Postprint: Detection of Methane Emissions from Ruminant Animals Using Infrared Spectroscopy

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Abstract

The establishment of a precise evaluation system for monitoring methane emissions from ruminants has consistently been challenging. One key reason is the inadequacy of conventional methods for detecting ruminant methane emissions; furthermore, detection instruments are susceptible to external environmental influences, making it difficult to ensure their accuracy and sensitivity. Currently, infrared spectroscopy detection technology is being widely applied to ruminant methane emission monitoring. Several state-of-the-art detection methods based on this technology, such as Laser Methane Detection (LMD), Fourier Transform Infrared Spectroscopy (FTIR), the GreenFeed (GF) system, and the Portable Automated Open-path Gas Quantification System (GQS), have also been widely employed. Compared with traditional methods, infrared spectroscopy detection technology possesses certain advantages. Based on existing literature, this paper discusses the current application status and future prospects of infrared spectroscopy detection methods in ruminant methane emissions from the perspectives of principles, reliability, and comparison with other common methods, aiming to provide a reference for the precise detection of methane emissions from ruminants.

Full Text

Preamble

Methane Emission of Ruminants Determined by Infrared Spectroscopy

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Abstract: Establishing an accurate assessment system for monitoring methane emissions from ruminants has remained challenging, primarily because conventional detection methods are inadequate and detection instruments are susceptible to external environmental influences, making it difficult to guarantee their accuracy and sensitivity. Currently, infrared spectroscopy detection technology is being widely applied to measure ruminant methane emissions. Several novel detection methods based on infrared spectroscopy, such as laser methane detection (LMD), Fourier transform infrared spectroscopy (FTIR), the GreenFeed (GF) system, and portable auto-open circuit gas quantification systems (GQS), have also gained widespread adoption.

Compared with traditional methods, infrared spectroscopy detection technology offers distinct advantages. Based on existing literature, this paper discusses the principles, reliability, and comparative performance of infrared spectroscopy detection methods for ruminant methane emissions, examining both current applications and future prospects to provide a reference for accurately detecting methane emissions from ruminants.

Keywords: ruminant; methane emission; infrared spectroscopy
Classification Codes: S823; S826

Introduction

Greenhouse gas emissions [including carbon dioxide (CO_2) and methane (CH_4)] constitute a major factor driving climate change. The global warming potential of CH_4 is 21 times that of CO_2. According to statistics, anthropogenic CH_4 emissions contribute 22.9% of total greenhouse effects, ranking second only to CO_2 [1]. Agricultural greenhouse gas emissions account for 58% of global anthropogenic emissions, with enteric CH_4 emissions from livestock representing 32% of agricultural greenhouse gas emissions [2]. China’ s enteric CH_4 emissions from animals constitute approximately 10% of the global total, of which ruminant CH_4 emissions account for 98.7% of animal enteric CH_4 emissions [3]. From an energy perspective, CH_4 emissions from ruminants represent a loss of 2%-12% of gross energy intake [4]. A survey of U.S. dairy production from 1944 to 2007 revealed that the energy loss from CH_4 per kilogram of milk produced exceeded half of the total energy consumption in dairy production, indicating that CH_4 production imposes certain limitations on dietary utilization efficiency [5]. In recent years, accurately detecting CH_4 emissions from ruminants has become a research focus worldwide. Currently, two major challenges exist: first, establishing a precise detection system is difficult; second, obtaining efficient and reliable data is complicated by multiple influencing factors [6].

Infrared spectroscopy technology for CH_4 detection offers high accuracy and sensitivity, along with advantages such as a large dynamic measurement range, rapid response time, and minimal interference from other gases [7-8]. This review summarizes the current status of infrared spectroscopy applications in detecting ruminant CH_4 emissions, providing new references for precise measurement.

1. Infrared Spectroscopy Methods and Principles for Detecting Ruminant CH_4 Emissions

Currently, spectral absorption methods represent one of the primary approaches for CH_4 emission detection, with infrared spectroscopy being widely applied. Technologies including portable indium gallium arsenide (InGaAs) laser methane detectors (LMD), Fourier transform infrared spectroscopy (FTIR), GreenFeed (GF) systems, and portable auto-open circuit gas quantification systems (GQS) are all based on this principle.

Spectral absorption methods are founded on the Lambert-Beer law. Most diatomic or polyatomic molecules exhibit characteristic absorption spectra in the infrared region. CH_4 has two strong absorption bands near 3.3 and 7.7 m, with other infrared bands formed through combinations of these fundamentals and two additional inactive fundamentals, creating rich overtone and combination bands. Based on the Lambert-Beer law, infrared spectroscopy quantifies CH_4 concentration by detecting changes in transmitted light intensity [8-9].

LMD employs wavelength modulation spectroscopy using a helium-neon laser beam as the excitation source. The InGaAs second-harmonic detection signal aligns with CH_4’ s two strong infrared absorption bands (3.3 and 7.7 m), while the gas absorption line shape approximates the Lorentzian formula, enabling CH_4 concentration calculation [10]. LMD can rapidly adjust temperature and drive current, offering high sensitivity for detecting average CH_4 concentrations between two points within several meters [11-12].

The FTIR principle relies on the unique near-infrared absorption spectrum of different molecules. A dual-source interferometer converts single-pass near-infrared light into double-pass light, creating two different optical path differences that generate interference patterns through Fourier transform mathematical calculations. Since absorbance in the Lambert-Beer law directly correlates with peak height and area in the spectrum, unknown sample concentrations can be determined by comparing peak characteristics with those of known concentration standards [13]. FTIR can typically detect various components and gas concentrations. Standard benchtop research-grade FTIR instruments achieve resolutions of 1\$×$10^{\{-4\}-25cm}${-1}\$, making them suitable for climate change research [14-15].

2.1 Research Status

Infrared spectroscopy technology has been extensively applied to detect CH_4 emissions in atmospheric environments, coal mines, natural gas production, livestock housing, soil respiration, and human respiration, demonstrating high precision and sensitivity. For instance, atmospheric CH_4 concentrations are low (approximately 1.80 mL/m^3) and uniformly mixed, making regional concentration differences difficult to detect. Using high-resolution (0.02 cm^{-1}) FTIR to retrieve atmospheric CH_4 concentration variations, Tian et al. [16] found retrieval errors below 1% with average daily variation less than 0.02 mL/m^3. Luo et al. [17] reported an infrared gas detection system for coal mine applications with a detection range of 0–1.00$×10^{6}$ mL/m^3 and measurement error below 2%, demonstrating high precision suitable for mining safety applications. In natural gas production safety monitoring, Li et al. [18] studied infrared detection of CH_4 and hydrogen sulfide (H_2S) with detection limits of 1.09$×10^{{2}−1.31×10}{3}$ mL/m^3, meeting safety requirements. Excessive harmful gases [such as CH_4, H_2S, and ammonia (NH_3)] in livestock housing severely affect animal health. Childers et al. [19-20] reported that open-path FTIR enables high-precision trace detection of harmful gases (0–3.00$×10^{-3}$ mL/m^3) with errors below 3%. For human respiration detection, infrared spectroscopy simultaneously measures H_2 and CH_4 with low error ($±2×10^{2}$ mL/m^3 [21]. Davidson et al. [22] also noted that infrared absorption spectroscopy offers rapid response, continuous measurement, and high sensitivity for soil respiration CO_2 and CH_4 emissions, with detection limits of 0.11–1.08 mL/m^3.

Although infrared spectroscopy has been studied and applied across various fields for CH_4 detection [8], its application in ruminant CH_4 emission measurement emerged relatively late. Chagunda et al. [23] first applied LMD to ruminant CH_4 detection in 2009. Madsen et al. [24] first reported in 2010 that FTIR could serve as a rapid, reliable, and low-cost method for ruminant CH_4 detection with good potential for broader adoption. Also in 2010, the GF system (Figure 1 [Figure 1: see original paper][25]) was successfully implemented for ruminant CH_4 emission detection. This system installs infrared sensors near the animal' s head to effectively detect exhalation flow velocity and CH_4/CO_2 concentrations. When the animal' s head is properly positioned, a radio frequency identification (RFID) reader automatically scans its ear tag for individual data sampling and analysis, ultimately calculating 24-hour CH_4 emissions [26].

More recently, Dorich et al. [27] reported a novel GQS feeding trough detection device (Figure 2 [Figure 2: see original paper]) that employs infrared spectroscopy for automatic real-time CO_2 and CH_4 detection during cattle feeding. In recent years, infrared spectroscopy applications for ruminant CH_4 detection have been frequently reported internationally, while domestic research remains limited and requires further development.

2.2.1 Applicable for Detecting Differences Between Dietary Treatments

In practical production, evaluating how dietary nutritional value affects ruminant CH_4 emissions requires direct measurement using infrared spectroscopy to enable multi-replicate detection across different dietary treatments. This facilitates optimization and screening of palatable diets that reduce CH_4 emissions. Using FTIR, Haque et al. [28] investigated the effects of high-energy versus conventional lactation diets on dairy cow CH_4 emissions, finding average daily CH_4 emissions of 13.9–14.2 g per kilogram of dry matter intake (DMI). However, CH_4 emissions showed a linear positive correlation with both intake and energy-corrected milk yield. Guyader et al. [29] reported infrared detection of CH_4 emissions from Holstein cows fed diets with a 50:50 concentrate-to-forage ratio, measuring 308.6 g/d. Adding linseed oil reduced emissions by 17% (252.7 g/d), nitrate addition reduced emissions by 19% (238.1 g/d), and combined addition reduced emissions by 32% (206.8 g/d), with minimal variation in dietary digestibility before and after supplementation.

2.2.2 Advantage of Not Disturbing Animal Behavior

Conventional CH_4 detection methods constrain animal behavior, such as respiration chambers and ventilated hoods, which prevent natural behavior studies and yield unrealistic CH_4 emission data. A major advantage of LMD is its non-interference with ruminant behavior. Chagunda et al. [12] reported significant effects of activity status on CH_4 emissions in Holstein cows (n=110): CH_4 concentration during rumination (279.0 mL/m^3) was 1.4 times and 1.1 times higher than during free movement (202.9 mL/m^3) and feeding (262.2 mL/m^3), respectively. Lactating cows (326.2 mL/m^3) also showed significantly higher emissions than dry cows (203.8 mL/m^3). Subsequently, Chagunda et al. [30] found that CH_4 emissions during drinking (368.0 mL/m^3) and feeding (284.0 mL/m^3) were significantly higher than during free movement (106.0 mL/m^3). In contrast, emissions during free movement and sleeping (186.0 mL/m^3) were lower.

2.2.3 Capability for Real-Time CH_4 Emission Detection

Infrared spectroscopy enables real-time CH_4 emission detection, quantifying emissions within specific time intervals. Goopy et al. [31] reported using GQS to detect CH_4 emissions from sheep, finding that measurements taken 2 hours post-feeding could predict 50%-82% of average daily CH_4 emissions. Bickell et al. [32] used infrared spectroscopy to examine the relationship between hourly feed intake and CH_4 emissions in sheep, finding a correlation coefficient of 0.22 under ad libitum feeding conditions. The correlation between 24-hour cumulative CH_4

emissions (measured hourly) and daily emissions was 0.89, with significant differences between hourly emission rates. Ricci et al. [33] used LMD to analyze CH_4 emissions 3-5 hours post-feeding in sheep, finding a correlation coefficient of 0.92 with DMI, higher than respiration chamber results (0.79). Recently, Ou et al. [34] reported an infrared gas detection capsule placed in the animal' s stomach (Figure 3 [Figure 3: see original paper]) capable of real-time gas detection, currently able to measure CH_4, CO_2, and H_2 concentrations per unit time, though gas flow rate detection remains under development. Real-time precise detection is a prerequisite for studying CH_4 mitigation during metabolic processes, as it provides metabolic quantification for individual animals and forms the basis for monitoring CH_4 emissions during metabolism.

2.2.4 Applicability for Grazing Conditions

The non-invasive nature and real-time detection capability of infrared spectroscopy provide a foundation for grazing condition applications. Madsen et al. [24] used FTIR to measure CH_4 emissions from dairy cows in barns, obtaining CO_2 emissions of 3,880.00 mL/m^3 and CH_4 emissions of 241.80 mL/m^3. The corrected CH_4/CO_2 ratio in exhaled breath was 0.08 (a method based on proportional detection). This ratio showed small fluctuation ranges, remained stable against external environmental influences, and demonstrated that FTIR could serve as a convenient, reliable, and economical method suitable for grazing conditions.

Jones et al. [36] reported open-path FTIR applications under grazing conditions, investigating the effect of residual feed intake (the difference between actual intake and predicted intake based on performance and maintenance requirements) on CH_4 emissions from Angus beef cattle. Low residual feed intake animals emitted 0.34 g CH_4 per kilogram of live weight (LW) daily, while high residual feed intake animals emitted 0.46 g/kg LW. Grobler et al. [37] found that under grazing conditions, forage type affected CH_4 emissions across cattle breeds. Jersey cows grazing natural pasture showed significantly lower CH_4 emissions during rumination (25.8 mL/m^3) compared to Bonsmara (32.7 mL/m^3) and Nguni cattle (30.6 mL/m^3), as measured by LMD. However, no significant differences existed between breeds when fed sorghum silage. McGinn et al. [38] used open-path LMD under grazing conditions, measuring average CH_4 emissions of 141.0 g/head/day with a recovery rate of 77%.

In summary, infrared spectroscopy technology ensures accurate detection data and meets specific CH_4 measurement requirements, demonstrating substantial potential for grazing condition applications.

3. Comparison Between Infrared Spectroscopy and Common Detection Methods

Common ruminant CH_4 emission detection methods include respiration chambers, ventilated hoods, sulfur hexafluoride (SF_6) tracers, micrometeorological techniques, prediction equations, and in vitro gas production methods. These methods possess distinct advantages and disadvantages, as summarized in Table 2 .

As shown in Table 2, common ruminant CH_4 detection methods each have merits and demerits, making it difficult to identify a method that is both low-cost and stable. Infrared spectroscopy technology, however, offers both cost-effectiveness and stability, as demonstrated through the following comparisons.

Open-circuit respiration chambers provide stable CH_4 emission monitoring, with Hellwing et al. [44] reporting average recovery rates of 101% for CO_2 and 99% for CH_4. However, respiration chambers incur high costs. Goopy et al. [31] compared open-circuit respiration chambers with GQS for sheep CH_4 emissions, finding a correlation coefficient of 0.71 between methods and a repeatability (intra-individual correlation coefficient) of 0.88 for respiration chambers. Chagunda et al. [40] analyzed dairy cow CH_4 emissions, reporting a positive correlation coefficient of 0.80 between LMD and indirect open-circuit respiration calorimetry, confirming LMD’ s feasibility and cost-effectiveness.

The SF_6 method is commonly used for dynamic CH_4 emission detection under grazing conditions. Hammond et al. [26] compared SF_6 (186.0 g/d) with GF system measurements (164.0 g/d) in Holstein cows, obtaining a correlation coefficient of 0.60. Dorich et al. [27] reported that GQS showed lower measurement variability (coefficient of variation: 14.1%-22.4%) and higher correlation with DMI (r=0.42) compared to SF_6 (coefficient of variation: 16.0%-111.0%; r=0.17). Furthermore, SF_6 is a greenhouse gas with 23,900 times the warming potential of CO_2 and an atmospheric lifetime of 3,200 years, presenting significant limitations [45]. Clearly, infrared spectroscopy holds advantages for grazing condition monitoring.

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4. Challenges and Future Prospects for Infrared Spectroscopy Detection of Ruminant CH_4 Emissions

Current challenges remain in infrared spectroscopy detection of ruminant CH_4 emissions. For instance, gas detection capsules, LMD, FTIR, and GF systems cannot yet measure gas flow rates or CH_4 concentrations during eructation [34,40,46-47]. In barn environments, Barrancos et al. [46] used open-path FTIR to measure annual CH_4 and NH_3 emissions of 167.0 kg/head and 1.3 kg/head, respectively—values that differ substantially from the European Air Pollution Emission Inventory (100.0 kg/head CH_4 and 8.7 kg/head NH_3 annually), possibly due to micrometeorological factors (airflow velocity, wind direction) affecting measurements. Such factors may also influence FTIR measurements under grazing conditions. GF systems can encounter issues such as improper animal head positioning relative to RFID readers [47]. During spectral modeling for CH_4 concentration retrieval, errors exceeding specific thresholds may occur, primarily from assuming constant temperature, humidity, and pressure parameters—issues requiring resolution in subsequent software development [16]. These factors can all affect CH_4 emission detection accuracy.

Recent studies have addressed these challenges. Wu et al. [48] used an improved FTIR system to analyze an artificial reference cow (ARC) device that simulates bovine exhalation, inhalation, and eructation, finding only a 2.8% difference between ARC and actual cow CH_4 emissions, suggesting ARC could develop into a practical reference system. As reported by Ou et al. [34], gas detection capsules placed in the stomach could avoid micrometeorological interference and RFID positioning issues under grazing conditions, providing design insights for precise internal CH_4 measurement. Future work should focus on optimizing model input values during spectral modeling to improve computational accuracy. Thus, infrared spectroscopy technology for ruminant CH_4 detection requires continued improvement.

Looking ahead, infrared spectroscopy technology shows excellent prospects for precise ruminant CH_4 emission measurement. From a holistic mitigation perspective, Wall et al. [49] proposed that if emissions can be accurately measured, breeding programs selecting for low-emission animals could reduce emissions per kilogram of animal product. Widespread infrared spectroscopy application could facilitate detection of difficult-to-measure phenotypic traits, reducing ecological footprints and improving production efficiency [50].

In conclusion, infrared spectroscopy technology demonstrates substantial potential for accurately detecting ruminant CH_4 emissions, offering advantages of non-disturbance, real-time monitoring, and particular suitability for grazing conditions. Practical applications must consider factors such as animal respiration rate, average CH_4 concentration during eructation, and micrometeorological influences. Furthermore, integrating infrared spectroscopy with other CH_4 detection methods could guide selection of low-CH_4 emission traits and animal breeds through genome-wide association studies.

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