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

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Abstract

Reducing methane (CH_4) emissions has become increasingly important in recent years due to its importance for radiative forcing. Fugitive emissions of CH_4 from natural gas distribution infrastructure are of particular interest as a mitigation target within the oil and gas sector. ~~One area of particular interest is the oil and gas sector, which often results in fugitive emissions of methane from natural gas distribution.~~ Previous studies have shown the ability to detect these emissions ~~,~~ by use of mobile surveys measuring ~~(~~CH_4~~)methane~~. Some of these previous studies use ratios to secondary co-emitted compounds as a means of predicting the source of emission. This study aims to adapt existing algorithm parameters by investigating the limitations of equipment within the platform used for mobile surveys. These changes suggest that previous methods may underpredict the number of Leak Indications (LIs) by 53.5 % with 27 LIs detected with the old methodology compared to 58 LIs detected with the new methodology.~~the number of LIs detected with the old methodology being 27 and the new methodology 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~~ determination was included as a core step within the algorithm ~~itself~~, which ~~this stage~~ was shown to reduce the misassignment of LIs, suggesting that when not using this step,~~the previous methodology may include~~ emissions from pyrogenics and biogenics are included within ~~their~~ LI assignments.

1 Introduction

Following COP26 and the Global Methane Pledge~~methane pledge~~ *(European Commission and United States of America 2021)*, CH_4 ~~methane~~ and its emissions have received increased attention. The pledge states that the signatories will attempt to reduce their CH_4 ~~methane~~ emissions by 30 % of their 2020 levels by 2030. This was brought about due to increasing concern over the potency of CH_4~~methane~~ as a greenhouse gas, with its warming potential 28 times greater than CO_2 over a 100-year timescale and 84 times ~~higher~~ greater over a 20-year timescale *(IPCC, 2021)*. Anthropogenic sources are estimated to contribute to 65 % of all CH_4 ~~methane~~ emissions, with ~~are thought to be anthropogenic in nature and~~ atmospheric CH_4~~methane has~~ seeing~~n~~ 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)*.~~,~~ T~~t~~herefore, understanding and mitigating anthropogenic CH_4~~methane~~ is a key ~~goal~~ step in ~~to~~ complying with the G~~g~~lobal M~~m~~ethane P~~p~~ledge.

Of ~~the~~ anthropogenic emissions, the agricultural sector has the largest contribution towards atmospheric emissions *(Saunois et al., 2025)*. Although there are means of reducing these emissions, ~~from these areas, including changing~~ such as changes to cattle, crop and~~or~~ land management as well as changing the feedstock of the cattle, ~~for example~~ from grass silage to maize silage (*Bačėninaitė et al., 2022; Nisbet et al., 2025)*.~~,~~ T~~t~~hese changes may still require time to implement, so this sector cannot be the sole focus in order to reach the 2030 deadline.

After agriculture, the largest ~~contribution~~ contributor to anthropogenic emissions is ~~from~~ the energy sector, with oil, natural gas and coal having relatively similar contributions to CH_4 ~~methane~~ emissions. Natural gas is of particular importance to the United Kingdom (UK), ~~with it being~~ which is the 19th largest country emitter of CH_4~~methane~~ from the natural gas network (*Scarpelli et al., 2022).*

One of the major sources of CH_4 ~~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 "g~~G~~as l~~L~~eaks".~~,~~ H~~h~~owever, 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~~ UK 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 UK's 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 CH_4 ~~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), a network of over 5,000 miles of high-pressure steel pipes and more than 500 above ground installations. ~~This n~~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. The GDN responsible for York covers 2.7 million homes and businesses across the northeast of England and northern Cumbria, ~~which means the operation of~~ resukting in tens of thousands of kilometres of pipework and therefore large ~~unknowns~~ uncertainties in the locations of fugitive emissions. To combat this, previous studies have implemented mobile measurement approaches centred around the detection of areas with elevated CH_4~~methane~~.

1.1 Previous Mobile Measurement Methodology

~~There have been many~~Multiple previous studies ~~that~~ have attempted to design algorithms to detect fugitive emissions of natural gas, all of which focus on locating enhancements in CH_4, 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 CH_4~~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~~ Additionally, OPs must not cover a distance greater than 160 m. Enhancements occurring within 5 seconds of each other are grouped together. Mobile surveys are repeated multiple times and Leak Indications (LIs) are determined ~~after~~ by grouping OPs that occur within 20 m of one another and ~~seeing~~ determining which of these grouped clusters contain OPs from more than one mobile survey. The LIs are then quantified into emission rates in L min^{-1}, using an equation derived from the results of a controlled release experiment, shown in ***Equation 1***.

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

Where:

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

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

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

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 out burning emissions. Source attribution was also used in *(Fernandez et al., 2022)*, using isotopic measurements of CH_4 in addition to ethane: methane (C_2:C_1) 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***.

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

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

Variations of this algorithm have been used in many major cities across the USA and Canada *(Ars et al., 2020; Weller et al., 2022)* ~~as well as European cities~~ Europe *(Defratyka et al., 2021; Fernandez et al., 2022; Wietzel et al., 2023; Vogel et al., 2024)* and Asia~~n 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 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 Wolfson Atmospheric Chemistry Laboratories (WACL) Air Sampling Platform (WASP) detailed in *(Wagner et al., 2021)* is the base for these measurements. The sampling inlet for the WASP 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),* the WASP has 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 CA~~a~~vity cE~~E~~nhanced Differential oO~~O~~ptical aA~~A~~bsorption sS~~S~~pectrometer (ICAD) for measurements of NO_x (NO_2 + NO), and an Aerodyne Tuneable Infrared Laser Direct Absorption Spectrometer (TILDAS) lL~~L~~aser tT~~T~~race gG~~G~~as aA~~A~~nalyser for measurements of ethane (C_2H_6) *(Yacovitch et al., 2014)*. Measurements of C_2H_6 ~~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 a calibration ~~is~~was performed before and after the mobile survey itself, a linear regression ~~is~~ was performed to find the slope and intercept of the calibration concentrations versus measured concentrations. The average ~~is taken for~~ of the two calibrations was taken to account for instrument drift during the mobile survey and the resulting equation, ***Equation 4***, ~~is~~was then used to apply a correction to C_2H_6 ~~ethane~~ concentrations.

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

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, a ~~S~~stainless ~~S~~steel ~~I~~ntegral ~~B~~onnet ~~N~~eedle ~~V~~alve, 0.37 Cv, 1/4 in. #SS-1RS4 and an electronic ~~U~~pstream ~~F~~low ~~C~~ontrol ~~V~~alve, 10,000 sccm, 0.25 in. ~~T~~ube, ~~V~~iton ~~S~~eal #0248A-10000SV which allows small changes to maintain the internal pressure at 70 Torr. With the two valves in series, the instrument was~~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~~which indicated 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 C_2H_6 ~~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 C_2H_6~~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)*.~~,~~ ~~a~~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 had~~ving~~ 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.

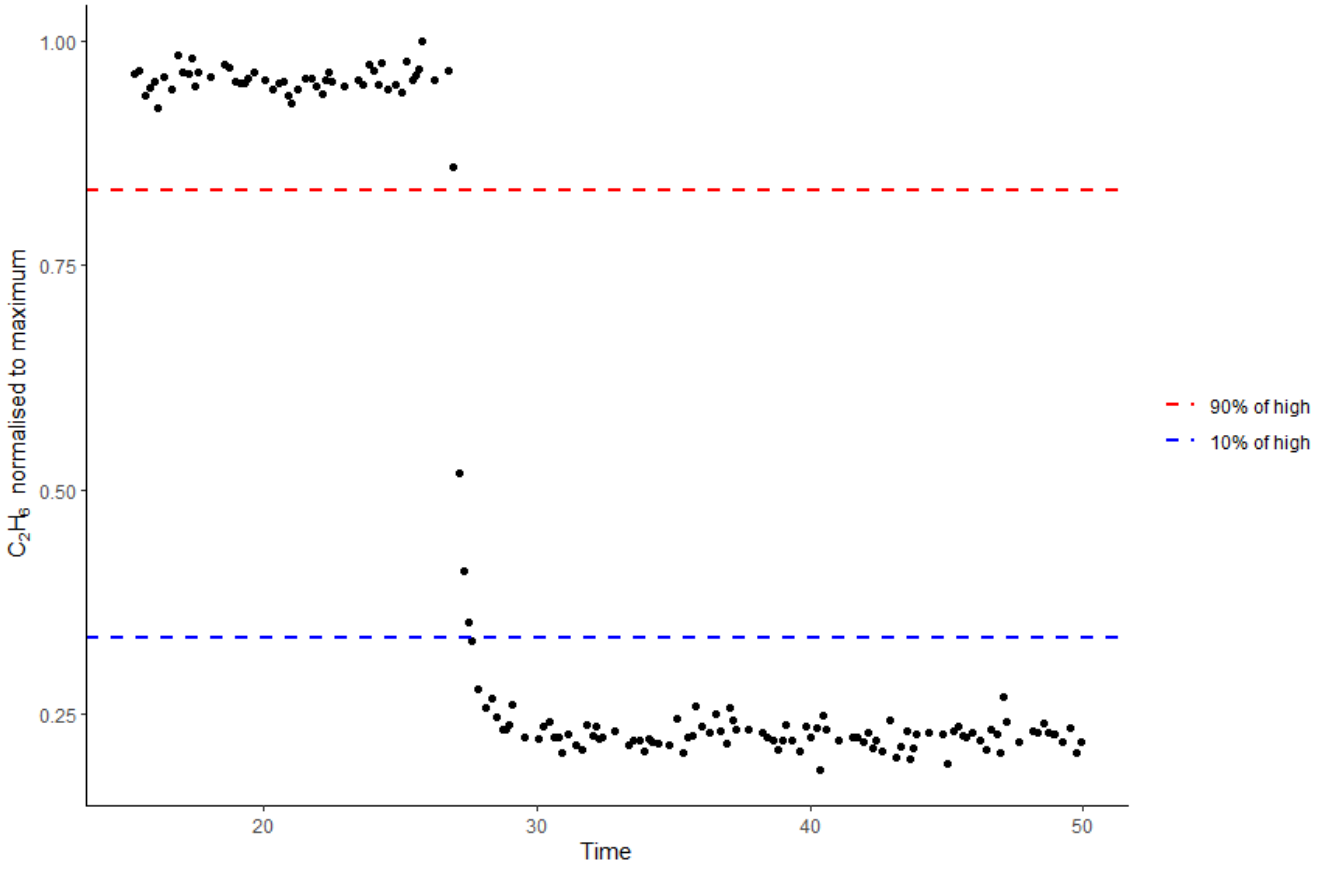


Figure 1: *Example response transition of TILDAS high concentration to low concentration, normalised to maximum recorded response*

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 was undertaken. The standard deviation of ~~methane~~ CH_4 measurements over ~~this~~ a 2 hour period was calculated to understand the minimum detectable enhancement for the CH_4 ~~methane~~ detection algorithm.

For 2 hours compressed air ~~was flown~~flowes through the Los Gatos MGGA, with an observed ~~measured at a~~ median measured value of 7.2 ppm, ~~the~~ and a standard deviation of ~~these results was found to be~~ 0.006 ppm. An enhancement criteria was proposed as 5 times this standard deviation divided by the median baseline, resulting in an enhancement criteria of 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 ledads to the detection of enhancements that werecare too small to be reliably quantified. Instead, the CH_4methane mixing ratios measured during each mobile survey, were collated and the standard deviation was calculated for each mobile survey. Again eEnhancement criteria was calculated as anything larger than 5 times the standard deviation divided by the median of CH_4methane mixing ratios., tThis was done repeated for each mobile survey and resulted in a median enhancement criteria of 1.01 times the baseline. However, as this cwould result in detection of very small, diffuse, or non-natural gas emission enhancements, 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.

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 mobile surveys of a "flower petal" route, shown in ***Figure 2***, staying within the outer ring roads of the A64 and A1237 and mainly focused primarily on sampling residential areas of the city. The 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 sampled regardless of the direction of travel. The route was driven 18 times as, in order 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 as it only covers 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)*.

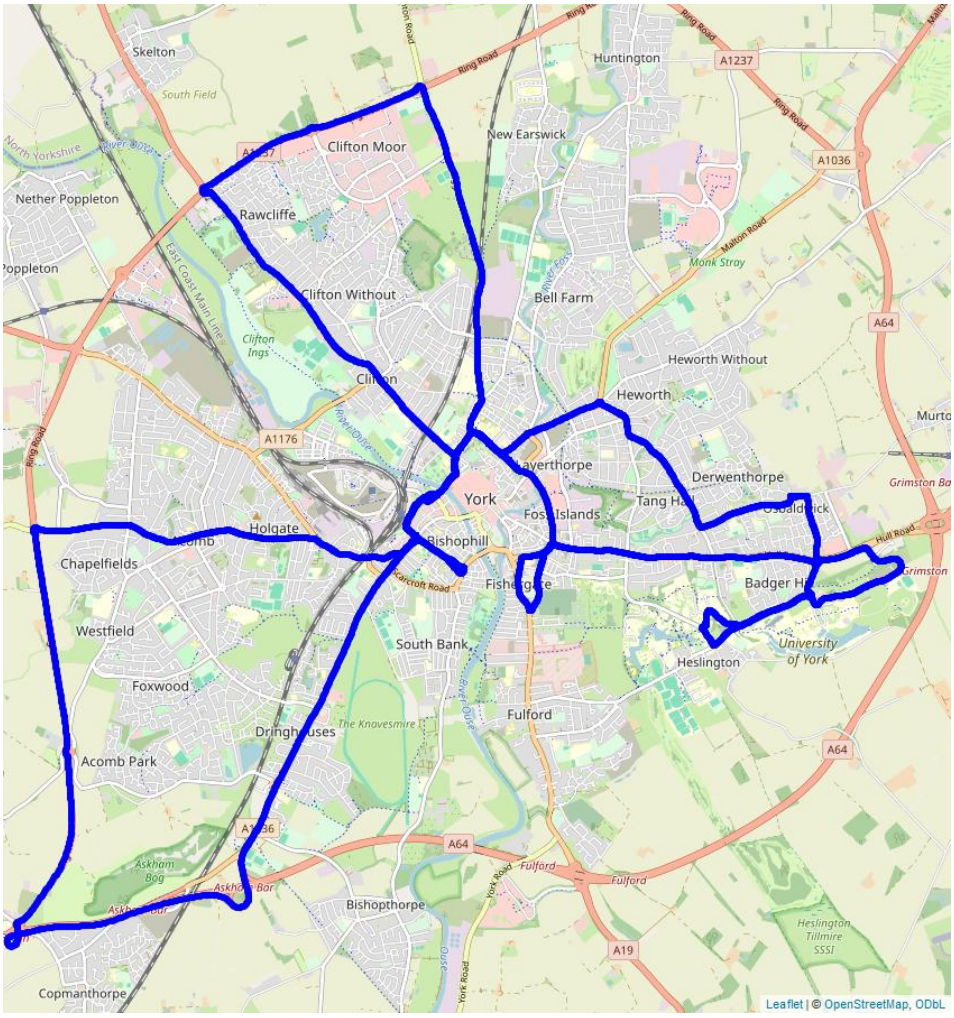


Figure 2: *Map of the route taken in WASP surveys. Produced using Leaflet (Cheng et al., 2025) with tiles taken from OpenStreetMap* (© OpenStreetMap contributors *https://www.openstreetmap.org/copyright*).

2.3 Enhancement Detection Algorithm

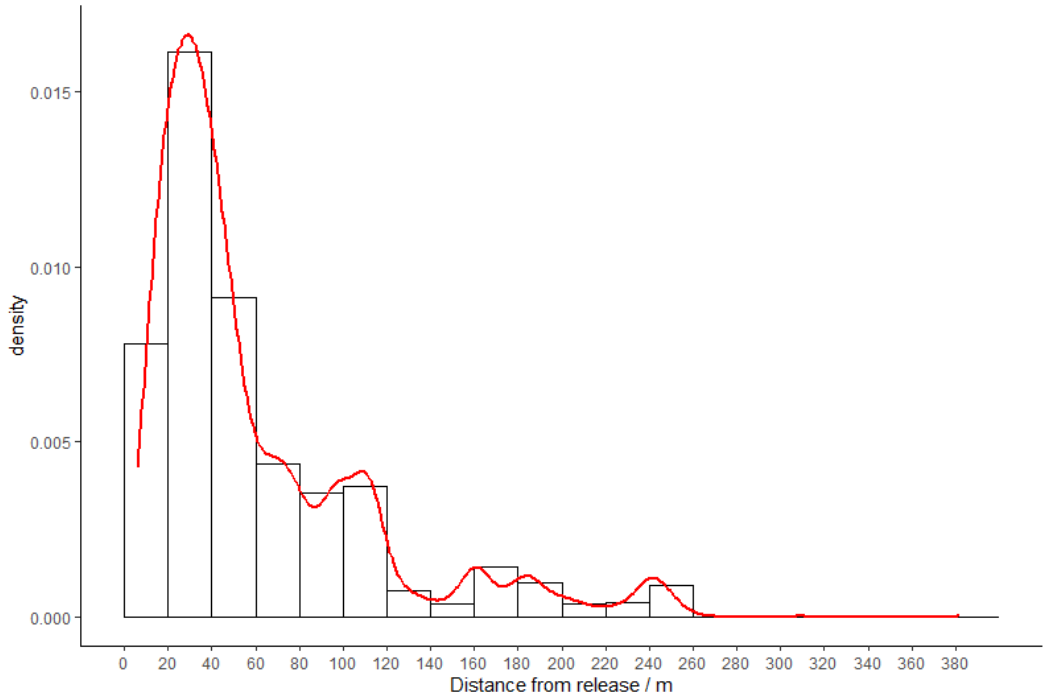
The original algorithm, used in *(Weller et al., 2019)*, ~~has been~~was adapted following the findings in ***2.1.1*** and ***2.1.2***. OPs ~~are~~ were clustered within 1 s as opposed to 5 s. With a faster instrument response, it was expected that the measurements would more readily distinguish between 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 44.5 m 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, thise 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~~ thermogenic source type, to further reduce the chance of comparing long standing thermogenic fugitive emissions with possible nearby single occurrence pyrogenic or biogenic emissions.

2.4 Controlled Release Experiment

To ~~attain~~ obtain a quantification equation specific to the equipment used ~~at~~in 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~~ May of 2024. A MiniCRF was deployed to manage releases of CH_4 ~~methane~~ and ammonia (NH_3), 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 CH_4 ~~methane~~ (0.2 - 70.48 L min^{-1}), ~~ethane~~ C_2H_6 (0 - 7.01 L min^{-1}) and ~~ammonia~~ NH_3 (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 multi 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 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~~ moved further away in distance for another set of 10 repeats.~~,~~ T~~t~~his continued until the plume was either lost, or a lack of driveable ground was left available. It was noted that larger releases were detectable further away, however, as the data from the controlled release was intended to be used in quantifying under-road and near-road fugitive emissions of natural gas, a maximum distance of 30 m from the point of release was applied for data analysis to reflect the maximum ~~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 analysis~~, this is~~ due to several reasons, including that some releases did not hav~~c~~~~ing~~ detectable enhancements. Within these 26 releases, 3525 CH_4 ~~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~~ enhancements were filtered to ~~include enhancements up to~~ a maximum distance of 30 m from the release point, this resulted in 1226 enhancements from 23 mobile ~~surveys~~releases. Density plots of the 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.



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

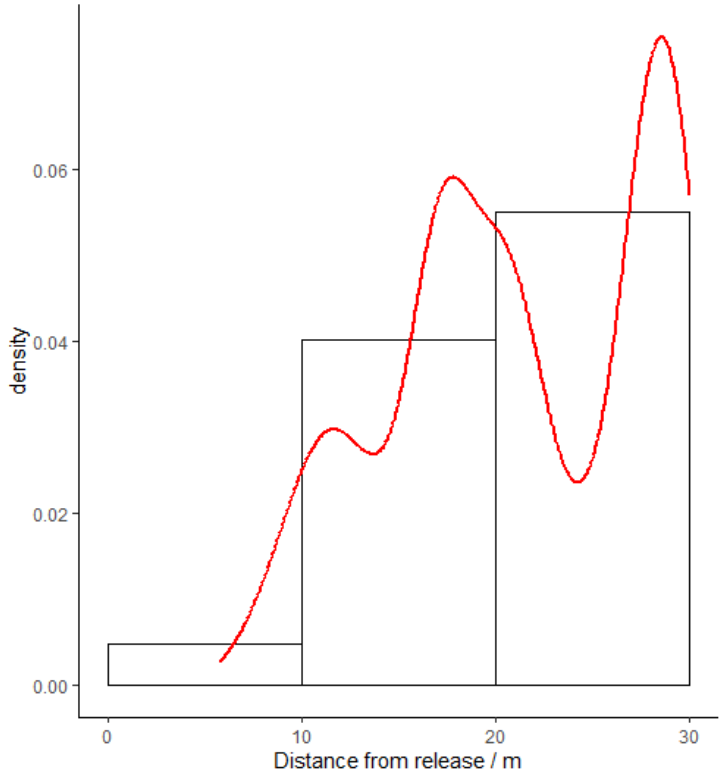

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Figure 4: *Density plot of number of detected enhancements against distance from release point (Limited to 0 – 30 m)*

2.4.1 Quantification equation

There has been much development and advancement in the last few years on the use and application of "aAdvanced mMobile lLeak dDetection" 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 asdue to external 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 a 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 Bedford 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.

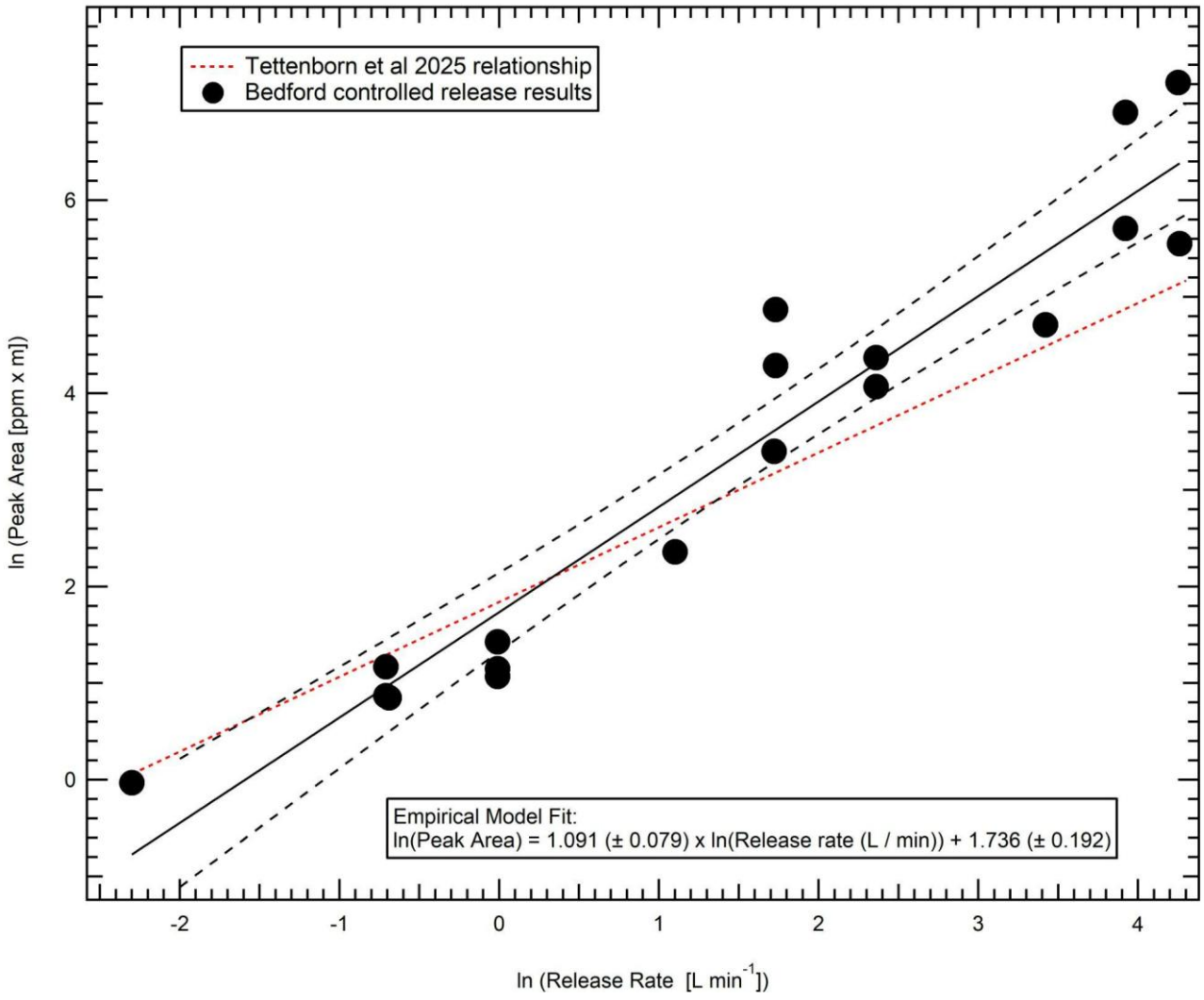


Figure 5: *Peak area vs actual release rate for plume transects within 30m of release. Data shown is an average of multiple transects (at least 10) for each release.*

~~As it can be seen in ***Figure 5***, w~~Whilst the general trend of increasing plume area with release rate is adhered to, as can be seen in ***Figure 5***, 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 g~~Given the importance of windspeed in ~~more distal~~ emissions modelling

(e.g. Gaussian plume modelling from vehicles *(Dowd et al. 2024))*, it would appear to have the potential for significant uncertainty in the resultant emissions quantification. To test this, 1 Hz wind data (averaged to 1 minute data) was taken from the 3 m mast located on site at the controlled release and incorporated into the analysis according to ***Equation 5***.

~~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.

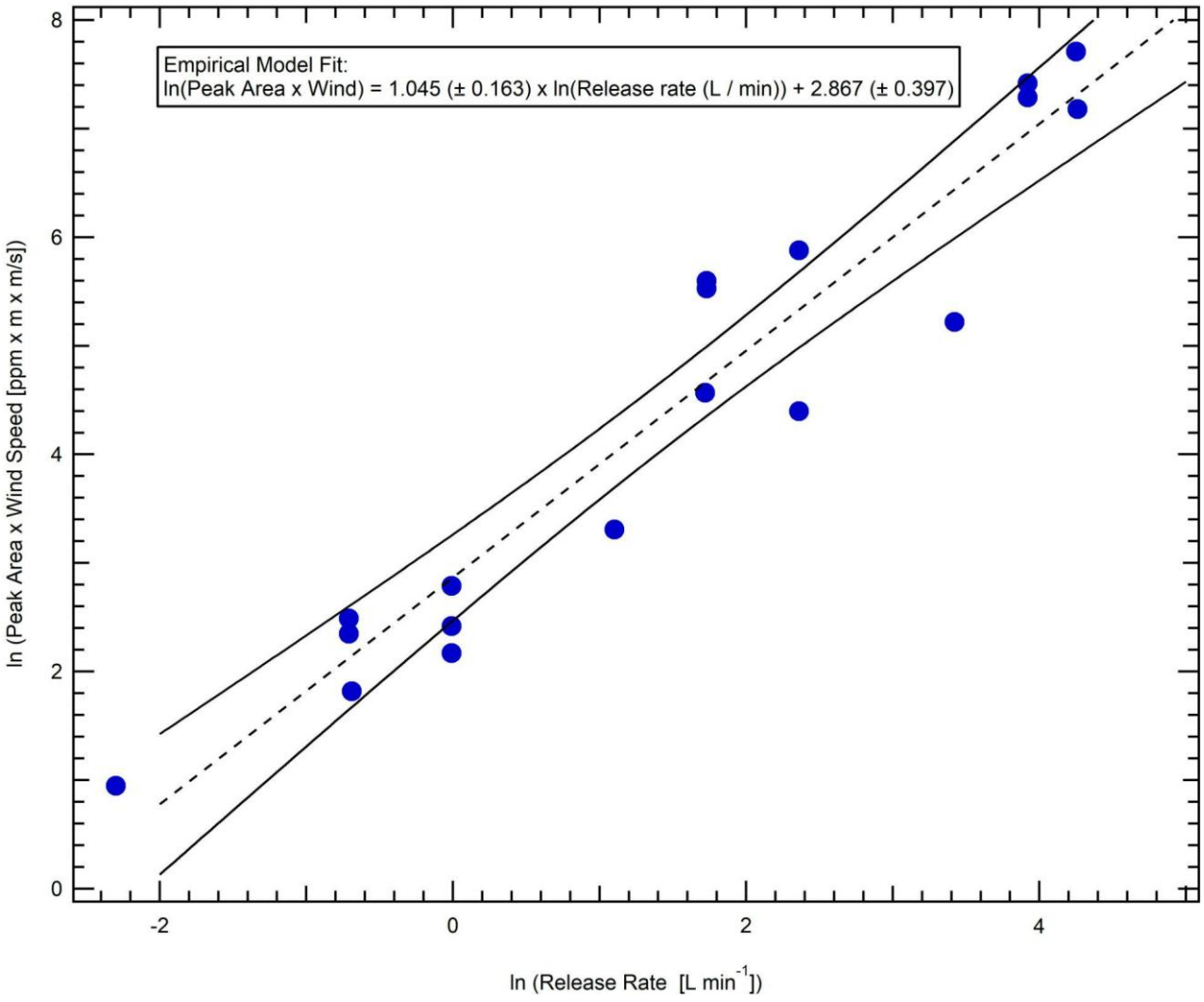


Figure 6: *Peak area x Wind speed vs actual release rate for plume transects within 30m of release. As with Figure 5, data shown is an average of multiple transects (at least 10) for each release.*

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

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

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 allowed~~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.

2.4.2 Instrument Lag Time

For each of the releases, the lag time between detecting an ~~ethane~~ C_2H_6 enhancement and a CH_4~~methane~~ enhancement was calculated. 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 the manifold to each instrument. To ~~find this~~ determine a more accurate lag time ~~of~~ for the instruments, the maximum CH_4~~methane~~ enhancement for each transect~~pass~~ was ~~found, following this~~ identified along with the maximum C_2H_6~~ethane~~ enhancement ~~was found (that occurred within 5 seconds of the methane)~~ occurring within 5 seconds of the CH_4 enhancement. The resulting 10 second window was selected ~~as~~based on ~~transects of the~~ vehicle speeds during the controlled release, where the ~~van~~ WASP travelled at roughly 20 miles $hour^{-1}$. Over the course of 10 s (5 s either side of the methane maximum) this would ~~mean~~ result in a distance of 85 m covered ~~in the van,~~ (the average length of a transect being 180 m). The time lag between C_2H_6~~ethane~~ and CH_4~~methane~~ showed that in most cases (88.1 %), maximum C_2H_6 ~~ethane~~ concentration preceded maximum CH_4~~methane~~ concentration with a mean ~~lag~~ of 2.7 s before and a median of 3.8 s before. Observing a window of time of maximum CH_4 ~~methane~~ to 5 s before maximum CH_4~~methane~~ resulted in a mean lag of 3.3 s from C_2H_6 ~~ethane~~ to CH_4~~methane~~ and a median lag of 3.9 s. This helped inform the detection algorithm to look for maximum C_2H_6~~ethane~~ within a window only up to 5 s before the maximum CH_4~~methane~~. Density plots showing the time lag of maximum C_2H_6 ~~ethane~~ from maximum CH_4~~methane~~ are shown in ***Figure S2*** for the full 10 second time window and ***Figure S3*** for up to 5 seconds before the time of maximum measured CH_4~~methane~~.

2.5 Source Appointment

Source determination using ethane :~~-~~methane (C_2:C_1) ratios has been shown to be effective, due in part to the knowledge that C_2H_6~~ethane~~ is present in measurable quantities in thermogenic gas but not biogenic gas ***(****Fernandez et al., 2022)*. These ~~ethane:methane (C_2:C_1)~~ ratios can be used in order to determine the source of a CH_4~~methane~~ emission. Demonstrated in *(Yacovitch et al., 2014; Lowry et al., 2020; Defratyka et al., 2021; Fernandez et al., 2022)*, C_2:C_1 < 0.005 may be 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 7***. In order to calculate these ratios CH_4~~methane~~ and C_2H_6~~ethane~~ values must first be aligned in time, due to them being measured on separate instruments, the criterion for time alignment was discussed in ***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.

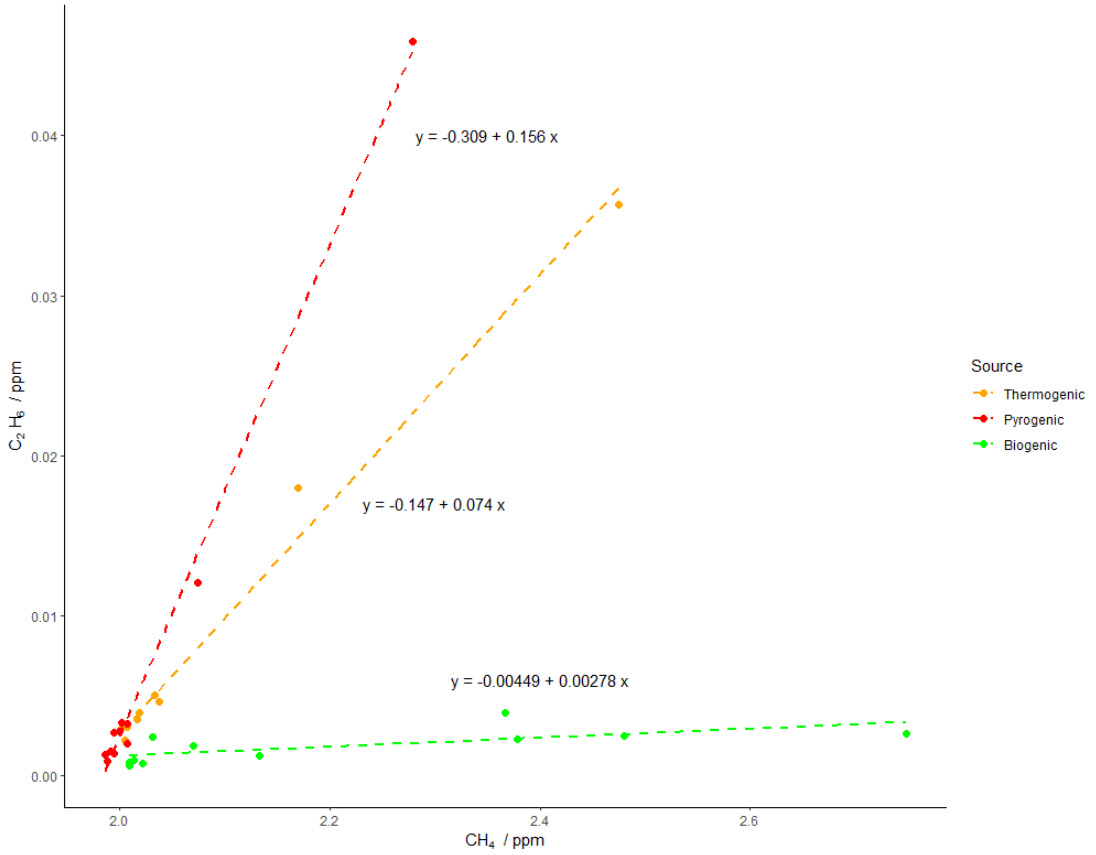


Figure 7: *Relationship between CH_4 and C_2H_6 for three OPs of different source types located during the sampling campaign.*

3 Results

3.1 Results of York mobile surveys

17 mobile surveys were conducted across the route of York, the raw data was taken from 10 Hz files for CH_4 ~~methane~~ (MGGA) and C_2H_6 ~~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 CH_4 ~~methane~~ concentration is shown in ***Figure 8***. The data was then processed to remove ~~data~~ measurements taken when vehicle speeds were 0 or > 40 miles hour^{-1} as well as removing data within the area of ~~Wolfson Atmospheric Chemistry Laboratories~~ WACL, ~~due to this also being the location within which~~as calibrations and other instrument tests were conducted in this location. A rolling 2.5-minute median background of CH_4 was then applied and enhancements were determined as any CH_4 measurement taken that was greater than 1.05 times the calculated background. The 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 468 OPs were detected over the course of the 18 mobile surveys.

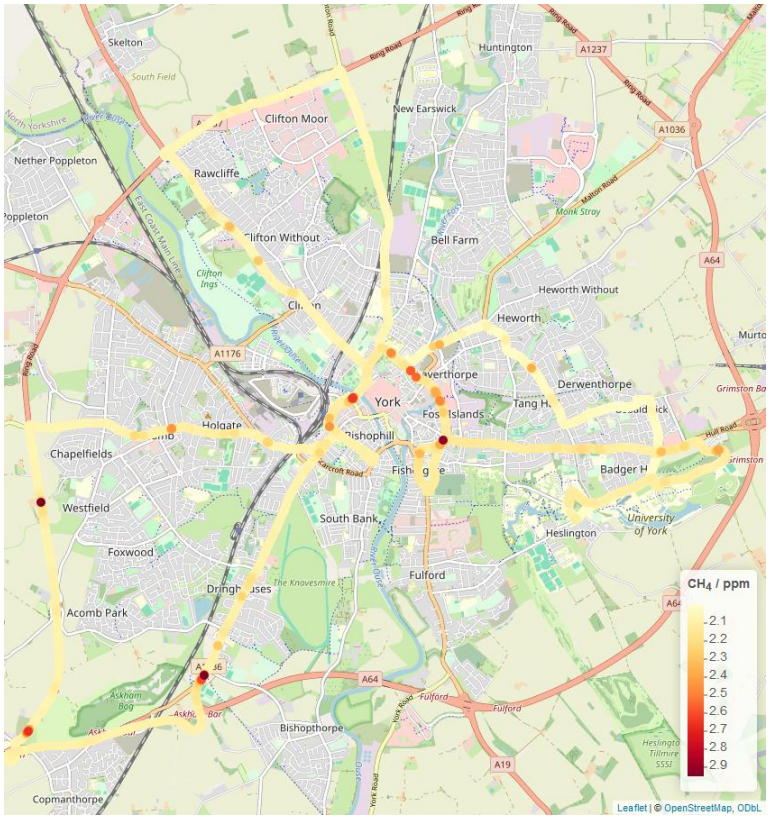


Figure 8: *Colour map of CH_4 concentration from one of the York mobile surveys.*

For each of these OPs the maximum C_2H_6 ~~ethane~~ value was found from the time of maximum CH_4 ~~methane~~ to 5 seconds prior. The two instruments' data were then aligned for each OP such that time of maximum CH_4 ~~methane~~ measurement was equal to time of maximum C_2H_6 ~~ethane~~ measurement. A linear regression was then taken of values from 5 seconds prior to the maximum CH_4 ~~methane~~ measurement 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 o~~Of the 468 OPs, 177 (37.8 %) were found to be thermogenic in origin. ~~Each~~ All~~T~~ thermogenic OP w~~ere as~~ then spatially clustered using a 30 m threshold, with the resulting clusters~~to find others within 30 m of one another and~~ filtered to ensure that each cluster~~s~~ contained OPs occurring on at least two separate mobile surveys, ~~to~~ remov~~inge~~ any OPs occurring from an event ~~happening~~ observed~~on~~ during only one mobile survey. The remaining clusters were then averaged into LIs such that the latitude and longitude were calculated as a weighted spatial average, resulting in 24 thermogenic LIs from the 177

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~~ each LI cluster. The smallest leak rate was determined to be 0.01 L min^{-1} and the largest being 4.13 L min^{-1}, when assigned to bins 2 were classified as small (2 - 6 L min^{-1}) and 22 were very small (0 - 2 L min^{-1}). When the source ~~type filter~~ determination step ~~is~~was omitted, ~~this~~ it resulted~~s~~ in 58 LIs with leak rates ranging from 0.01 to 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}).

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 emission~~s~~ was~~ere~~ ~~identified~~ detected over the course of the campaign.

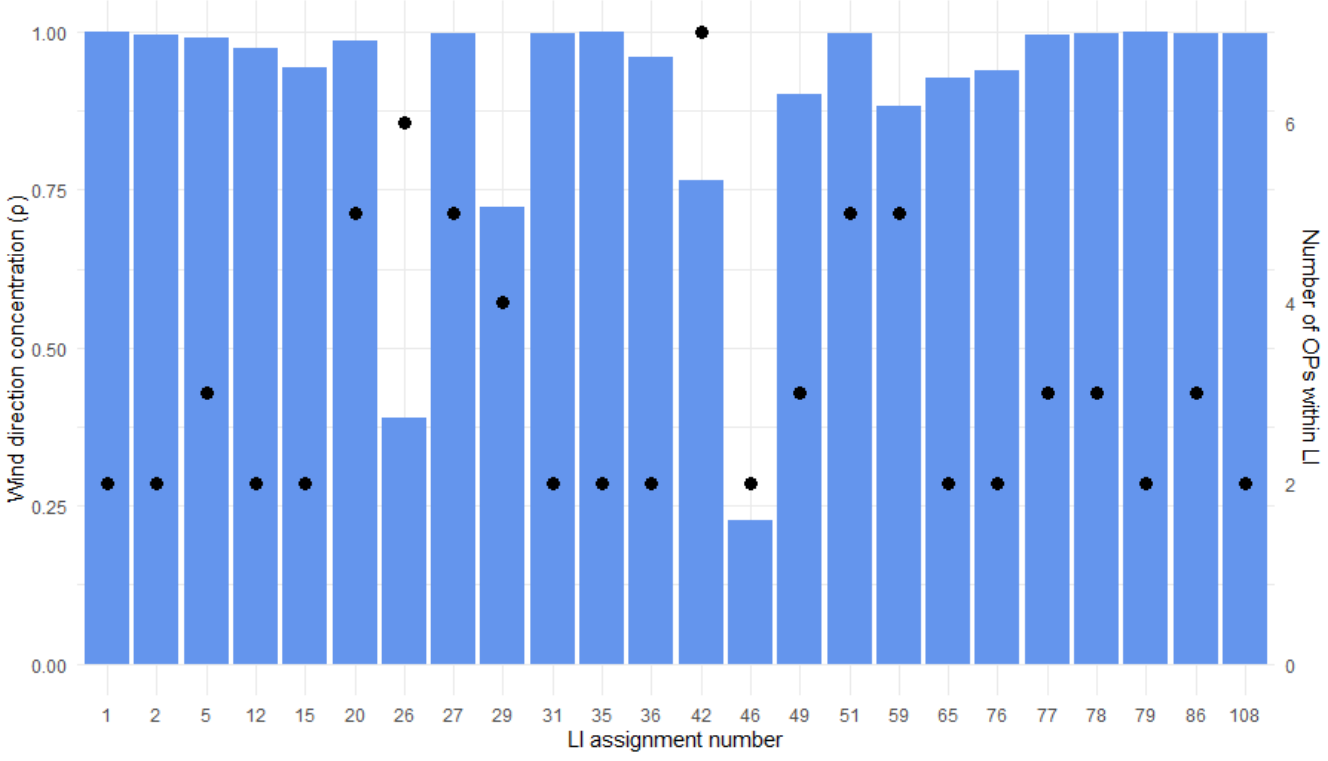


Figure 9: *Wind direction consistency and number of OPs per t~~T~~hermogenic leak indication*

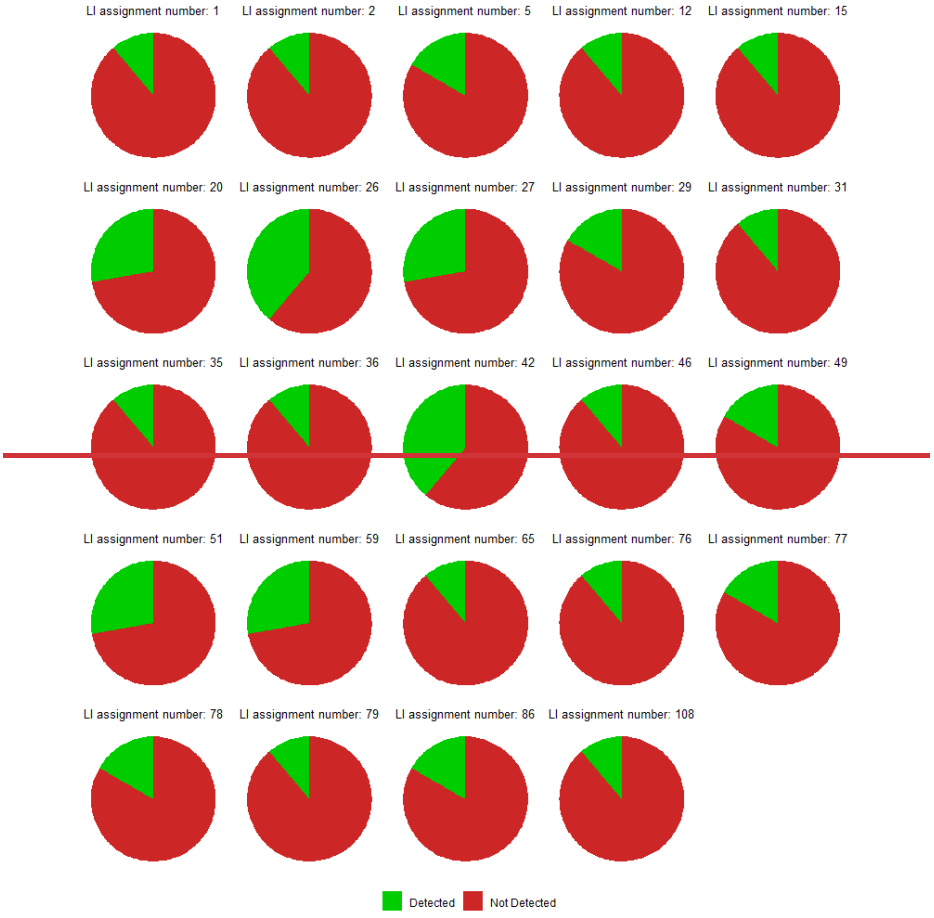
The effect of wind on detection of LIs was initially investigated by calculating the mean resultant length of wind directions when a t~~T~~hermogenic OP was detected. This was calculated using ***Equation 7***.

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

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 9*** 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~~to be detected.



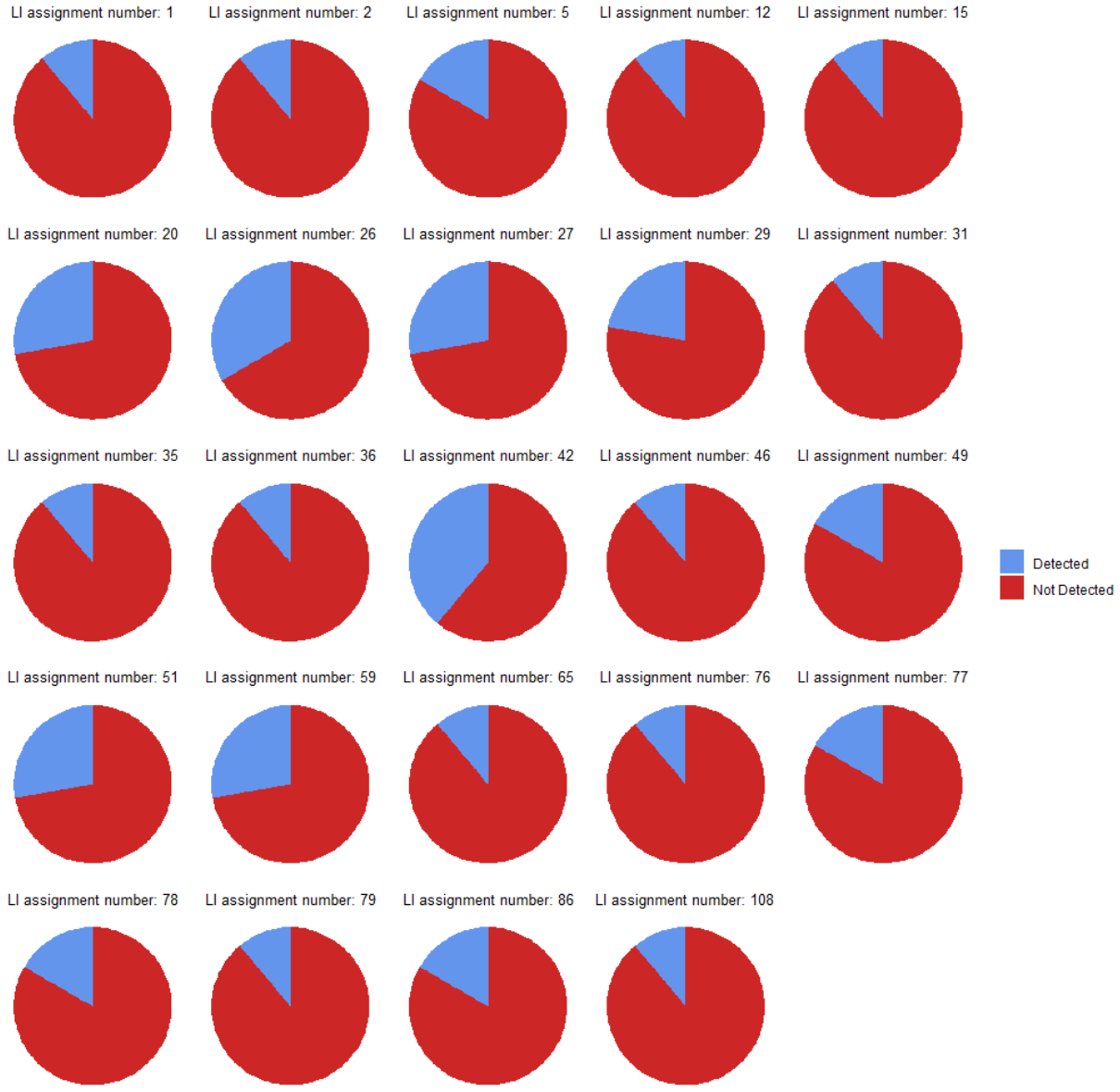


Figure 10: *Pie charts of each LI showing ~~number of mobile surveys they were detected vs not~~detection frequency.*

The number of mobile surveys 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 surveys~~, for the~~ .Of the 24 LIs detected over the course of this campaign, 12 LIs were detected on 2 mobile surveys, 6 were detected on 3 mobile surveys, 4 on 5 mobile surveys and 2 on 7, ~~this meant that the~~ resulting in an average probability of detection ~~was~~ of 0.18.~~,~~ D~~d~~etection versus non detection for each LI is demonstrated in ***Figure 10***. This low probability of detection highlights the need for surveys with multiple ~~passes~~repeats.

3.2 Emissions from other sources

While 177 of the 468 OPs were determined to be thermogenic, 39 were assigned as biogenic (8.3 %), 199 were pyrogenic (41.8 %) and 53 were not able to be assigned a source type. NO_x:CO_2 ratios were investigated for the pyrogenic OPs using the same methodology used for the ~~CH_4:C_2H_6~~ C_2:C_1 source assignment. 115 of the 199 pyrogenic OPs were able to be analysed in this way, 87 of these 115 OPs (75.7 %) 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 were more likely emissions from domestic heat and power generation (such as emissions from domestic boilers) *(Cliff et al., 2025)*.

Emissions from pyrogenic and biogenic sources ~~are~~ were compared to thermogenic ~~source~~ emissions at the OP stage on a mobile survey by mobile survey 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 11***.

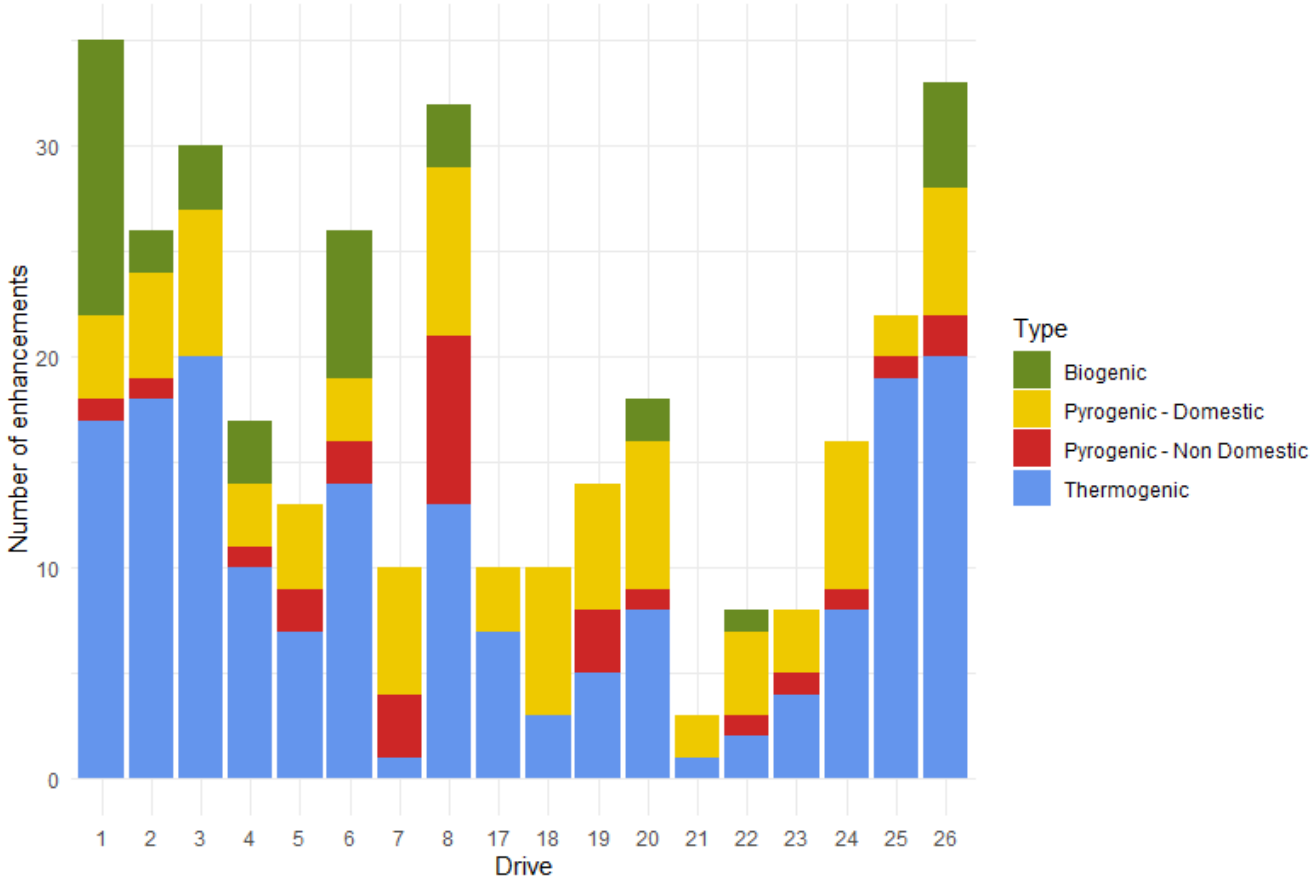


Figure 11: *Total number of enhancements from each source type for each ~~of the~~ mobile survey~~s~~.*

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.

3.3 Comparison to previous methods

The main alterations to this methodology from that presented in (*Weller et al., 2019)* (and other studies that were based off this method) ~~were that~~was the decrease in enhancement criteria ~~was changed~~ from 1.1 times the baseline to 1.05 times the baseline, a decrease in the clustering time window from 5 s to 1 s~~clustering by time was changed such that emissions within 1 s of each other were clustered instead of those within 5 s. Finally~~ and the addition of 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~~*1* shows the effect ~~that all~~ of each of these changes ~~to the methodology make~~ on the resulting detection of OPs and LIs.

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	468	58
		Yes	177	24

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

~~This shows~~These results show the new methodology could locate more LIs. 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~~ For the 27 LIs ~~from the old non filtered~~ detected in the original unaltered methodology 10 were small and 17 were very small. Finally, for the 6 LIs detected when applying the source determination step to the original unaltered methodology,~~from the old source filtered methodology,~~ 1 was small and 5 were very small. This shows the original~~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 included in

the method.; Tthis 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.

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~~ LIs. 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 12***.

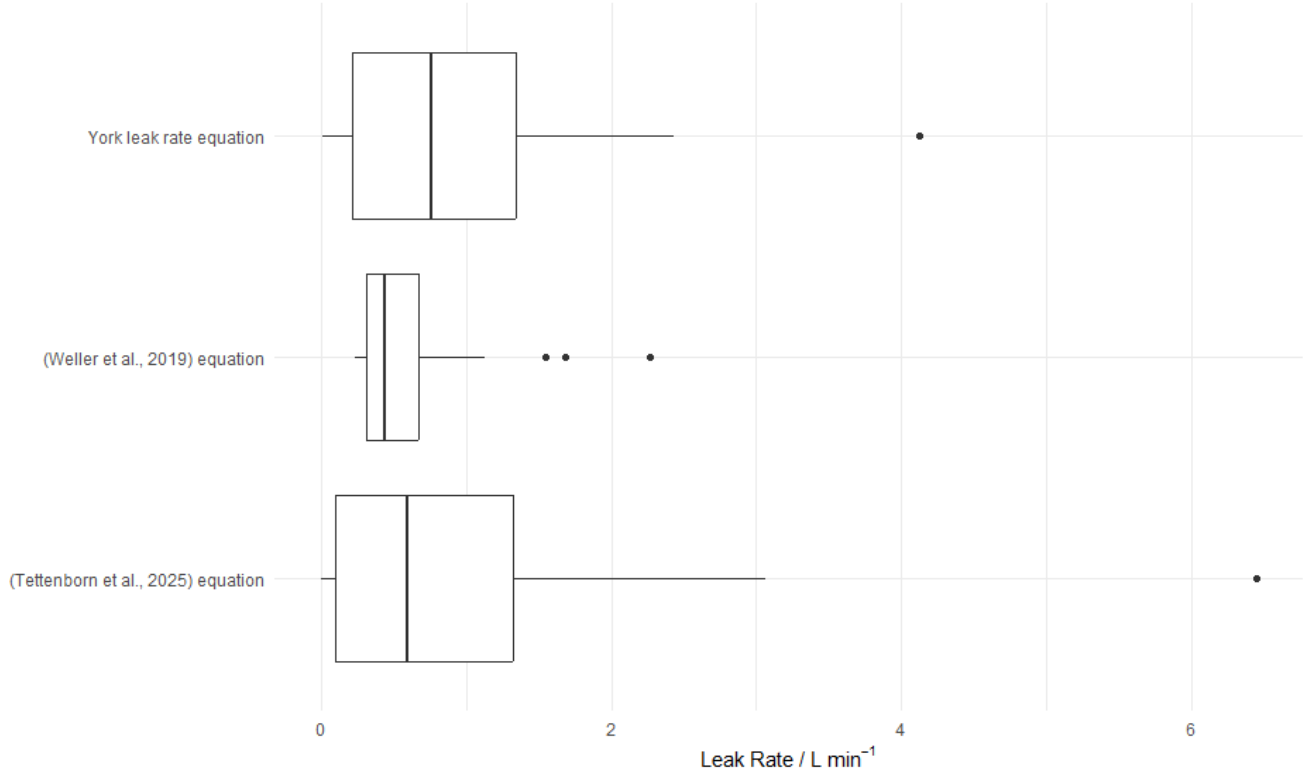


Figure 12: *Comparison of calculated leak rates of LIs from each of the 3 quantification equations.*

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 limitations of ~~surveying~~ instrumentation 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 an 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 times the background was selected so as to not incorporate small, diffuse emissions within the analysis. Response rate of the instruments was calculated to inform the time window for clustering, with both MGGA and TILDAS having sub ~~one second~~ 1 s response rate, the time clustering was ~~instead~~ limited to ~~one second~~ 1 s due to the limitations of GPS data collection speed. Employing the parameters used in previous methodologies, resulted in the~~These changes show previous methodologies would result in~~ detection of 27 LIs compared to the 58 LIs detected 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.

Source ~~appointment~~ determination 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, source ~~filtering~~ determination has helped to highlight that although t~~T~~hermogenic emissions from natural gas are the highest contributor to CH_4 ~~methane~~ emissions, pyrogenic emissions related to domestic heat and power generation also provide a high, but often overlooked contribution to a city's CH_4 ~~methane~~ emissions.

~~The~~ U~~u~~pdating ~~of~~ the quantification equation from a peak height approach to a peak area approach result~~ed~~s in a much wider range of leak rates being calculated in the study.~~,~~ H~~h~~owever, these values ~~are~~ were not as high as when quantified using the original equation presented in *(Tettenborn et al., 2025)*.

This new method has shown that ~~with~~ by changing enhancement criteria and time clustering parameters, it is ~~able~~ possible to detect many more LIs, ~~and~~ but that by applying a source ~~type filter~~ determination step at the OP detection stage there is a reduction in the number of detected LIs due to the reduction in~~it is capable of reducing~~ the incorrect assignment of ~~LIs~~OPs. However, the methodology has ability to improve further, primarily by ~~using~~ employing instrumentation that is capable of

detecting both CH_4 ~~methane~~ and C_2H_6 ~~ethane on one instrument,~~ so as to remove uncertainty related to time lag between ~~the~~ two instruments.~~, but s~~Secondly, improvement can be made by having all instrumentation and hardware able to operate at a sub ~~one second~~ 1 s ~~time~~ response 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, JF, JL. Initial draft of paper: TM. Subsequent drafts and/or revisions to 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.

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