See discussions, stats, and author profiles for this publication at: https://www.researchgate.net/publication/223701943

# Livestock methane emission: From the individual grazing animal through national inventories to the global...

#### Article in Agricultural and Forest Meteorology · February 2007

DOI: 10.1016/j.agrformet.2006.03.028

CITATIONS	READS
133	701

1 author:

Keith Richard Lassey

Lassey Research & Education

120 PUBLICATIONS 2,709 CITATIONS

SEE PROFILE

All content following this page was uploaded by Keith Richard Lassey on 06 May 2015.



Available online at www.sciencedirect.com





Agricultural and Forest Meteorology 142 (2007) 120-132

www.elsevier.com/locate/agrformet

## Livestock methane emission: From the individual grazing animal through national inventories to the global methane cycle

Keith R. Lassey \*

National Institute of Water and Atmospheric Research, P.O. Box 14-901, Wellington, New Zealand Received 11 November 2004; received in revised form 15 November 2005; accepted 15 March 2006

#### Abstract

Methane is a potent greenhouse gas whose atmospheric abundance has grown 2.5-fold over three centuries, due in large part to agricultural expansion. The farming of ruminant livestock, which generate and emit methane during digestion ('enteric fermentation'), is a leading contributor to this growth. This paper overviews the measurement or estimation of enteric methane emissions at a range of spatial scales. Measurement of individual animal emissions focuses particularly on grazing livestock for which the SF<sub>6</sub> tracer technique is uniquely appropriate. Gaining insight into factors that influence methane production requires that feed intake and feed properties be determined, enabling the methane emitted to be expressed per unit of intake. The latter expression is commonly encapsulated in the 'methane conversion factor',  $Y_{\rm m}$ , an entity that enables small-scale methane emission estimates to be extrapolated to national and global enteric methane inventories. The principles of this extrapolation and sources of uncertainty are discussed, along with the significance of this global source within the global methane cycle. Micrometeorological and similar measurement techniques over intermediate spatial scales are also surveyed.

© 2006 Elsevier B.V. All rights reserved.

Keywords: Atmospheric greenhouse gas; Methane; Ruminant livestock; Microbiological fermentation

#### 1. Introduction

As an atmospheric greenhouse gas, methane assumes a climatic importance that belies its low abundance of ~1750 ppb (parts per billion, or nanomoles per mole dry air). This abundance represents a 2.5-fold growth through the industrial era. Currently, methane contributes about 20% of anthropogenic 'radiative forcing', second behind carbon dioxide at 60%. With its strong infrared absorbance more than off-setting its short atmospheric residence time when compared to carbon dioxide, its 'global warming potential' (GWP) is estimated at 23 kg CO<sub>2</sub>/kg CH<sub>4</sub> (Ramaswamy et al.,

\* Fax: +64 4 386 2153.

E-mail address: k.lassey@niwa.co.nz.

2001), though an earlier estimate of 21 kg CO<sub>2</sub>/kg CH<sub>4</sub> applies to inventories reported to the UN Framework Convention on Climate Change (UNFCCC) and to Kyoto Protocol accounting. Thus, each incremental kg of methane injected into the atmosphere absorbs the same amount of infrared energy from Earth's outgoing radiation spectrum as 23 kg CO<sub>2</sub> (or 8.4 mol CO<sub>2</sub>/mol CH<sub>4</sub>) over a 'time horizon' standardized at 100 years.

Analyses of air trapped in polar ice show that over at least the past 450,000 years and four glacial cycles the methane mixing ratio, while correlating strongly with temperature, has not exceeded  $\sim$ 700 ppb (Delmotte et al., 2004). Etheridge et al. (1998) have documented a relatively stable atmospheric methane of  $693 \pm 10$  (1 S.D.) ppb over 1010–1700 AD, followed by a steadily rising mixing ratio reaching 1750 ppb in 2000 (Fig. 1).

<sup>0168-1923/\$ –</sup> see front matter  $\odot$  2006 Elsevier B.V. All rights reserved. doi:10.1016/j.agrformet.2006.03.028



Fig. 1. A reconstruction of the global-mean methane mixing ratio in the troposphere (lower atmosphere) over three centuries based on measurements in air trapped in Antarctic ice and firn (Etheridge et al., 1998) and on contemporary measurements based on the global NOAA/CMDL monitoring network (Dlugokencky et al., 1998, 2003). The atmospheric record for the centuries preceding 1700 AD is relatively featureless (Etheridge et al., 1998). 1 ppb = 1 nmol/mol dry air, measured against 1983 NOAA/CMDL gas standards.

The rate of methane accumulation has slowed in recent years, while exhibiting appreciable inter-annual variability (Simpson et al., 2002; Dlugokencky et al., 2003). There is no doubt that this 2.5-fold increase in atmospheric methane over three centuries is caused by human activities, with agriculture, most notably livestock and rice farming, prominent among them.

This paper discusses the role of farmed livestock as a direct source of methane via 'enteric fermentation'. Indirect sources from manure management are also important but not discussed herein. Methods used to quantify enteric methane emissions over a full range of spatial scales, from animal to global scale and linkages between them, are surveyed. The main focus is on grazing livestock, which account for the majority of the world's farmed livestock yet are the least amenable to investigation of their emissions and their emission determinants. Section 2 discusses the measurement of emissions from individual animals on a daily or similar time scale. In conjunction with concurrent measurements of feed intake and feed quality, they provide direct estimates of the 'methane conversion factor', denoted  $Y_{\rm m}$  and defined below. Section 3 overviews 'top-down' techniques based on measuring and interpreting spatial gradients of methane concentration in the atmosphere. In extrapolating to national emission inventories (i.e., annual emissions by country, as reported for example to the UNFCCC), Section 4 examines inventory methods and rationale and the linkage through  $Y_{\rm m}$  to small-scale measurements. Section 5 further extrapolates to the global livestock source again emphasizing the linkages through  $Y_{\rm m}$ , followed in Section 6 by a discussion of this global source in the context of the global methane cycle.

Section 7 supplies some numerical perspectives on methane emission prior to conclusions in Section 8.

#### 2. Direct per-animal emission measurements

Of all livestock, ruminants dominate methane production. This is a result of the complex microbiological fermentation that breaks down cellulose and other macro-molecules in the rumen (fore-stomach), generating methane (CH<sub>4</sub>) in the process and expelling it via eructation through the mouth and nose (Moss et al., 2000). This digestive step increases the efficiency at converting fibrous forages to nutrients, relative to nonruminant herbivores. The long-established method for determining methane emission rates from individual animals encloses each animal in a respiration chamber; for an open-circuit chamber the compositions of the inflowing and out-flowing air are analyzed and compared, or for a closed-circuit chamber the methane build-up is measured (reviewed by Johnson and Johnson, 1995; Johnson et al., 2000a). An alternative approach uses a hood that encloses the animal's head during intervals between feeding sessions (e.g., Boadi et al., 2002b). The aim of those methane determinations, at least prior to ca 1990, was to better understand the energetics of ruminant metabolism, recognising that methane eructation represents an unproductive loss of dietary energy. The greenhouse issue has since introduced another imperative that has stimulated a newer method, the 'SF<sub>6</sub> tracer technique' (Johnson et al., 1994), which enables emissions to be determined from individual animals whether confined or not. This technique, first applied to grazing cattle, has since been applied to grazing sheep (Lassey et al., 1997; Ulyatt et al., 2002a, 2005), deer (M. Krause, formerly AgResearch, New Zealand; S. Hoskin, Massey University, New Zealand, personal communication, 2004), and alpaca (Pinares-Patiño et al., 2003d). It remains the only viable technique for determining enteric methane emissions from individual grazing animals.

#### 2.1. The SF<sub>6</sub> tracer technique

The SF<sub>6</sub> tracer technique is based on inserting a calibrated source of SF<sub>6</sub> (sulfur hexafluoride) into the rumen of each participating animal. This inert tracer, which discharges from a 'permeation tube' (Lassey et al., 2001), has the virtue of being quantitatively detectable in gas samples at very low levels (parts per 10<sup>12</sup>). Time-integrated breath samples are collected, usually over 24 h, and the ratio of the  $CH_4$  to  $SF_6$  release rates is equated to the ratio of their backgroundcorrected concentrations as measured in the breath sample (Johnson et al., 1994; Lassey et al., 1997). Repeated 24-h samples collected over  $\sim$ 5 successive days generally display good day-to-day consistency in inferred daily emission for each animal, such that the variance in per-animal daily emission averaged across the herd or flock is dominated by inter-animal variation (Lassey et al., 1997).

Uncertainties inherent in the SF<sub>6</sub> tracer technique arise from: extrapolation of permeation tube performance (Lassey et al., 2001); variations in breath collection efficiency throughout the collection period (important only if the methane production rate also varies); concerns that the imposition of sampling equipment may affect feeding behaviour; and a dearth of data on the proportion of methane released from the anus (undetected by the SF<sub>6</sub> tracer technique).

As validation support for their proposed SF<sub>6</sub> tracer technique, Johnson et al. (1994) reported a comparison with chamber-methods for a single heifer over three daytime periods, obtaining good agreement:  $8.3 \pm 2.6$ and  $9.2 \pm 0.4$  g CH<sub>4</sub> h<sup>-1</sup> for the tracer-based and chamber-based measurements, respectively. Ulyatt et al. (1999) have summarized then-available data comparing the SF<sub>6</sub> tracer technique with alternative measurement protocols, including the use of chambers. Although there were some contradictory data, Ulyatt et al. cited the results of two independent comparisons between the tracer-based and chamber-based techniques that could not detect significant differences between measurements of emission rates. Boadi et al. (2002b) have compared in more detail the  $SF_6$  tracer technique with direct measurements using ventilated hoods enclosing the heads of six heifers. They obtained

insignificantly different mean emission rates of  $93 \pm 3$ and  $98 \pm 3$  g CH<sub>4</sub> day<sup>-1</sup>, for the tracer-based and hoodbased measurements, respectively. However, interanimal variability was significant only for the tracerbased data, suggesting that a repeat of the comparison with a greater number of animals might be warranted.

The favourable comparison between the SF<sub>6</sub> tracer technique and chamber (but not hood) measurements would appear to affirm that methane efflux from the anus (flatus) is a minor component of enteric methane. Nevertheless, the sole determination of flatus-mediated methane release that is available,  $\sim 2\%$  based on measurements on four ewes (Murray et al., 1976), does not preclude this source being significant in an overall methane inventory.

The SF<sub>6</sub> tracer technique is widely adopted in many countries, including the U.S.A. (Pavao-Zuckerman et al., 1999; Johnson et al., 2000b; Westberg et al., 2001; DeRamus et al., 2003), Canada (McCaughey et al., 1997, 1999; Boadi et al., 2002a, 2004), New Zealand (Lassey et al., 1997; Judd et al., 1999; Lassey and Ulyatt, 2000; Lassey et al., 2002; Ulyatt et al., 2002a,b, 2005; Pinares-Patiño et al., 2003d), Australia (Leuning et al., 1999), Ireland (F. O'Mara, University College Dublin, personal communication, 2001), France (Pinares-Patiño et al., 2003a), Brazil (Primavesi et al., 2004), India (A. K. Srivastava, National Dairy Development Board, Gujarat, India, personal communication, 2003), China (H. Dong, Agrometeorology Institute, Beijing, China, personal communication, 2003).

#### 2.2. Supplementary feed intake determinations

Because the methane is derived from ingested feed, measuring methane emitted without also measuring feed ingested limits both data utility and opportunities to investigate emission determinants. A more universal measure of emission is the dimensionless 'methane conversion factor', also known as the 'methane yield',  $Y_{\rm m}$ , which is the methane emitted per unit of feed intake with both methane and intake expressed as energies of combustion. Most feeds contain about 18.4 MJ of gross energy (GE) per kg of dry matter (DM) and methane has energy content 55.65 MJ/kg, so that a typical  $Y_{\rm m}$  value of 6% corresponds to 19.8 g CH<sub>4</sub>/kg DM intake. However, determining feed intake by grazing animals is particularly difficult (discussed below), and intake estimates will usually be the biggest source of uncertainty in SF<sub>6</sub>-based estimates of  $Y_{\rm m}$  for individual animals. When averaged across a herd or flock, a confounding uncertainty will be inter-animal variation

in  $Y_{\rm m}$  (Lassey et al., 1997). While confining the animals under controlled feeding conditions will markedly reduce intake uncertainty, it may also alter the feeding behaviour and feed selection relative to freely grazing animals.

Determining feed intake by a grazing animal is perforce indirect and fraught with uncertainty. It is usually determined by estimating the fraction of the feed that is not digested and therefore voided, together with the daily faecal output of each animal. The former is usually taken as a property of the feed alone (the complement of feed digestibility) and determined for example by near-infrared reflectance spectroscopy (Norris et al., 1976). Collecting daily faecal output is feasible only for small male animals such as sheep (i.e., not to cattle because of the quantity voided), but the burden of a collection bag plus the need for regular mustering can affect grazing behaviour. A biologically inactive marker such as a compound of chromium or ytterbium can be used in place of total faecal collection (Prigge et al., 1981): from the marker concentration in intermittent faecal samples, together with the dose rate or intra-ruminal release rate of the marker, the faecal production can be inferred. However, the concentration of such markers can show marked diurnal variation and lead to unreliable or biassed feed intake estimates, difficulties which can be overcome by using slow intraruminal release capsules of *n*-alkanes, typically  $C_{32}$ (Dove and Mayes, 1991). However, there remain concerns that with some *n*-alkane formulations the pre-calibrated release rate may not be matched intraruminally (G. Waghorn, Dexcel, N.Z., personal communication, 2003). With such concerns in mind, some investigators have preferred to compute the feed intake for individual cattle by applying an energy requirements model (Section 4) in conjunction with easily measured characteristics such as liveweight and milk production (Lassey et al., 1997; Ulyatt et al., 2002a,b), arguing that this provided the more dependable feed-intake estimate (Ulyatt et al., 2002a).

The SF<sub>6</sub> tracer technique not only measures a peranimal methane emission rate and, with co-determined feed consumption rate, a corresponding  $Y_m$  value, but also facilitates the study of those factors that influence methane emissions. Examples of such studies include examining the role of feed characteristics in methane emission (e.g., Pavao-Zuckerman et al., 1999; Woodward et al., 2001, 2002; Boadi and Wittenberg, 2002; Boadi et al., 2002a, 2004; Lassey et al., 2002; Waghorn et al., 2002; DeRamus et al., 2003), and identifying animal and digestion characteristics that may be associated with inter-animal contrasts in methane production (Pinares-Patiño et al., 2003b,c). Such experiments would commonly impose feeding conditions that enable feed intakes to be directly and accurately measured.

An important goal when investigating methane determinants is the development of methane abatement strategies. Many such strategies target a lower methane emission per unit product through greater efficiency of feed utilization, requiring higher intakes of higherquality feed (Mosier et al., 1998). (However, since that strategy increases methane emission per head, its efficacy depends on a concomitant reduction in animal numbers.) The SF<sub>6</sub> tracer technique is thus a valuable tool in methane mitigation research.

Many measurements of methane emissions by grazing livestock have yet to be reported in peerreviewed literature. Table 1 summarizes  $Y_m$  values reported in experiments where both methane emission and feed intakes were determined for grazing cattle, except for the Brazilian experiments (Primavesi et al., 2004). Table 2 reports the same for grazing sheep.

It is inappropriate to draw conclusions about methane determinants based solely on the data of Tables 1–2. An individual experiment may be unintentionally atypical of grazing conditions that it seeks to represent, such as a kikuyu pasture experiment, unreported in those tables, which revealed extraordinarily low methane emissions by both sheep and cattle for reasons that remain undetermined (Ulyatt et al., 2002b). In addition, the uncertainty and possible biases of the various feed-intake determinations should be taken into account.

Despite the above caveats that could account for some  $Y_{\rm m}$  outliers in Tables 1 and 2, it is noteworthy that  $Y_{\rm m}$  values reported in those tables broadly support recommendations in the range 6–7% by the IPCC Good Practice Guidance (IPCC, 2000, Tables 4.8 and 4.9).

#### 2.3. Application to housed livestock

Application of the  $SF_6$  technique to individual animals housed indoors raises questions about identifying background concentrations of both  $SF_6$  and  $CH_4$ , and re-inhaling air locally enriched in these gases.

Breath collected from an inlet near the nose is perforce diluted with local background air. The efficacy of the SF<sub>6</sub> tracer technique requires that  $CH_4$  and SF<sub>6</sub> mixing ratios in each sample be much larger than those in background air, which should be representative of that inhaled. A standard out-door practice would be to measure  $CH_4$  and SF<sub>6</sub> mixing ratios in background air samples upwind of the herd/flock under test, and

Table 1
Survey of methane and intake measurements for grazing cattle using the SF <sub>6</sub> tracer technique

ID <sup>a</sup>	Country	#Animals	Age (year)	Liveweight (kg/hd)	Pasture <sup>b</sup>	Feed digest'y <sup>c</sup> (%)	Voluntary (kg/hd day)	Dry matter intake how measured? <sup>d</sup>	Milk product'n (kg/hd/d)	CH <sub>4</sub> emission	
										(g/hd/d)	$Y_{\rm m}~(\%)$
1	NZ	10	Mature	483	PRG/WC	77.3	12.9	Req'ments model	14.1	263	6.2
2a	NZ	10	6.6	475	PRG/WC	82.0	19.3	Req'ments model	31.1	431	6.8
2b	NZ	10	6.8	489	PRG/WC	75.5	18.1	Req'ments model	23.5	248	4.2
2c	NZ	10	7.1	505	PRG/WC	68.4	14.8	Req'ments model	12.1	182	3.7
2d	NZ	10	7.3	489	PRG/WC	62.8	6.8	Req'ments model	0.0	137	6.1
					+ hay						
3a	NZ	9	6.3	438	Kikuyu	61.4	15.6	Req'ments model	11.2	363	7.1
3b	NZ	8	7.2	585	Crabgrass	67.0	18.9	Req'ments model	14.5	422	6.7
4	CA	16	1.1	398	ALF/MB	59.3	13.9	$Cr_2O_3$ marker 0.0		195	4.5
5a	CA	16	Heifer	506	ALF/MB	51.2	11.4	$Cr_2O_3$ marker Early lact		267	7.1
5b	CA	16	Heifer	516	MB	45.0	9.7	Cr <sub>2</sub> O <sub>3</sub> marker Early lac		294	9.5
6a	FR	6	8	712	Timothy	83.8	22	$Yb_2O_3$ marker 0		204	5.9
6b	FR	6	8	712	Timothy	72.7	17	Yb <sub>2</sub> O <sub>3</sub> marker	0	273	6.7
6c	FR	6	8	712	Timothy	57.9	16	Yb <sub>2</sub> O <sub>3</sub> marker	0	232	6.6
6d	FR	6	8	712	Timothy	48.7	15	Yb <sub>2</sub> O <sub>3</sub> marker	0	228	6.5

<sup>a</sup> The numerical character refers to the references as follows, and the alphabetical character identifies a separate experiment: 1, Lassey et al. (1997); 2, Ulyatt et al. (2002a); 3, Ulyatt et al. (2002b); 4, McCaughey et al. (1997); 5, McCaughey et al. (1999); 6, Pinares-Patiño et al. (2003a).

<sup>b</sup> Pasture abbreviations (common and botanical names) are as follows—PRG: perennial ryegrass (*Lolium perenne*); WC: white clover (*Trifolium repens*); Kikuyu: kikuyu grass (*Pennisetum clandestinum*); Crabgrass: crabgrass (*Digitaria sanguinalis*); ALF: alfalfa/lucerne (*Medicago sativa*); MB: meadow brome (*Bromus biebersteinii*); Timothy: timothy (*Phleum pratense*).

<sup>c</sup> Feed digestibility is either DM (or energy) digestibility, or in the case of ID 4–6 organic matter digestibility (all measured *in vitro*); the latter exceeds the former by typically 1–3% points.

<sup>d</sup> 'Req'ments model' refers to calculating individual intakes via an energy requirements model such as the IPCC tier 2 methodology; other entries refer to the identified slow-release runnial marker detected quantitatively in faecal samples.

Table 2	
Survey of methane and intake measurements for grazing sheep using the SF <sub>6</sub> tracer technique	
	-

ID <sup>a</sup>	Country	#Animals	Age (month)	Liveweight (kg)	Pasture <sup>b</sup>	Feed digest'y <sup>c</sup> (%)	Voluntary dry (kg/day)	Matter intake how measured? <sup>d</sup>	CH <sub>4</sub> emission	
									(g/day)	$Y_{\rm m}~(\%)$
1	NZ	50	8	37.0	PRG/WC	75.3	1.27	Whole faeces	18.9	4.6
2a	NZ	12	48	54.3	PRG/WC	82.0	1.51	n-Alkane marker	30.6	6.1
2b	NZ	12	50	53.6	PRG/WC	72.2	1.46	n-Alkane marker	33.2	6.9
2c	NZ	12	54	62.0	PRG/WC	74.5	1.35	n-Alkane marker	27.0	6.1
2d	NZ	12	58	65.5	PRG/WC	82.0	1.89	n-Alkane marker	27.9	4.6
3	NZ	10	6	34.5	Kikuyu	61.2	0.76	Whole faeces	15.6	6.3
7a	NZ	12	8	37.9	Mixed	79.7	1.39	Whole faeces	19.3	4.1
7b	NZ	12	9	41.2	grasses PRG/WC	81.4	1.70	Whole faeces	21.9	3.9
7c	NZ	12	7	46.9	Standing dead	54.0	1.21	Whole faeces	21.4	5.3
7d	NZ	12	>24	69.0	BT/CF	73.2	1.69	n-Alkane marker	35.2	6.3
8	NZ	11	8	ca. 35	PRG/WC	81.2	1.68	Whole faeces	19.4	3.6
9	AU	7	12	27.1	ALF/RC/RG	69.5	0.51	<i>n</i> -Alkane marker	11.7	6.9

<sup>a</sup> The numerical character refers to the references as in Table 1 and as follows, and the alphabetical character identifies a separate experiment: 7, Ulyatt et al. (2005); 8, Judd et al. (1999); 9, Leuning et al. (1999).

<sup>b</sup> Pasture abbreviations (common and botanical names) are as in Table 1 and as follows—BT: browntop (*Agrostis lanatus*); CF: cocksfoot (*Dactylis glomerata*); RC: rose clover (*Trifolium hirtum*); RG: ryegrass (*Lolium rigidum*).

<sup>c</sup> Feed digestibility DM (or energy) digestibility throughout (all measured *in vitro*).

<sup>d</sup> 'Whole faeces' refers to the collection of all faecal production as a measure of the indigestible feed throughput (amenable only to male sheep); '*n*-Alkane marker' refers to a slow release alkane runnial marker detected quantitatively in faecal samples.

125

subtract them from those measured at the mouth (Lassey et al., 1997). Results can be confounded if there is significant re-inhalation of the same air parcels, unless the air is stirred and thereby sampled by the background collectors. The re-inhalation problem would be at its most severe for animals housed indoors in close proximity, though the problem could also be present in principle for animals intimately grazing in very still air. The indoor problem can be alleviated by using forced ventilation and deploying multiple background collectors appropriately positioned to test for indoor concentration gradients. This approach was adopted by Woodward et al. (2001), though not directly reported, and no concentration heterogeneities significant enough to influence emission rate calculations were detected.

To measure collective emissions from a small herd (or flock) confined indoors at high density, a tracer such as SF<sub>6</sub> can be released within the enclosure, and gas for subsequent CH<sub>4</sub> and SF<sub>6</sub> analysis collected at or near a ventilation outlet (Marik and Levin, 1996; Kaharabata et al., 2000). Such techniques are helpful for assessing the daily emission pattern by the enclosed herd, and can also provide estimates of herd-average  $Y_m$  if corresponding feed information is collected. (Strictly, this strategy does not provide a value for  $Y_m$  averaged across the herd, but the quotient of herd-aggregate methane emission rate to herd-aggregate GEI; the quotient of two averages is not identical to the average quotient.) However, such  $Y_m$  estimates are not necessarily transferable to differently managed livestock.

### 3. Top-down emission estimates based on concentration gradients

Herd-average (or flock-average) estimates of surface fluxes can be made using 'top-down' techniques based on measured atmospheric concentrations. These include micrometeorological techniques useful for paddockscale estimates, through to air sampling from aircraft for regional-scale estimates, and ultimately to remotely sensed atmospheric constituents, an approach still in its infancy (Frankenberg et al., 2005). The techniques are all based on measuring methane concentration gradients to sufficient precision, and using interpretive models to infer the causal surface fluxes. The concentration gradients may be either upwind–downwind differences or vertical profiles or both. Artificially high stocking densities may be deployed to assure large gradients and attain sufficient precision in inferred fluxes.

The footprint of a particular sampling point ('receptor') varies with the position of that point,

including its elevation (Schuepp et al., 1990). The higher the receptor, the further upwind and larger in extent is the footprint. Thus, as the receptor moves up a vertical profile the footprint moves upwind and expands, and in the process may envelope areas of different stocking density or indeed may envelope other methane sources such as waste management facilities. These considerations introduce uncertainty into flux estimates that rely on analysis of how the footprint overlaps a heterogeneous source area (e.g., flux gradient methods).

Various modelling approaches suitable for spatial scales of  $\sim 10-100$  m have been examined and compared by Denmead and collaborators (Denmead, 1995; Denmead et al., 1998, 2000; Harper et al., 1999; Leuning et al., 1999; Griffith et al., 2002; Desjardins et al., 2004). In particular, the mass balance (MB) approach, introduced into this context by Denmead (1995) and well suited to optical detection techniques, relates the methane flux from a confined area to the net methane outflow assessed through measuring horizontal and vertical gradients at or near the confinement boundary. Leuning et al. (1999) obtained good agreement between this approach and the SF<sub>6</sub> tracer technique applied to 7 of the 14 sheep that were confined at high density in a  $24 \text{ m} \times 24 \text{ m}$  enclosure in NSW, Australia. Mean methane emissions of the 14 sheep by MB and the 7 sheep by tracer technique were  $11.9 \pm 1.5$  (S.E.M.) and  $11.7 \pm 0.4$  (S.E.M.) g day<sup>-1</sup> sheep<sup>-1</sup>, respectively. Desjardins et al. (2004) demonstrate that a deliberate release of methane from a 9  $m^2$  grid can be 'recovered' to within 5% using an open-path laser detection system (crosswind path length  $\sim$ 50 m at six heights to 6 m, at 10–12 m downwind of the release) through a MB analysis.

In a novel MB approach, Lockyer and Jarvis (1995) covered a grazing area ( $\sim 40 \text{ m}^2$ ) with a polythene tunnel and determined emissions based on methane concentration in the forced airflow at the inlet and outlet. Although they could 'recover' a known methane release, emission rates from 5 to 12 grazing ewes during confinement periods of 7.2–25.5 h proved very low, possibly related to feeding patterns being disturbed by the confinement.

For paddock-scale flux estimates, Judd et al. (1999) applied flux gradient (FG) analysis to vertical profile data collected downwind of a paddock stocked with 6-month old sheep at 20 sheep ha<sup>-1</sup> under normal farm management in New Zealand (NZ). A sub-flock of the sheep were individually assessed using the SF<sub>6</sub> tracer technique. In a 5-day experiment, the tracer-based emission estimate averaged over 99 sheep-days was  $19.5 \pm 0.5$  (S.E.M.) g day<sup>-1</sup> sheep<sup>-1</sup>, which compared

favourably with a mean of 23 g day<sup>-1</sup> sheep<sup>-1</sup> based on FG analyses for 102 half-hour intervals throughout the 5 days and nights. A significant source of uncertainty was the varying flux heterogeneity caused by the sheep roaming and flocking within the enclosure.

Laubach and Kelliher (2004) conducted a comparison similar to that of Judd et al. (1999) for a paddock stocked with lactating dairy cows in three seasonal experiments (January, March, October 2002) in NZ. Each experiment was analyzed using both FG and a variant of MB known as the "integrated horizontal flux" (IHF) approach. The IHF and FG flux estimates were found to be consistent, but the former was adjudged superior through having a smaller measurement error, due principally to the FG estimate being more sensitive to cow movements within the paddock. Both flux estimates were consistent with the tracer-based estimates from a sub-herd of 20 cows for 5 days during each experiment. Combining the three experiments, the IHF, FG and tracer-based emission estimates were  $330 \pm 153$ ,  $344 \pm 187$  and  $365 \pm$  $61 \text{ g day}^{-1} \text{ cow}^{-1}$ , respectively, in which cited errors are standard deviations and the FG estimate uses the two most favourable receptor altitudes.

Flesch et al. (2004) presented a "backward-Lagrangian stochastic" (BLS) model, an inverse modelling approach that directly infers emissive gas fluxes from an areal source based on upwind-downwind contrasts. This approach is well suited to a detection system that measures line-average concentrations such as a laser detection system. The BLS model explicitly simulates source-receptor relationships and avoids the need to measure the complete vertical concentration profile. Flesch et al. (2004) tested the model using a known methane release distributed over a  $6 \text{ m} \times 6 \text{ m}$ square and measuring up to 100 m upwind. They tabulated extensively the release rates and comparative 'recoveries', reporting satisfactory recoveries except during periods of strong atmospheric instability (Obukhov stability length <2 m in magnitude) and during transition periods. Even with wind obstructions introduced, this approach worked well except very close to the obstructions (Flesch et al., 2005).

Laubach and Kelliher (2005) extended their earlier work as summarised above (Laubach and Kelliher, 2004) to include a BLS model interpretation of the same data. They concluded that the BLS model corroborates the IHF approach, requiring no extra data. In cases where the extra data demands of IHF (downwind vertical concentration and wind-speed profiles), cannot be met, BLS would be an acceptable alternative, at a small cost to measurement error (typically 20% for BLS, 15% for IHF under favourable conditions). The mean per-cow emission rate inferred using the BLS model that can be compared with the three-experiment means reported above was  $368 \pm 171 \text{ g day}^{-1} \text{ cow}^{-1}$ .

On a spatial scale of  $\sim 100 \text{ m}$  to a few km, downwind vertical profiles can be determined through sampling from masts, from towers, or from tethered balloons or helikites. Gimson et al. (2004) have presented some preliminary results for grazing dairy cattle in regions of NZ, including interpretation through inverse modelling. Work is presently under way to validate these interpretations using known methane release rates.

On a regional scale (20-50 km along the wind direction), Wratt et al. (2001) estimated methane emissions from livestock grazing within that region based on vertical concentration profiles upwind and downwind of the region. The profiles were determined from analyses of air sampled from light aircraft, typically between 150 and 1500 m altitude. The region selected was a coastal plain in NZ during on-shore (westerly) winds, assuring no sources upwind of the coast. The approach was similar to that adopted by Choularton et al. (1995) who measured methane fluxes from Scottish peatlands. Wratt et al. (2001) employed a range of modelling techniques to check the influence of NZ topography, including a nested meso-scale meteorological model that simulated airflows over complex terrain, validated against local meteorological data. Simulated profiles based on livestock distribution data and dispersion modelling were consistent with observation. Gimson and Uliasz (2003) subsequently developed inverse modelling techniques that provided both best-fit emission fluxes and confidence limits. In these nonideal terrains with heterogeneous fluxes and with restrictions on altitudes attainable by aircraft there are significant challenges to attaining precisions of  $\sim 20\%$  in area-averaged fluxes.

Top-down techniques are unsuitable for providing estimates of  $Y_{\rm m}$  due to the limited precisions attainable in both emission and feed-intake estimates, if the latter estimates are obtainable at all. Thus such techniques should be viewed as tools to validate, verify or up-scale smaller scale determinations, to supply verification support for methane abatement implementations, or to provide areal budgets of methane from all natural and anthropogenic sources, rather than as a technique for delivering definitive emission rate estimates for use in inventories. To date, micrometeorological measurements can achieve precision of ~15–20% in estimates of herd-average per-animal emission rates under favourable meteorological conditions and on favourable terrain. Such estimates are fully consistent with those based on the  $SF_6$  tracer technique, within the limitations of this precision.

#### 4. National inventory estimation

Ratifying parties to the UNFCCC agree to report annually their national emission inventory following guidelines promulgated by IPCC (1996, 2000). Key requirements for compiling a national enteric methane inventory are a characterization of the national livestock population and of their feedstock, and appropriate  $Y_m$ values which provide the linkage to small-scale measurements. The characterization involves disaggregation by livestock category, viz: dairy and non-dairy cattle, sheep, buffalo, etc, each by gender, breeding status, age cohort, husbandry regime, season, as appropriate. Seasonal emission variation might follow from pregnancy, birthing and lactation seasons, seasonal variation in feed quality, or from seasonal management regimes (e.g., grazing in summer, feedlots in winter).

Two methodologies are recommended by IPCC good practice guidance (GPG): tier 2 for countries with sufficient data to allow the livestock and feed to be characterized (cattle, sheep, buffalo only); or tier 1 otherwise (IPCC, 2000, Chapter 4.2). A country may develop its own tier 2 methodology, appropriately documented and peer-reviewed. A tier 2 methodology should be used for 'key source categories' (sources of sufficient prominence in the national inventory).

The tier 2 methodology for enteric methane applies an energy requirements model to calculate an 'emission factor' (EF: annual methane emission per animal) for each livestock category, disaggregated as far as is appropriate to capture variations by gender, age, management regime or season. This model matches the realised energy demand (of maintenance + productivity) by a representative animal in each category to the energy supply (feed), taking account of the inefficiencies of feed conversion (gross energy ingested to net energy utilized). The energy demand is calculated by summing the energy needed to maintain body condition or to achieve growth, to sustain pregnancy, to produce milk and/or fleece, and to deliver work (e.g., ploughing by bullocks). In assessing energy supply, the single most important feed property is its digestibility (the proportion of the feed, usually measured by its DM, that is digested). With digestibility specified, the annual gross energy intake (GEI) of the animal that upon utilization will satisfy the net energy demand, can be estimated using the IPCC (1996, Appendix C, p. 4.49) or alternative requirements model. Such alternative models may use different measures of energy, and Fig. 2

Ruminant energy throughput, typical of a high-quality forage diet

Of every 100 MJ of gross energy intake (GEI):  $\sim 25$  MJ excreted as faeces (indigestible energy) 75 MJ digestible energy  $\sim 6$  MJ eructated as methane ( $Y_m \approx 6\%$ )  $\sim 5$  MJ excreted as urine 63 MJ metabolizable energy  $\sim 5$  MJ radiated as heat 58 MJ net energy, available for maintenance, production

Fig. 2. A representation of typical energy flows and energy losses during ruminant digestion of a high-quality forage diet, illustrated with a digestibility of 75%.

illustrates the relationship between the variously used measures: gross, digestible, metabolizable, and net energy.

The loss of energy as emitted methane is assessed as the fraction  $Y_{\rm m}$  of the computed GEI, with recommended values for  $Y_{\rm m}$  tabulated in GPG (IPCC, 2000, Tables 4.8 and 4.9). Thus, in effect the national feed intake is calculated and the methane emission estimated by applying Y<sub>m</sub> for each disaggregated livestock/feed combination. Recognising the large scatter in  $Y_{\rm m}$  values reported in the literature that neither strongly correlate with forage quality nor differentiate ruminant categories on the same diet (Johnson and Johnson, 1995; Johnson et al., 2000a), the GPG recommendations are 'broadbrush'  $Y_{\rm m}$  values: either 0.06 or 0.07 for all cattle except feedlot cattle fed with a diet comprising at least 90% concentrates (0.04) and for all sheep except juveniles (0.05), each with range of uncertainty  $\pm 0.005$ . As a result,  $Y_{\rm m}$  will often be applied beyond the limited range of livestock categories, husbandry regimes and diets for which it has been measured. (Strictly,  $Y_{\rm m}$  values selected for this purpose should be calculated from experimental data as the quotient of mean CH<sub>4</sub> to mean GEI, each averaged across the measured herd/flock, rather than as the mean of individual  $Y_{\rm m}$  quotients. However, the uncertainties and broad-brush choice of  $Y_{\rm m}$  values do not warrant this distinction being heeded, despite established inter-animal variations in  $Y_{\rm m}$ .)

The much simpler 'tier 1' methodology calculates the emission from a specific animal category by applying a fixed EF for which default values are tabulated (IPCC, 1996, Tables 4-3 and 4-4). The tier 1 default EFs for cattle are themselves calculated using a tier 2 methodology in which attributes such as bodyweight, milk productivity, feed digestibility, and  $Y_{\rm m}$  are assigned values adjudged to be appropriate for each of several regions (IPCC, 1996, Appendix A, pp. 4.29–4.35). Consequently, where local values for some or all of these attributes are known, it is always possible to use tier 2 to calculate EFs that are superior to the tier 1 defaults. Furthermore, uncritical adoption of tier 1 EFs over successive years will fail to capture emission trends that result from increasing productivity. Some tier 1 default EFs can also be unrealistic (e.g., those for Oceania when applied to Australian or NZ cattle) and can mislead attempts to calculate global enteric emissions (see Section 5).

The above assessment of an enteric methane inventory relies on the livestock population and the feedstock being well characterized. In some countries, climatic extremes or variability (and regional variations of these) will effect diet quality and availability, and impose changes in animal husbandry, that will be difficult (and costly) to characterize. In addition, livestock population censuses may be infrequent, be unreliable or fail to capture seasonal population variations (see Section 5). All of these considerations challenge the inventory compiler and introduce appreciable inventory uncertainty.

The less that is known about livestock and feed characteristics, the more uncertain the inventory is likely to be. In addition, the applied  $Y_{\rm m}$  values introduce compounding uncertainty, including that due to extrapolating those  $Y_{\rm m}$  values beyond their measurement circumstances. Accordingly, national enteric methane inventories will rarely have less than ~20% uncertainty.

#### 5. The global livestock methane source

Crutzen et al. (1986) were the first to make a comprehensive estimate of the global methane source from livestock. They used an energy-requirements approach to estimate feed intakes and EFs, applying these to FAO-sourced animal population data. Several of these EFs survive as tier 1 defaults in GPG. The global enteric methane source was estimated at 74 Tg year<sup>-1</sup> for ca 1982 of which 74% were contributed by cattle and 8-9% by each of buffalo and sheep. More recent estimates, summarized to 1994 by Johnson and Ward (1996), have involved re-categorizing global animal populations and combining these with the best EFs available. Such estimates include:  $84 \text{ Tg year}^{-1}$  by Anastasi and Simpson (1993) for 1990; 80 Tg year<sup>-1</sup> by Mosier et al. (1998) for 1994; 71 Tg year<sup>-1</sup>, including 44 Tg year<sup>-1</sup> from grassland-derived feed, by Clark et al. (2005) for 2003. A global source strength of  $\sim$ 80 Tg year<sup>-1</sup> makes livestock arguably the largest anthropogenic methane source (Prather et al., 1995, 2001).

While the FAO dataset on animal populations is the best that is widely available, the data may contain some biases, and the characterization of both livestock and feed would depend upon access to local knowledge. In some countries, census data may be biased either low or high depending on perceived or actual advantages in under-reporting or over-reporting herd/flock sizes (e.g., due to tax impositions or tax relief). There may also be reporting issues for farms adjacent to or straddling international borders, and due to international livestock trading. For some countries the census may be timed to coincide with a stable breeding population and by not taking account of births the census value is lower than the season-weighted mean population. An example of the last-mentioned is the sheep population in NZ: at the census date of 30 June (winter) the sheep population of about 40 million (ca. 2000) is dominated ( $\sim$ 75%) by breeding ewes, the remainder being mainly immature replacement stock and "hoggets" to be slaughtered at about 1-year of age. In August-September about 35 million lambs are born and reared ( $\sim 120$  per 100 ewes), which are either slaughtered for the meat trade a few months later at  $\sim 2/3$  mature weight or are retained as replacement stock or hoggets. While emissions by this 'lamb pulse' are accommodated in the national inventory, they would not be captured by a sole reliance on FAO population data.

In principle, reliance on FAO data can be minimized by estimating the global methane source as an aggregation of national inventories that are reported to UNFCCC. In practice this is complicated by a need to complete many gaps and ambiguities in the record, usually without access to sufficient local information. Nevertheless, the USEPA do maintain datasets of such aggregations (Scheehle et al., 2002; Scheehle and Kruger, in press) in which the global enteric methane source is assessed at 76.6, 79.0, and 80.9 Tg for 1990, 1995 and 2000, respectively. These estimates agree well with those based directly on FAO data and little countryspecific information.

Uncertainties should therefore be recognised in both the animal population estimates and their categorization, and in the various applied  $Y_{\rm m}$  values. These uncertainties cannot be quantified rigorously. If the population data and thence global feed intake, and the applied  $Y_{\rm m}$  values, each have ~15–20% uncertainty, then the corresponding estimate of the global enteric source would be ~20–24% uncertain. This figure is consistent with the uncertainty estimate supplied by Prather et al. (1995) in the IPCC Second Assessment Report of  $65-100 \text{ Tg yr}^{-1}$ .

#### 6. The global methane budget

The most dependable estimate of the total methane source to the atmosphere comes from balancing the global methane budget. By comparison, aggregating individual source categories (wetlands, biomass burning, coal mining, oil and gas mining, enteric fermentation, manure management, rice agriculture, landfills, etc.) provides a more uncertain measure of the global source.

Comprehensive global measurement networks (e.g., Dlugokencky et al., 1994, 1998, 2003) provide spatial patterns of atmospheric methane mixing ratio which give confidence in an estimate of the contemporary global tropospheric burden at  $\sim$ 4900 Tg, with a growth rate varying in the range  $\sim 10 \pm 10$  Tg year<sup>-1</sup>. The rate of methane removal from the troposphere (lower atmosphere up to the 'tropopause' at  $\sim 10$  km) is believed to be dominated by reaction with the photo-chemically produced OH radical (e.g., Crutzen and Zimmerman, 1991). This same mechanism and others almost completely remove methane from the stratosphere so that troposphere-stratosphere exchange is effectively a uni-directional removal ( $\sim 40 \text{ Tg year}^{-1}$ ). Methanotrophic soil microbes remove  $\sim 30 \text{ Tg vear}^{-1}$  methane from air entrained in pores. The resulting best estimate of the global sink is: 506 (tropospheric OH) + 30 (soils) + 40 (stratosphere)  $\approx 576 \text{ Tg year}^{-1}$  (Prather et al., 2001). Thus a methane molecule resides in the atmosphere for  $\sim 4900/576 = 8.4$  years on average. If a varying growth rate averages  $\sim 10 \text{ Tg year}^{-1}$ , then the global source is  $\sim$ 586 Tg year<sup>-1</sup>. Of the global source, around 220 Tg year<sup>-1</sup> is believed to be natural (Houweling et al., 2000), leaving an anthropogenic source of  $\sim$  370 Tg year<sup>-1</sup>. While this estimate of anthropogenic source carries as much as  $75 \text{ Tg year}^{-1}$  uncertainty (Schimel et al., 1996), enteric fermentation is arguably the largest individual component at 20-25%.

#### 7. Some numerical perspectives

Ruminant livestock (cattle, buffalo, sheep, goats, camels, deer, etc.) are prolific methane generators. One way to put the magnitude into a quantitative perspective is to view the methane released as a hypothetical energy source. The energy output of a single dairy cow ( $\sim$ 300 g CH<sub>4</sub> day<sup>-1</sup>) is equivalent to a power wastage of  $\sim$ 0.2 kW. In some countries motor vehicles have been modified to use natural gas (methane) as a fuel.

The same cow's methane output could power such a motor car for about  $5 \text{ km day}^{-1}$ .

On a global scale, the annual methane emission from all farmed livestock is about 80 Tg which has a total combustion energy of 4.5 EJ (etajoule or  $10^{18}$  J) at a mean generation rate of 140 GW. The 80 Tg CH<sub>4</sub> would occupy 112 km<sup>3</sup> or 4 trillion ft<sup>3</sup> at standard temperature and pressure (0 °C and 1 atm.). This represents about 20% of natural gas production in the USA (19 trillion ft<sup>3</sup> in ca. 2000), and would provide sufficient fuel for about 95 power stations each of 600 MW(e), assuming 40% efficiency typical of single-cycle thermal generation.

It should be emphasized that the writer is not advocating the collection of enteric methane for its energy value. However, improved feed utilization could result in some of this energy being channelled into production of milk, meat, etc.

For a greenhouse perspective of enteric methane, compare the  $CO_2$ -equivalent release that accompanies the production of one litre of milk with the  $CO_2$  as a combustion product of one litre of gasoline. For typical dairying systems in both NZ (Lassey et al., 1997) and USA (Westberg et al., 2001), the production of methane and milk are in the ratio of about 19 g CH<sub>4</sub> per litre milk, despite the greater per-cow production of both in USA. This is equivalent to 0.44 kg CO<sub>2</sub> emitted per litre of milk produced, which is about 19% of the ~2.3 kg CO<sub>2</sub> produced upon combustion of 11 of gasoline.

Recognising that  $SF_6$  is a very potent greenhouse in its own right (global warming potential 22,200 kg CO<sub>2</sub>/ kg SF<sub>6</sub>: (Ramaswamy et al., 2001)), it could also be asked whether SF<sub>6</sub> released from the intra-ruminal permeation tube is of greater concern for global warming than the methane emitted by its host. For experiments undertaken with sheep in NZ, permeation rates of  $\sim 0.8 \text{ mg}$  $SF_6 day^{-1}$  are selected, equivalent to ~18 g CO<sub>2</sub> day<sup>-1</sup>. 1. This is less than 20% of the CO<sub>2</sub> equivalent of  $\sim 30 \text{ g CH}_4 \text{ day}^{-1}$  ( $\approx 700 \text{ g CO}_2 \text{ day}^{-1}$ ). For experiments undertaken with cows, permeation rates are ~4 mg SF<sub>6</sub> day<sup>-1</sup> ( $\approx$ 90 g CO<sub>2</sub> day<sup>-1</sup>), which is less than 15% of ~300 g CH<sub>4</sub> day<sup>-1</sup> ( $\approx$ 7000 g CO<sub>2</sub> day<sup>-1</sup>). Even though there are inefficiencies in charging and utilizing  $SF_6$  permeation tubes, the use of  $SF_6$  as a tracer in this context appears to be justifiable.

#### 8. Summary and conclusions

Increasing attention is being paid to the build-up of greenhouse gases in the atmosphere that are believed to foreshadow global warming. The 2.5-fold growth in methane abundance over the past 300 years is largely a

result of food production (livestock farming, rice cultivation), waste decomposition (e.g., landfills), and fossil-fuel mining, and makes methane the second largest contributor after carbon dioxide to putative human-induced warming.

Farmed ruminant livestock are arguably the largest single source of methane to the atmosphere from human activities. Since methane emission, generated by microbial action in the rumen, represents an unproductive loss of dietary energy, reducing the methane emitted per unit intake or per unit product will benefit both the farmer and the environment. That energy loss, encapsulated in the 'methane conversion factor'  $Y_{\rm m}$ , is in the range 4–10% of gross energy intake, but typically exceeds 10% of the net energy available for milk or other production (Fig. 2). As  $Y_{\rm m}$  is also the key parameter for extrapolating to national and global emission inventories encompassing a wide range of animal husbandries, it necessary to better understand origin of the variability in reported  $Y_{\rm m}$  values, and in particular the deterministic role of diet quantity and quality upon  $Y_{\rm m}$ .

Experiments to measure  $Y_{\rm m}$ , whether from grazing or housed animals, commonly employ the SF<sub>6</sub> tracer technique (Johnson et al., 1994) in tandem with feed intake determinations. Not only is this technique uniquely able to measure methane emissions by individual grazing animals, but it is also well suited to investigating the dependence of  $Y_{\rm m}$  on animal and feed characteristics. Because intakes by grazing animals are notoriously difficult to measure reliably and accurately, a controlled feed supply would commonly be used to investigate determinants of  $Y_{\rm m}$ . Currently adopted in many countries, the SF<sub>6</sub> tracer technique is proving pivotal to establishing more accurate inventories and to examining the efficacy of potential methane mitigation strategies.

The standard compilation of national and global inventories for enteric methane assesses the feed required to sustain the animal populations and to enable those populations to be productive (pregnancy, milk, meat, fleece, work). Methane emission is equated to the fraction  $Y_{\rm m}$  of that feed requirement, estimated separately for disaggregated livestock categories. This explicitly links  $Y_{\rm m}$  to small scale experiments where it is measured. The global enteric source so assessed, ~80 Tg year<sup>-1</sup> (uncertainty 65–100 Tg year<sup>-1</sup>), contributes 20–25% of the observed 2.5-fold growth in atmospheric methane.

#### Acknowledgements

I have benefited greatly through collaboration with Marc Ulyatt on applications of the SF<sub>6</sub> tracer technique since its introduction into NZ in 1995 with assistance from Kris Johnson, Hal Westberg (Washington State University) and Pat Zimmerman (formerly, NCAR, Boulder, CO). Many discussions over several years with Marc Ulyatt, Garry Waghorn (formerly AgResearch, NZ), Don Johnson (Colorado State University, Fort Collins, CO) and Harry Clark (AgResearch) have enhanced my knowledge of animal sciences. Discussions with Neil Gimson (NIWA, NZ) on meteorological modelling, and Elizabeth Scheehle on the USEPA source compilation, have been valuable. Marc Ulyatt and Neil Gimson provided timely critiques of the manuscript, which was further improved following anonymous reviews. Funding support is from the NZ Foundation for Research, Science and Technology under contract C01X0204 and through co-funding of the NZ Pastoral Agricultural Research Institute (AgResearch), and from WMO Commission for Agricultural Meteorology.

#### References

- Anastasi, C., Simpson, V.J., 1993. Future methane emissions from animals. J. Geophys. Res. 98, 7181–7186.
- Boadi, D.A., Wittenberg, K.M., 2002. Methane production from dairy and beef heifers fed forages differing in nutrient density using the sulphur hexafluoride (SF<sub>6</sub>) tracer gas technique. Can. J. Anim. Sci. 82, 201–206.
- Boadi, D.A., Wittenberg, K.M., McCaughey, W.P., 2002a. Effects of grain supplementation on methane production of grazing steers using the sulphur (SF<sub>6</sub>) tracer gas technique. Can. J. Anim. Sci. 82, 151–157.
- Boadi, D.A., Wittenberg, K.M., Kennedy, A.D., 2002b. Validation of the sulphur hexafluoride (SF<sub>6</sub>) tracer gas technique for measurement of methane and carbon dioxide production by cattle. Can. J. Anim. Sci. 82, 125–131.
- Boadi, D.A., Wittenberg, K.M., Scott, S.L., Burton, D., Buckley, K., Small, J.A., Ominski, K.H., 2004. Effect of low and high forage diet on enteric and manure pack greenhouse gas emissions from a feedlot. Can. J. Anim. Sci. 84, 444–453.
- Choularton, T.W., Gallagher, M.W., Bower, K.N., Fowler, D., Zahniser, M., Kaye, A., 1995. Trace gas flux measurements at the landscape scale using boundary-layer budgets. Phil. Trans. R. Soc. Lond. A 351, 357–369.
- Clark, H., Pinares-Patiño, C., deKlein, C., 2005. Methane and nitrous oxide emissions from grazed grasslands. In: McGilloway (Ed.), Grassland: A Global Resource. Wageningen Academic Publishers, Wageningen, The Netherlands. Proceedings 20th International Grassland Congress, University College Dublin, 26 June–1 July 2005, pp. 276–293.
- Crutzen, P.J., Zimmerman, P.H., 1991. The changing photochemistry of the troposphere. Tellus 43AB, 136–151.
- Crutzen, P.J., Aselmann, I., Seiler, W., 1986. Methane production by domestic animals, wild ruminants, other herbivorous fauna, and humans. Tellus 38B, 271–284.
- Delmotte, M., Chappellaz, J., Brook, E., Yiou, P., Barnola, J.M., Goujon, C., Raynaud, D., Lipenkov, V.I., 2004. Atmospheric methane during the last four glacial-interglacial cycles: Rapid

changes and their link with Antarctic temperature. J. Geophys. Res. 109, D12104, doi:10.1029/2003JD004417.

- Denmead, O.T., 1995. Novel meteorological methods for measuring trace gas fluxes. Phil. Trans. R. Soc. Lond. A 351, 383–396.
- Denmead, O.T., Harper, L.A., Freney, J.R., Griffith, D.W.T., Leuning, R., Sharpe, R.R., 1998. A mass balance method for non-intrusive measurements of surface-air trace gas exchange. Atmos. Environ. 32, 3679–3688.
- Denmead, O.T., Leuning, R., Griffith, D.W.T., Jamie, I.M., Esler, M.B., Harper, L.A., Freney, J.R., 2000. Verifying inventory predictions of animal methane emissions with meteorological measurements. Boundary-Layer Meteor. 96, 187–209.
- DeRamus, H.A., Clement, T.C., Giampola, D.D., Dickison, P.C., 2003. Methane emissions of beef cattle on forages: efficiency of grazing management systems. J. Environ. Qual. 32, 269–277.
- Desjardins, R.L., Denmead, O.T., Harper, L., McBain, M., Massé, D., Kaharabata, S., 2004. Evaluation of a micrometeorological mass balance method employing an open-path laser for measuring methane emissions. Atmos. Environ. 38, 6855–6866.
- Dlugokencky, E.J., Steele, L.P., Lang, P.M., Masarie, K.A., 1994. The growth rate and distribution of atmospheric methane. J. Geophys. Res. 99, 17021–17043.
- Dlugokencky, E.J., Masarie, K.A., Lang, P.M., Tans, P.P., 1998. Continuing decline in the growth rate of the atmospheric methane burden. Nature 393, 447–450.
- Dlugokencky, E.J., Houweling, S., Bruhwiler, L., Masarie, K.A., Lang, P.M., Miller, J.B., Tans, P.P., 2003. Atmospheric methane levels off: Temporary pause or a new steady-state? Geophys. Res. Lett. 30, 1992, doi:10.1029/2003GL018126.
- Dove, H., Mayes, R.W., 1991. The use of plant wax alkanes as marker substances in studies of the nutrition of herbivores. Aust. J. Agric. Res. 42, 913–952.
- Etheridge, D.M., Steele, L.P., Francey, R.J., Langenfelds, R.L., 1998. Atmospheric methane between 1000 A.D. and present: evidence of anthropogenic emissions and climate variability. J. Geophys. Res. 103, 15979–15993.
- Flesch, T.K., Wilson, J.D., Harper, L.A., Crenna, B.P., Sharpe, R.R., 2004. Deducing ground-to-air emissions from observed trace gas concentrations: a field trial. J. Appl. Meteor. 43, 487–502.
- Flesch, T.K., Wilson, J.D., Harper, L.A., 2005. Deducing ground-toair emissions from observed trace gas concentrations: a field trial with wind disturbance. J. Appl. Meteor. 44, 475–484.
- Frankenberg, C., Meirink, J.F., van Weele, M., Platt, U., Wagner, T., 2005. Assessing methane emissions from global space-borne observations. Science 308, 1010–1014.
- Gimson, N.R., Uliasz, M., 2003. The determination of agricultural methane emissions in New Zealand using inverse modelling techniques. Atmos. Environ. 37, 3903–3912.
- Gimson, N., Brailsford, G., Bromley, T., Lassey, K., Martin, R., Moss, R., Uliasz, M., 2004. Determination of methane emissions from livestock at the farm scale using observed meteorological and concentration profiles and inverse modelling techniques. In: Proceedings 16th Symposium on Boundary Layers and Turbulence. American Meteorological Society, Portland, Maine, USA.
- Griffith, D.W.T., Leuning, R., Denmead, O.T., Jamie, I.M., 2002. Airland exchanges of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O measured by FTIR spectrometry and micrometeorological techniques. Atmos. Environ. 36, 1833–1842.
- Harper, L.A., Denmead, O.T., Freney, J.R., Byers, F.M., 1999. Direct measurements of methane emissions from grazing and feedlot cattle. J. Anim. Sci. 77, 1392–1401.

- Houweling, S., Dentener, F., Lelieveld, J., 2000. Simulation of preindustrial methane to constrain the global source strength of natural wetlands. J. Geophys. Res. 105, 17243–17255.
- IPCC, 1996. Houghton et al. (Ed.), IPCC Guidelines for National Greenhouse Gas Inventories, vol. 3: Greenhouse Gas Inventory Reference Manual. IPCC WG1 Technical Support Unit, Hadley Centre, Meteorological Office, Bracknell, United Kingdom.
- IPCC, 2000. Good practice guidance for uncertainty management in national greenhouse gas inventories. Penman et al. (Ed.), IPCC National Greenhouse Gas Inventories Programme Technical Support Unit, Institute for Global Environmental Strategies, Hayama, Kanagawa, Japan.
- Johnson, D.E., Ward, G.M., 1996. Estimates of animal methane emissions. Environ. Monit. Assess 42, 133–141.
- Johnson, D.E., Johnson, K.A., Ward, G.M., Branine, M.E., 2000a. Ruminants and other animals. In: Khalil (Eds.), Atmospheric Methane: Its Role in the Global Environment. Springer-Verlag, Berlin, pp. 112–133.
- Johnson, K., Huyler, M., Westberg, H., Lamb, B., Zimmerman, P., 1994. Measurement of methane emissions from ruminant livestock using a SF<sub>6</sub> tracer technique. Environ. Sci. Technol. 28, 359–362.
- Johnson, K.A., Johnson, D.E., 1995. Methane emissions from cattle. J. Anim. Sci. 73, 2483–2492.
- Johnson, K.A., Westberg, H.H., Lamb, B.K., Kincaid, R.L., 2000b. An overview of the SF<sub>6</sub> technique for measuring methane emissions from cattle. In: Proceedings Second International Conference on Methane Mitigation, Novosibirsk, Russia, pp. 103–108.
- Judd, M.J., Kelliher, F.M., Ulyatt, M.J., Lassey, K.R., Tate, K.R., Shelton, I.D., Harvey, M.J., Walker, C.F., 1999. Net methane emissions from grazing sheep. Global Change Biol. 5, 647–657.
- Kaharabata, S.K., Schuepp, P.H., Desjardins, R.L., 2000. Esimating methane emissions from dairy cattle housed in a barn and feedlot using an atmospheric tracer. Environ. Sci. Technol. 34, 3296– 3302.
- Lassey, K.R., Ulyatt, M.J., 2000. Methane emission by grazing livestock: A synopsis of 1000 direct measurements. In: van Ham, et al. (Eds.), Non-CO<sub>2</sub> Greenhouse Gases: Scientific Understanding, Control and Implementation. Kluwer, Dordrecht, The Netherlands, pp. 101–106.
- Lassey, K.R., Ulyatt, M.J., Martin, R.J., Walker, C.F., Shelton, I.D., 1997. Methane emissions measured directly from grazing livestock in New Zealand. Atmos. Environ. 31, 2905–2914.
- Lassey, K.R., Walker, C.F., McMillan, A.M.S., Ulyatt, M.J., 2001. On the performance of SF<sub>6</sub> permeation tubes used in determining methane emission rates from grazing livestock. Chemosphere: Global Change Sci. 3, 367–376.
- Lassey, K.R., Pinares-Patiño, C.S., Ulyatt, M.J., 2002. Methane emission by grazing livestock: Some findings on emission determinants. In: van Ham, et al. (Eds.), Non-CO<sub>2</sub> Greenhouse Gases: Scientific Understanding, Control Options and Policy Aspects. Millpress, Rotterdam, The Netherlands, pp. 95–100.
- Laubach, J., Kelliher, F.M., 2004. Measuring methane emission rates of a dairy cow herd by two micrometeorological techniques. Agric. For. Meteorol. 125, 279–303.
- Laubach, J., Kelliher, F.M., 2005. Measuring methane emission rates of a dairy cow herd (II): results from a backward-Lagrangian stochastic model. Agric. For. Meteorol. 129, 137–150.
- Leuning, R., Baker, S.K., Jamie, I.M., Hsu, C.H., Klein, L., Denmead, O.T., Griffith, D.W.T., 1999. Methane emission from free-ranging sheep: a comparison of two measurement methods. Atmos. Environ. 33, 1357–1365.

- Lockyer, D.R., Jarvis, S.C., 1995. The measurement of methane losses from grazing animals. Environ. Pollut. 90, 383–390.
- Marik, T., Levin, I., 1996. A new tracer experiment to estimate the methane emissions from a dairy cow shed using sulfur hexafluoride (SF<sub>6</sub>). Global Biogeochem. Cycles 10, 413–418.
- McCaughey, W.P., Wittenberg, K., Corrigan, D., 1997. Methane production by steers on pasture. Can. J. Anim. Sci. 77, 519–524.
- McCaughey, W.P., Wittenberg, K., Corrigan, D., 1999. Impact of pasture type on methane production by lactating beef cows. Can. J. Anim. Sci. 79, 221–226.
- Mosier, A.R., Duxbury, J.M., Freney, J.R., Heinemeyer, O., Minami, K., Johnson, D.E., 1998. Mitigating agricultural emissions of methane. Clim. Change 40, 39–80.
- Moss, A.R., Jouany, J.-P., Newbold, J., 2000. Methane production by ruminants: its contribution to global warming. Ann. Zootechnol. 49, 231–253.
- Murray, B.R., Bryant, A.M., Leng, R.A., 1976. Rates of production of methane in the rumen and large intestine of sheep. Br. J. Nutr. 36, 1–14.
- Norris, K.H., Barnes, R.F., Moore, J.E., Shenk, J.S., 1976. Predicting forage quality by infrared reflectance spectroscopy. J. Anim. Sci. 43, 889–897.
- Pavao-Zuckerman, M.A., Waller, J.C., Ingle, T., Fribourg, H.A., 1999. Methane emissions of beef cattle grazing tall fescue pastures at three levels of endophyte infestation. J. Environ. Qual. 28, 1963– 1969.
- Pinares-Patiño, C.S., Baumont, R., Martin, C., 2003a. Methane emissions by Charolais cows grazing a monospecific pasture of timothy at four stages of maturity. Can. J. Anim. Sci. 83, 769–777.
- Pinares-Patiño, C.S., Ulyatt, M.J., Lassey, K.R., Barry, T.N., Holmes, C.W., 2003b. Persistence of differences between sheep in methane emission under generous grazing conditions. J. Agric. Sci. 140, 227–233.
- Pinares-Patiño, C.S., Ulyatt, M.J., Lassey, K.R., Barry, T.N., Holmes, C.W., 2003c. Rumen function and digestion parameters associated with differences between sheep in methane emissions when fed chaffed lucerne hay. J. Agric. Sci. 140, 205–214.
- Pinares-Patiño, C.S., Ulyatt, M.J., Waghorn, G.C., Lassey, K.R., Barry, T.N., Holmes, C.W., Johnson, D.E., 2003d. Methane emission by alpaca and sheep fed on lucerne hay or grazed on pastures of perennial ryegrass/white clover or birdsfoot trefoil. J. Agric. Sci. 140, 215–226.
- Prather, M., Derwent, R., Ehhalt, D., Fraser, P., Sanhueza, E., Zhou, X., 1995. Other trace gases and atmospheric chemistry. In: Houghton, et al. (Eds.), Climate Change 1994: Radiative Forcing of Climate Change. Cambridge University Press, Cambridge, UK, Chapter 2, pp. 73–126.
- Prather, M., Ehhalt, D., Dentener, F., Derwent, R., Dlugokencky, E., Holland, E., Isaksen, I., Katima, J., Kirchhoff, V., Matson, P., Midgley, P., Wang, M., 2001. Atmospheric chemistry and greenhouse gases. In: Houghton, et al. (Eds.), Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, UK, pp. 239– 287.
- Prigge, E.C., Varga, G.A., Vicini, J.L., Reid, R.L., 1981. Comparison of ytterbium chloride and chromium sesquioxide as fecal indicators. J. Anim. Sci. 53, 1629–1633.
- Primavesi, O., Frighetto, R.T.S., Dos Santos Pedreira, M., de Lima, M.A., Berchielli, T.T., Barbosa, P.F., 2004. Metano entérico de

bovinos leiteiros em condições tropicais brasileiras. Pesq. Agropec. Bras. 39, 277–283 (In Portuguese with English Abstract).

- Ramaswamy, V., Boucher, O., Haigh, J., Hauglustaine, D., Haywood, J., Myhre, G., Nakajima, T., Shi, G.Y., Solomon, S., 2001. Radiative forcing of climate change. In: Houghton, et al. (Eds.), Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, UK, pp. 349–416.
- Scheehle, E.A., Kruger, D., in press. Global anthropogenic methane and nitrous oxide emissions. Energy J.
- Scheehle, E.A., Irving, W.N., Kruger, D., 2002. Global anthropogenic methane emissions. In: van Ham, et al. (Eds.), Non-CO<sub>2</sub> Greenhouse Gases: Scientific Understanding, Control Options and Policy Aspects. Millpress, Rotterdam, The Netherlands, pp. 257–262.
- Schimel, D., Alves, D., Enting, I.G., Heimann, M., Joos, F., Raynaud, D., Wigley, T.M.L., Prather, M., Derwent, R., Ehhalt, D., Fraser, P., Sanhueza, E., Zhou, X., Jonas, P., Charlson, R., Rodhe, H., Sadasivan, S., Shine, K., Fouquart, Y., Ramaswamy, V., Solomon, S., Srinivasan, J., Albritton, D., Derwent, R., Isaksen, I., Lal, M., Wuebbles, D., 1996. Radiative forcing of climate change. In: Houghton, et al. (Eds.), Climate Change 1995: The Science of Climate Change. Cambridge University Press, Cambridge, UK, (Chapter 2), pp. 65–131.
- Schuepp, P.H., Leclerc, M.Y., MacPherson, J.I., Desjardins, R.L., 1990. Footprint prediction of scalar fluxes from analytical solutions of the diffusion equation. Boundary-Layer Meteor. 50, 355–373.
- Simpson, I.J., Blake, D.R., Rowland, F.S., Chen, T.-Y., 2002. Implications of the recent fluctuations in the growth rate of tropospheric methane. Geophys. Res. Lett. 29, 1479, doi:10.1029/2001GL014521.
- Ulyatt, M.J., Baker, S.K., McCrabb, G.J., Lassey, K.R., 1999. Accuracy of SF<sub>6</sub> tracer technology and alternatives for field measurements. Aust. J. Agric. Res. 50, 1329–1334.
- Ulyatt, M.J., Lassey, K.R., Shelton, I.D., Walker, C.F., 2002a. Seasonal variation in methane emission from dairy cows and breeding ewes grazing ryegrass/white clover pasture in New Zealand. N. Z. J. Agric. Res. 45, 217–226.
- Ulyatt, M.J., Lassey, K.R., Shelton, I.D., Walker, C.F., 2002b. Methane emission from dairy cows and wether sheep fed subtropical grass-dominant pastures in mid summer in New Zealand. N. Z. J. Agric. Res. 45, 227–234.
- Ulyatt, M.J., Lassey, K.R., Shelton, I.D., Walker, C.F., 2005. Methane emission from sheep grazing four pastures in late summer in New Zealand. N. Z. J. Agric. Res. 48, 385–390.
- Waghorn, G.C., Tavendale, M.H., Woodfield, D.R., 2002. Methanogenesis from forages fed to sheep. Proc. N. Z. Grassland Assoc. 64, 167–171.
- Westberg, H., Lamb, B., Johnson, K.A., Huyler, M., 2001. Inventory of methane emissions from U.S. cattle. J. Geophys. Res. 106, 12633–12642.
- Woodward, S.L., Waghorn, G.C., Ulyatt, M.J., Lassey, K.R., 2001. Early indications that feeding *Lotus* will reduce methane emissions from ruminants. Proc. N. Z. Soc. Anim. Prod. 61, 23–26.
- Woodward, S.L., Waghorn, G.C., Lassey, K.R., Laboyrie, P.G., 2002. Does feeding sulla (*Hedysarum coronarium*) reduce methane emissions from dairy cows? Proc. N. Z. Soc. Anim. Prod. 62, 227–230.
- Wratt, D.S., Gimson, N.R., Brailsford, G.W., Bromley, A.M., Lassey, K.R., Bell, M.J., 2001. Estimating regional methane emissions from agriculture using aircraft measurements of concentration profiles. Atmos. Environ. 35, 497–508.