Beef cattle methane emissions measured with tracer-ratio and inverse dispersion modelling techniques

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Abstract. The development and validation of management practices to mitigate greenhouse gas (GHG) emissions from livestock requires accurate emission measurements. This study assessed the accuracy of a practical inverse dispersion (IDM) micrometeorological technique to quantify methane (CH_4) emitted from a small cattle herd (16 animals) confined to a 63 x 60 m experimental pen. The IDM technique calculates emissions from the increase in CH_4 concentration measured downwind of the animals. The measurements were conducted for 7 days. Two types of open-path (OP) gas sensors were used to measure concentration in the IDM calculation: a Fourier transform infrared spectrometer (IDM-FTIR) or a CH_4 Laser (IDM-Laser). The actual cattle emission rate was measured with a tracer-ratio technique using nitrous oxide as the tracer gas. We found very good agreement between the two IDM emission estimates (308.1 ± 2.1 (mean ± s.e) and 304.4 ± 8.0 g CH_4 head^{-1} d^{-1} for the IDM-FTIR and IDM-Laser, respectively) and the tracer-ratio measurements (301.9 ± 1.5 g CH_4 head^{-1} d^{-1}). This study shows that a practical IDM measurement approach can provide an accurate method of estimating cattle emissions.

Keywords: micrometeorological techniques, GHG emissions, beef cattle, spectroscopy, open-path gas sensors

1 Introduction

Agriculture is the main source of anthropogenic methane (CH_4) emitted to the atmosphere, which includes emissions from ruminants, rice agriculture, waste treatment, and biomass burning (Solomon et al., 2007). Methane is an important greenhouse gas (GHG) with a global warming potential that is 28 times that of carbon dioxide (CO_2) in a 100 year time (Myhre et al., 2013). Enteric CH_4 from livestock is a major source of GHG emissions. A significant effort is being made to mitigate these emissions through diet modification feed supplements, farm management, grazing strategies, and animal breeding (Min et al., 2020; Vyas et al., 2018); with ruminant nutritional management strategies seen as the most direct impact mitigation option (Cottle et al., 2011). Increasingly there is a requirement for mitigation claims to be validated when these practices are applied on-farm (DoE, 2014), and simple and accurate methods for on-farm emission measurements are required.

On-farm enteric emissions have been measured using three main techniques. 1) Portable respiration hoods for tethered and non-tethered animals (Garnsworthy et al., 2012; Zimmerman and Zimmerman, 2012) directly measure the gas concentration of incoming and exhaust air from individual animals. However, this technique limits the animal's movements, requires intensive training for animals and labor, and it does not account for emissions from the animal rectum. 2) Tracer-ratio gas releases from the animal (Johnson et al., 1994), such as SF_6 (Grainger et al., 2007), assumes the tracer gas and the emitted CH_4 have similar transport paths, so that a tracer measurement can establish the CH_4 emission rate. This is a simple technique, but there are challenges with logistics and handling animals similar to the respiration hood technique. 3) Micrometeorological techniques are typically considered a herd-scale measurement, where the emission rate is calculated from the measurement of enhanced gas concentrations downwind of an animal herd (Harper et al., 2011), and these include the mass balance technique (Laubach et al., 2008; Lockyer and Jarvis, 1995), eddy covariance (Dengel et al., 2011; Felber et al., 2015), and inverse dispersion techniques (Flesch et al., 2005; Todd et al., 2014). The main advantage of micrometeorological techniques is that they do not interfere with the animals or the environment.

The objective of this study was to examine the accuracy of a practical inverse dispersion modelling (IDM) technique for measuring CH_4 emissions from beef cattle. The IDM technique offers the possibility of relatively simple emission measurements, without the need for animal handling or modifying animal behavior. In this study two IDM techniques are used to measure emissions from a small herd of confined cattle, and the results tested against a robust tracer-ratio based measurement.

2 Materials and Methods

2.1 Experimental design

The study took place at the Chiswick pastoral research laboratory (30° 37' S, 151° 33' E) in Armidale, New South Wales, Australia in February 2013. Methane emissions were measured from 16 Angus steers placed in a temporary 63 × 60 m pen (Fig. 1) located in a flat and open field. There were no other cattle or animal manure storages nearby during the study, and the nearest trees (30 m height) were at least 300 m from the site. Vegetation in the field was removed prior to the study and no pasture was available to graze.

The study cattle had an average body weight of 373 kg (standard deviation = 59 kg). The animals were fed a blended oaten/lucerne chaff ration (90.2% of dry matter, 15.1% crude protein) dispensed from automated feeders (Bindon, 2001) that recorded the individual animal intakes. The feeders were cleaned daily, and any remaining feed was weighed to check that the total consumed amount matched the sum of the individual animal intake. Feed and water were offered *ad libitum*. This feeding regime began four weeks prior to the emission measurements. During the seven-day emission measurement period, the average dry matter intake (DMI) was 11.9 kg $head^{-1}$ d^{-1}. Cattle manure was not removed during the measurement period. Approximately two weeks before the measurements, each animal was fitted with a backpack (glued to their back) to hold a small nitrous oxide (N_2O) gas canister used for the tracer-ratio emission measurements (Jones et al., 2011).

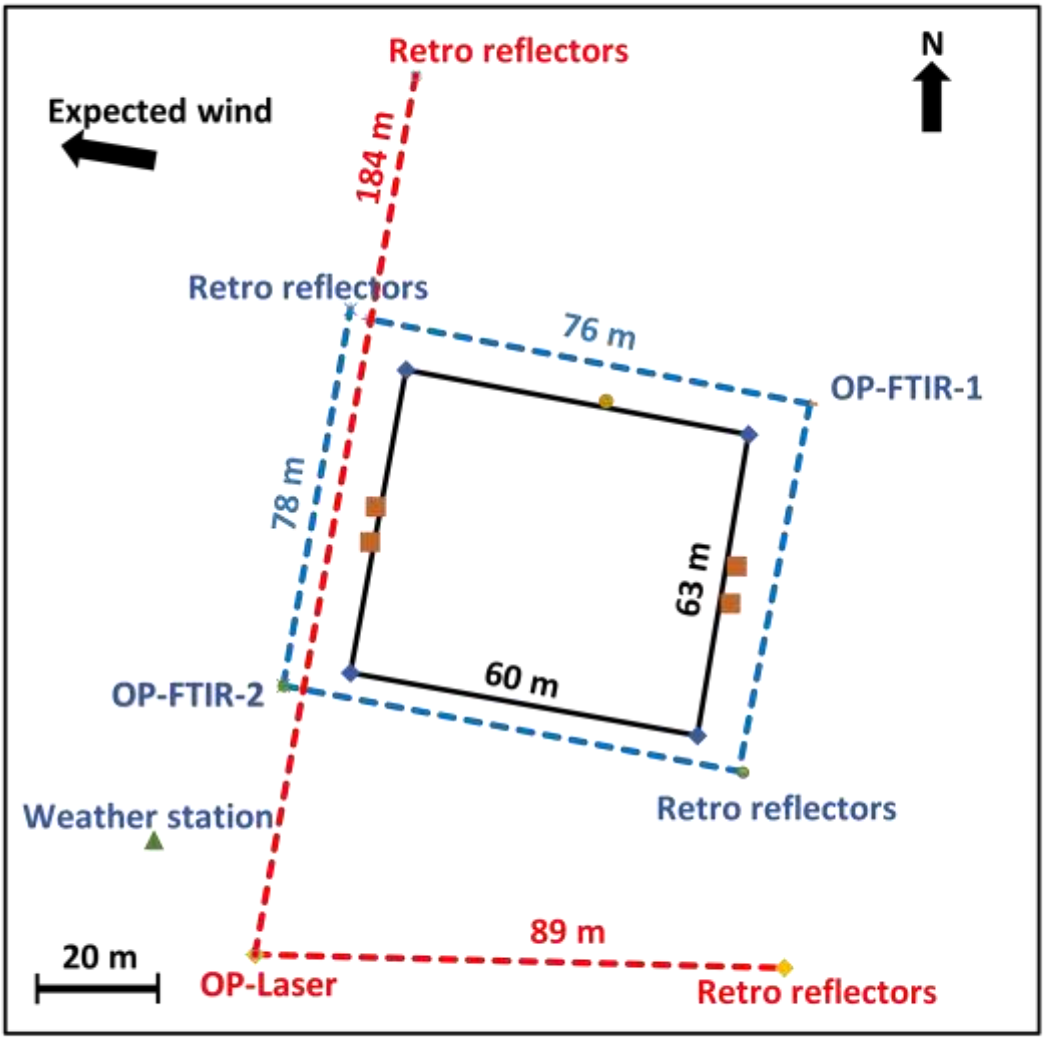

Figure 1: Schematic layout of the experimental site, showing an animal pen in the center, two OP-FTIR systems (blue dashed lines) and the OP-Laser system (red dashed lines). Two feeding troughs (brown squares) were on both sides of the pen, and one water trough (brown circle) was on the north of the pen. A weather station (green triangle) was 50 m away from the SW corner of the animal pen.

During the emission measurement period (14 to 21 February 2013) each study animal carried a N_2O canister in a backpack, and controlled rates of N_2O were released as part of the tracer-ratio measurement technique. At 9:00 daily during the measurement period, the 16 study animals were walked from the cattle pen to the adjacent yards (80 m north), and the N_2O gas canister in the backpack was replaced with a fully filled canister. Cattle were absent from the study pen for approximately 15 to 30 min while this occurred. Other than during the canister replacement period, the animals moved and ate freely in the pen while emissions were measured.

2.2 Concentration sensors

2.2.1. OP-FTIR

Atmospheric concentrations of CH_4 and N_2O were measured upwind and downwind of the cattle pen using two open-path Fourier transform infrared (OP-FTIR) spectrometers. OP-FTIR can quantify a wide range of real-time gas concentrations simultaneously with high resolution (Smith et al., 2011). The details of the OP-FTIR system used in this study can be found in Bai (2010) and Paton-Walsh et al. (2014). Briefly, the modulated infrared (IR) beam from the Bruker IRcube spectrometer (Matrix-M IRcube, Bruker Optics, Ettlingen, Germany) is transferred through the optics to a modified Meade Schmidt-Cassegrain telescope (25.4 cm diameter, Model LX200R, Meade Instrument Corp., Irvine, California, USA) and a secondary mirror, and diverged to 250 mm parallel beam and extended to a distant retro reflector (up to 500 m from the spectrometer) (PLX Industries, Deer Park, New York, USA). The parallel beam is then reflected by the retro reflector and returned to a Mercury Cadmium Telluride (MCT) detector (Infrared Associates Inc., Stuart, Florida, USA) where temperature is controlled by a Stirling cycle mechanical refrigerator cooling system (-196 °C) (Ricor K508, Salem, New Hampshire, USA), as described further in (Bai, 2010). A Zener-diode thermometer (type LM335) and a barometer (PTB110, Vaisala, Helsinki, Finland) provide real-time ambient temperature and pressure data (at the same height of the measurement path) for the analysis of the measured spectra. The spectrometer is operated at 1 cm^{-1} resolution, and one spectrometer scan takes approximately 4 secs (13 scans min^{-1}). For acceptable signal to noise ratios, a minimum measurement period of 1 min is required. The measured spectra are quantitatively analyzed using the MALT analysis program and a nonlinear least squares fitting procedure described in Griffith (1996), based on the reference spectra from the molecule absorption databases (HITRAN) (Rothman et al., 2009). The best fitted spectrum is used to retrieve the line-average gas concentrations of CH_4 and N_2O over the measurement path. The sensitivity of the OP-FTIR units for CH_4 and N_2O is 1 part per billion (ppb), corresponding to 2 and 0.4 ppb for a 100 m path, respectively. To achieve good spectra, parameters including instrument field-of-view (FOV), spectral signal intensity (spec. max), and the residual spectrum between the measured and modelled spectra (RMSresid) are examined. A software “Spectronous” (Ecotech, Knoxfield, Victoria Australia) automatically controls spectrometer, sample collecting, spectrum analysis, data logging and display of the calculated concentrations in real time, together with ambient pressure and temperature.

The OP-FTIR spectrometers were mounted on a motorized aiming system (custom made at the University of Wollongong) to allow the spectrometer to be aimed at different retro reflectors. The two OP-FTIR units were positioned on opposite corners outside the cattle pen, and each unit was alternatively aimed at two reflectors so that gas concentration was measured along the four sides of the pen (Fig. 1). This configuration allowed the downwind CH_4 and N_2O enhancements to be measured for any wind direction. The OP-FTIR measurement sequence was repeated automatically so that every 5-min the line-average gas concentration on each path was measured. The average gas concentrations on each of the four paths were averaged over a series of 15-min intervals, from which we calculated a timeseries of CH_4 emissions. The OP-FTIR measurement-paths fell approximately 7 m outside the fence line. The distance between the OP-FTIR sensor and retro reflector was either 76 or 78 m, and the measurement path was 1.4 m above the ground.

2.2.2. OP-Laser

The open-path laser system used a single laser unit (GasFinder2, Boreal Laser Inc., Edmonton, AB, Canada) located outside the animal pen, mounted on a pan-tilt scanning motor (PTU D300, FLIR Motion Control

Systems, Burlingame, CA, USA). The OP laser contains a transceiver that houses the laser diode, drive electronics, detector module and micro-computer subsystems. Collimated light emitted from the transceiver traverses the open measurement path to a distant retro reflector (up to 500 m) and back. A portion of the beam passes through an internal reference cell. The ratio of measured external and reference signals is used to determine the gas concentration from the open path. The retro reflector mounted on a tripod consists of an array of six gold-coated 6 cm corner cubes with effective diameters of approximately 20 cm. The scanning motor was programmed to sequentially measure CH_4 concentration on two paths. The paths ran along two sides of the pen, and their location was chosen to provide upwind and downwind concentrations during the prevailing easterly winds (Fig. 1). The two-paths were 89 and 184 m in length, and the laser measurement path was approximately 5 m outside the fence line. The laser alternated between the two paths with a dwell time of 1-min on each path. Line-average CH_4 concentration was recorded approximately once a second, and the path average concentrations were averaged into 15-min intervals. The sensitivity of the laser units is 1 part per million-meter (ppm-m), corresponding to 10 ppb for a 100-m path.

2.3 Methodologies

A tracer-ratio technique was used to measure CH_4 emissions from the study animals. This is a conceptually simple and defensible method for measuring emissions, and we will consider this technique as giving the “true” CH_4 emission rate from the animals. Two different implementations of the IDM technique were compared with the tracer ratio measurements.

2.3.1 Tracer-ratio technique (N_2O Tracer)

The tracer-ratio measurements followed the procedure described in Bai (2010), Griffith et al. (2008), and Jones et al. (2011), with N_2O used as the tracer gas and released through a canister at a controlled release rate. The N_2O release point was closed to cattle mouth and nose where the majority CH_4 was emitted. The N_2O tracer gas followed the emitted CH_4 downwind of the animal pen, and both concentrations of N_2O and CH_4 were measured simultaneously by an OP-FTIR (Fig. 1).

The N_2O tracer (> 99%, BOC Instrument grade, Australia) was released from pressurized canisters (Catalina Cylinders) located in insulated backpacks on each animal. Each canister was fitted with a head encompassing capillary tube (0.025 mm inner diameter, SGE Analytical Science Pty Ltd, Australia) to control the N_2O flow rate. The canister was filled with approximately 300 g of N_2O to provide an average flow rate of 10 g h^{-1} over a 24 h period. The temperature of the canisters was recorded every 5 minutes (Thermochron Temperature model TCS, OnSolutions, Australia). The canisters and temperature sensors were exchanged every 24 h at a nearby yard. Following the procedure in Bai (2010), the canister flow rate was calibrated with a gas temperature dependent factor determined from the measured canister temperature. Canisters were also weighed at the start and end of each 24 h period to get the actual daily N_2O release rate.

The calculation for each pressurized canister N_2O flow rate follows three steps:

1) The N_2O flow rate of each canister was calculated following Bai (2010) (Eq.1):

$$Q_{N2O}(t) = Q_0 + \alpha T(t) \quad (1)$$

Where Q_{N2O} (t) is the individual canister flow rate (g h^{-1}) at temperature T (°C), t = time, T = temperature °C at time (t), Q_0 is a constant canister flow rate at temperature 0°C, g h^{-1}, α is the N_2O flow rate temperature dependent factor, g h^{-1} $°C^{-1}$. The temperature was measured at 5-min intervals.

2) The integrated N_2O flow rate over the total release time (RT, ~24h) equals the mass loss of N_2O gas (Δm_{N2O}, g) (Eq.2):

$$Q_0 = (\Delta m_{N2O} / RT) - (\Sigma (\alpha T (t)))/RT \qquad (2)$$

Where $\Delta m_{N2O} = WN_2O_{start} - WN_2O_{end}$

The mass loss of N_2O was determined by the initial and the end weight of the canister (g), WN_2O_{start}, WN_2O_{end}, respectively. The integrated N_2O flow rate of each canister was then interpolated to a 15-min interval flow rate using linear interpolation function (Igor 6.3.7.2). The total N_2O flow rate of the 16 canisters (Q_{N2O}) was used for the CH_4 emission rate calculation.

3) Following the procedure described in Bai (2010), Griffith et al. (2008), and Jones et al. (2011), the herd emission rate of CH_4 was calculated (Eq.3):

$$Q_{CH4} = Q_{N2O} * (\Delta CH_4 / \Delta N_2O) * (M_{CH4} / M_{N2O}) / N_{animal} \qquad (3)$$

Where Q_{CH4} is the CH_4 emission rate, g $head^{-1}$ h^{-1}, Q_{N2O} is the integrated N_2O flow rate of total canisters in the animal backpacks, determined by mass loss of N_2O at canister temperature T and release time t, g h^{-1}, is multiplied by 24 to calculated g $head^{-1}$ d^{-1}. The ΔCH_4 and ΔN_2O parameters are the CH_4 and N_2O concentration enhancements (above the local background level) measured downwind of the animal pen using the OP-FTIR spectrometers, M_{CH4} is the molecular mass of CH_4, 16 g mol^{-1}, M_{N2O} is the molecular mass of N_2O, 44 g mol^{-1}, N_{animal} is animal number, 16.

During the study we collected a number of air samples using volumetric flasks (600 mL). Samples were spaced along each measurement path and taken when animals were absent from the pen. These samples were later analyzed in the laboratory using a closed-path FTIR spectrometer (Griffith, 1996) and the CH_4 and N_2O values were used to cross-calibrate the two OP-FTIR sensors.

Tracer-ratio emission measurements were excluded for periods when the canisters outlets were blocked or had dropped off the animals, when there was optical misalignment of the OP-FTIRs, or when the enhanced CH_4 and N_2O concentration was less than 50 and 10 ppb, respectively.

2.3.2 Inverse Dispersion Modelling technique

Herd CH_4 emissions were calculated using the IDM technique (Flesch et al., 2004). This micrometeorological technique estimates emissions based on the enhancement of CH_4 measured downwind of the animal pen. The link between the concentration enhancement and the pen emission rate is calculated using an atmospheric dispersion model. The freely available software WindTrax (www.thunderbeachscientific.com) is used for that calculation. WindTrax combines a backward Lagrangian stochastic dispersion model with mapping software and takes as input: the upwind and downwind CH_4 concentration measurements, wind information from a sonic anemometer, and a map of the pen and gas sensor locations. General information on WindTrax applications is given in Flesch and Wilson (2005).

The upwind and downwind CH_4 concentration was measured using either the OP-FTIR system previously described (designated IDM-FTIR) or by an open-path CH_4 laser system (designated IDM-Laser).
Air samples collected during the study were used to cross-calibrate the laser and the OP-FTIR sensors (applying a retroactive correction multiplier to the laser concentrations). Air samples were collected at 2-min intervals to get 15-min average concentrations for the period from 9:15 to 9:30 when the cattle were not in the paddock. The samples were analyzed using a gas chromatograph at the University of Melbourne laboratory. Three positions were sampled: 1) directly west of the paddock along the laser/FTIR line, 2) near the laser, southwest of the paddock, and 3) far south of the paddock along the southerly laser line. Winds were from light and from the east. We assumed the CH_4 and N_2O concentrations at these positions would be similar (as cattle were absent) and would provide the basis for calibration of the lasers and FTIRs.

A weather station southwest of the cattle pen (Fig. 1) included a 3-dimensional sonic anemometer (CSAT-3, Campbell Scientific Inc, Logan Utah, USA) mounted 2.45 m above the ground. The anemometer provided the wind information needed for the IDM calculation, including the friction velocity (u_*), Obukhov stability length (L), average windspeed and wind direction, and the standard deviation of the velocity fluctuations in the three directional components ($\sigma_{u,v,w}$). The surface roughness length (z_0) was calculated from these variables (Garratt, 1992). The wind variables were averaged into 15-min intervals matched to the gas concentration dataset.

2.3.3 Data filtering criteria

The CH_4 emissions were calculated in 15-min intervals using the WindTrax software. We defined the CH_4 as coming from an elevated area source 0.8 m above ground, which overlaid the pen area. In the IDM analysis we followed the procedure of Flesch et al. (2005) to remove error-prone intervals when either $u_* < 0.15$ m s^{-1}, $|L| < 5$ m, $z_0 < 0.9$ m, or when the fraction of WindTrax trajectory touchdowns inside the pen source covered < 10% of the pen area. Intervals were also removed when the concentrations measured by the OP-FTIR or the laser corresponded to low signal levels: i.e., FOV < 35, RMSresid < 0.2%, spec.max was < 0.25 in the spectral region of 2200 cm^{-1} for the OP-FTIR, or the light level reported by the laser fell outside the 2000 to 13000 range, or the laser quality parameter $R^2 < 0.97$.

2.3.4 Calculating Average Emissions

The tracer-ratio and IDM measurements are a discontinuous time series of 15-min average emission rates lasting for seven days. In order to create a properly weighted daily average emission rate, these discontinuous data were used to create an ensemble 24-h diel emission “curve” for each technique. Each emission observation was binned into one of the 96 15-min periods making up the ensemble day. We used Generalized Additive Models (GAM) fitted to the time series of gas emission to impute missing measurements (Bai et al., 2020). The time series of gas emission and associated GAM fit for each measurement method are shown in Appendices (Fig. A1). The average daily emission rate was calculated by summing the 15-min emission intervals over the 24 h day.

Following IPCC (2006) recommendation, CH_4 emission using were also calculated based on DMI (Eq. 10.21). This assumes CH_4 energy content = 55.65 MJ (kg CH_4)$^{-1}$, DMI energy content = 18.45 MJ (kg DMI)$^{-1}$, and CH_4 conversion factor Y_m = 6.5%.

3 Results

3.1 Climate condition

During the seven-day emission measurement period the total rainfall was 0.4 mm, the average minimum and maximum ambient temperature was 12.9 and 22.4 °C, respectively. The wind speeds (at 2.45 m above ground) varied from 2 to 8 m s^{-1}, and the wind direction was predominately from the east (Fig. 2). This period had excellent conditions for the micrometeorological measurements due to the lack of precipitation, the absence of light wind periods, and the steady easterly winds.

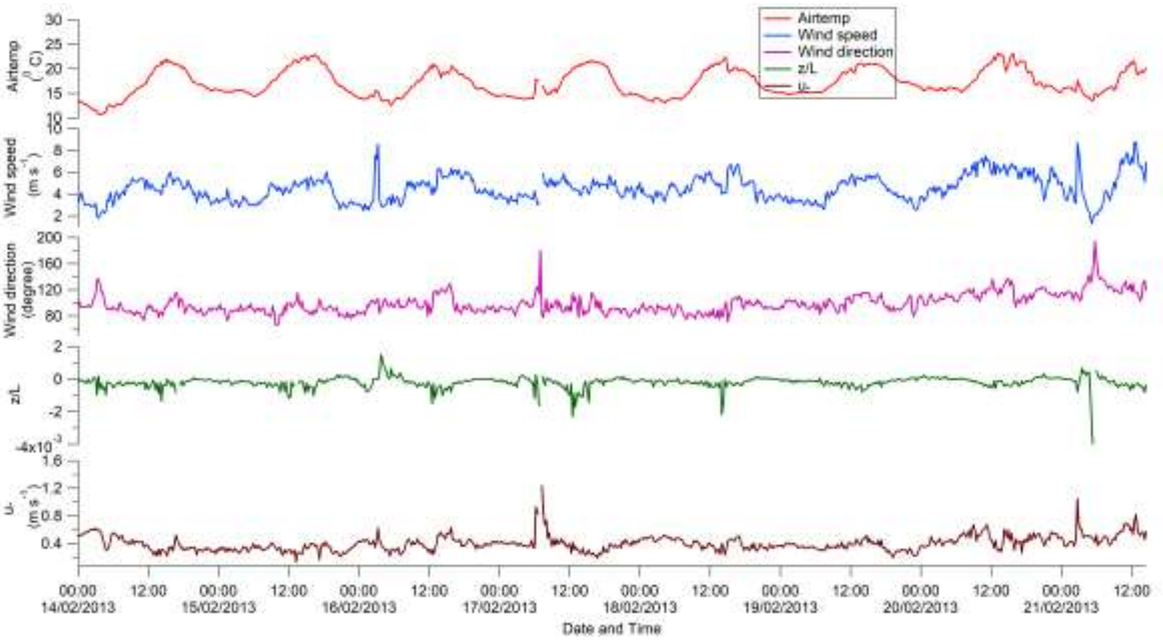

Figure 2 Ambient temperature (Airtemp), wind speed, wind direction was measured during the study. Atmospheric stability parameter (z/L) and wind friction velocity (u_*) are also plotted.

3.2 Methane emission measurements

3.2.1 Tracer-ratio measurements

The OP-FTIR system measured downwind CH_4 enhancements between 50 and 150 ppb, and N_2O enhancements between 12 and 30 ppb over the study (Fig. 3). These enhancements are well above the minimum sensitivity of the OP-FTIR given by Bai (2010) of 2 ppb for CH_4 and < 0.4 ppb for N_2O. Over the seven study days, emissions were measured during 90% of the ensemble 24 h day (i.e., 86 of the 96 possible 15-min periods). The average daily emission rate (± standard error) from the tracer-ratio technique was 301.9 (± 1.5) g CH_4 head^{-1} d^{-1} (Table 1).

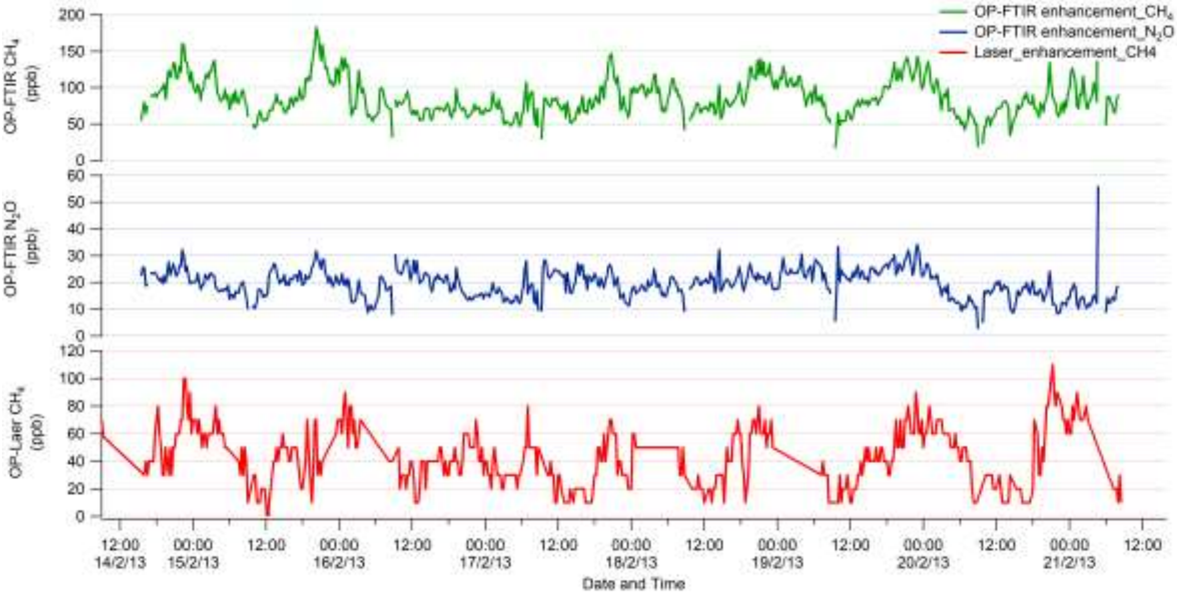

Figure 3. The concentration enhancement of N_2O and CH_4 from OP-FTIR and CH_4 from OP-Laser over the measurement period of 14−21 February 2013.

Table 1. Methane emission rates from the three micrometeorological measurements (Tracer-Ratio, IDM-FTIR, IDM-Laser) and an emission estimate based on the dry matter intake of the animals (using an IPCC recommended calculation§). Methane yield (g CH_4 kg^{-1} DMI) is also shown.

	Emission Rate (g CH_4 $head^{-1}$ d^{-1})	Methane Yield (g CH_4 kg^{-1} DMI)
Tracer-Ratio	301.9 (1.5)	27.0
IDM-FTIR	308.1 (2.1)	27.0
IDM-Laser	304.4 (8.0)	27.1
IPCC§	254§	21.3

§IPCC (2006) calculation based on DMI (Eq. 10.21). Assumes CH_4 energy content = 55.65 MJ (kg CH_4)$^{-1}$, DMI energy content = 18.45 MJ (kg DMI)$^{-1}$, and CH_4 conversion factor Y_m = 6.5%.

3.2.2 The inverse-dispersion modelling (IDM) emissions

Over the seven-day study, 90% of the ensemble was represented with the IDM-FTIR measurements, and 79% was represented by the IDM-Laser measurements. The majority of missing periods resulted from instrumental issues (e.g., low signals caused by condensation on mirrors, power failure), and to a lesser extent by inappropriate meteorological conditions (e.g., low wind speed, $u_* < 0.15$ m s^{-1}). The 24-h diel CH_4 flux over the measurement period is shown in Figure 4. There are differences between the three ensemble emission relationships in Figure 4. We assume the tracer-FTIR data is most accurate data set. Differences between the tracer and IDM approaches are due to a combination of a less-sensitive laser sensor (compared to the OP-FTIR) and the incorrect assumption that animals were spread evenly over the pen (which effects the FTIR and laser estimates differently due to different measurement locations). Both of the IDM-FTIR and tracer-ratio measurements show a similar emission pattern: emission rates at a minimum around 9:00 local time, and at a maximum during the early evening. This emission peak pattern reflected the time when animals were fed, or the pellets were topped up. However, IDM-Laser shows a late minimum emission at 12:00 local time, likely due to a solar related alignment of the retro reflector. We calculated average daily emission rates of 308.1 (± 2.1) and 304.4 (± 8.0) g CH_4 head^{-1} d^{-1} for the IDM-FTIR and IDM-Laser measurements, respectively (Table 1). These results are not statistically different from each other. Both IDM estimates were not statistical different from the tracer-ratio results.

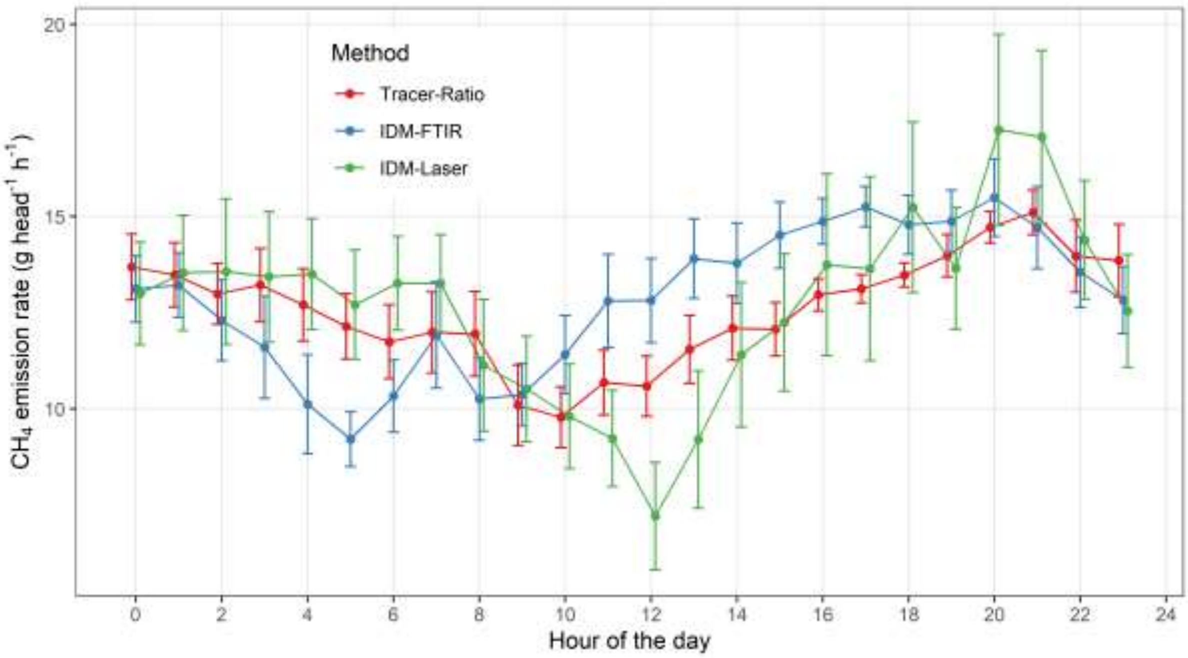

Figure 4: Ensemble 24-h diel CH_4 emission pattern measured by IDM-Laser, IDM-FTIR, and Tracer-Ratio method (hourly values based on 7-d of measurements). Error bars denote the standard error of mean.

4 Discussion

There was excellent agreement between the tracer-ratio and the IDM measurements of cattle CH_4 emissions (there were no statistical differences between the different techniques). For potential users of the IDM technique, these results are an important finding. When previously applied to cattle environments, some recent IDM studies have monitored animal positions assuming this information is critical to getting accurate calculations (e.g., McGinn et al., (2011)). Alternatively, other studies constrained animal locations by fencing to minimize the errors when animal positions were not monitored (Flesch et al., 2016). However, our IDM calculations assuming cattle were evenly distributed across the paddock were nearly identical to the tracer-ratio results that implicitly include the impact of animal positions. This indicates that IDM studies like ours can use the much simpler approach where the whole paddock is treated as a gas source, and animal positions need not be monitored. This seems to confirm a similar finding from McGinn et al. (2015). The effect of this simplification on measurement accuracy is likely to depend on animal density and the size of the paddock. For example, the measurement of a small number of animals in a large paddock is likely to be very sensitive to the exact animal positions. But in the modest sized paddock studied here (and in McGinn et al., (2015)) this is not the case.

It is interesting to compare our measured CH_4 emission rates with estimates made using the IPCC (2006) suggested relationships based on DMI. Using the IPCC recommendations that CH_4 emissions represent 6.5 % of the gross energy intake of the cattle (Y_m) and with our DMI = 11.9 kg d^{-1}, we calculate (Eq. 10.21) an emission rate of 254 g CH_4 $head^{-1}$ d^{-1}. Using the equation from Charmley et al. (2016) and with the yield of 20.7 g CH_4 kg^{-1} DMI, the estimated CH_4 emissions rate is 246 g CH_4 $head^{-1}$ d^{-1}. The DMI based CH_4 estimates were lower than the tracer-ratio measurement of 321 g CH_4 $head^{-1}$ d^{-1}. What might explain this difference?

- Weather conditions during our study were nearly ideal for the micrometeorological calculations, resulting in a large and representative set of emission calculations over the study, and a good estimate of the 24-h ensemble daily emission rate. A time-of-day sampling bias in the tracer-ratio measurements is unlikely to cause the difference.
- Differences between the tracer-ratio and IPCC estimated rates would occur if there were significant manure or rectal emissions that are measured by the micrometeorological techniques, but not reflected in the IPCC estimates. However, the general view is that these emissions are small in comparison to enteric emissions (Flessa et al., 1996; Kebreab et al., 2006; McGinn et al., 2019). In addition, when animals were absent from the pen, we did not observe enhanced CH_4 levels downwind of the pen, indicating low emission rates from the pen manure. There were no manure stockpiles nearby during the study. This suggests that IPCC estimates may have larger uncertainties.
- Based on the tracer-ratio measurements, the CH_4 conversion factor Y_m in this study is higher than the IPCC suggested value, that is: our measured Y_m of 8.3 % is outside the 6.5 ± 1 % range suggested by IPCC (2006). However, the IPCC suggestion is a rough estimate, and several grazing studies have found Y_m values higher than our 8.3 % (e.g., Tompkins and Charmley (2015); McGinn et al. (2011); Ominski et al. (2006)).

5 Conclusions

We are very confident in the tracer-ratio measurements given the conceptual simplicity of the approach (where each animal is a tracer gas source), given that the OP-FTIR is a very sensitive gas sensor, and given the agreement between the associated IDM measurements. We thus view the relatively high emission rates we observed to be representative of the conditions of the study.

The (external) tracer ratio technique is a “gold standard” for measuring cattle emissions in an ambient outdoor environment. However, this technique is difficult to use given the need to outfit the animals with tracer sources, and to monitor tracer gas concentrations downwind. Encouragingly, our results indicate that a logistically simple IDM technique can provide an accurate tool for measuring emissions from cattle, with far greater practicality than the tracer-ratio technique. It is worth noting that micrometeorological methods like IDM represent one of the major approaches for measuring cattle emissions (in addition to internal SF_6 tracer technique and respiration chambers). Our results should give users added confidence that a practical micrometeorological technique can provide an accurate method of estimating emissions at farm scales.

6 Appendices

Appendix A

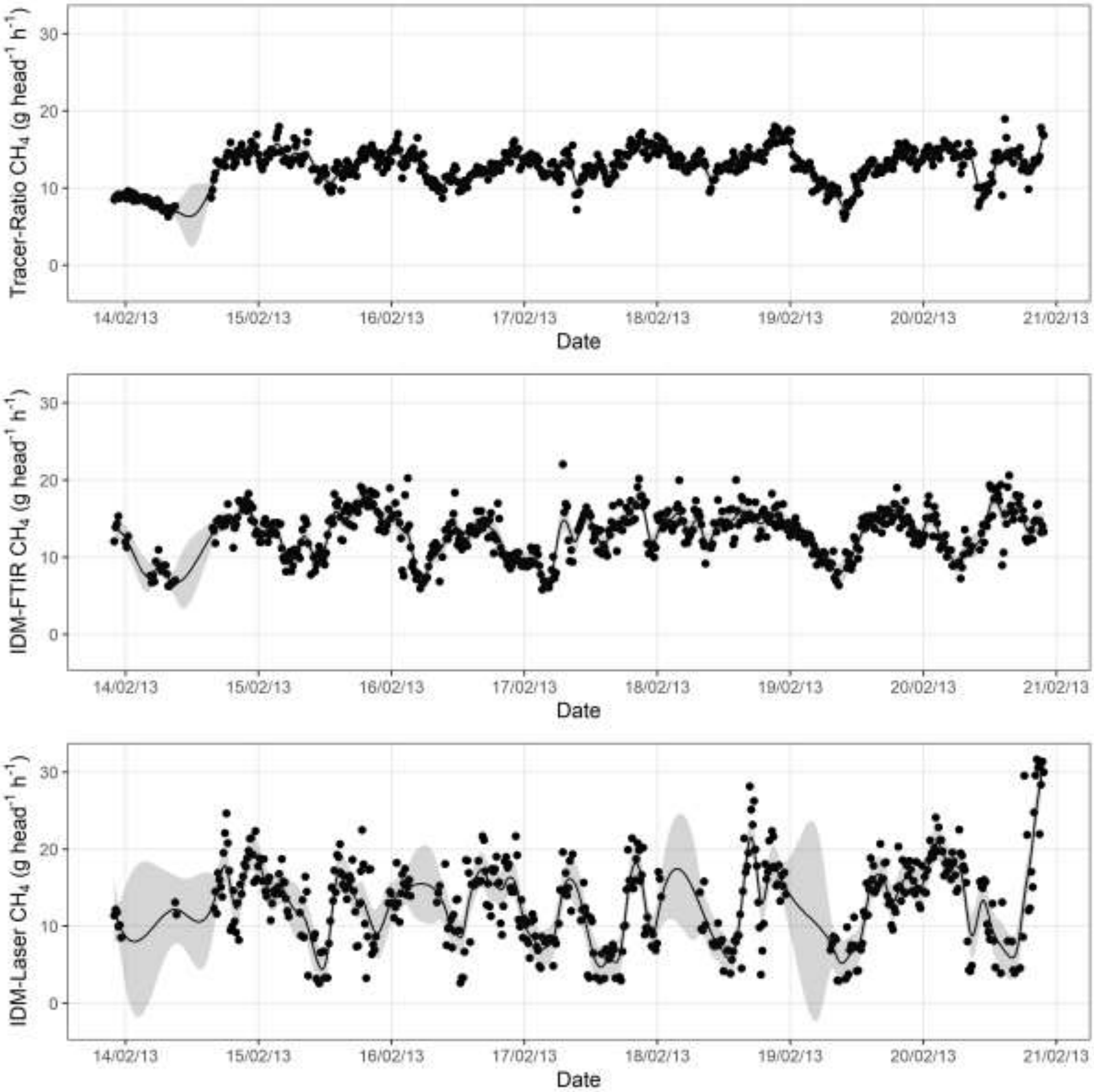

Figure A1: Time series of CH_4 emissions measured using the Tracer-Ratio, IDM-FTIR, and IDM-Laser methods. Black dots show the 15 minutes measurements. The solid black line shows the mean value of gas emission estimated from a GAM fit to the measurement data. The shaded area represents the 95% credible intervals of the mean gas emission from the GAM fit (i.e., it contains 95% of the potential mean values of gas emission at a given time).

7 Data availability

The raw data are not available to the public. For any inquiry about the data, please contact the corresponding author (mei.bai@unimelb.edu.au).

8 Author contributions

All authors contributed to the conceptualization, methodology, draft writing, and original draft preparation. TC, JIV, TF, FP, MB, and NT contributed to writing, reviewing and editing. TF, JIV, TC, FP, and MB contributed to formal analysis. DC, RH, NT, JH, DM contributed to funding acquisition and investigation.

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10 Declaration of interests

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.

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