Atmospheric Environment 230 (2020) 117440

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: http://www.elsevier.com/locate/atmosenv

The calibration and deployment of a low-cost methane sensor

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HIGHLIGHTS

- Low-cost, low-power methane sensor is investigated for use in a measurement network.
- Calibration shows a non-linear response to methane change between 2 and 6 ppm.
- Long-term deployment shows sensor is reliable and can be used to measure autonomously.
- Sensor can detect methane close to background but requires routine calibration.
- A network of low-cost sensors could help identification and mitigation of methane.

ARTICLE INFO

Keywords:
Methane
Mixing ratio
Measurement
Inexpensive
Calibration

ABSTRACT

Since 1850 the atmospheric mixing ratio of methane (CH_4), a potent greenhouse gas, has doubled. This increase is directly linked to an escalation in emissions from anthropogenic sources. An inexpensive means to identify and monitor CH_4 emission sources and evaluate the efficacy of mitigation strategies is essential. However, sourcing reliable, low-cost, easy-to-calibrate sensors that are fit for purpose is challenging. A recent study showed that CH_4 mixing ratio data from a low-power, low-cost CH_4 sensor (Figaro TGS2600) agreed well with CH_4 mixing ratios measured by a high precision sensor at mixing ratios between 1.85 ppm and 2 ppm. To investigate, as a proof of concept, if this low-cost sensor could be used to measure typical ambient CH_4 mixing ratios, we operated a TGS2600 in conjunction with a Los Gatos Ultra-portable Greenhouse Gas Analyzer (UGGA) in controlled laboratory conditions. We then explored the sensor's long-term reliability by deploying the TGS2600 near an onshore gas terminal to calculate emissions from May to July 2018. Our initial studies showed that previously published linear algorithms could not convert TGS2600 output to CH_4 mixing ratios measured by the UGGA. However, we derived a non-linear empirical relationship that could be used to reliably convert the output of a TGS2600 unit to CH_4 mixing ratios over a range of 1.85–5.85 ppm that agree to a high-precision instrument output to ±0.01 ppm. Our study showed that the TGS2600 could be used to continuously measure variability in CH_4 mixing ratios from 1.82 to 5.40 ppm for three months downwind of the gas terminal. Using a simplified Gaussian Plume approach, these mixing ratios correspond to an emission flux range of 0–238 g CH_4 s^{-1}, with average emission of 9.6 g CH_4 s^{-1} from the currently active North Terminal and 1.6 g CH_4 s^{-1} from the decommissioned South Terminal. Our work here demonstrates the feasibility of utilizing a low-cost sensor to detect methane leakage at concentrations close to ambient background levels, as long as the device is routinely calibrated with an accurate reference instrument. Having a widely deployed network of such low-cost CH_4 sensors would allow improved identification, monitoring and mitigation of a variety of CH_4 emissions.

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https://doi.org/10.1016/j.atmosenv.2020.117440
Received 11 November 2019; Received in revised form 10 February 2020; Accepted 21 March 2020

1. Introduction

Methane (CH_4) is a greenhouse gas that is also partially responsible for production and loss of tropospheric ozone. Since 1850 atmospheric CH_4 mixing ratios have increased from 715 ppb to 1865 ppb in 2005 (NOAA, 2019). This increase in mixing ratio is largely attributed to increased anthropogenic emissions (Turner et al., 2019). The ability to estimate the size and location of CH_4 emissions is essential for all mitigation strategies and associated policies (de Coninck et al., 2018). Current greenhouse gas emission inventories are principally compiled using industry-standard or recommended emission factors, which are based on measurements made at a limited number of sites, combined with estimates of activity levels (BEIS, 2018). Despite their widespread use, recent studies suggest that the use of emission factors may be insufficient to describe CH_4 emissions from complex processes and direct measurements are preferable (Cerri et al., 2017; Riddick et al., 2019b, 2019a; Turner et al., 2015; Yang et al., 2017). Emission estimates from direct measurements are generally calculated using gas mixing ratios measured downwind of the source.

For CH_4, current options for measuring near-ambient mixing ratios include spectroscopic instruments, such as the Los Gatos UGGA or Picarro G2301 CRDS instruments which cost between $50,000 and $100,000 each, or lower cost gas chromatographs costing around $10,000 each. These instruments are high precision (1 standard deviation < 2 ppb at 1 Hz) and have been used on long-term measurements campaigns for autonomous measurements (Connors et al., 2018; Riddick et al., 2018, 2017). However, power consumption demands (~260 W) mean they require a continuous electricity supply for deployment longer than an hour at most and insurance requirements for these expensive instruments demand locked and secure premises, which means that many remote locations cannot be measured.

Various government and industry/non-profit initiatives, such as the U.S. Department of Energy's MONITOR program and the Environmental Defense Fund's Methane Detector Challenge, support the research and development of new technologies to measure CH_4 mixing ratios. These methods range from satellite-based methods to new laser-based methods. However, current systems cost between $10,000 and $20,000 and the security of the instruments during measurement campaigns remain an issue. Due to the importance of CH_4 emission reduction strategies (IPCC, 2018), testing and deployment of low-cost CH_4 measurement devices is needed. This study investigates the use of very low-cost sensors (~$10) as an alternative to high-cost, high-precision instruments.

One example of a low power (~0.5 W), very low-cost sensor (~$10 US dollars) is the Taguchi Gas Sensor TGS2600(Figaro Engineering Inc., Osaka, Japan) that is designed to measure ambient CH_4 mixing ratios between 1 and 100 ppm (Figaro, 2005). In 2012, Eugster and Kling (2012) reported that CH_4 mixing ratios calculated from output of a TGS2600 at Toolik Lake, Alaska, USA were in good agreement ($R^2 = 0.85$) with CH_4 mixing ratios measured by a Los Gatos Research FMA 100 CH_4 analyzer. However, the range of CH_4 mixing ratios reported by Eugster and Kling (2012) was small, between 1.85 and 2 ppm, relative to mixing ratios observed near active sources of CH_4.

In this study, we report the findings of a measurement campaign to investigate the use of a TGS2600 low-cost CH_4 sensor as an alternative to a high-cost, high-precision instrument. Our goal is to assess the potential of deploying such sensors in large networks to identify a variety of methane leakage sources in order to improve greenhouse gas emission inventories. Our objectives are to: 1) Investigate if a TGS2600 sensor output can be used to estimate realistically observed CH_4 mixing ratio measurements of between 2 and 10 ppm; 2) Make long term measurements of CH_4 mixing ratios downwind of a natural gas point source using a TGS2600 without a mains power source or security measures; and 3) Estimate the emissions from a gas terminal using measured ambient methane mixing ratios and meteorological data. To our knowledge this is the first time that the low-cost TGS2600 sensor has been calibrated to quantify mixing ratios between 1.8 and 5.8 ppm and then used to calculate fugitive CH_4 emissions from a natural gas point-source.

2. Methods

2.1. Calculating methane mixing ratios from the TGS2600 output

The TGS2600 is a solid-state sensor that uses titanium dioxide (TiO_2) as the sensing material. When the TiO_2 is heated, gases in the air adsorb to its surface and as the concentration of CH_4 in air increases the resistance of TiO_2 decreases (Figaro, 2005). The schematic diagram showing the setup of the TGS2600 in this application can be found on the TGS2600 datasheet (Figaro, 2005) and in Fig. 2 in Eugster and Kling (2012). The TiO_2 has a resistance in clean air (R_0, Ω), i.e. air with ambient methane, which becomes lower in the presence of methane (R_s, Ω) and the ratio of these resistances (R_s/R_0) gives a measure of the CH_4 mixing ratio in air. However, the resistance of TiO_2 is also affected by the air temperature (T_a, °C) and relative humidity (rH, %) and the ratio of resistance must be corrected for these factors (Eq. (1)). The uncalibrated CH_4 mixing ratio ($[CH_4]_{raw}$, ppm) can be calculated as a linear function of the corrected ratio of these resistances $(R_s/R_0)_{corr}$ following the equation of Eugster and Kling (2012) (Eq. (2)).

$$\left(\frac{R_s}{R_0}\right)_{corr} = \left(\frac{R_s}{R_0}.(0.024 + 0.0072.rH + 0.0246.T_a)\right) \tag{1}$$

$$[CH_4]_{raw} = 1.8280 + 0.0288.\left(\frac{R_s}{R_0}\right)_{corr} \tag{2}$$

2.2. Calibrating the TGS2600

As Eugster and Kling (2012) reported, the TGS2600 does not measure CH_4 well in low relative humidity, i.e. $< 40\%$. Because of this, we could not easily calibrate the TGS2600 against cylinder reference standard gases of known (certified) concentrations, as these standards are typically very dry (often only a few ppm H2O). As an alternative, we calibrated the TGS2600 by running it alongside a Los Gatos Research (Mountain View, CA, USA) Ultra-portable Greenhouse Gas Analyzer (UGGA).We conducted three side-by-side experiments: 1.21st to April 22, 2018 in an indoor laboratory at the University of Manchester, UK; 2.24th June at St Michael's Church, Rampside, UK and; 3. Between the 24th August and September 3, 2018 at University of Manchester's measurement site at Plumpton Hall Farm, Lancashire, UK. In addition to side-by-side measurements with the UGGA, a second TGS2600 was run on the September 3, 2018 to test for differences in output between sensors from the same manufacturer.

Using Equations (1) and (2), we calculated CH_4 mixing ratios using TGS2600 voltage output, temperature, and relative humidity data. Equation (2), the algorithm for calculating $[CH_4]_{raw}$, was then tuned for optimum performance using the UGGA CH_4 mixing ratio data as a reference. The metrics for selecting the most appropriate algorithm are the gradient and R^2 of the mixing ratios compared to the UGGA mixing ratios and the area under the mixing ratio time series. The area under the mixing ratio time series is used as a measure of mass inferred by the measurements and thought to be the best metric for choosing the most appropriate algorithm because it is expected that the because of the passive nature of the TGS2600 it may not to respond to changes in mixing ratio exactly at the same time as the UGGA, which has air pumped through the measurement cavity.

2.3. Case study – measuring CH_4 emissions from a natural gas terminal

2.3.1. Methane emission source

The Rampside gas terminal in Barrow-in-Furness, UK was chosen as the test site for the TGS2600 which was set up 1.5 km downwind at St. Michael's church. This site was chosen because of its accessibility and the relatively low emissions from the gas terminal, which provided a good test of the detection ability of the TGS2600. The Rampside gas terminal collects and processes natural gas from platforms in Morecambe Bay (Fig. 1). It has two terminals - the North Terminal and the South Terminal. In 2016 the South Terminal was decommissioned and all gas produced in South Morecambe Bay was re-routed to the North Terminal. Spirit Energy, which operates the terminal, expected that in 2018 the North Terminal would operate more efficiently than in previous years and closer to design capacity, while the shuttered South Terminal would be expected to have zero emissions (R. Davidson, Spirit Energy, pers. comm.).

Air from the North Terminal arrives at St Michael's church when the wind blows from between 270 and 315°. During May, June and July 2018, when our measurements were made, the Morcambe Bay platforms collectively produced an average of 0.49 Gg CH_4^{-1} day^{-1}(OGA, 2018). The latest CH_4 emission estimate published for the terminal was 0.68 Gg yr^{-1}, or 0.4% of annual production, in 2015, with the largest source of emissions associated with natural gas processing (DEFRA, 2019). To put the emissions from this site in context, a landfill of area 0.1 km^2 emits 2 Gg CH_4 yr^{-1}(Riddick et al., 2017), while in 2017 larger UK gas terminals, Bacton and Easington, emitted 1.6 and 1.0 Gg CH_4 yr^{-1}, respectively (BEIS, 2018).

2.3.2. *Deployment of TGS2600*

In this application, the TGS2600 was configured to give a DC voltage output corresponding to CH_4 mixing ratios of between 1 and 10 ppm. Sampling was controlled by an Arduino Uno microcomputer (Arduino, Ivrea, Italy), which digitizes and logs the voltage output from the sensor, and records the date and time, temperature, and relative humidity to a SD card at 1 min intervals. The sensor was installed in the grounds of St Michael's Church, Rampside on May 4, 2018 (Fig. 1) and was powered by a 35 Ah lead acid battery which had sufficient capacity to operate the sampling and logging hardware for 7 days. The site was chosen because of proximity to the gas terminal, low background mixing ratios, and ease of access.

2.3.3. *Meteorological data*

Meteorological data were also collected at St Michael's Church, Rampside using a wireless weather station (Maplin, UK) attached to a mast 200 m from the nearest building and 2 m above the ground. The weather station was position 10 m away from the gas sensor and the location was chosen to jointly ensure an obstruction free wind field and security. Meteorological data were sampled and recorded at 1-min intervals and included: wind speed (u, m s^{-1}), wind direction (*WD*, ° to North), air temperature (T_a, K), relative humidity (*RH*, %), rain rate (R, mm hr^{-1}), irradiance (I, W m^{-2}) and air pressure (P, Pa).

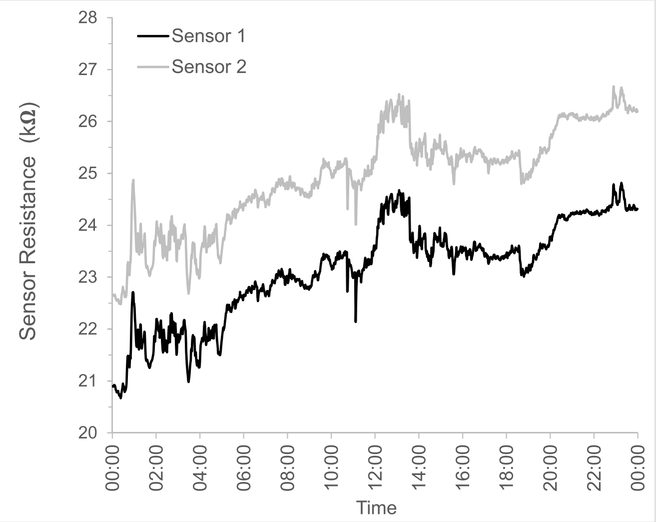

Fig. 2. The sensor resistance from the original TGS2600 sensor (Sensor1) and a second TGS2600 (Sensor 2) run beside each other on the September 3, 2018 at Plumpton Hall Farm, Lancashire, UK.

2.3.4. *Gaussian Plume model*

A Gaussian plume (GP) model was used to calculate the emissions from the gas terminal. A GP model describes the mixing ratio of a gas as a function of distance downwind from a point source (Seinfeld and Pandis, 2016). As a gas is emitted, it is entrained in the prevailing ambient air flow and disperses in the y and z directions (relative to a mean horizontal flow in the x direction) with time, forming a dispersed concentration cone. The concentration of the gas (X, μg m^{-3}), at any point x metres downwind of the source, y metres laterally from the centre line of the plume and z metres above ground level can be calculated (Eq. (3)) using the source strength (Q, g s^{-1}), the height of the source (h_s, m) and the

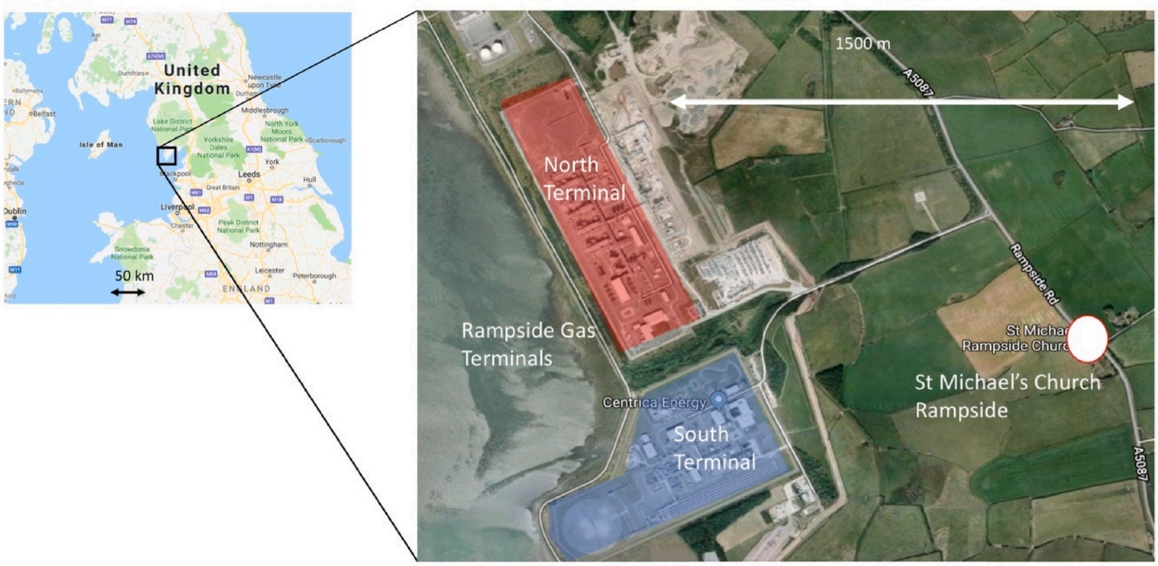

Fig. 1. Location of the North and South Terminals at the Rampside gas terminal site in relation to the measurement location at St Michael's Church, Rampside. Images courtesy of Google Maps.

Pasquill-Gifford stability class (PGSC) as a measure of air stability. The standard deviation of the lateral (σ_y, m) and vertical (σ_z, m) mixing ratio distributions are calculated from the Pasquill-Gifford stability class (PGSC) of the air (Pasquill, 1975; Busse and Zimmerman, 1973; US EPA, 1995). The GP model assumes that the vertical eddy diffusivity and wind speed are constant and there is total reflection of CH_4 at the surface.

$$X(x,y,z) = \frac{Q}{2\pi u \sigma_y \sigma_z} e^{-\frac{y^2}{(2\sigma_y)^2}} \left(e^{-\frac{(z-h_s)^2}{(2\sigma_z)^2}} + e^{-\frac{(z+h_s)^2}{(2\sigma_z)^2}} \right) \tag{3}$$

2.3.5. Gaussian Plume model parameterization

Data used as input to the GP model are filtered by wind direction and only air from the North Terminal (270°–315°) in the analysis, these include: wind speed, wind direction, temperature, CH_4 mixing ratio at Rampside church, background CH_4 mixing ratio and the PGSC. The PGSC is estimated from wind speed and irradiance data (Seinfeld and Pandis, 2016), as measured by the meteorological station (Supplementary Material Section 1).

2.3.6. Uncertainty in emissions

We conducted an uncertainty analysis for the emissions using the GP approach. Scenarios were run in which individual input variables were changed and the resulting changes in average CH_4 emissions calculated for the entire measurement period were tracked. Individual uncertainties were determined by the precision of the instrument: the TGS2600 (calculated below in Section 3.2); the wind speed (the result of a ± 0.5 m s^{-1} measurement uncertainty); the air temperature (± 0.5 °C); and the uncertainty in relative humidity ($\pm 0.5\%$). Ordinarily, we would expect there to be uncertainty in assigning a PGSC value for use in the GP model. However, we found this not to be the case here and we discuss this further in Section 3.5. An additional uncertainty is that the TGS2600 is cross-sensitive to carbon monoxide (CO), iso-butane, ethanol, and hydrogen. However, these gases are not expected to pose a significant problem of contamination at a coastal site in marine inflow conditions. An overall uncertainty for the CH_4 emission estimate is presented as the as the root mean square deviation (RMSD) of the individual uncertainties.

3. Results

3.1. TGS2600 output reproducibility

To test for differences in TGS2600 output between sensors from the same manufacturer, a second TGS2600 (Sensor 2, Fig. 2) was run next to the original TGS 2600 sensor (Sensor 2, Fig. 2) on the September 3, 2018 at Plumpton Hall Farm, Lancashire, UK. Despite the resistance of the two sensors having nearly identical temporal response to changes in CH_4 concentrations, the resistance of the original and second sensor correlate with $R^2 = 0.995$, $m = 1.015$, p-value = 0, there is an offset of 1.45 kΩ between the resistance of the two sensors at the same CH_4 concentration (between 1.9 and 3.3 ppm) (Fig. 2).

3.2. TGS2600 calibration

After the calibration periods of April 21, 2018, 24th June and between the August 24, 2018 and September 3, 2018,the output data of the TGS2600were used to calculate $(R_s/R_0)_{corr}$ using Eq. (1). This $(R_s/R_0)_{corr}$ value was then used to calculate the mixing ratio $[CH_4]_{raw}$ (Eq. (2)). The calculated CH_4 mixing ratios ranged from 1.85 ppm to 1.86 ppm (Red line, Fig. 3) while the UGGA measurements varied between 1.85 ppm and 6.0 ppm (Black line, Fig. 3). This strongly suggests that our sensor did not behave the same way as the sensor used by Eugster and Kling (2012).

As our setup and operation were exactly the same as Eugster and Kling (2012), we suggest there could either be manufacturing differences between our TGS2600 and the sensor used by Eugster and Kling (2012) or the very narrow range of CH_4 mixing ratios measured by Eugster and Kling (2012), 1.85–2 ppm, means that Eq. (2) gives a poor fit at higher mixing ratios. To better calibrate the TGS2600 for a larger mixing ratio range, we calculated alternative linear and heuristically derived non-linear empirical relationships for our TGS2600 sensor using data from the UGGA to tune the algorithms (Fig. 3; Table 1).

The gradient, m, and R^2 values of the linear regression between the $[CH_4]_{raw}$ values and the UGGA mixing values (Table 1) suggests the linear relationship (Eq. (4) Table 1; orange line Fig. 3) generates the best fitting $[CH_4]_{raw}$ estimates when compared to the UGGA mixing ratios. However, when using the area under the mixing ratio curve as a metric, the non-linear algorithm (Eq. (5) Table 1; grey line Fig. 3) agrees best with the area under the UGGA mixing ratios (black line Fig. 3), while the linear relationship (Eq. (4) Table 1) overestimates the mass emitted by 25%. This suggests that, even though the $[CH_4]_{raw}$ mixing ratios calculated by Eq. (5) are not directly comparable, (i.e. do not occur at exactly the same time as the UGGA mixing ratios), it gives the best agreement when calculating the time averaged mixing ratio.

Methane mixing ratios calculated using Eq. (5) were generally slightly lower than those measured by the UGGA, with the mean difference between the TGS2600 and UGGA CH_4 mixing ratios of -0.004 ppm. Using the 95% uncertainty intervals, by comparing the UGGA CH_4 mixing ratios to those calculated from the TGS2600 output we report an uncertainty in CH_4 mixing ratios at $\pm$ 0.01 ppm. The TGS2600 also took longer to respond to changes in mixing ratio than the UGGA for the range of measured mixing ratios (background concentrations of about 2 ppm–7 ppm).We also found that over time the TGS2600 drifted by 0.002 ppm per day and the TGS2600 output was very uncertain when relative humidity was less than 40%.

3.3. Methane mixing ratios in air from the gas terminal

Methane mixing ratios were calculated using TGS2600 measurements made at Rampside church and calculated using Eq. (5). These calculated CH_4 mixing ratios indicate enhancements can be observed most of the time when the wind comes from the North Terminal (Fig. 4). The largest enhancement, 5.4 ppm, was observed on the June 2, 2018 (grey dots; Fig. 4) with the mean CH_4 mixing ratio of 2.0 ppm detected over the three months. The background mixing ratio was taken as the minimum mixing ratio over a rolling 24 h period to capture intermittent enhancements from other sources in the "background" measurement.

3.4. Methane emissions from the gas terminal

Fifteen-minute averaged CH_4 emissions from the Rampside gas terminal were calculated with the Gaussian plume model using measured mixing ratios and matching meteorological data (as described in Section 3.3). As the location of emissions within each terminal is unknown, for the purpose of this calculation we assume the mixing ratio measured by the sensor corresponds to the centre of a Gaussian plume (i.e. the peak value) whenever the wind direction is between 270° and 315° for the North Terminal and between 225° and 270° for the South Terminal. For other wind directions no emission rate is calculated for either terminal.

Under this assumption we calculate a maximum emission from the North Terminal of 238 g s^{-1}, observed on the 13th July (black dots; Fig. 4), with a mean emission from the North Terminal of 9.6 g s^{-1}. This is over six times higher than the mean emission of 1.6 g s^{-1} calculated for the South Terminal during the same period. Our results suggest that even though gas is not passing through the South Terminal, residual CH_4 continues to be emitted from the site.

Assuming the measured mixing ratio in air that has passed over either terminal is representative of the centre of a Gaussian plume is clearly a significant simplification. Emissions within each terminal are likely to emanate from one or several point sources. Therefore, our

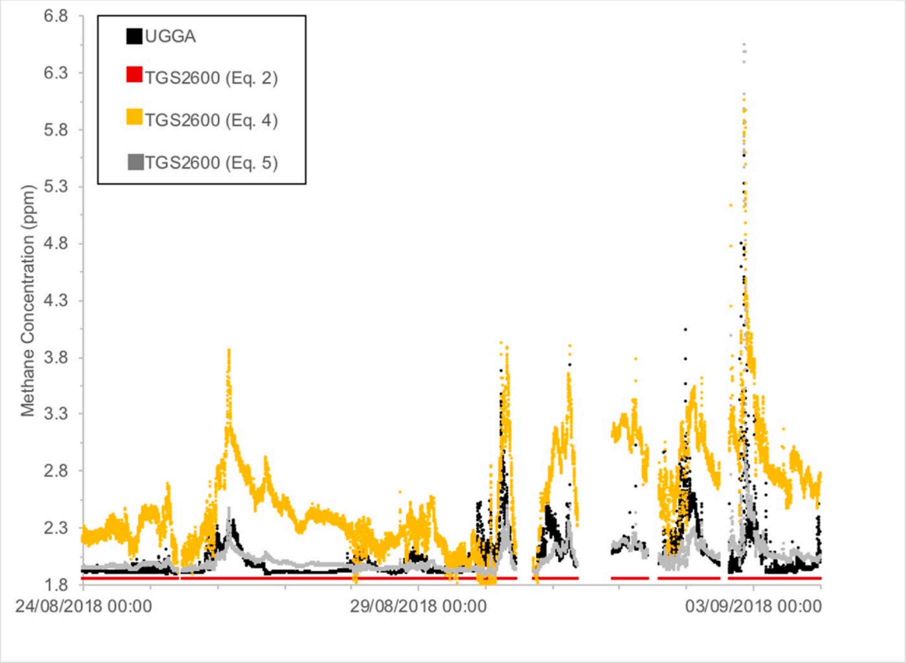

Fig. 3. Methane mixing ratios calculated by the TGS2600 output and the method of Eugster and Kling (2012) (Eq. (2); Red dots), TGS2600 output and a linear relationship (Eq. (4); orange dots), TGS2600 output and a non-linear relationship (Eq. (5); grey dots) and the UGGA (black dots) between the 24th August and September 4, 2018 at University of Manchester's Site at Plumpton Hall Farm. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1

Comparison of algorithms to derive $[CH_4]_{raw}$ using gradient, R^2 and the area under the mixing ratio line in Fig. 3.

Algorithm	Eq. #	Line colour Fig. 3	Equation to calculate $[CH_4]_{raw}$	m	R^2	Area under line
UGGA		Black				34,619
Eugster & Kling	2	Red	$1.8280 + 0.0288.\left(\frac{R_s}{R_0}\right)_{corr}$	0.003	0.27	31,537
Linear	4	Orange	$-7.37 + 12.74.\left(\frac{R_s}{R_0}\right)_{corr}$	1.19	0.27	43,833
Non-linear	5	Grey	$1.8 + 0.09 \cdot \exp\left(11.669 \cdot \left(\left(\frac{R_s}{R_0}\right)_{corr} - 0.7083\right)\right)$	0.57	0.23	34,690

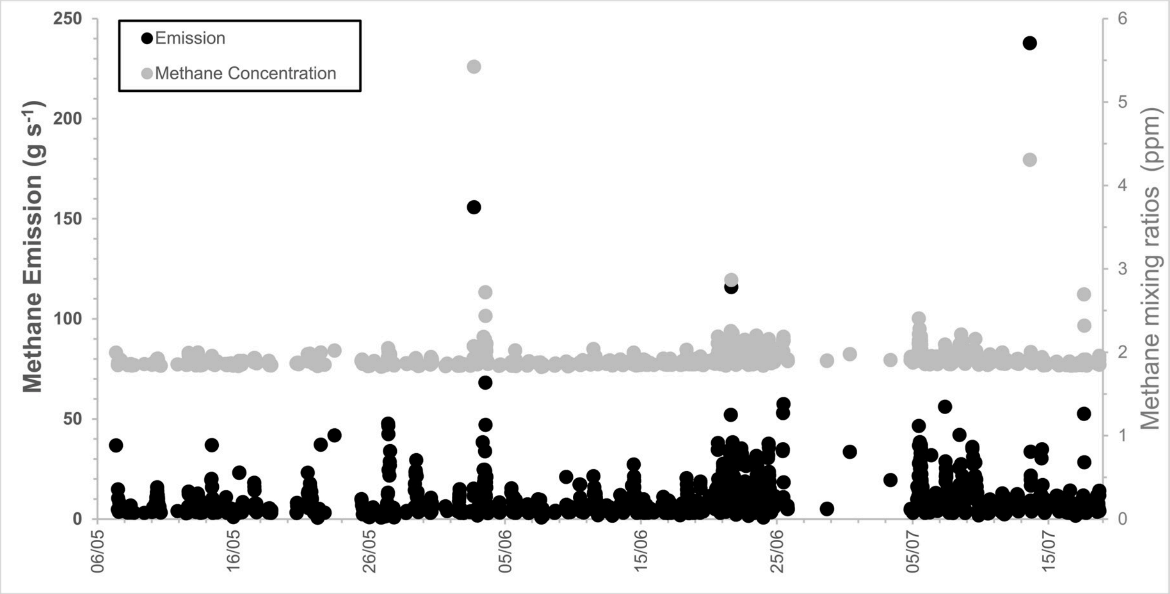

Fig. 4. Time series of 15-min averaged CH_4 concentrations measured by the TGS2600 at St Michael's Church and calculated CH_4 emissions using Eq. (5) from Rampside gas terminal, Rampside, Cumbria.

calculated mean emission rate will be biased low, as in reality many of our measurements will not represent the central (maximum) mixing ratio within the plume. Using only a single sensor we do not have enough information to constrain both spatial and temporal emission patterns. We present the emission estimates above to demonstrate the potential utility of these sensors; they are not intended to be considered as accurate estimates of typical emissions from this site on annual timescales.

3.5. *Uncertainty in emission estimates*

As discussed in Section 2.3.6, we would expect there to be uncertainty in assigning a PGSC when using a GP model. Here we have found little uncertainty at the site as it was very windy and the wind from the gas terminal to the church was from the open ocean. This meant that for 75% of the measurements the wind was greater than 6 m s^{-1}, i.e. corresponding to neutral conditions (Supplementary Materials Section 1), with the remaining 25% in slightly unstable conditions.

We estimate that contamination by CO, iso-butane, ethanol, and hydrogen will not affect the TGS2600 CH_4 mixing ratio measurements, assuming the gas terminal is not the source of the contamination. Any background increase in contaminant mixing ratio will result in an increase in the rolling background mixing ratio which will be included when calculating the CH_4 emission using the GP model.

Scenarios were run using the GP model to reflect variability in the TGS2600 measured CH_4 mixing ratio (±0.01 ppm), air temperature, wind speed and relative humidity. Uncertainties in air temperature (±0.5 °C) affected the calculated average emission the most (±13%). This was similar to the uncertainties in wind speed ±9%) and TGS2600 CH_4 mixing ratio (±8%), while the uncertainty in relative humidity (±0.5%) affected the average calculated emission the least (±3%). We estimate the RMSD in average CH_4 emission calculated over the measurement period to be ±18%.

4. Discussion

4.1. *Low-cost sensor*

Our direct comparison of the CH_4 mixing ratios measured by the TGS2600 and the UGGA indicate that the TGS2600 can be used to reliably measure CH_4 mixing ratios from 1.8 up to 6 ppm using an empirical correction. The main drawback to the TGS2600 is that the sensor output representing low CH_4 mixing ratios (between 1 and 10 ppm) appears to be highly variable between sensors. However, since only two sensors were tested, we cannot yet assess the typical variability for a larger number of sensors. The differences between individual sensors may be due to differences in manufacturing that affect R_s at low CH_4 mixing ratios, and requires a high-precision instrument to calibrate the sensor. The algorithm Eugster and Kling (2012) used to calculate CH_4 mixing ratios (Eq. (2)) was different from the non-linear empirical relationship used in this study (Eq. (5)) and may reflect significant differences in individual sensor response to changes in CH_4 concentrations. This means that the TGS2600 may only be useful to those with access to high-precision CH_4 instruments. We suggest manufacturers could make these simple sensors more consistently if they were looking to market the TGS2600 as an accurate “off the shelf” CH_4 sensor. Without this, we conclude that empirical corrections may need to be derived for individual sensors to yield meaningful data.

In addition to the calibration, the TGS2600 did not to respond to changes in mixing ratio exactly at the same time as the UGGA. This is expected to be related to the passive nature of the TGS2600 sensor in contrast to the UGGA, where air is pumped through the measurement cavity. The TGS2600 sensor cannot be calibrated using calibration gases as the very low humidity of these gases (<40%) result in unstable output. The most accurate method for calibration was found to be running the TGS2600 next to a high-precision instrument for a period of time. The TGS26000 was also noted to drift over time. Our measurements estimate the output varied by 0.002 ppm per day indicating that calibration checks should be made frequently to ensure that any drift is corrected and remains linear over time. We suggest that, for a sensor with similar output to the one used in the study, calibrations every two months should be adequate to quantify any drift, i.e. the drift over two months of 0.12 ppm should be observable.

Aside from these negatives, the TGS2600 succeeded in measuring CH_4 mixing ratios, within ±0.01 ppm to mixing ratios measured by the UGGA, autonomously and continuously over a period of three months. The power consumption meant that it could be run for seven days from a 35 Ah lead acid battery. The TGS2600 output was logged to an SD card, the CH_4 mixing ratio calculated by post-processing, and the CH_4 emissions from a source calculated using a Gaussian Plume model. These data confirm the proof of concept that individual TGS2600 sensors could be run as part of a network to estimate changes in the CH_4 emission landscape, however their measurements are only inter-comparable and reliable if they are frequently calibrated with an high precision instrument.

4.2. *Methane emissions from the rampside gas terminal*

The measurements made at St Michael’s Church, Rampside, 1.5 km from the source, estimated the average CH_4 emission from the Rampside gas North Terminal between May and August 2018 at 9.6 g CH_4 s^{-1} with a peak emission of 238 g CH_4 s^{-1}. In addition, we measured an average CH_4 emission from the decommissioned South Terminal of 1.6 g s^{-1}. The identification of non-zero emissions from the South Terminal demonstrates the utility of direct emission monitoring using continuous ground-based measurement. To give these emissions some context, the average emission from the North Terminal can be used to extrapolate up to an annual estimate of 0.30 Gg CH_4 yr^{-1}, which is comparable to the 2018 UK National Atmospheric Emissions Inventory (NAEI) emission estimate of 0.45 Gg CH_4 yr^{-1}. The difference between our estimate and the NAEI estimate is expected as the single sensor method and simple Gaussian analysis used in this study was predicted to underestimate the emission.

Overall, this study shows that a low-cost sensor can be used to make direct CH_4 mixing ratio measurements and the data collected can be used to calculate realistic CH_4 emissions from an onshore gas terminal. In contrast to emission factor generated values, which only estimate emission from known sources, direct measurements can show temporal and geographical variability in emissions and can be used to indicate where and when unknown leakage of CH_4 occurs. Many sensors surrounding the perimeter of the site networked together could be used to explicitly identify the size and location any source of leakage in almost real-time and the problem could be fixed before significant CH_4 is lost to the atmosphere.

4.3. *Implications for large low-cost sampling networks for methane*

Although some additional uncertainty in emission estimates may be unavoidable when low-cost sensors are used, this study indicates that low-cost sensors, when properly calibrated against high-precision instruments, can overcome many of the logistical and cost issues associated with higher-cost, high-precision sensors and can be used to monitor emissions locally and at a distance from a source. We suggest that networks of low-cost sensors could be deployed and provide a “first-look” at local emission landscapes over a wider area and longer time period than is possible with costly sensors and can be used to identify emission hotspots that should be investigated further using high-precision instruments. Such a wide deployment of low-cost sensors would facilitate more realistic greenhouse gas inventories than those currently developed using emission factors and activity levels that do not fully capture the actual leakage processes that may be occurring.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Stuart N. Riddick: Conceptualization, Investigation, Data curation, Formal analysis, Writing - original draft, Writing - review & editing. **Denise L. Mauzerall:** Conceptualization, Supervision, Writing - original draft, Funding acquisition, Writing - review & editing. **Michael Celia:** Supervision, Funding acquisition, Writing - review & editing. **Grant Allen:** Investigation, Data curation, Formal analysis, Writing - review & editing. **Joseph Pitt:** Investigation, Data curation, Formal analysis, Writing - review & editing. **Mary Kang:** Funding acquisition, Writing - review & editing. **John C. Riddick:** Investigation, Data curation, Formal analysis, Writing - review & editing.

Acknowledgements

We would like to thanks the curators of St Michaels Church, Rampside, Jim and Brenda Webster for their assistance during the measurement campaign as well as the Reverend Lucie Lunn for allowing us to measure at the church. We also acknowledge funding from the National Oceanic and Atmospheric Administration (Grant # AWD1004141) and Princeton University's Science, Technology, and Environmental Policy (STEP) program for research support.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2020.117440.

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