected in York ρ is close to 1, suggesting that most LIs occur away from the road and require correct wind direction in order to detect them. For LIs where ρ is lower it can also be noted that the mean wind speeds also tend to be lower, suggesting that these LIs may occur closer to the mobile platform therefore not needing a specific direction of wind in order to detect them.

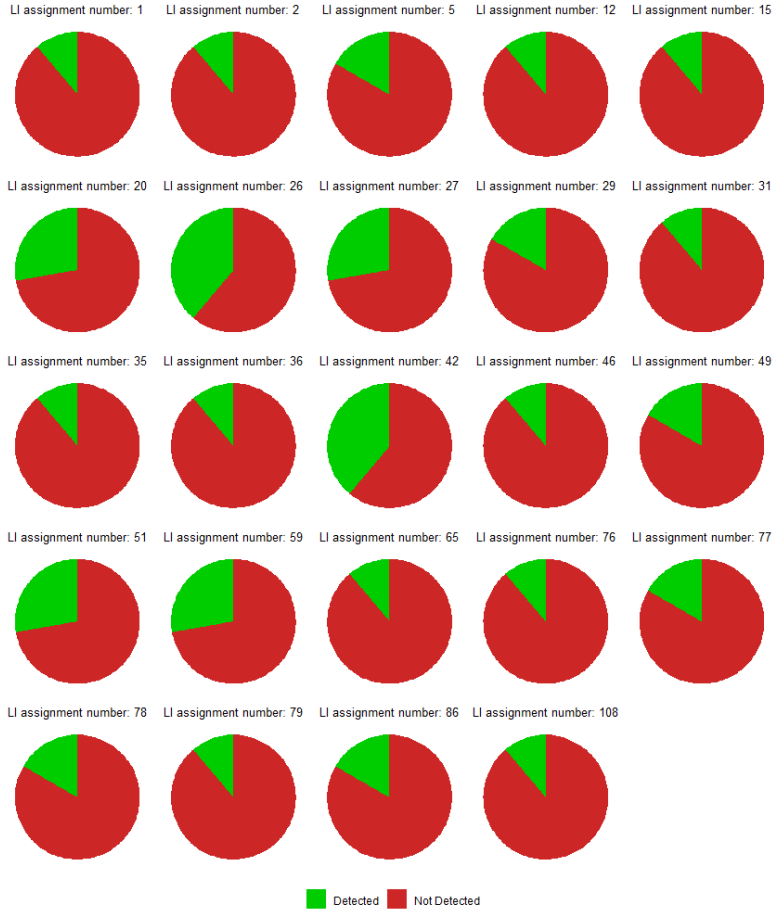


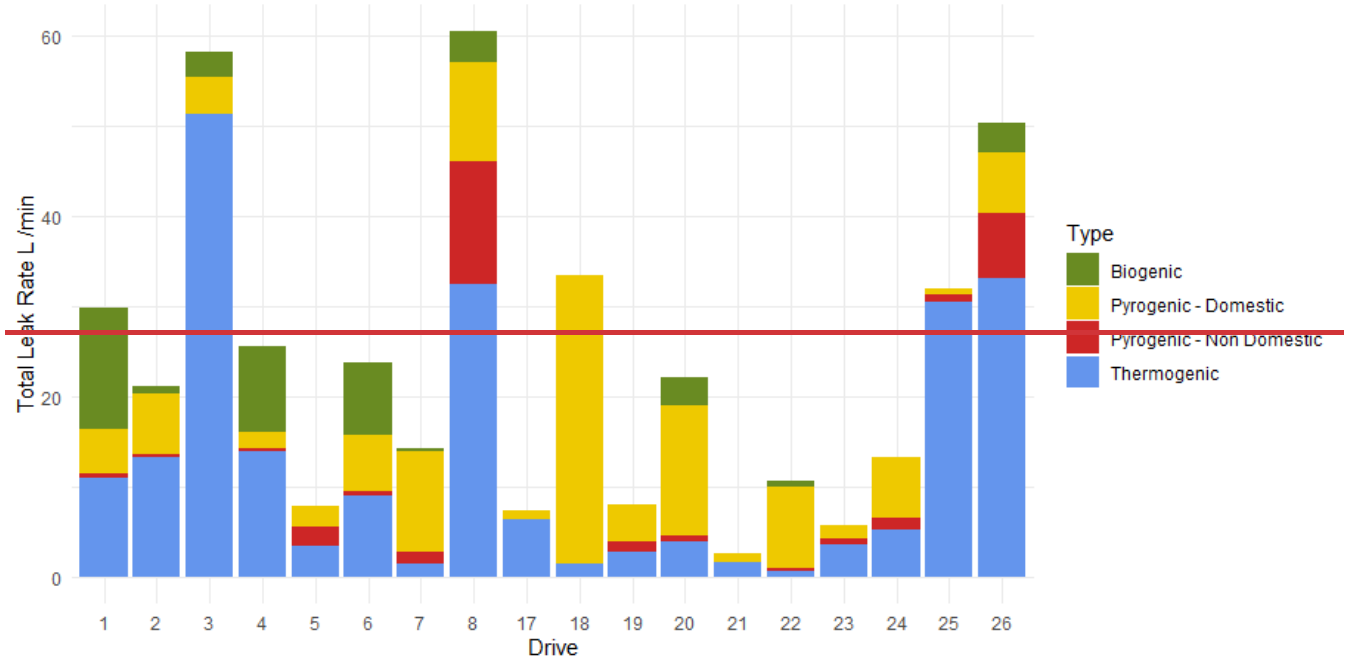
Figure 12: *Pie charts of each LI showing number of drives mobile surveys they were detected vs not*

The nNumber of mobile surveys drives is a large factor in the probability of detecting an LI. Each LI requires the enhancement to be detected on at least 2 separate mobile surveysdrives, for the 24 LIs detected over the course of this campaign 12 LIs were detected on 2 drives, 6 were detected on 3 drives, 4 on 5 drives and 2 on 7, this meant that the average probability of detection was 0.18., detection versus non detection for each LI is demonstrated in ***Figure 12***. This low probability of detection highlights the need for surveys with multiple passes.

3.2 Emissions from other sourcesPyrogenic emissions

While 1778 of the 4687 OPs were determined to be Thermogenic, 3941 were assigned as biogenic (8.3 8%) and, 1996 were Pyrogenic (41.8 2.0%) and 53 were not able to be assigned a source type. NO_x:CO_2 ratios were investigated for the pyrogenic Ops these OPs using the same methodology used for the CH_4:C_2H_6 source assignment. 1152 of the 1996 pyrogenic OPs were able to be analysed in this way, 875 of these 1152 OPs (75.7 9%) had a NO_x:CO_2 ratio < 0.88 x 10^{-3}. This implied that the majority of pyrogenic emissions did not originate from traffic but more likely emissions from domestic heat and power generation (such as emissions from domestic boilers) *(Cliff et al., 2025)*. When these 85 Pyrogenic boiler OPs were clustered over 30m and filtered to only include multiple drives (much like how LIs were determined from Thermogenic OPs) it resulted in only 5 recurring emissions of this type, this is to be expected, as emissions from boilers are less likely to be a consistent emission source than a fugitive emission from a natural gas pipe.

Emissions from pyrogenic sources are compared at the OP stage on a mobile survey by mobile survey drive by drive basis due to the high unlikelihood of pyrogenics and biogenics being persistent emission sources, the number of times each source type was detected per mobile survey is shown in ***Figure 13***.these emissions being detected on multiple drives.



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432 ~~***Figure 12:*** *Total leak rate (L min^{-1}) contribution from each source type for each of the drives.*~~

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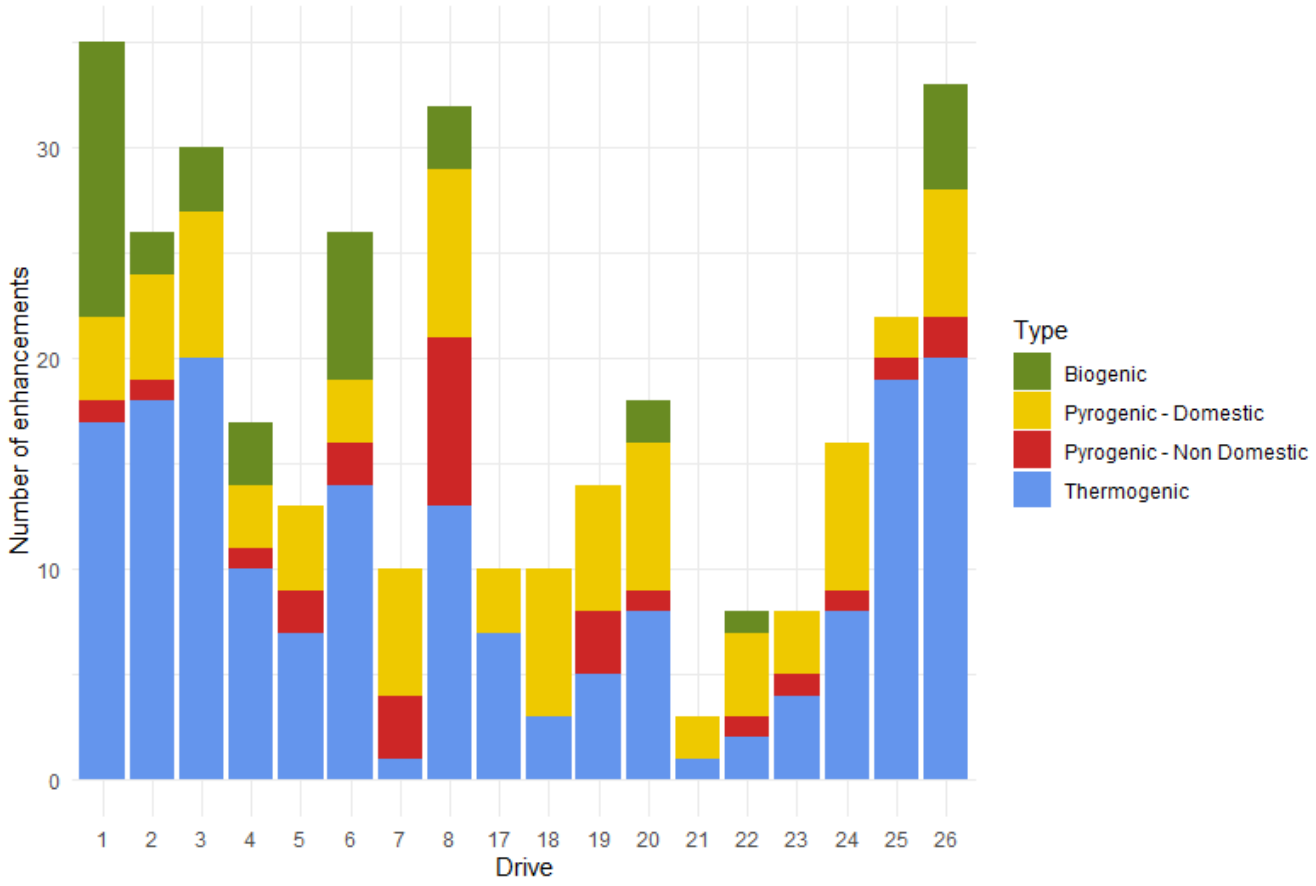
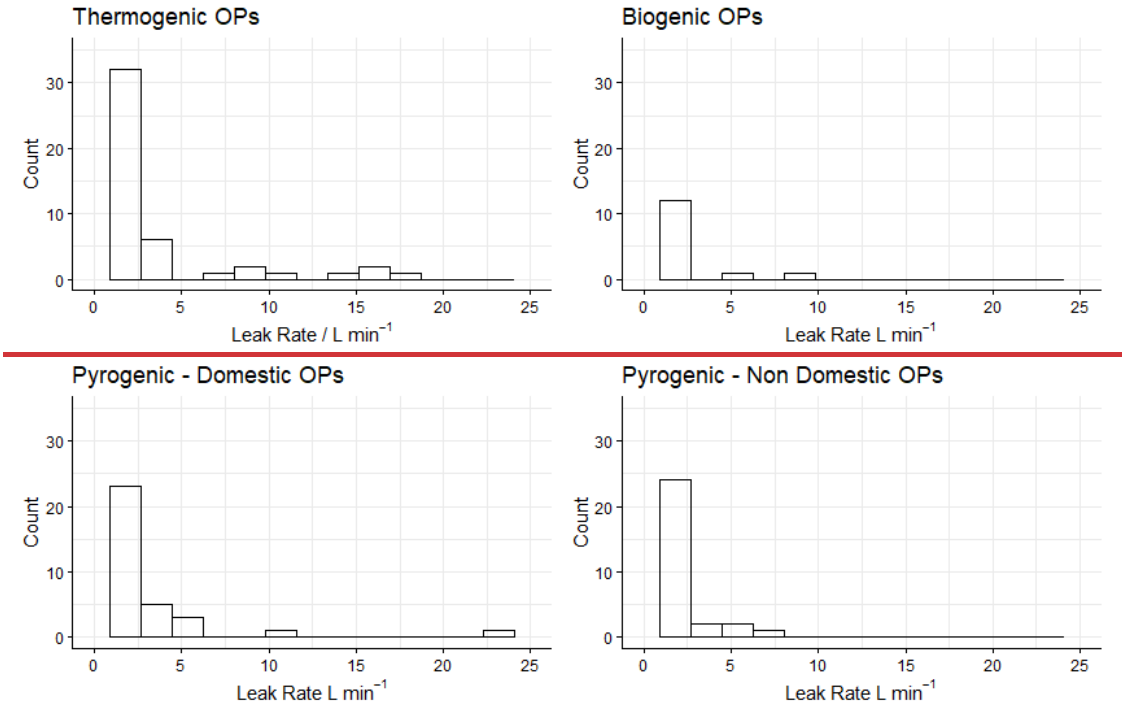


Figure 13: *Total number of enhancements from each source type for each of the mobile surveys.*

Thermogenics were the most frequently located source type on 13 of the 18 surveys, with mobile surveys 7, 18, 19, 21 and 22, finding pyrogenic emissions related to heating and cooking were the most frequently occurring source type. ~~Thermogenic emissions from natural gas had the highest overall contribution to OPs across all drives and had the highest individual contribution on 10 of the 18 drives. However, for 7 of 18 drives pyrogenic emissions related to heating and cooking had the second highest overall contribution to methane emissions along the sampling route. For all source types, the majority of enhancements detected were of leak rates less than 5 L min^{-1}, no non-domestic pyrogenic or biogenic emission was detected with a leak rate greater than 10 L min^{-1}. Although a few higher leak rate outliers were detected for domestic pyrogenics, thermogenic emissions were the only source type with emissions seen consistently at leak rates up to 20 L min^{-1}.~~



~~***Figure 13:*** *Histograms of leak rate distributions by source type.*~~

3.3 Comparison to previous methods

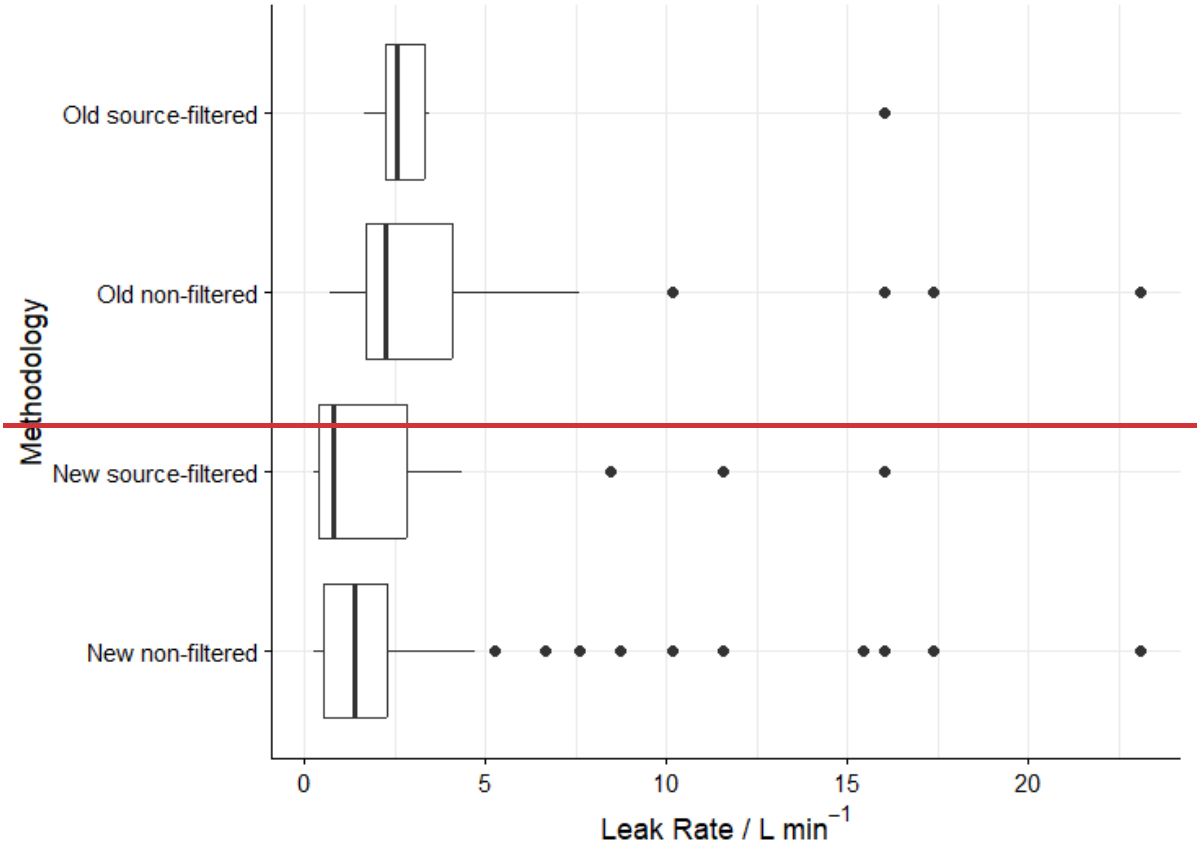
The main alterations to this methodology from that present in (*Weller et al., 2019*) (and other studies that were based of this method) were that enhancement criteria was changed from 1.1 times the baseline to 1.05 times the baseline, clustering by time was changed such that emissions within 1 s of each other were clustered instead of those within 5 s. F~~ and f~~inally ~~that~~ a source determination stage was added as a core step in the algorithm as opposed to previous iterations that either had no source determination stage or one that came later in the analysis. ***Table 1*** shows the effect that all of these changes to the methodology make on the resulting detection of OPs and LIs.~~filter was applied as opposed to no source determination occurring or that this came later in the analysis. By comparing the methodology with the new one derived from the instrument limitations we find the following:~~

Enhancement Criteria	Time Clustering Criteria / s	Source Determination Included?	Number of OPs	Number of LIs
110% of baseline	5	No	179	27
		Yes	66	6
	1	No	216	29
		Yes	79	7
105% of baseline	5	No	357	58
		Yes	144	23
	1	No	4687	58
		Yes	1778	24

Table 1: *Number of detected OPs and LIs depending on changing algorithm parameters*

This shows ~~Showing~~ the new methodology could locate more LIs. ~~Additionally, when the predicted leak rates were compared from the non-source filtered LIs it showed that in addition to being able to locate more LIs, the changes to enhancement criteria and time clustering have also led to the ability to locate LIs of a greater range of leak rates.~~ Binning into the leak rate categories of very small (0 - 2 L min^{-1}), small (2 - 6 L min^{-1}), medium (6 - 40 L min^{-1}) and high (> 40 L min^{-1}) showed that of the 24 LIs in the new source filtered methodology, 2 were small and 22 were very small. For the 58 LIs of the new non filtered methodology, 9 were small and 49 were very small. For the ~~previous method the~~ = 27 ~~detected~~ = from the old non filtered methodology 10 were small and 17 were very small.~~LIs range in leak rate between 0.70 and 23.10 L min^{-1}, with a mean Leak Rate of 4.53 L min^{-1}, 75% of LIs have a leak rate between 1.71 and 4.10 L min^{-1}, whereas with the new enhancement criteria and time clustering parameter there was a range in leak rate of 0.24 to 23.10 L min^{-1}, a mean of 3.19 L min^{-1} and 75% of LIs having a Leak rate between 0.51 and 2.26 L min^{-1}. The total methane emissions from LIs were 122.43 L min^{-1} for the old method compared to 185.10 L min^{-1} for the new method. This however changes when the source filtering is factored in where the Leak Rates ranged from 0.26 to 16.04 L min^{-1} with a mean leak rate of 2.51 L min^{-1} and 75% of LIs having a leak rate between 0.38 and 2.85 L min^{-1} additionally the cumulative emissions was 60.23 L min^{-1}. If this source filtering is applied to the old method however we see that for the 6 LIs, leak rates ranged from 1.64 to 16.04 L min^{-1} with a mean of 4.74 L min^{-1} and a cumulative emission of 28.48 L min^{-1}. Showing the old methods underprediction in total methane emissions of 33.9% when not source filtered and 59.4% when source filtering is considered.~~ Finally the 6 LIs from the old source filtered methodology, 1 was small and 5 were very small. This shows the old methodology requiring an enhancement of 1.1 times the baseline with 5 s time clustering misses a large proportion of LIs that the newer methodology, requiring an enhancement of

1.05 times the baseline with 1 s time clustering, detects. A large proportion of these missed LIs occur in the very small category as expected with a smaller enhancement criteria. Source filtering shows that regardless of criteria used less LIs will be detected with this method, this suggests previous methodologies that do not use this stage may be mischaracterising some thermogenic enhancements as being permanent as they may instead be detecting methane enhancements of differing source types that occur within the same vicinity of one another.



~~***Figure 14:** Box plot showing the varying detected Leak Rates depending on methodology used*~~

3.4 Comparison of alternate quantification approaches

As previously described, the quantification equation used within this body of work is based on the *(Tettenborn et al., 2025)* approach of using peak area to calculate the leak rate of a LI. However, previous works have used the quantification equation present in *(Weller et al., 2019)* which instead quantifies release rate based on peak height. This campaign's results were reprocessed using each of these previous quantification equations in order to compare the effects of the updated parameters in the York quantification equation to the original, present in *(Tettenborn et al., 2025)* but also to explore the difference in

quantified leak rates from a peak height approach. As previously mentioned, the results of the York quantification approach resulted in the 24 LIs being assigned to leak rate bins such that 2 were small and 22 were very small, the *(Tettenborn et al., 2025)* equation results in 1 medium, 1 small and 22 very small and the *(Weller et al., 2019)* equation results in 1 small and 23 very small. The specific leak rates of LIs calculated with these three equations are presented in box-plots in ***Figure 14***.

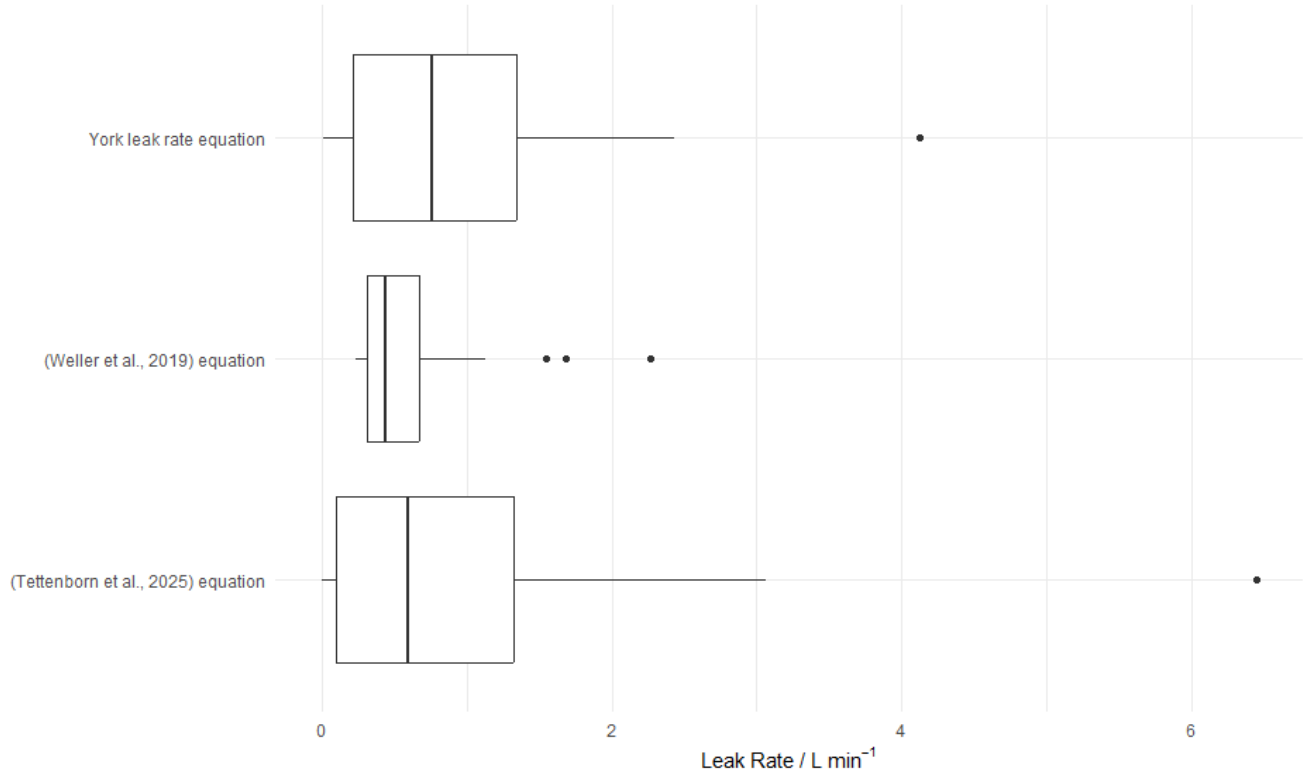


Figure 14: *Comparison of calculated leak rates of LIs from each of the 3 quantification equations.*

This shows that both the peak area approaches result in a much larger range of calculated leak rates from the LIs than from the peak height approach present in *(Weller et al., 2019)*. This suggests that the instrumentation used to detect CH_4 enhancements, may result in low, wide peaks as opposed to higher, sharper peaks, thus explaining why leak rates are weighted much lower from this method. The *(Tettenborn et al., 2025)* equation appears to be mostly consistent with the equation determined from the York methodology, however there is slightly higher weighting of leak rates with the *(Tettenborn et al., 2025)* equation, resulting in the 24 LIs changing from the assignments of 5 small and 19 to 1 medium, 5 small and 18 very small.

4. Conclusions

This study focused on using the specifications of the equipment used for the surveying to better inform a detection algorithm. Enhancement criteria was determined by investigating the variance of the MGGA, although laboratory experiments suggested the instrumentation was capable of detecting enhancements at a minimum of 1.005 times the baseline was possible, in field experiments showed that enhancement criteria of 1.01 times the baseline was more likely the lower limit for detection. However, for the surveys a criteria of 1.05 was selected so as to not incorporate small, diffuse emissions within the analysis. ~~, in field experiments showed that a preferable limit was 1.05 times the baseline.~~ Response rate of the instruments was calculated to inform the time window for clustering, with both MGGA and TILDAS having sub one second response rate, the time clustering was instead limited to one second due to the limitations of GPS data collection speed. These changes show previous methodologies would result in detection of 27 LIs compared to the 58 LIs using updated parameters (53.5 % less), the parameter change has also shown the ability to detect more LIs in all leak rate categories, but in particular, the very small (0 - 2 L min^{-1}) category, where 17 of 27 LIs were located in the previous methodology, but 46 of the 58 were located in the new methodology.

~~These changes show previous methodologies would result in detection of 33.9% lower cumulative emissions.~~

Source appointment proved to be a useful tool for predicting emissions directly related to natural gas, when source filtering was introduced at the OP stage of detection, it resulted in only 41.4% of LIs still being detected as opposed to the non-source filtered method. ~~Additionally, this meant a reduction of 67.5% in the total estimated emissions of methane.~~ Additionally source filtering has helped to highlight that although Thermogenic emissions from natural gas are the highest contributor to methane emissions, pyrogenic emissions related to domestic heat and power generation also provide a high but often overlooked contribution to a ~~cities~~city's methane emissions.

The updating of the quantification equation from a peak height approach to a peak area approach results in a much wider range of leak rates being calculated in the study, however these values are not as high as when quantified using the original equation presented in *(Tettenborn et al., 2025)*.

This new method has shown that with changing enhancement criteria and time clustering parameters, it is able to detect many more LIs and a wider range of leak rates and that by applying a source type filter at the OP detection stage it is capable of reducing of reducing the incorrect assignment of LIs.~~the overprediction of methane emissions from natural gas~~. However, the methodology has ability to improve further, primarily by using instrumentation that is capable of detecting methane and ethane on one instrument, so as to remove uncertainty related to time lag between the two instruments, but secondly by having all instrumentation and hardware able to operate at a sub one second time rate in order to reduce the time clustering parameter limit and further improve spatial resolution.

Code / Data availability

Code and data will be made available upon request.

Author Contribution

Contributed to conception: TM, JH, WD, JL. Contributed to data acquisition: TM, JH, WD, SY, SHB, MS, JL. Contributed to analysis and interpretation of data: TM, JH, WD, SY, MLV, JF, JL. Drafted and/or revised the paper: TM, JH, WD, SY, DL, JF, JL. Approved the submitted version of this paper for publication: TM, JH, WD, SY, SHB, MS, JF, DL, JL.

Competing Interests

The authors declare that they have no conflict of interest.

Acknowledgements

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