



On-farm beef cattle methane emissions measured with tracer-1 ratio and inverse-dispersion modelling techniques 2

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14 Abstract. The development and validation of management practices to mitigate greenhouse gas (GHG) 15 emissions from livestock requires accurate emission measurements. This study assessed the accuracy of a 16 practical inverse dispersion (IDM) micrometeorological technique to quantify methane (CH₄) emitted from a 17 small cattle herd (16 animals) confined to a 63 x 60 m pen. The IDM technique calculates emissions from the 18 increase in CH₄ concentration measured downwind of the animals. Two types of open-path (OP) gas sensors 19 were used to measure concentration in the IDM calculation: a Fourier transform infrared spectrometer (IDM-20 FTIR) or a CH₄ Laser (IDM-Laser). The actual cattle emission rate was given by a tracer-ratio technique using 21 nitrous oxide as the tracer gas. We found very good agreement between the two IDM emission estimates (316 22 and 322 g CH₄ head⁻¹ d⁻¹ for the IDM-FTIR and IDM-Laser, respectively) and the tracer-ratio measurements 23 (315 g CH₄ head⁻¹ d⁻¹). This study shows that a practical IDM measurement approach can provide an accurate 24 method of estimating cattle emissions.

25 Keywords: micrometeorological techniques, GHG emissions, grazing cattle, spectroscopy, OP-FTIR

26 **1** Introduction

27 Enteric methane (CH₄) from livestock is a major contributor to agricultural greenhouse gas (GHG) emissions. A 28 significant effort is being made to mitigate these emissions through diet modification, feed supplements, and 29 breeding (Vyas et al., 2018). Increasingly there is a requirement for mitigation claims to be validated when these 30 practices are applied on-farm (DoE, 2014), and simple and accurate methods for on-farm emission 31 measurements are required.

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33 On-farm enteric emissions have been measured by portable respiration hoods for tethered and non-tethered 34 animals (Garnsworthy et al., 2012; Zimmerman and Zimmerman, 2012); by use of internal or external tracer-gas 35 releases from the animal (Johnson et al., 1994); and by micrometeorological techniques based on the enhanced 36 gas concentrations measured downwind of animals (Harper et al., 2011). The objective of this study was to 37 examine the accuracy of a practical inverse dispersion modelling (IDM) technique for measuring CH₄ emissions 38 from a herd of confined animals. The IDM technique offers the possibility of a relatively simple emission





- measurement, without the need for animal handling or modifying animal behavior. In this study the IDMtechnique is used to measure emissions from a small herd of cattle, and these measurements are tested against a
- 41 robust tracer-ratio based measurement.

42 2 Materials and Methods

43 2.1 Experimental design

44 The study took place at the Chiswick pastoral research laboratory (30° 37' S, 151° 33' E) in Armidale, New 45 South Wales, Australia in February 2013. Methane emissions were measured from 16 Angus steers placed in a 46 temporary 63 × 60 m pen (Fig. 1) located in a flat and open field. There were no other cattle nearby, and the 47 nearest trees (30 m height) were at least 300 m from the site. Vegetation in the field was removed prior to the 48 study and no pasture was available to graze.

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50 The study cattle had an average body weight of 373 kg (standard deviation = 59 kg). The animals were fed a 51 blended oaten/lucerne chaff ration (90.2% of dry matter, 15.1 % crude protein) dispensed from automated 52 feeders (Bindon, 2001) that recorded the individual animal intakes. The feeders were cleaned daily and any 53 remaining feed was weighed to check that the total consumed amount matched the sum of the individual animal 54 intakes. Feed and water were offered ad libitum. This feeding regime began four weeks prior to the emission 55 measurements. During the seven-day emission measurement period, the average dry matter intake (DMI) was 56 11.9 kg head-1 d-1. Approximately two weeks before the measurements, each animal was fitted with a backpack 57 (glued to their back) to hold a small nitrous oxide (N2O) gas canister used for the emission measurements.









60 Figure 1: Schematic layout of the experimental site, showing the OP-FTIR and the laser paths. The figure is not to scale.

62

63 During the emission measurement period (14–21 February 2013) each study animal carried an N₂O canister in 64 their backpack, from which gas was released at a controlled rate as part of the tracer-ratio measurement 65 technique. At 9:00 each morning during the measurement period the 16 study animals were walked from the 66 cattle pen to the adjacent yards (80 m north), and the N₂O gas canister in the backpack was replaced with a fully 67 filled canister. Cattle were absent from the study pen for approximately 30 min while this occurred. Other than 68 during the canister replacement period, the animals moved and ate freely in the pen while emissions were 69 measured.

70 2.2 Methodologies

A tracer-ratio technique was used to measure CH₄ 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" CH₄ emission rate from the animals. Two different implementations of the IDM technique were compared with the tracer ratio measurements.

75 2.2.1 Tracer-ratio technique (N₂O Tracer)

76 The tracer-ratio measurements followed the procedure described in Bai (2010), Griffith et al. (2008), and Jones

et al. (2011) with the herd emission rate of CH_4 calculated following Eq. (1):

78 $Q_{CH4} = Q_{N2O}^* (\Delta CH_4 / \Delta N_2 O)$

(1)

79 Where Q_{N2O} is the known N₂O release rate from the canisters in the animal backpacks, and ΔCH_4 and ΔN_2O are 80 the CH₄ and N₂O concentration enhancements (above the local background level) measured downwind of the 81 animal pen.

82

83 The N₂O tracer was released from pressurized canisters (Catalina Cylinders) located in insulated backpacks on 84 each animal. Each canister was fitted with a head encompassing capillary tube (0.025 mm inner diameter, SGE 85 Analytical Science Pty Ltd, Australia) to control the N2O flow rate. The canister was filled with approximately 86 300 g of N₂O to provide an average flow rate of 10 g h^{-1} over a 24 h period. The temperature of the canisters 87 was recorded every 6 minutes (Thermochron Temperature model TCS, OnSolutions, Australia). The canisters 88 and temperature sensors were exchanged every 24 h at a nearby yard. Following the procedure in Bai (2010), 89 the canister flow rate was calibrated with a gas temperature dependent factor determined from the measured 90 canister temperature. Canisters were also weighed at the start and end of each 24 h period to get the actual daily 91 N₂O release rate.

92

93 Atmospheric concentrations of CH₄ and N₂O were measured upwind and downwind of the cattle pen using two 94 open-path Fourier transform infrared (OP-FTIR) spectrometers. The details of the OP-FTIR system can be 95 found in Bai (2010). The OP-FTIR spectrometers were mounted on a motorized aiming system (custom made at 96 the University of Wollongong) to allow the spectrometer to be aimed at different retro reflectors. The two OP-97 FTIR units were positioned on opposite corners outside the cattle pen, and each unit was alternatively aimed at





98	two reflectors so that gas concentration was measured along the four sides of the pen (Fig. 1). This configuration
99	allowed the downwind CH_4 and N_2O enhancements to be measured for any wind direction. The OP-FTIR
100	measurement-paths fell approximately 7 m outside the fence line. The distance between the OP-FTIR sensor and
101	retro reflector was either 76 or 78 m, and the measurement path was 1.4 m above the ground. The OP-FTIR
102	measurement sequence was repeated automatically so that every 5-min the line-average gas concentration on
103	each path was measured. The collected OP-FTIR spectra were analyzed using the MALT (Multiple Atmospheric
104	Layer Transmission) analysis program to derive the line-average concentrations of CH4 and N2O (Griffith,
105	1996). Ambient temperature and pressure were measured and used in the MALT analysis. The average gas
106	concentrations on each of the four paths were averaged over a series of 15-min intervals, from which we
107	calculated a timeseries of CH ₄ emissions.
108	
109	During the study we collected a number of air samples using volumetric flasks (600 mL). Samples were spaced
110	along each measurement path and taken when animals were absent from the pen. These samples were later
111	analyzed in the laboratory using a closed-path FTIR spectrometer and the CH4 and N2O values were used to
112	cross-calibrate the two OP-FTIR sensors, with retroactive correction factors applied to all OP-FTIR data.
113	
114	Tracer-ratio emission measurements were excluded for periods when the canisters outlets were blocked or had
115	dropped off the animals, when there was optical misalignment of the OP-FTIRs, or when the enhancement of
116	CH ₄ and N ₂ O concentration was less than 50 and 10 ppb, respectively.
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- 137 (*L*), average windspeed and wind direction, and the standard deviation of the velocity fluctuations in the three 138 directional components ($\sigma_{u,v,w}$). The surface roughness length (*z*₀) was calculated from these variables. The wind 139 variables were averaged into 15-min intervals matched to the gas concentration dataset.
- 140

141 The CH₄ emissions were calculated in 15-min intervals using the WindTrax software. In the calculations we 142 defined the CH₄ as coming from an elevated area source 0.8 m above ground, which overlaid the pen area. In the 143 IDM analysis we followed the procedure of Flesch et al. (2005) to remove error-prone intervals when either $u_* <$ 144 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 145 covered < 10% of the pen area. Intervals were also removed when the concentrations measured by the OP-FTIR 146 or the laser corresponded to low signal levels: i.e., spec.max was < 0.25 in the spectral region of 2200 cm⁻¹ for 147 the OP-FTIR, or the light level reported by the laser fell outside the 2000-13000 range, or the laser quality 148 parameter $R^2 < 0.97$.

149 2.2.3 Calculating Average Emissions

150 The tracer-ratio and IDM measurements are a discontinuous time series of 15-min average emission rates lasting 151 for seven-days. In order to create a properly weighted daily average emission rate, these discontinuous data were 152 used to create an ensemble 24-h diel emission "curve" for each technique. Each emission observation was 153 binned into one of the 96 15-min periods making up the ensemble day. Any 15-min interval with no 154 observations was assigned an emission rate estimated by interpolation from the surrounding intervals. The 155 average daily emission rate was calculated by summing the 15-min emission intervals over the 24 h day.

156 3 Results

157 3.1 Climate condition

158 During the 7-d emission measurement period the total rainfall was 0.4 mm, the wind speeds (at 2.45 m above 159 ground) varied from 2 to 8 m s⁻¹, and the wind direction was predominately from the east. This period had 160 excellent conditions for the micrometeorological measurements due to the lack of precipitation, the absence of 161 light wind periods, and the steady easterly winds.

162 3.2 Methane emission measurements

163 3.2.1 Tracer-ratio measurements

The OP-FTIR system measured downwind CH₄ enhancements between 50 and 150 ppb, and N₂O enhancements
between 12 and 30 ppb over the study. These enhancements are well above the minimum sensitivity of the OPFTIR given by Bai (2010): 2 ppb for CH₄ and < 0.3 ppb for N₂O). 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 calculated
average daily emissions (± standard error) from the tracer-ratio technique was 315.1 (± 4.3) g CH₄ head⁻¹ d⁻¹
(Table 1).

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 is also shown.





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	Emission Rate	Yield
	$(g CH_4 head^{-1} d^{-1})$	(g CH ₄ kg ⁻¹ DMI)
Tracer-Ratio	315	27.0
IDM-FTIR	316	27.0
IDM-Laser	322	27.1
IPCC [§]	254 [§]	21.3
Charmley et al. $(2016)^*$	246	20.7

[§]IPCC (2006) calculation based on DMI (Eq. 10.21). Assumes CH₄ energy content = 55.65 MJ (kg CH₄)⁻¹, DMI energy content = 18.45 MJ (kg DMI)⁻¹, and CH₄ conversion factor $Y_m = 6.5\%$. ^{*}Methane production (g/day) = 20.7 (±0.28) ×DMI (kg d⁻¹)

175 3.2.2 The inverse-dispersion modelling (IDM) emissions

176 Over the seven-day study, 90% of the ensemble day was represented with the IDM-FTIR measurements, and 177 79% was represented by the IDM-Laser measurements. The majority of missing periods resulted from 178 instrumental issues (e.g., low signals caused by condensation on mirrors, power failure), and to a lesser extent 179 by inappropriate meteorological conditions (e.g., low wind speed). The 24-h diel CH4 flux over the 180 measurement period is shown in Figure 2. There are differences between the three ensemble emission 181 relationships in Fig. 2. We assume the tracer-FTIR data is most accurate data set. Differences between the tracer 182 and IDM approaches is due to a combination of a less-sensitive laser sensor (compared to the OP-FTIR) and the 183 incorrect assumption that animals were spread evenly over the pen (which effects the FTIR and laser estimates 184 differently due to different measurement locations). Both IDM-FTIR and Tracer-Ratio measurement show a 185 similar emission pattern: emission rates at a minimum at 9:00 local time, and at a maximum during the early 186 evening. This emission peak pattern reflected the time when animals were fed, or the pellets were topped up. 187 However, IDM-Laser shows a late minimum emission at 12:00 local time, likely due to a solar related alignment 188 of retro reflector. From the 24-h ensemble diel emission curve we calculate average daily emission rates of 189 316.4 (\pm 12.1) and 321.6 (\pm 26.9) g CH₄ head⁻¹ d⁻¹ for the IDM-FTIR and IDM-Laser measurements, 190 respectively (Table 1). These results are not statistically different from each other (p < 0.05). Both IDM 191 estimates were not statistical different from the tracer-ratio results (p < 0.05).

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193

194Figure 2: Ensemble 24-h diel CH4 emission pattern measured by IDM-Laser, IDM-FTIR, and Tracer-Ratio method195(hourly values based on 7-d of measurements). Error bars denote the standard error of mean.

196 4 Discussion

197 There was excellent agreement between the tracer-ratio and the IDM measurements of cattle CH₄ emissions 198 (there were no statistical differences between the different techniques). For potential users of the IDM 199 technique, these results are an important finding. When previously applied to cattle environments, some recent 200 IDM studies have monitored animal positions assuming this information is critical to getting accurate 201 calculations (e.g., McGinn et al. (2011)). Alternatively, other studies constrained animal locations by fencing to 202 minimize the errors when animal positions were not monitored (Flesch et al., 2016). However, our IDM 203 calculations assuming cattle were evenly distributed across the paddock were nearly identical to the tracer-ratio 204 results that implicitly include the impact of animal positions. This indicates that IDM studies like ours can use 205 the much simpler approach where the whole paddock is treated as a gas source, and animal positions need not be 206 monitored. This seems to confirm a similar finding from McGinn et al. (2015). The effect of this simplification 207 on measurement accuracy is likely to depend on animal density and the size of the paddock. For example, the 208 measurement of a small number of animals in a large paddock is likely to be very sensitive to the exact animal 209 positions. But in the modest sized paddock studied here (and in McGinn et al., (2015)) this is not the case.

210

211It is interesting to compare our measured CH_4 emission rates with estimates made using the IPCC (2006)212suggested relationships based on DMI. Using the IPCC recommendations that CH_4 emissions represent 6.5 % of213the gross energy intake of the cattle (Y_m) and with our DMI = 11.9 kg d⁻¹, we calculate (Eq. 10.21) an emission214rate 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 215kg⁻¹ DMI, the estimated CH_4 emissions rate is 246 g CH_4 head⁻¹ d⁻¹. The DMI based CH_4 estimates were lower216than the tracer-ratio measurement of 321 g CH_4 head⁻¹ d⁻¹. What might explain this difference?





217	٠	Weather conditions during our study were nearly ideal for the micrometeorological calculations,
218		resulting in a large and representative set of emission calculations over the study, and a good estimate
219		of the 24-h ensemble daily emission rate. A time-of-day sampling bias in the tracer-ratio measurements
220		is unlikely to cause the difference.
221	•	Differences between the tracer-ratio and IPCC estimated rates would occur if there were significant
222		manure or rectal emissions that are measured by the micrometeorological techniques, but not reflected
223		in the IPCC estimates. However, the general view is that these emissions are small in comparison to
224		enteric emissions (Flessa et al., 1996; Kebreab et al., 2006; McGinn et al., 2019). In addition, when
225		animals were absent from the pen, we did not observe enhanced CH4 levels downwind of the pen,
226		indicating low emission rates from the pen manure.
227	•	Based on the tracer-ratio measurements, the Y_m in this study is higher than the IPCC suggested value:
228		our measured Y_m of 8.3 % is outside the 6.5 \pm 1 % range suggested by IPCC (2006). But the IPCC
229		suggestion is a rough estimate, and several grazing studies have found Y_m values higher than our 8.3 %

230 (e.g., Tompkins and Charmley (2015); McGinn et al. (2011); Ominski et al. (2006)).

231 **5** Conclusions

232 We are very confident in the tracer-ratio measurements given the conceptual simplicity of the approach (where 233 each animal is a tracer gas source), given that the OP-FTIR is a very sensitive gas sensor, and given the 234 agreement between the associated IDM measurements. We thus view the relatively high emission rates we 235 observed to be representative of the conditions of the study.

236

237 The (external) tracer ratio technique is a "gold standard" for measuring cattle emissions in an ambient outdoor 238 environment. However, this technique is difficult to use given the need to outfit the animals with tracer sources, 239 and to monitor tracer gas concentrations downwind. Encouragingly, our results indicate that a logistically 240 simple IDM technique can provide an accurate tool for measuring emissions from cattle, with far greater 241 practicality than the tracer-ratio technique. It is worth noting that micrometeorological methods like IDM 242 represent one of the major approaches for measuring cattle emissions (in addition to internal SF₆ tracer 243 technique and respiration chambers). Our results should give users added confidence that a practical 244 micrometeorological technique can provide an accurate method of estimating emissions.

245 6 Data availability

246 The raw data are not available to the public. For any inquiry about the data, please contact the corresponding 247 author (mei.bai@unimelb.edu.au).

248 7 Author contributions

249 All authors contributed to the conceptualization, methodology, and draft writing-original draft preparation. TC, 250 TF, FP, MB, and NT contributed to writing-reviewing and editing. TF, TC, FP, and MB contributed to formal 251 analysis. DC, RH, NT, JH, DM contributed to funding acquisition and investigation.

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256 9 Declaration of interests

- 257 The authors declare that they have no known competing financial interests or personal relationships that could
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