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Enteric methane emission rates determined by the SF_6 tracer technique: Temporal patterns and averaging periods

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ABSTRACT

The sulphur hexafluoride (SF_6) tracer technique has been widely applied to determine CH_4 emission rates by ruminants since its development in the mid-1990s. It remains the only viable method for determining emission rates from individual grazing animals. Essential parts of the method for each participating animal are pre-insertion into the rumen of a source of SF_6 with known release rate and breath sample collections near the nose and mouth for CH_4 and SF_6 analysis. Breath samples are accumulated over an 'averaging period' of usually 24 h to yield estimates of CH_4 emissions. As a tracer, SF_6 is biologically inert and has a very low detection limit (*i.e.*, 10^{-12}), enabling release rates of a few tens of μl/h to be sustained for many months by an initial SF_6 charge of ~1 g. Any departure from a uniform SF_6 collection rate, such as through SF_6 interactions in the digestive tract, could introduce variability into the inferred CH_4 emission rate, which has the potential to explain reports of higher variability in CH_4 emission rates estimated with this technique compared with whole animal chamber techniques. Our study examined SF_6 and CH_4 excretion rates for their variability using a novel automated gas chromatography system that isolated and analysed 20 min breath samples collected successively for 6 d from each of 9 housed sheep. We found that that SF_6 was not excreted into the breath stream at a uniform rate, but its daily pattern of excretion was strongly correlated with that of CH_4, suggesting that some SF_6 is retained within the digestive tract and later ventilated with eructated gases following feeding. Methane emission rates can be estimated for different averaging periods through different combinations of the 20 min data. Methane emission rate estimates for each sheep are independent of averaging period between 3 h and 6 d, although inter-period variability is highest for averaging periods less than 1 d. Improved understanding of the SF_6 tracer technique supports it as a reliable unbiased estimator of enteric CH_4 emission rate in ruminants.

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Abbreviations: ECD, electron capture detector; FID, flame ionisation detector; GC, gas chromatography.

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1. Introduction

Methane emitted by the world's farmed ruminant livestock accounts for about one quarter of all anthropogenic CH_4 emissions, typically estimated at 80–90 Tg/yr of a total of ~350 Tg/yr (Denman et al., 2007; Lassey, 2007, 2008). This makes ruminant emitted CH_4 important in radiative forcing of climate, and a target for abatement measures, which have so far had limited success (Beauchemin et al., 2008).

Development of CH_4 emission abatement measures requires detailed study of emission determinants, such as feed composition and intake level, in conjunction with techniques for accurate and precise measurement of emissions from animals. Two types of techniques are in wide use for such measurements, being enclosure and tracer techniques. The most common form of enclosure technique employs an open circuit respiration chamber with the CH_4 emission rate from the enclosed animal determined from the difference in CH_4 content between inflow and outflow air (Johnson and Johnson, 1995). In a variation of the enclosure technique, the animal's head is enclosed in a head box, recognising that enteric CH_4 is emitted through the mouth and nose. The tracer technique, pioneered by Johnson et al. (1994), uses sulphur hexafluoride (SF_6) as the tracer of choice released from an intra-ruminal pre-calibrated permeation tube. Methane emission rates at the nose and mouth are estimated from integrated breath samples (*i.e.*, respired plus eructed gases) that are collected and analysed for CH_4 and SF_6, usually by off line gas chromatography (GC). The SF_6 tracer technique, now widely used (Lassey, 2007), is the only technique available to measure emissions from individual animals while grazing. The technique can also be used in studies to link feed characteristics with CH_4 emissions, wherein animals would normally be fed under controlled conditions to avoid uncertainties of feed intake.

In their seminal paper on the SF_6 tracer technique, Johnson et al. (1994) reported good agreement between the technique and chamber measurements, citing an illustrative comparison for a single heifer. Several researchers have subsequently compared enclosure and tracer techniques in more detail (Boadi et al., 2002; Grainger et al., 2007; McGinn et al., 2006; Pinares-Patiño et al., 2008a, this issue), applying both techniques to the same animals fed the same diet at the same intake level. A common finding of these comparisons is that estimates of average CH_4 emission rates over several days do not differ between techniques, but that intra- and inter-animal variability in emission rates can be appreciably higher for the tracer technique. This suggests that the tracer technique introduces additional variability without an accompanying bias in emission estimates.

The introduction of unbiased variability is explainable by SF_6 being a non-ideal tracer of CH_4 on a daily time scale, recognising that CH_4 generated in the rumen responds to feeding pattern (Grainger et al., 2007), whereas it is assumed that SF_6 is released into the rumen at a steady rate. Technique induced variability could be due to different sites of CH_4 and SF_6 generation/release, and of their respective excretions, differences between CH_4 and SF_6 dynamics within the digestive tract and fluctuations in SF_6 permeation rate from its host tube.

Using four ewes fed lucerne chaff, Murray et al. at (1976) deduced that about 87% of CH_4 was generated in the rumen, and 13% in the hind-gut; with respective excretion sites being eructation (95%) plus respiration (5%), and respiration (89%) plus flatus (11%). Thus, at least 98% of CH_4 is excreted into the breath stream and emitted at the nose and mouth. However, the proportion of feed digested in the hind gut is likely to vary with diet nutritional quality and quantity. A ruminal source of SF_6 cannot ideally trace all these routes. McGinn et al. (2006) found that CH_4 emission rate estimated by the SF_6 and head box techniques differed most, but not significantly, for those diets and feeding regimes with more hind gut digestion, consistent with CH_4 in flatus increasing with the proportion of hind-gut digestion. However, Boadi et al. (2002) reported more inter-animal variability in CH_4 emission rates for cattle measured by tracer than when measured using head boxes, neither of which detect flatus derived emissions.

A common view is that any SF_6 that migrates down the digestive tract is absorbed in the blood and respired from the lungs, as Murray et al. (1976) also found for CH_4. Such SF_6 would be detected at the nose and mouth indistinguishably from SF_6 excreted in eructed gas. There are some lines of evidence to support this view. Levitt and Levitt (1973) demonstrated that both SF_6 and CH_4 infused at different sites within the rat's digestive system are nearly completely recovered via pulmonary excretion (*i.e.*, >90% within 8 h), but diffusion and absorption rates of SF_6 are lower than those of CH_4. More detailed study of gas exchange in the pulmonary airways of sheep support the view that blood borne inert gases with a range of solubilities readily out-gas in the lung (Schimmel et al., 2004).

Recognising that typical SF_6 permeation rates of ~1–5 mg/d equate to ~6–32 μl/h (with 1 μl≡1 mm^3), the fate of such tiny quantities (c.f. 10^5-fold higher sheep eructation rates ~1 L CH_4/h) would be controlled by rumen disposition and could easily become sequestered in the digestive tract. Influences could include the role of rumen gases as sparging agents (*i.e.*, bubbled gases that strip poorly soluble gases out of solution; Law et al., 1994), and physical disturbances caused by feeding or physical activity.

The release rate of SF_6 is controlled by the physics of permeation (Namiešnik, 1984). Driven by an internal SF_6 vapour pressure of ~3.2 MPa, the permeation rate would remain constant in a fixed temperature environment for at least a few months (Lassey et al., 2001). Thus any variation in the rate of SF_6 emission would be a result of either variable rumen temperature or variable SF_6 dynamics within the digestive tract. Rumen temperature variations may result from feeding and drinking patterns, according to the temperature dependence of permeation (~7%/°C: Namiešnik, 1984). However, it seems unlikely that such variations could induce an appreciable or prolonged perturbation to the permeation rate, although a mean rumen temperature different from the calibration temperature of 39 °C could potentially introduce bias. While little is known about SF_6 dynamics within the digestive system, it is unlikely to be an ideal tracer for CH_4 that is generated during