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Methane emission measurements in a cattle grazed pasture: a comparison of four methods

T. Tallec[1,*], K. Klumpp[1], A. Hensen[2], Y. Rochette[3], and J.-F. Soussana[1]

[1]INRA, UR874 Grassland Ecosystem Research, 234 Avenue du Brézet, 63100
Clermont-Ferrand, France
[2]Energy Research Centre of the Netherlands (ECN) P.O. Box 1 1755 ZG Petten,
The Netherlands
[3]INRA, UR1213 Herbivores, 63122 St Genes Champanelle, France
[*]now at: Centre d'Etudes Spatiales de la BIOsphère (CESBIO), 18 avenue Edouard Belin bpi
2801, 31401 Toulouse cedex 9, France

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Abstract

Methane (CH_4) is considered to be the second main contributor to the global greenhouse gas effect, with major CH_4 emissions originating from livestock. Accurate measurements from ruminating herds are required to improve emission coefficients used in national emission inventories, and to evaluate mitigation strategies. Previous measurements of enteric methane emissions from domestic animals have been carried out in artificial conditions such as laboratory chambers, or by fitting individual animals with capillary tubes and using SF_6 as a tracer. Here we evaluated the reliability of eddy covariance technique (EC), already used for CO_2 fluxes, for continuous CH_4 measurements over a grazed field plot. Analyzer accuracy and reliability of eddy covariance technique were tested against field scale measurements with the SF_6 tracer technique, Gaussian plume model and emission factors (i.e. IPCC). Results indicate a better agreement between EC and SF_6 method when grazing heifers were parked close to the EC setup. However, a systematic underestimation of EC data appeared and even more when the distance between the source (ruminating heifers) and EC setup (mast) was increased. A two-dimensional footprint density function allowed to correct for the dilution effect on measured CH_4 and led to a good agreement with results based on the SF_6 technique (on average 231 and 252 g CH_4 ha^{-1} over the grazing experiment, respectively). Estimations of the CH_4 budgets for the whole grazing season were in line with estimates (i.e. emission factor coefficients) based on feed intake and animal live weight as well as SF_6 technique. IPCC method Tier 2, however, led to an overestimation of CH_4 fluxes on our site.

1 Introduction

With a global warming potential of 25, methane (CH_4) is considered to be the second most important greenhouse gas after carbon dioxide (CO_2). Since the pre-industrial era CH_4 concentration has increased worldwide by 150 % (IPCC, 2007). The livestock

production sector (i.e enteric fermentation and manure management) represented 37 % of global anthropogenic CH_4 emissions (FAO, 2006). Enteric fermentation by ruminants is estimated to reach 85 million tonnes CH_4 per year and 82 % of total livestock emissions (FAO, 2006). Grazed systems contribute with one third compared to two third from mixed farming systems (i.e. paddock and barn) to these total methane emissions, indicating the significant contribution during grazing (FAO, 2006). Although several national and international reports (e.g. EPA, 2006; FAO, 2006) provide numbers on amount of CH_4 emitted from the livestock sector, there are only a few reports of CH_4 emission measurements from grazing ruminants. Consequently, IPCC (2006) Tier 1 emission factors for enteric CH_4 are often used as default values to estimate emissions (IPCC, 2006). However, CH_4 production is both very variable in space and time, and between animals (Vermorel et al., 2008; Hegarty et al., 2007; Sauvant and Giger-Reverdin, 2009; Martin et al., 2010; Archimède et al., 2011; Eugene et al., 2011). The simple use of IPCC emission factors for grazing livestock may not only lead to an under-/overestimation of CH_4 emissions, but also reduce the scope for developing mitigation strategies at the field scale. Other mitigation options such as soil carbon sequestration are developed at field scale (i.e. $g\,C\,m^{-2}\,yr^{-1}$) and trade-offs with non-CO_2 GHG emissions need to be assessed on the same scale (Soussana et al., 2007, 2010).

There are number of techniques to quantify methane emissions from individual or groups of animals. In the past, most of available data on cattle CH_4 emissions derived from calorimetric studies were collected using closed respiration chambers. These enclosure techniques are precise but involve artificial conditions with restricted animal movement, which may not accurately predict the CH_4 production in real environments such as in pasture. An alternative to the chamber method is the sulphur hexafluoride tracer method (SF_6) (Johnson et al., 1994; Pinares-Patino et al., 2007; Giger et al., 2000; Vermorel, 1995). This tracer method allows CH_4 emissions of individual grazing animals to be determined over a time period of one or two days. However, variation between animals is strong and repeatability of this "animal effect" has been questioned (Münger and Kreuzer, 2008; Vlaming et al., 2008). In addition, there are significant

uncertainties in CH_4 measurements due to spatial and temporal variation in feed intake quality and quantity (Martin et al., 2010), as well as potential CH_4 emissions from dung which can not be captured by SF_6 technique.

The development of a new generation of fast analysers (e.g. tuneable laser diode, cavity ring down spectroscopy analyser) has made it possible to apply the eddy covariance (EC) technique – already used for CO_2 exchanges between ecosystems and the atmosphere – to CH_4 fluxes (see Kroon et al., 2007; Hendriks et al., 2008; Smeets et al., 2009; Dengel et al., 2011). The eddy covariance technique offers precise nonintrusive concentration measurements at a high sampling rate (10 to 20 Hz) over a larger measure area (e.g. several hectares) and over long time periods. Recent studies have reported the accurate use of EC technique in wet grassland (Hendriks et al., 2008; Kroon et al., 2010), rice fields (Detto et al., 2011) and pine plantations (Smeets et al., 2009). So far, only a few studies have applied the EC method to ruminating animals (i.e. restored wetland; Detto et al., 2011; Herbst et al., 2011) on permanent grasslands (Dengel et al., 2011). However, no particular attention was paid on the reliability and magnitude of EC measurements with respect to presence/absence of animals in the footprint area and their distance to the EC setup. These discrepancies may lead to misinterpretation of EC measurements given the large variability in CH_4 emissions resulting from animal behavior: animals do not behave at random and grazing and ruminating is separated in time and space. Moreover, the paddock is in most cases larger than the measured footprint, which might make it necessary to either gapfill emissions for periods where animals are outside the footprint or to track animals (e.g. using webcams or laser systems; see Detto et al., 2011; Herbst et al., 2011). In other cases, the area of interest may be smaller than the measured footprint, which makes it necessary to filter for data outside the boundaries of the paddock, as the adjacent paddock may have different stocking rate and animal species from the measured paddock.

Here we liked to investigate the applicability of the EC method for CH_4 fluxes in grazed grasslands. More specifically, the performance of EC technique was analysed by (i) testing effects of distance, footprint localisation and night atmospheric stability in

a method-comparison experiment and (ii) by assessing the temporal scale and CH_4 budget over an annual grazing period. In the present study we compared EC method with dual tracer method (SF_6) (Pinares-Patino et al., 2007), a dispersion model based on the Gaussian plume method (Hensen and Scharff, 2001) and with emission factors such as IPCC (2007) guidelines.

2 Material and methodology

2.1 Experimental area, design and climatic conditions

The study was carried out at the French semi-natural upland grassland site Laqueuille (45°38′ N, 2°44′ E; 1040 m a.s.l.). The mean annual precipitation reaches 1100 mm with a mean annual temperature of 8 °C. The experimental field (2.81 ha), is continuously grazed by heifers from May to October and receives 213 kg N ha^{-1} yr^{-1} in 3 splits. The stocking rate is intensive, compared to regional agricultural practices, and comprises 1.16 Livestock Units ha^{-1} yr^{-1} (1.93 animal ha^{-1} yr^{-1}) (for further details see Klumpp et al., 2011). Methane emissions were measured continuously during grazing period in 2010 and 2011 (25 May to 18 October 2010 and 27 April to 13 October 2011).

2.2 Instrumentation and data processing

The grassland site is part of the global FLUXNET observation network and integrates European projects (i.e. CarboEurope, GHG-Europe, ICOS). The site is equipped with a meteorological station, providing 30 min averaged values of global radiation, air temperature, soil temperature (at 5, 10, 30 cm depths), soil water content (at 10 and 30 cm depths) and precipitation, and with an eddy covariance flux measurement system (EC) situated in a fenced area in the middle of the paddock. The EC system comprises a fast response (20 Hz) sonic anemometer (Gill Instruments, Lymington, UK, Model Solent R3) and an open path CO_2-H_2O analyzer (LI-Cor Inc., Lincoln Nebraska, USA, Model LI-7500) installed at a height of 2 m. CO_2-flux (i.e. net ecosystem exchange,

NEE) calculation are done following Carboeurope-IP guidelines (Aubinet et al., 2000) (for further details see Allard et al., 2007 and Klumpp et al., 2011).

Methane fluxes were measured by an off-axis integrated cavity output spectroscopy methane analyzer (CRDS, DLT-100 Los Gatos Research Inc. is located in Mountain View, California, USA) installed in a closed-path set-up with a dry vacuum scroll pump (XDS35i, BOC Edwards, Crawly, UK) providing a maximum air flux of $583 \, l \, min^{-1}$ (i.e. $375 \, l \, min^{-1}$ at a required pressure of 170 hPa) to obtain 10 Hz measurements (for set up details see also Hendriks et al., 2008). Air was sucked to the analyser by a 4.8 m long PTFE tube (internal diameter of 6.5×10^{-3} m), with an inlet installed at a 20 cm distance from the sonic anemometer.

EC measurements are logged with EdiSol software (Moncrieff et al., 1997), which performs rotational corrections of wind direction and calculates a 30 min mean flux of all constituents using a 200 s running mean to detrend raw data. EdiRe software (Clement, 2004; University of Edinburgh) was used to calculate fluxes on 5 and 30 min intervals following CarboEurope-IP recommendations (Aubinet et al., 2000). A 2-D rotation was applied in order to align the streamwise wind velocity component with the direction of the mean velocity vector. Calculation of surface-atmosphere CH_4 exchange by EC method (Aubinet et al., 2000) involves the estimation of two kinds of term: the turbulent fluxes and the storage term (Finnigan et al., 2009). Assuming horizontal homogeneity and a flat terrain within the averaging time of 30 min, the net final flux of the trace gas CH_4 is given by:

$$F_{CH_4}^{EC} = \int_0^h \overline{\frac{\delta \chi_c}{\delta t}} dz + \overline{w' \chi_c'}(h),$$ (1)

where $F_{CH_4}^{EC}$ is the measured EC flux of CH_4 in $\mu mol \, s^{-1}$. The first term on the left-hand side corresponds to the storage flux, i.e. the time-rate-of-change in CH_4 concentration (χ_c) in ppm ($\mu mol \, mol^{-1}$) below the height (h) at which measurements are made (z referring to the vertical coordinate). However, U^* threshold analyses (see below,

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i.e. Supplement, Fig. S1) revealed that the storage term was negligible and contributed very little to the total fluxes. Accordingly, storage term was not further considered in our calculations. The second term of Eq. (1) corresponds to the vertical turbulent exchange given as the covariance between the vertical wind speed (w) and the CH_4 concentration. The primes denote the instantaneous deviation from the temporal mean of wind speed (w) and CH_4 concentration calculated by Reynolds decomposition as:

$$w' = w - \overline{w} \qquad \chi_c = \chi_c - \overline{\chi_c}$$

Fluxes were corrected for spectral frequency loss (Moore, 1986). Latent heat fluxes were corrected for air density variations (Webb et al., 1980). Although much smaller than in open-path EC systems, the Webb-correction theory has to be considered, in order to avoid underestimation of the absolute flux magnitudes of upward-directed fluxes. The Webb-correction for density fluctuations was not performed since there was a constant temperature and pressure in the sampling cell. However, the Webb-correction for the influence of water vapor fluctuations on trace gas fluxes was applied to the data since the sample was not dried to a constant humidity before the molar concentration was measured. Since, no low-pass filtering effect was observed on the water signal the true free atmospheric water vapor cospectra were calculated from the open-path LI-7500 data by applying corrections for lateral separation and sensor line averaging only. The cospectra of the water vapor flux inside the measurement cell of the CRDS analyzer were then simulated by decreasing the free atmospheric cospectra with the inverted transfer functions for cell volume averaging.

According to Ibrom et al. (2007), we applied a *phase effect* that can lead to additional delay in travelling time for CH_4 and water vapor compared to the dry air component and implied a decoupling between gases. In other words we applied the same time lag for the covariance of water vapor and vertical wind velocity than the time lag calculated for CH_4.

2.3 Data quality assessment and gapfilling

Methane and carbon dioxide flux values associated with spikes resulting from signal loss or instrument malfunctioning were removed, as well as short periods when maintenance and instruments cleaning were carried out and power failure occurred. Gaps of up to 1.5 h were filled by applying a simple interpolation and gaps of several hours were filled using the mean diurnal variation (MDV) method (Falge et al., 2001), a method where a missing value is replaced by the mean for that time period based on adjacent days. This gap-filling method was considered to be valid for CH_4 (atmospheric and soil) flux at Laqueuille. Nevertheless, on consecutive rainy periods the MDV method was not applicable to methane fluxes and these gaps were not filled.

2.4 Footprint analyses

For CH_4 and CO_2 annual budgets calculation, in order to attribute measured fluxes to our experimental field (2.81 ha), and to avoid integration of fluxes belonging to adjacent paddocks, we applied the analytical footprint model by Kljun et al. (2004). The footprint analyses was projected to the measure area by rotating the footprint information into the wind direction and overlaid with measurement area representing the field limits. These field limits are the distance between the EC-setup and the measured area (e.g. agricultural fields, paddock) in clockwise 10° steps (in total for 360°). The footprint function was calculated to integrate 80 % of flux, where fluxes data coming from outside the boundary of the measure area, (i.e. experimental field) were excluded.

For the SF_6 experiment, which liked to investigate the reliability of EC method for CH_4 emissions from animals (see Sect. 2.6), we applied a two-dimensional footprint density function based on Kormann and Meixner (2001). This function (for details see Neftel et al., 2008 and Hendriks et al., 2010) allows determining the relative footprint contribution of heifers confined (see Supplement, Fig. S2) in two different distances to the EC setup. The so obtained dilution factor was applied to CH_4 fluxes measured during the SF_6 experiment (see Sect. 2.6).

2.5 CH$_4$ flux measurement with Gaussian Plume method

The Gaussian plume distribution assumes that air plumes follow a normal probability distribution. Accordingly, Gaussian models are most often used for predicting the dispersion of air plumes originating from ground-level or elevated sources. Here we analyzed if modeled CH$_4$ release plumes from cows and a defined artificial CH$_4$ source strength were in line with measurements done by the EC setup, i.e. the CRDS anal**y**zer, in the footprint area. To do so, during autumn 2009, a herd of 5 cows was confined in 10 × 10 m enclosure in the EC-footprint area (see Fig. 1). Downwind of the herd ($\sim$ 20 m distance), the CH$_4$ concentration was measured along a 90 m transect perpendicular to two methane plumes: the confined herd and a defined artificial CH$_4$ source strength ($\sim$ 0.15 g CH$_4$ s^{-1} provided by a gasflask and mass flow controller), separated in space (30 m) to obtain two distinct signals. Air was sampled at 2 m height using a handheld inlet tubing system (i.e. 100 m) connected to the CRDS analyzer, while wind direction and speed was monitored by the EC setup (i.e. sonic anemometer). Sampling frequency was 10 Hz. Each plume transect took $\sim$ 45 s to walk through. A lag time response of 10 s was registered due to tube length. In total 10 measurements were done. At our field site, a typical background CH$_4$ concentration was $\sim$ 1860 ppb.

During experiment (late summer), wind direction was south-western. The mobile measurements took, thus, place on the path north-east of the herd (see Fig. 1).

The measured concentrations in the plume transects were compared with the output of the multiple gauss plume model (Hensen and Scharff, 2001). For each measurement, animal distribution within the enclosure was noted using a grid (5 × 5 m, 4 points), further used as source map to determine separate plumes for each grid point (see Hensen and Scharff, 2001). For each cow-analyzer combination (i.e. source-receptor combination) the receptor concentration is:

$$\text{Concentration}(x,y,z) = \frac{Q}{2\pi u \sigma_y \sigma_z} e^{\frac{-y^2}{(2\sigma y)^2}} \left[e^{\frac{-(z-H)^2}{(2\sigma z)^2}} + e^{\frac{-(z+H)^2}{(2\sigma z)^2}} \right]$$

with

$$\sigma_y = A x^B z_0^{0.2} T^{0.35}, \quad \sigma_z = C x^D (10 z_0)^{0.53 E} \quad \text{and} \quad E = x^{-0.22}$$

where x is the distance along the plume axis, y the axis perpendicular to the plume axis, z the height above ground level, Q the source strength, u the wind speed measured on top of the paddock, and H the height of the emission (cows head). σ_y and σ_z are dispersion parameters that depend on distance to the source, on the degree of turbulence of the atmosphere, the roughness length of the surface z_o, and on the timescale used for averaging. A, B, C and D are dependent on the stability class (Pasquill, 1974). Then the herd emission was equal to the source strength needed in the model to achieve an agreement between the integral of the modelled and measured concentration pattern along the plume transect. During the experiment, average wind speed was $2.5 \, \text{m s}^{-1}$ and stability class and roughness length of the surface (z_o), were set to D and 0.05. The reported herd emission is the average of a set of emissions estimates for individual plume transects.

2.6 CH_4 flux measurement with SF_6 method

CH_4 emissions by heifers were measured in two 4-days measurement campaigns (autumn 2009), using the SF_6 tracer technique as described by Martin et al., (2008). Measurements were carried out on 5 heifers confined in a $20 \times 20 \, \text{m}$ enclosure (Fig. 2). Enclosures were set up in the four main wind directions (i.e. N, S, W, E) to ensure an animal presence in the footprint area. In order to test the effect of distance, four enclosures were setup at $10 \, \text{m}$ (D1) and $30 \, \text{m}$ (D2) distance from the EC setup (see Fig. 2). Depending on the main instantaneous wind direction, the herd was placed in one of the four respective enclosures and distances to the EC setup. Daytime and night time herd positions are shown in Supplement, Table S3.

To apply the tracer technique, a calibrated SF_6 permeation tube was dosed orally into the rumen of each cow 2 weeks before measurement campaigns. Representative breath samples from each animal were collected in pre-evacuated yoke-shaped

polyvinyl chloride collection devices by means of capillary and Teflon tubing fitted to a halter. The collection devices were changed every 12 h to get daytime (from 08:00 h to 20:00 h) and night time (from 20:00 h to 08:00 h) measurements, as during night time low turbulences can lead to stratification of the atmosphere which can make impossible to measure CH_4 by the EC-method. Decoupling of day- and night time measurements, allows quantifying possible losses of CH_4 emission during night-time.

Absolute CH_4 emissions from each animal were calculated according to Johnson et al. (1994), using a known permeation rate of the hexafluoride (SF_6) tracer and the concentrations of SF_6 and CH_4 in the breath samples:

$$F_{Heif}^{SF_6}(g\,d^{-1}) = SF_6\ permeation(g\,d^{-1}) \times \frac{[CH_4]}{[SF_6]}$$

A CH_4 budget per unit ground area ($F_{heif}^{SF_6}$) was calculated by adding the measured CH_4 emission rate per animal of 5 cows and dividing the sum per unit ground area (2.81 ha).

2.7 Comparison with CH_4 emission factors

CH_4 emissions measured by the EC setup and the SF_6 method were compared to CH_4 emissions estimated by three frequently-used CH_4 emission factors based on ingested biomass, stocking rate and animal live weight, respectively (i.e. Giger et al., 2000; Pinares-Patino et al., 2007 and IPCC, 2006, Chapter 10: Emissions from Livestock and Manure Management; Tier 2 – Eq. 10.19).

3 Results and discussion

3.1 EC-setup

3.1.1 Spectral analyses

Ruminating animals create CH_4 plumes of warm, humid air, which are expected to be-
⁵ have differently to CH_4 emissions from soil-vegetation. In a first step, the reliability of
the EC set-up was examined by comparing power spectra of T_s [sonic temperature],
[H_2O], [CO_2] and [CH_4]. Co-spectra signals give no further information since the cor-
rection factor is instrument related (Ibrom et al., 2007). For those analyses we randomly
selected twenty half-hour data sets (i.e. between June and September) identified with a
¹⁰ presence of animals in the footprint. The results of the spectral analysis were averaged
and the logarithmic spectral densities were plotted against frequency (Fig. 3). Com-
parisons showed that at low frequencies ($< 0.01\,Hz$) the normalized power spectra of
both CO_2 and, to a lesser degree, H_2O had relatively higher spectral power than the
temperature spectrum (Fig. 3). On the contrary, the normalized power spectra of CH_4
¹⁵ showed a slight tendency to extend downwards in the low frequency range, indicating
that the low range was instrument-related. In the high-frequency domain spectra were
very similar, confirming that no physical low-pass filtering (i.e. EC closed-path system)
had affected the H_2O and CH_4 spectra. This is certainly due to the combination of
a relatively short tube and a high flow rate compared to other closed path systems.
²⁰ Accordingly, our EC-setup delivered reliable measurements of CH_4 emissions from ru-
minants.

3.1.2 Quality performance of EC measurements for nighttime periods

During night time periods with low friction velocity (u^*), the turbulence of the atmo-
sphere can become too low to perform EC measurements correctly. To determine the
²⁵ critical u^* threshold value for EC measurements at our site, the CH_4 flux data were

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plotted against u^* data from night periods ($R < 20\,\mathrm{W\,m^{-2}}$). Nightly CH_4 fluxes showed a significant decrease for periods with $u^* < 0.06\,\mathrm{m\,s^{-1}}$ (Supplement, Fig. S1). This result is lower than the critical u^* value for CO_2 fluxes (of $0.13\,\mathrm{m\,s^{-1}}$) at the same site and lower than the critical u^* value of $0.09\,\mathrm{m\,s^{-1}}$ found for CH_4 fluxes over peat meadow in the Netherlands (Hendriks et al., 2008). Due to the u^* threshold, 12 % of night time fluxes could not be accurately measured with the turbulent flux term solely. To complete the flux calculation in these conditions, the storage term (i.e. the time-rate-of-change in CH_4 concentration below the height at which measurements are made) should be added to the turbulent flux term. However, those turbulent conditions were quite rare at our site (7 % for the total data set). Moreover, the storage term had very low values between -0.5 and $0.5\,\mathrm{nmol\,m^{-2}\,s^{-1}}$ for u^* up to $0.06\,\mathrm{m\,s^{-1}}$ below 2 m, indicating that this term was negligible at our site.

3.2 Data comparison between CH_4 measurements and Gaussian plume modeling

Figure 4 shows the dynamics of averaged CH_4 plumes along the measured transect. The line shows the measured excess concentration. CH_4 concentration increased progressively and reached a maximum value of 200 and 270 ppb when walking close by the herd and the defined artificial CH_4 source (i.e. gasflask), respectively. The ratio of the integrated measured animal plume vs. integrated modelled animal plumes was 1.01 (± 0.07), indicating that CH_4 emissions from the animals measured by the analyzer were in agreement with those calculated by the Gaussian plume model. Using this ratio, the absolute estimated CH_4 emissions reached in average $280\,\mathrm{g}\,(\pm 18)\,\mathrm{day^{-1}}\,\mathrm{animal^{-1}}$ which was in line with values obtained by SF_6 technique (176 to $275\,\mathrm{g\,CH_4\,day^{-1}\,animal^{-1}}$ see Pinares-Patino et al., 2007; Allard et al., 2007). Possible uncertainty between measured and estimated CH_4 emissions, determined through the ratio between integrated modelled and measured gasflask data, respectively, showed an uncertainty of about 30 %. However, this result should be taken with

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precaution as measurement accuracy is closely related to meteorological conditions (i.e. changing wind direction, instable downwind, etc.) and possible longer lag times or analyzer failure, leading to scatter in the measured emission data.

3.3 Data comparison between the EC method and the SF$_6$ method

In general, footprint calculations carried out for average daytime conditions during the SF$_6$ measurement campaign showed that animals were downwind within the footprint area. The main source location contributing to the measured CH$_4$ flux was between 10 and 30 m from the EC setup (see Supplement, Fig. S2). Ninety per cent of the flux came from within 80 m of the EC setup.

CH$_4$ concentration dynamics showed that CH$_4$ production by enteric fermentation was more important during the 1st period of measurement (Supplement, Fig. S3), ranging from a background concentration of 1.87 to 2.15 ppm. During the 2nd period, maximum values reached around 1.9 ppm as CH$_4$ emissions from animals were very low. Meteorological conditions also varied significantly between measurement campaigns, with warmer temperatures and lower friction velocities during the first campaign (15 to 25 °C, 0.03 to 0.4 m^2 s^{-2}) compared with the second measurement campaign (6 to 14 °C, 0.4 to 0.7 m^2 s^{-2}).

According to the SF$_6$ method, CH$_4$ emissions showed strong variation among animals (data not shown) both during the day and at night, which underlines that a simple extrapolation of emission factors is likely to lead to misleading extrapolations of CH$_4$ emissions. CH$_4$ emissions from heifers varied between 90.5 and 149.3 g ha^{-1} and between 69.5 and 159.3 g ha^{-1} for day- and night-time periods, respectively (Table 1). Daily CH$_4$ emissions (i.e. 24 h) were in the range of 160.2 to 290.2 g ha^{-1}, which is similar to a previous report for our study site (200 to 242 g CH$_4$ ha^{-1}, Pinares-Patiño et al., 2007).

According to the EC method, the herd emission rates and their contribution to measured flux varied depending on the distance between the herd and EC setup. The

dilution effect due to increasing distance between the EC system and fenced area was corrected for using a two-dimensional footprint density function (see Neftel et al.., 2008; Tuzson et al., 2010) based on Korman and Meixner 2001 (see Supplement, Fig. S2). However, we found that the relative footprint contribution of the fenced animals to the measured flux were lower than those registered by Tuzson et al. (2010). This discrepancy between studies may reflect the higher position of the EC instrumentation in our setup (2 m versus 1.2 m in the former study), leading to a "higher" footprint. Nevertheless method comparison showed that at short distances to the EC setup (D_1, 10 to 30 m), measurements were in agreement between methods, with mean emissions of 241 and 225 g ha^{-1} d^{-1} for the SF_6 and EC method respectively (i.e. EC was 6.5 % lower than SF_6; Table 1). Lower values (i.e. 10 % lower) were found at greater distances D_2; with mean emissions of 263 and 237 g ha^{-1} d^{-1} for the SF_6 and EC methods. These deviations from expected values (i.e. SF_6 method) were independent from meteorological variability and day/night time measurements. Additionally, animals were free to move within the enclosure minimizing effects of unnatural animal behaviour leading to low CH_4 emissions. Across the whole dataset, the EC method sometimes revealed higher CH_4 emissions than the SF_6 method, suggesting losses of CH_4 emissions measured by SF_6 method due to climatic conditions (i.e. high windspeed) and technical problems.

Overall, our results suggest a systematic error of the EC method due to dilution of the CH_4 signal in air (though within the footprint area), leading to lower values for animals far from the EC setup. The two-dimensional footprint density function correcting for this dilution effect resulted in 8.3 % lower emissions for the EC compared with the SF_6 method on average (Table 1). However, it should be noted that such a correction factor can only be applied for paddocks where animal localisation is known (e.g. individual geographic information system, camera) throughout the grazing period, which is technically and economically difficult to perform (see Detto et al., 2011) and does not always deliver reliable data (e.g. Herbst et al., 2011). In order to estimate the effect of

these CH_4 emissions "losses" (i.e. bias) on the annual CH_4 budget, CH_4 fluxes were analyzed during the grazing period May–September over two years (2010 and 2011).

3.4 CH_4 emission patterns

Over the 147 days grazing period in 2010, (i.e. 7680 half-hourly data sets), only 25 days (10 %) were not recorded due to power failure and maintenance operations. By filtering further using hard (footprint, range, spikes, u^*) and soft filters (gapfilling quality) we excluded 24 % of CH_4 flux data. As an example of flux pattern, CH_4 and CO_2 fluxes were plotted over eight weeks in 2010 (Fig. 5). Although the CH_4 fluxes were rather variable over time, a diurnal pattern could be observed with increasing CH_4 between 09:00 a.m. and 08:00 p.m. (maximum values occurring around 03:00 p.m., Fig. 5). This agrees with daily periodicity in the grazing and behaviour pattern of heifers observed in our own data as well as for sheep in other studies (Harris and O'Connor, 1980; Champion et al., 1994; Lockyer and Champion, 2001; Dengel et al., 2011). Daily mean methane emissions were related to the number of heifers in the field; the number of heifers decreased over the summer, and CH_4 emissions decreased in parallel (Fig. 6). There was considerable variability in CH_4 fluxes resulting from (i) variation in the number of animals present in the flux footprint and, (ii) variation in rumination pattern of heifers (see Dengel et al., 2011).

3.5   Annual CH_4 budgets

In our study we were not able to separate CH_4 fluxes (emissions) originating from CH_4 absorption (see below), presence/absence of animals in the footprint and daily periodicity in the grazing behaviour, respectively. Possible mismatches between "real" and measured CH_4 emissions might, thus, represent a systematic bias as it does in studies, where emissions appear in hotspots variable in space and time (e.g. N_2O, Flechard et al., 2007; Hendriks et al., 2008; Schrier-Uijl et al., 2010). To analyse such a systematic error CH_4 emissions measured by the EC setup were compared to CH_4

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emissions estimated by three frequently-used CH_4 emission factors based on ingested biomass, stocking rate and animal live weight, respectively.

Measured net CH_4 fluxes reflect the balance between methane production from ruminants and soil, and consumption by methanotrophic bacteria in the soil. Chamber measurements at the Laqueuille site showed that the soil component is negligible as orders of magnitude smaller than animal emissions (data not shown).

The EC method resulted a budget of 124 and 151 kg CH_4 ha^{-1} over the grazing period in 2010 (147 days) and 2011 (169 days) respectively (mean animal emissions of 199 and 206 g CH_4 day^{-1} animal^{-1} in 2010 and 2011). These results are close to values obtained previously at our site using SF_6 method (126 kg CH_4 ha^{-1} or 204 g CH_4 day^{-1} animal^{-1}; Allard et al., 2007). In situ CH_4 emission measurements also proved to be in line with theoretical calculation methods (i.e. based on ingested biomass and animal weight) developed by Giger et al. (2000) and Pinares-Patino et al. (2007) estimating annual CH_4 budgets of 160 and 126 kg ha^{-1} (255 and 204 g CH_4 day^{-1} animal^{-1}) in 2010 and of 198 and 157 kg ha^{-1} (261 and 206 g CH_4 day^{-1} animal^{-1}) in 2011 for our paddock. However, the IPCC (2006) calculation method Tiers 2 appeared to overestimate CH_4 emissions, producing values of 269 and 332 kg ha^{-1} yr^{-1}. According to our EC-based results, cumulated CH_4 emissions (EC method) over the grazing season seem to offset the CH_4 emissions "losses" (i.e. bias) observed during the short-term comparison experiment. Under real field conditions, animal distribution within the paddock was probably more random than during the comparison experiment where all animals were concentrated at a small surface. These "unrealistic" conditions may have partly interfered with surface upwind properties necessary for reliable EC measurements.

The potential carbon dioxide sink of the field over the measurement period was 559 g CO_2 m^{-2} in 2010 (Supplement, Fig. S4). When estimating the net greenhouse gas balance (i.e. considering CH_4 and a global warming potential of 25, IPCC, 2007), the sink activity was reduced by 310 g CO_2 eq m^{-2}, leading to a net carbon dioxide sink of 249 g CO_2 m^{-2} over the grazing period. As the calculation did not include a complete

year, we would expect an improved net greenhouse gas sink due to mean annual C sequestration activity of $795 \, g \, CO_2 \, m^{-2}$ at the study site (see Klumpp et al., 2011). Notably, estimating CH_4 emissions via IPCC Tier 2 emissions factors, CH_4 emissions would be twice as important, reducing the potential net carbon sink by 37 instead of 16 % indicating a need for direct field measurement.

4   Conclusions

Here we have shown that EC measurements are a convenient tool to investigate long-term dynamics of CH_4 fluxes of ruminants over a large area (hectare). We found that accuracy of the EC method varied depending on distance between animals and the measurement mast, and animal feeding activity, indicating that results may be improved by animal tracking. Nevertheless, CH_4 budget estimations for the whole grazing season were in good agreement with results of emission factors based on both feed intake and animal live weight and the SF_6 technique. In contrast, the IPCC method Tier 2 clearly overestimated the CH_4 fluxes at our site.

We like to underline that the EC method and associated detailed measurements offer original research opportunities to study in situ effects of management and vegetation structure at field and animal scales on adjacent paddocks, and may contribute to the development of more adapted mitigation options.

Supplementary material related to this article is available online at:
http://www.biogeosciences-discuss.net/9/14407/2012/
bgd-9-14407-2012-supplement.pdf.

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Table 1. Comparison of mean methane (CH_4) flux measured according to distances and methods, with SF_6 tracer and eddy covariance (EC) technique over two study periods. Dilution effect correction was applied using the two-dimensional footprint density function (see Neftel et al., 2008).

Distance	Date	Day ($g\,ha^{-1}$)		Night ($g\,ha^{-1}$)		24 h ($g\,ha^{-1}$)	
		SF_6	EC	SF_6	EC	SF_6	EC
10–30 m	28/09/2009	90.5	133.5	69.6	114.5	160.2	248.0
	29/09/2009	110.3	140.6	129.8	110.7	240.1	251.3
	12/10/2009	137.6	120.9	136.0	34.8	273.7	155.8
	13/10/2009	129.1	51.7	159.3	193.3	288.4	245.1
Mean D1		116.9	111.7	123.7	113.3	240.6	225.0
30–50 m	30/09/2009	116.8	108.0	118.3	112.2	235.0	220.2
	01/10/2009	149.3	131.2	108.6	103.5	257.9	234.7
	14/10/2009	128.2	140.3	140.8	118.0	269.1	258.3
	15/10/2009	140.8	135.6	149.4	98.0	290.2	233.6
Mean D2		133.8	128.8	129.3	107.9	263.1	236.7
Mean D1 + D2		125.3	120.2	126.5	110.6	251.8	230.9

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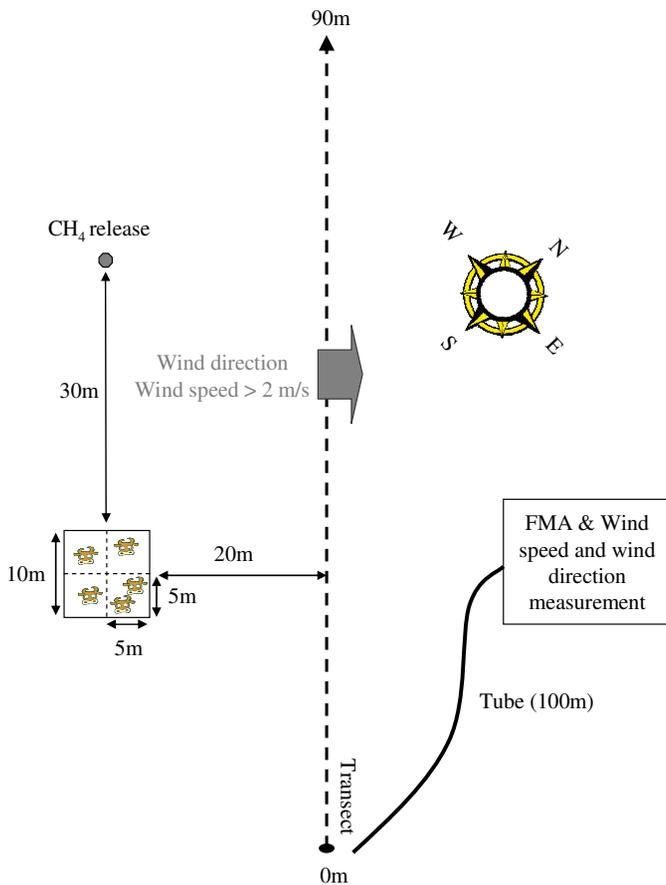
Fig. 1. Scheme of experimental setup carried out in autumn 2009 for comparison of CH_4 flux modelled with Gaussian Plume method against CH_4 flux measured with EC method. Wind direction was predominantly from the SW to W sectors.
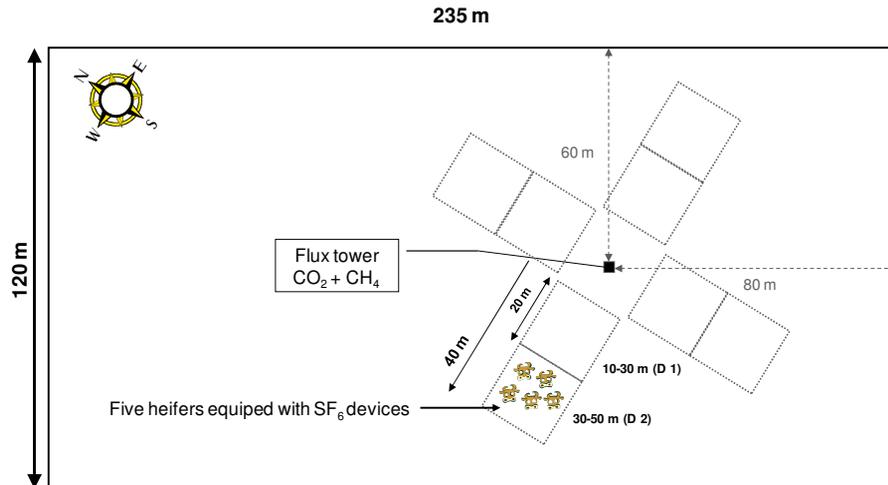
Fig. 2. Scheme of experimental setup for comparison of CH_4 flux measured with SF_6 method EC method. Experiment was carried out during autumn 2009 for 8 days and 8 nights. The paddocks in close and fare distance to the EC setup were grazed on 2 consecutive days (48 h) over 2 periods, by 5 heifers equipped with SF_6 devices. Wind direction was predominantly from the North to East sectors.
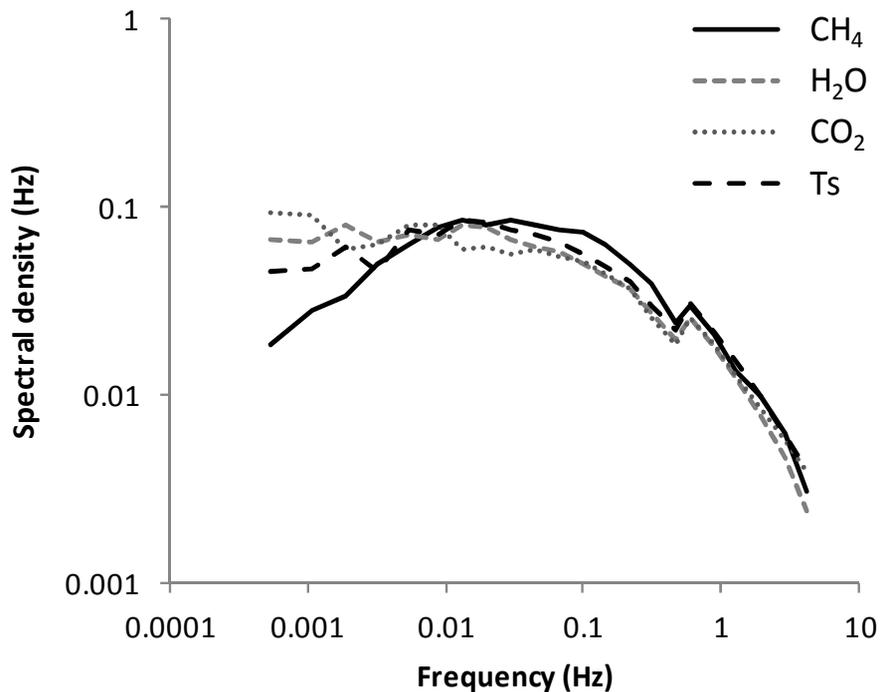
Fig. 3. Comparison of distribution in frequency of averaged normalized spectra of water vapour (H$_2$O, LI-7500), carbon dioxide (CO$_2$, LI-7500), sonic temperature (T_s) and methane (CH$_4$, CRDS analyzer with scroll pump) for signals identified with cows present in the near fetch of the tower. All the spectra are normalized so the area beneath the curve is equal to one.

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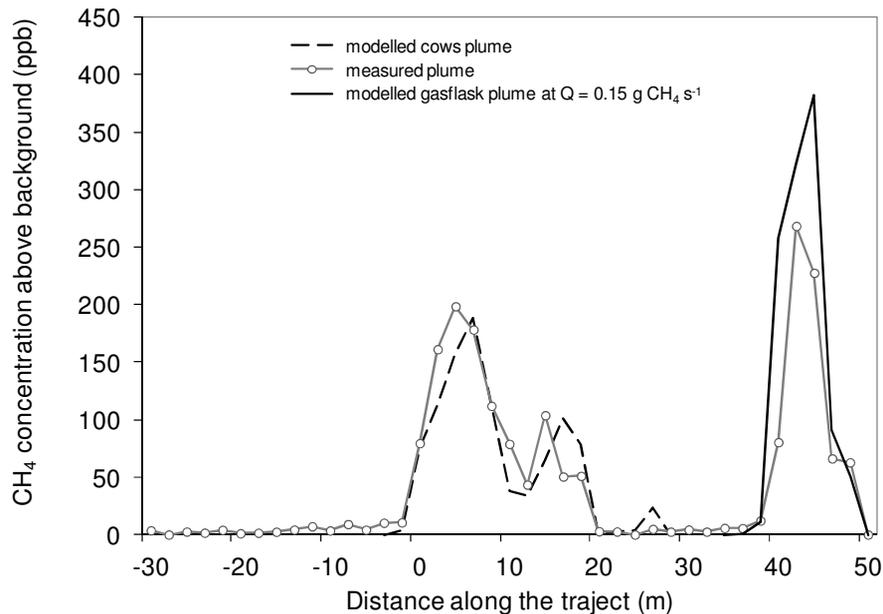
Fig. 4. Comparison of average dynamic of ten modelled CH_4 plumes for animals (black dashed line) and gasflask (black solid line) with average dynamic of ten measured CH_4 plumes (grey line) along a transect of 90 m.
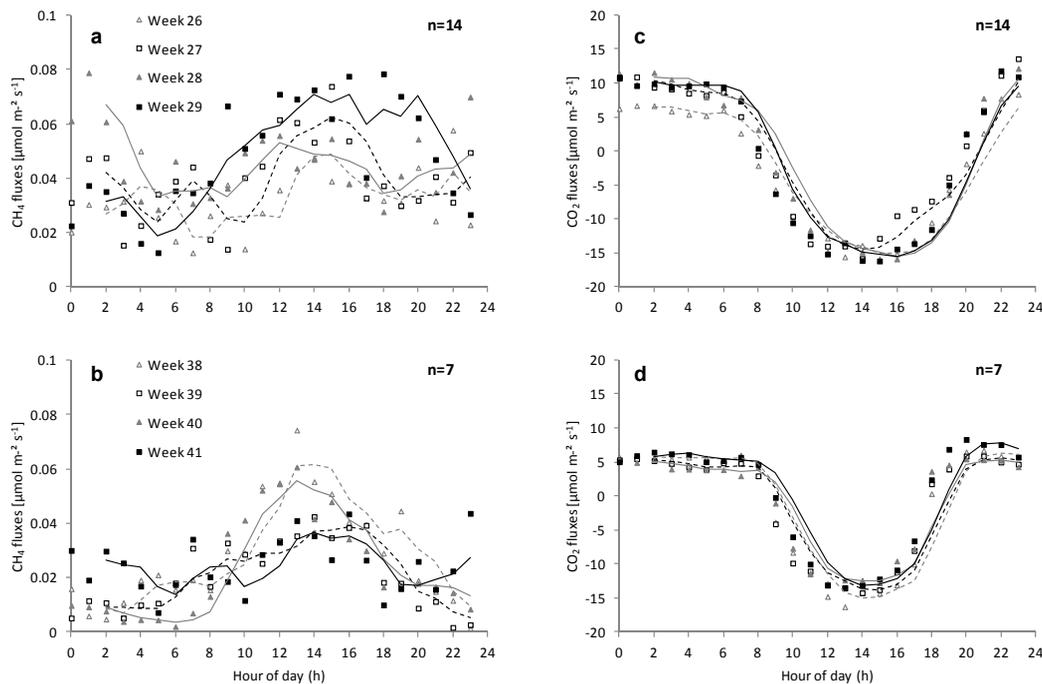
Fig. 5. Fluxes of CH_4 **(a, c)** and CO_2 **(b, d)** for 8 weeks of the year 2010. The numeral, *n*, refers to the number of heifers in the field. Lines are mobile means of 3 h intervals.

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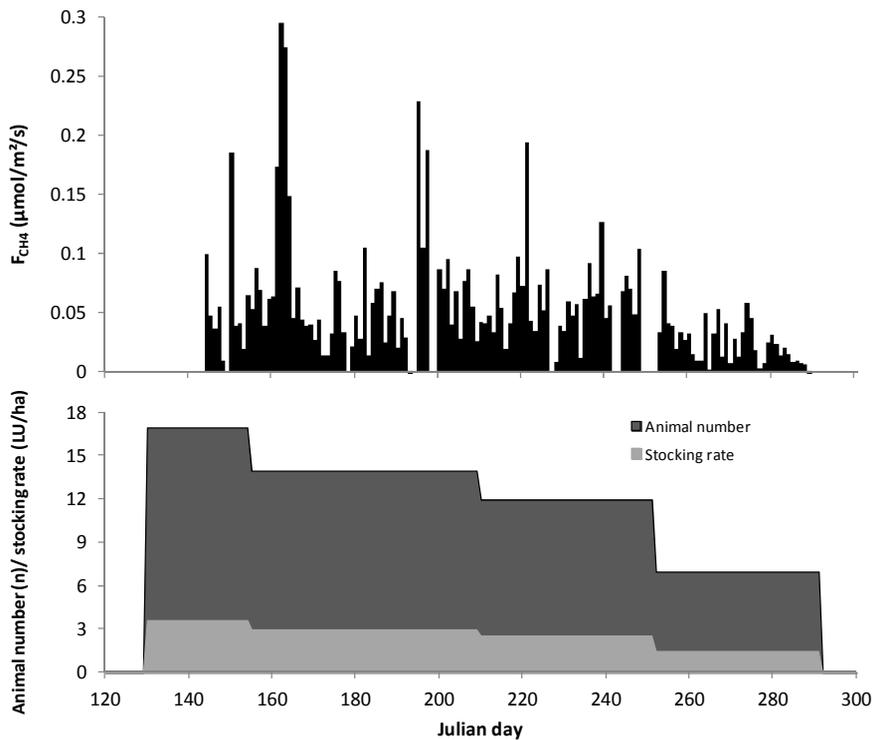
Fig. 6. Mean daily CH_4 fluxes in context of number and stocking rate of heifers of the year 2010.