Animal Feed Science and Technology 170 (2011) 265–276

Contents lists available at SciVerse ScienceDirect

Animal Feed Science and Technology

journal homepage: www.elsevier.com/locate/anifeedsci

Background matters with the SF_6 tracer method for estimating enteric methane emissions from dairy cows: A critical evaluation of the SF_6 procedure

S.R.O. Williams[a,*], P.J. Moate[a], M.C. Hannah[a], B.E. Ribaux[a], W.J. Wales[a], R.J. Eckard[a,b]

[a] Department of Primary Industries, Ellinbank Centre, 1301 Hazeldean Road, Ellinbank 3821, Australia
[b] Melbourne School of Land and Environment, University of Melbourne, Parkville 3010, Australia

ARTICLE INFO

Article history:
Received 14 June 2011
Received in revised form 18 August 2011
Accepted 21 August 2011

Keywords:
Sulfur hexafluoride
Dairy cows
Housing
Tracer gas

ABSTRACT

Since its inception, the sulfur hexafluoride (SF_6) tracer technique for estimating ruminal methane (CH_4) emissions has undergone several refinements. One key divergence from the original description of the method has been its use with animals housed indoors. Given the different molecular masses of CH_4 (16 g/mol) and SF_6 (146 g/mol) it is possible that these gases could disperse and accumulate differentially within animal houses. The purpose of this study was to examine and compare the ambient outdoor concentrations of CH_4 and SF_6 with background concentrations measured during indoor experiments. A literature search found 52 scientific papers which reported use of the SF_6 tracer technique with 17 reporting use indoors, 31 outdoors and 4 were desktop reviews or an uncommon implementation of SF_6 as a tracer. Complete details of where background concentrations were measured, and how they were used, were not provided in any of the papers. Concentrations of CH_4 in open air at Department of Primary Industries, Ellinbank, Victoria, Australia (38°14′S, 145°56′E) were variable at about 2.6 μmol/mol which was about 50% higher than those of 1.73 measured at the Cape Grim Baseline Air Pollution Station (40°41′S, 144°41′E). This difference was thought to be due to the CH_4 emissions from cows in the Ellinbank area. During the same period, the SF_6 concentration in open air at DPI Ellinbank increased from 4.9 pmol/mol in November 2003 to 6.8 pmol/mol in March 2010. This trend was similar to those measured at Cape Grim. Inside the DPI Ellinbank animal house, which is open to atmosphere on 2 sides, the accumulation of gases during experiments varied in a quadratic manner along the line of feeding stalls with the CH_4 concentration ranging from 4 to 10 μmol/mol and SF_6 ranging from 4 to 26 pmol/mol. Vertically, background concentration of CH_4 trended from 4.6 μmol/mol at 225 mm above the floor to 12.3 μmol/mol at 1775 mm while SF_6 trended from 8.2 to 14.9 pmol/mol at the same heights. Calculations showed that use of inappropriate background values to calculate CH_4 emissions could lead to discrepancies ranging from −6.2% to +0.8% on an emission of 500 g CH_4/cow/d. Thus, we recommend use of distributed sentinel canisters for monitoring accumulation of gases within animal houses, and using local background values to correct CH_4 and SF_6 measurements from individual animals.

Abbreviations: BW, body weight; DPI, Department of Primary Industries Victoria Australia; ppm, μmol/mol; ppt, pmol/mol; RSD, residual standard deviation.

* Corresponding author. Tel.: +61 3 5624 2222; fax: +61 3 5624 2248.
E-mail address: richard.williams@dpi.vic.gov.au (S.R.O. Williams).

0377-8401/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.anifeedsci.2011.08.013

1. Introduction

The sulfur hexafluoride (SF_6) tracer technique developed by Zimmerman (1993) has become the preferred method for estimating ruminal CH_4 emissions from large numbers of animals. The technique, which was first used in ruminant nutrition research by Johnson et al. (1994), relies on use of a permeation tube to release a small amount of SF_6 at a known rate once it has been dosed into the reticulo-rumen. The released SF_6 mixes with rumen fermentation gas and acts as a tracer for ruminally produced CH_4. A capillary tube mounted on a head-harness and attached to an evacuated container is used to collect a representative sample of the rumen fermentation gases from the animal as well as the SF_6 tracer eructated during the course of a day. In the method described by Johnson et al. (1994), the known daily release rate (Q_{SF_6}, mol/d) of SF_6 from the permeation tube and the ratio of the mixing ratios (sometimes designated as concentrations) of CH_4 and SF_6 in the collected sample of gas were used to estimate the daily rate of CH_4 emissions (Q_{CH_4}, mol/animal/d, Eq. (1)). Note, in Eq. (1), the concentration of CH_4 and SF_6 must be expressed in the same units, for example pmol/mol (ppt).

$$Q_{CH_4} = Q_{SF_6} \frac{[CH_4]}{[SF_6]} \tag{1}$$

Johnson et al. (1994) reported that they determined Q_{SF_6} by placing each permeation tube in a "temperature bath" at 39 °C and the permeation tubes were "routinely weighed until an accurate loss rate was determined". They validated their method by comparison with data obtained from animals in open-circuit respiration chambers. In their original experiments, Johnson et al. (1994) used the technique on individual beef cattle held outdoors in individual pens.

Recent uses of the SF_6 technique (*e.g.*, Grainger et al., 2010; Morgavi et al., 2008; Ramirez-Restrepo et al., 2010) have evolved from the technique described by Johnson et al. (1994). Lassey et al. (1997) were more thorough than Johnson et al. (1994) in their description of the technique, being apparently the first to explicitly report that the CH_4 emission calculation should use the concentrations of CH_4 and SF_6 "in excess of background". The intent of this correction procedure is stated explicitly in Eq. (2), where the M subscript indicates a measured sample, and the BG subscript indicates a background concentration.

$$R_{CH_4} = R_{SF_6} \frac{[CH_4]_M - [CH_4]_{BG}}{[SF_6]_M - [SF_6]_{BG}} \times \frac{MW_{CH_4}}{MW_{SF_6}} \times 1000 \tag{2}$$

In Eq. (2), R_{CH_4} is the calculated emission rate of ruminal CH_4 (g/animal/d), R_{SF_6} is the measured release rate of SF_6 from the permeation tube (mg/d), MW_{CH_4} is the molecular mass of methane (16), and MW_{SF_6} is the molecular mass of SF_6 (146). For convenience, the concentrations of CH_4 are generally expressed in μmol/mol (ppm) and the concentrations of SF_6 in ppt, and this is the case in Eq. (2). The factor of 1000 in Eq. (2) is a unit converter taking into account the disparate units for $[CH_4]$ in ppm, $[SF_6]$ in ppt, and R_{SF_6} in (mg/d) so that R_{CH_4} will have the desired units of g/d.

Since the research of Johnson et al. (1994) and Lassey et al. (1997) there have been more than 50 scientific publications where the background correction shown in Eq. (2) was used to make estimates of CH_4 emissions (*e.g.*, Boadi et al., 2002b; Grainger et al., 2010; Pinares-Patiño et al., 2007a). Contrasting with this is the work by Pavao-Zuckerman et al. (1999) who monitored background concentrations of CH_4 and SF_6 but deemed them insignificant so did not include them in their calculations. Several researchers mention using a background correction as per Johnson et al. (1994) (*e.g.*, Cavanagh et al., 2008; Chaves et al., 2006; Pinares-Patiño et al., 2008c) yet Johnson et al. (1994) made no mention of doing so, only stating that they "measured CH_4 and SF_6 concentrations".

Although Lassey et al. (1997) specified that background corrections should be made for CH_4 and SF_6, they did not describe how or where the background measurement should be made, nor did they report the background concentrations of CH_4 and SF_6 during their experiment. Furthermore, a cursory examination of a few recent investigations using the SF_6 technique (*e.g.*, Foley et al., 2009b; Holtshausen et al., 2009; Ramirez-Restrepo et al., 2010), revealed that although many reported that background samples were collected, none reported background concentrations of CH_4 and SF_6 nor the complete procedure for collecting these background samples, such as the location relative to the animals.

The most significant modification to the method of Johnson et al. (1994) has been the use of the technique indoors (*e.g.*, Boadi and Wittenberg, 2002; Molano et al., 2008; Vlaming et al., 2008). In many cases it is not clear from the scientific literature dealing with the SF_6 technique in housed animals where the background gas samples were collected in relation to the location of the animals. If background gas samples were collected indoors, it is possible that structural characteristics and ventilation of buildings could influence measured background concentrations of these gases compared to those measured outdoors and this, in turn, will influence calculated values of CH_4 emissions. Given that CH_4 (16 g/mol) is much lighter than SF_6 (146 g/mol) it is possible that these gases could disperse and accumulate differentially within animal houses, yet investigations of this possibility have not been reported.

The research described here consists of three main efforts: (1) a comprehensive survey of the scientific literature to ascertain all the known issues related to background concentrations of CH_4 and SF_6 with respect to the SF_6 technique for measuring enteric CH_4 emissions from ruminants, (2) documentation of outdoor, background CH_4 and SF_6 concentrations in a rural area and (3) experiments to investigate the horizontal and vertical distribution of CH_4 and SF_6 inside an animal house.