



Interactive comment on “On-farm beef cattle methane emissions measured with tracer-ratio and inverse-dispersion modelling techniques” by Mei Bai et al.

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Dear reviewer (Anonymous Referee #1), Re: Revision of manuscript Number: AMT-2020-445, Title: Beef cattle methane emissions measured with tracer-ratio and inverse dispersion modelling techniques

We thank your positive feedback on the manuscript. We have addressed the comments thoroughly, our response to every issue raised is given point by point in blue text below.

**** Abstract **** 1. Should be reconsidered based on the remark of the other sections. I would suggest to add the details of the experiment's management, e.g. duration of the

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trial, pointing that was an experimental pen. Adding the type of the FTIR and the Laser used can be useful to understand the method used. I would suggest also to add the uncertainty of the cumulative flux.

The information for the trial period and experimental pen as suggested has been added in the revised manuscript. The details of the type of instrument (OP-FTIR and OP-Laser), tracer-ratio and inverse dispersion modelling technique have been described in the Materials and Methods section, therefore we did not add this information in Abstract. Since we already documented the uncertainty in the average daily emission rate, no additional information in the cumulative uncertainty is needed.

****Introduction**** 2. The introduction is somewhat lacking and should be extended. Moreover, some elements missing to understand the novelty of this work compared to literature. First, I suggest adding some data from the inventories related to the contribution of the livestock to GHG emissions in Australia and the World. This Reviewer suggest a series of questions to be answered with the purpose to improve the introduction section: What is CH₄ and how it affects the climate? Where does CH₄ emissions come from? How agriculture (and livestock) contributes to CH₄? Which are the most prominent mitigation options?

We have added the information in the revised manuscript. “Agriculture is the main source of anthropogenic methane (CH₄) 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 (CO₂) in a 100 year time (Myhre et al., 2013). Enteric CH₄ 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).” Page 1, lines 28-35.

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3. Secondly, the section describing the different available methods and techniques to measure enteric CH₄ is, in my opinion, poor and should be improved. I suggest to add examples from literature, which can be used also in the discussion section (this Reviewer add here a non-exhaustive list: Felber et al., 2015; Dengel et al., 2011; Lockyer and Jarvis, 1995; Grainger et al., 2007; Laubach et al., 2008; Todd et al., 2014). The strengths and weaknesses of each measurement method or technique should be stressed in order to defend the type of methodologies used by this study. I suggest also adding some experience about the tracer-ratio technique, since it is defined as “true” in this paper (L 69).

It is beneficial to give some broad context to different methods, mainly to support our (critical) assertion that the tracer-ratio technique is the most accurate method in principle. The information has been added in the revised manuscript. Page 2, lines 38-51.

“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 SF₆ (Grainger et al., 2007), assumes the tracer gas and the emitted CH₄ have similar transport paths, so that a tracer measurement can establish the CH₄ 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

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is that they do not interfere with the animals or the environment.”

Following the suggestions of adding some experience about the tracer-ratio technique, the information can be found in the Materials and Methods section. Page 5, lines 169-192.

4. Third, the novelty of this study. If the IDM technique has been already applied to perform the quantification of CH₄ from grazing animals, I would encourage adding these information and stressing how the work you are presenting has some novelty (technical, methodological, or environmental conditions) compared to the literature and previous studies (i.e. Bai 2010). Finally, why do you compared two different concentration measurement tools?

The reviewer asks a good question – why include the laser system in the method comparison? It is true that the laser results are not needed to compare the IDM and Tracer Ratio techniques. But one reason for including the laser results is to justify the relatively high emission rates found with the FTIR measurements (in relation to the IPCC estimates and expectations). The fact that an independent system (with emissions calculated independently from the FTIR system) gave similar results supports the unexpectedly high emissions rates found by the FTIR system.

****Materials and Methods**** 5. This section should be reorganised. I suggest providing a detailed section of the laser and FTIR, along with their working principle and field setup, just below the Experimental Design section, then describe the two methods (tracer-ratio and IDM). The information about the FTIR and Laser is scattered and not well organised, disadvantaging readability and understanding. See also the comments #10 to 12; and #22. I suggest including here the details of the calculation of the losses made with IPCC's guidelines (Table 1); comment #21 Agree. As suggested, we reorganized the Materials and Methods section in the revised manuscript. page 8, lines 249-251. We also added the information of methane emission calculated using IPCC recommendation based on the dry matter intake (DMI).

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“Following IPCC (2006) recommendation, CH₄ emission using were also calculated based on DMI (Eq. 10.21). This 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%.”

****Experimental design**** 6. I suggest adding (here or in the Results section, see comment #19) more details about the experimental site, as the meteorological variables measurements from the weather station (e.g. rain, temperatures, wind direction and speed). This will help the reader to understand the validity of the measurements (e.g. wind direction), this particular environment and, of course, the results. Agree with the reviewer. Figure 2 with the ambient temperature, wind speed, wind direction, z/L and u* is added to the revised manuscript. Insert Figure 2

7. Can the Authors detail here more about the dejections management during the experiment in order to better understand the field set-up and neglecting further sources of methane? We have added a sentence in the revised manuscript “There were no other cattle or animal manure storages nearby during the study ...” page 2, lines 61-62.

8. L 54-55 I would suggest to add a reference to the part of the tracer description (2.2.1) Agree with reviewer’s suggestion. A reference has been added to the revised manuscript.

9. Figure 1. I warmly suggest re-making the picture with a proper scale. This will help better understating the field setting and the distances of the probes (and the weather station) from the fences. Agree. we have modified the figure (Fig. 1). Insert Figure 1.

*** Methodologies *** **** Tracer-ratio technique (N₂O Tracer) *** 10. I would suggest to explain with more detail and in a few line how is the principle of this method. L75, please explain what is QCH₄ in the text. The information has been added in the Introduction, as well as the methodologies section. Pages 5-6, line 169-192.

The calculation for each pressurized canister N₂O flow rate follows three steps: 1)

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The N₂O flow rate of each canister was calculated following Bai (2010) (Eq.1): $Q_{N2O}(t) = Q_0 + \dot{E}S T(t)$ (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), Q₀ is a constant canister flow rate at temperature 0°C, g h⁻¹, $\dot{E}S$ is the N₂O flow rate temperature dependent factor, g h⁻¹ °C⁻¹. The temperature was measured at 5-min intervals.

2) The integrated N₂O flow rate over the total release time (RT, ~24h) equals the mass loss of N₂O gas (Δm_{N2O} , g) (Eq.2): $Q_0 = (\Delta m_{N2O} / RT) - (\sum (\dot{E}S T(t))) / RT$ (2) Where $\Delta m_{N2O} = W_{N2Ostart} - W_{N2Oend}$ The mass loss of N₂O was determined by the initial and the end weight of the canister (g), $W_{N2Ostart}$, W_{N2Oend} , respectively. The integrated N₂O 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 N₂O flow rate of the 16 canisters (Q_{N2O}) was used for the CH₄ 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 CH₄ was calculated (Eq.3): $Q_{CH4} = Q_{N2O} * (\Delta CH_4 / \Delta N_2O) * (M_{CH4} / M_{N2O}) / N_{animal}$ (3) Where Q_{CH4} is the CH₄ emission rate, g head⁻¹ h⁻¹, Q_{N2O} is the integrated N₂O flow rate of total canisters in the animal backpacks, determined by mass loss of N₂O 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 CH₄ and N₂O 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 CH₄, 16 g mol⁻¹, M_{N2O} is the molecular mass of N₂O, 44 g mol⁻¹, N_{animal} is animal number, 16.

11. Can the Authors add the details of the producer of the FTDIR, the measurement range (to justify also lines L112-113 and L142-145), the uncertainty and sensitivity (to justify lines L162, L178), and all the technical parameters that can help to characterise this measurement. Could the Authors detail where the measurements were recorded ? Yes. The details of the information have been added in the revised manuscript. Pages 3-4, lines 91-128.

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2.2 Concentration sensors 2.2.1. OP-FTIR Atmospheric concentrations of CH₄ and N₂O 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 IRTube spectrometer (Matrix-M IRTube, 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 CH₄ and N₂O over the measurement path. The sensitivity of the OP-FTIR units for CH₄ and N₂O 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

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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."

12. Can the Authors add some information about the close-path FTIR used in laboratory in this section, or in the section of the concentration measurements (see comment #5)? The close-path FTIR analysis was operated by a laboratory staff, the information required by the reviewer is out of this study.

13. This section should represent one of the main methodological parts of the paper and, I retain, it can be improved. I suggest adding the principle behind the backward application of the short-range Lagrangian dispersion model used in the study (equation, number of trajectories used and principles of the MOST). This will improve the understanding of the scientific ground, the application of this technique in the case study presented here and better understand the Equation 1. Any reference to other study using IDM in the short range is recommended. Agree. we have added the information in the revised manuscript. Pages 6-7, lines 203-210.

"Herd CH₄ emissions were calculated using the IDM technique (Flesch et al., 2004). This micrometeorological technique estimates emissions based on the enhancement of CH₄ 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 CH₄ 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)."

14. Please, detail how the roughness length was calculated (reference or equation, and

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the results). Furthermore, can the Authors detail if they're using a constant or a variable z_0 . A reference (Garratt, 1992) is added. For z_0 , state that it is a variable inferred from the sonic anemometer measurements, as described in Flesch et al. (2004).

15. L139-142. Can you address why these thresholds were imposed for this case study and why these are different from Flesch et al 2005? Please, refer here to the methodological part requested in the comment #13. Furthermore, how many "15-mins" data were excluded from the dataset with these thresholds and in which part of the day? The meteorological (bLS) thresholds used in Flesch et al. (2005) were not presented as universal. Other studies have used different values.

"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 data were discarded were due the filtering criteria "In the IDM analysis we followed the procedure of Flesch et al. (2005) to remove error-prone intervals when either $u^* < 0.15 \text{ m s}^{-1}$, $|L| < 5 \text{ m}$, $z_0 < 0.9 \text{ m}$, 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., $FOV < 35$, $RMS_{resid} < 0.2\%$, $spec.max$ 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 $2000-13000$ range, or the laser quality parameter $R2 < 0.97$."

16. L 143. Please, explain what "spec.max" stands for. Spec.max stands for spectral signal intensity, which has been described in the revised manuscript. Page 4, line 114.

17. I would suggest to rewrite this part more clearly, giving some reference to other studies which use the same calculation. This will greatly help the reader. A gap-filling procedure has been used? Please add these details. We have several literature examples where emission rate observations have been grouped by time-of-day to come up with an ensemble 24-hour emission curve. For example, Bai et al., (2015), Loh et al., (2008) and Laubach et al. (2013).

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We also added the information for gap-filling procedure on page 7, lines 243-246. "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)."

18. Were the periods when the animals were not in the pen excluded from the measurement dataset? This point should be better described. We calibrated the measurement with air sample measurement during this period when animals were absent. See page 4, lines 194-197.

"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."

**** Results ** *Climate condition***

19. This part should be improved and extended. I would suggest adding a figure with the dynamics of air temperature, wind speed and rain, at least. Moreover, I warmly recommend to add a figure with the trends of u^* and the turbulence parameter z/L . see the response to # 6.

20. To better understand the measurement performed, given that two different methods are compared in this study (Laser and FTIR), it might be interesting to evaluate the concentrations observed over time by the two systems and by the tracer, before evaluating the final daily cumulative emissions. I suggest to provide these results. Agree. As suggested by the reviewer, we have plotted the enhanced concentrations of CH_4 and N_2O from OP-FTIR and the enhanced concentration of CH_4 from OP-Laser system. Insert Figure 3.

21. Table 1. I suggest putting the measurement uncertainty for each of the measurements. I would also suggest removing the reference (Charmley) from the table and

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keep it exclusively in discussions section along with the other sources cited to defend your findings. Furthermore, I would better explain the calculation with the IPCC's guidelines in materials and methods (see comment #5). Yes, agree. We removed the reference from the table and added the calculation information in the Materials and Methods section.

22. L161-163. What about the sensitivity of the laser source? We added the information in the revised manuscript. Page 5, lines 144-145.

"The sensitivity of the laser units is 1 part per million-metre (ppm-m), corresponding to 10 ppb for a 100-m path."

* The inverse-dispersion modelling (IDM) emissions * 23. L175. I would suggest to detail better what "low wind speed" means for the Authors. Or, if these percentages are comprehensive of the periods not considered because of the MOST conditions failure (L139-142)? Low wind speed refers to the wind velocity (u^*) is less than 0.15 m s⁻¹ when the MOST conditions failed. This has been described in the text "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."

24. I cannot see any comparison about the "sensitivity" of the two sensors. I suggest to address this part on the Materials and Methods section (see comment #5) and in the results (comment #19). See the response to #22

25. The lowest emission value is at 9 am, the time when the animals left the pen. How did this event affect the dataset? Are these gaps filled and how? Number of 15-min observations is used to create the average (i.e. 4 obs per hour X 7 days = ~28 is the maximum). The cattle were scheduled to be out at around 9 am, and it took about 15 to 30 min to change the canisters. There was maximum 4 observation an hour and minimum 1 observation.

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26. I would warmly suggest to insert a further figure about the trend of 15-mins emissions over the 7 days of measurement. This will give the real picture of the dataset, without the period of failures (technical), filtered because of the MOST failure. 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 Appendix A (Fig. A1). Insert Figure A1.

Furthermore, we have assumed that the animals were well adjusted to the site and feeding regime (ad libitum), and we expect that relatively consistent emission rates. Given the discontinuous nature of the emission measurements, it is better to look at averaged emission rates.

27. Figure 2. IDM-FTIR does not have the measurement at 11pm We thank the reviewer for pointing out the mistake. The figure has been modified (Figure 4). Insert Figure 4.

28. The discussions should be better set up and expanded with other literature studies to defend the validity of the measurement, i.e. defending that the conditions of the experiment were always suitable for the application of the IDM. It seems that the reliability of the IDM method is related only to the final cumulative emissions (Table 1). In order to define that the source was homogeneous, and therefore the monitoring of the animals is not needed, as stated, further results from this study - or results from other studies - should be provided. In this study the evaluation standard for IDM is agreement with the Tracer-Ratio. We assume, with good reason, that the Tracer-Ratio approach is the most accurate means of measuring emissions in ambient conditions. In this case, agreement with other studies (using other methods, in other animal situations) is not useful.

29. Referring only to the method of the IPCC guidelines is, in my opinion, limited. I would suggest broadening the discussions with other case studies, reporting their

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characteristics and results to make the measurement more robust (e.g. references cited online 226). The introduction of an IPCC emission value is a recognition that our measured emission rates are high. This gives some interesting context to our measurements (and I think it is something the audience would question), but it is not crucial to our primary objective of comparing IDM and Tracer-Ratio.

30. The conclusions, with respect to the use of IDMs, should be much more cautious given that this is an experiment of only 7 days, performed in micrometeorological conditions not detailed in the paper, without a real defence of the validity of the application of the method itself (homogeneity of the source). While seven days may not be sufficient to document long-term cattle emissions, seven days of near-continuous measurements is not an insignificant period when comparing micrometeorological techniques. One should not discount the very close agreement over a range of meteorological and animal position conditions.

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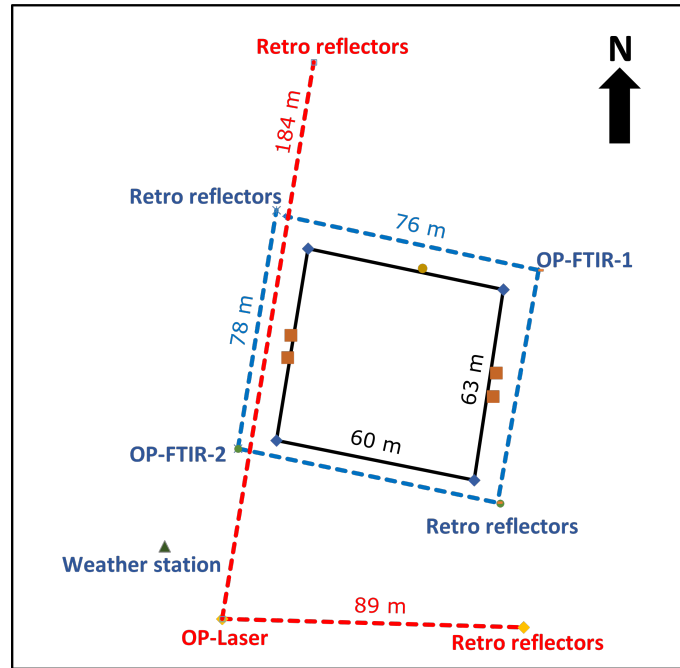


Fig. 1. Figure 1: Schematic layout of the experimental site, showing an animal pen in the center, two OP-FTIR systems (blue dashed lines) and the OP-Laser system (red dashed lines). Two feeding troughs (brown

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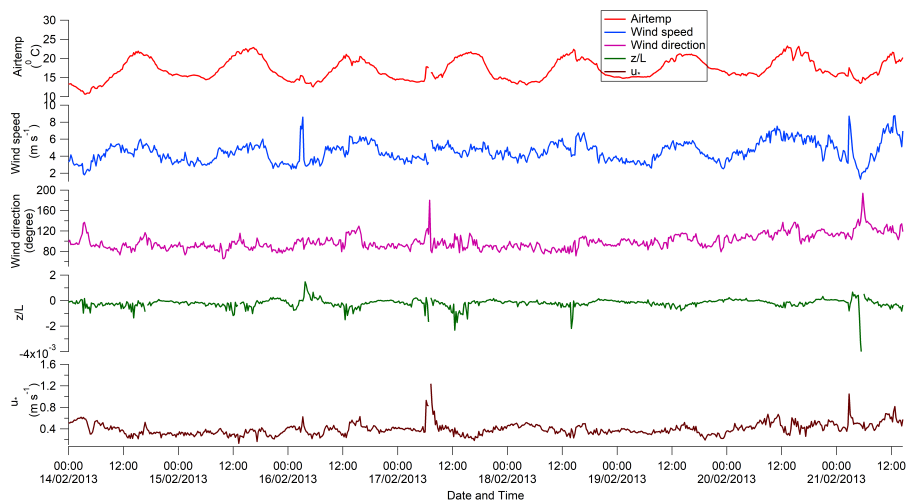


Fig. 2. Figure 2 Ambient temperature (Airtemp), wind speed, wind direction was measured during the study. Atmospheric stability parameter (z/L) and wind friction velocity (u^*) are also plotted.

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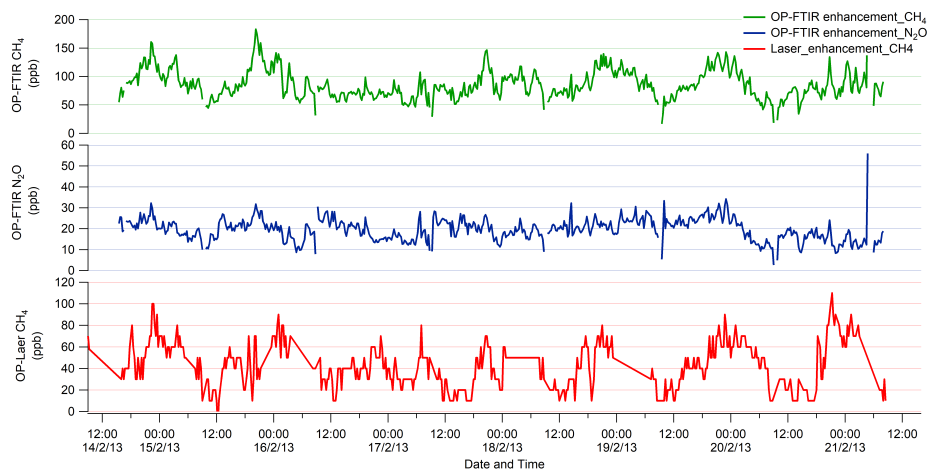


Fig. 3. Figure 3. The concentration enhancement of N₂O and CH₄ from OP-FTIR and CH₄ from OP-Laser over the measurement period of 14–21 February 2013.

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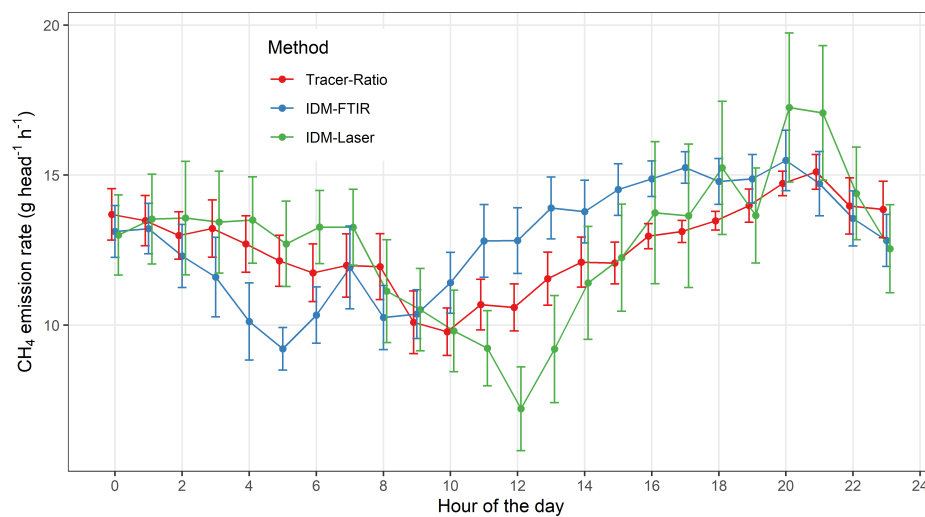


Fig. 4. Figure 4: Ensemble 24-h diel CH₄ emission pattern measured by IDM-Laser, IDM-FTIR, and Tracer-Ratio method (hourly values based on 7-d of measurements). Error bars denote the standard error of mean.

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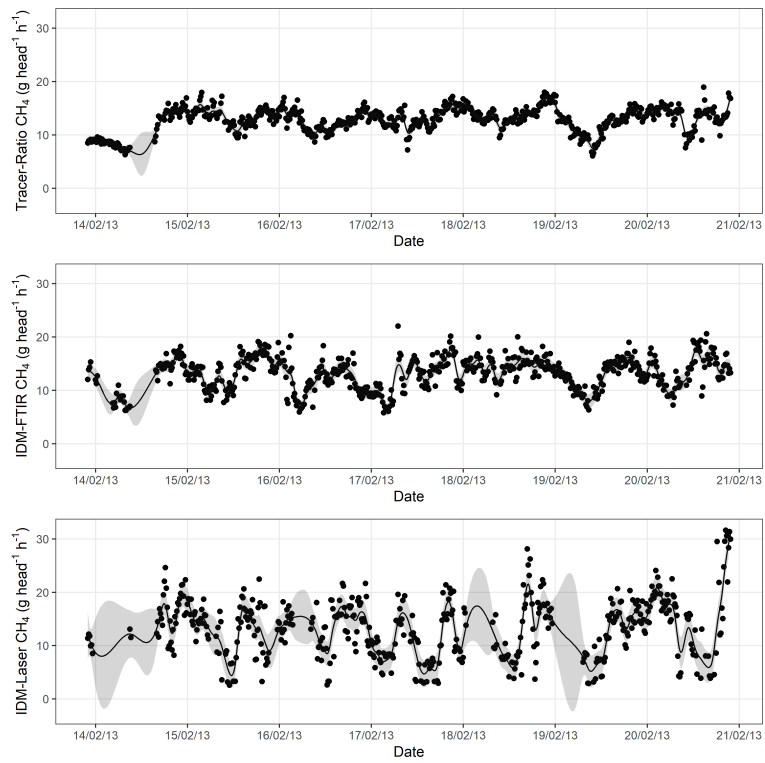


Fig. 5. Figure A1: Time series of CH₄ emissions measured using the Tracer-Ratio, IDM-FTIR, and IDM-Laser methods. Black dots show the 15 minutes measurements. The solid black line shows the mean value of gas