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Measurement of greenhouse gas emissions from agricultural sites using open-path optical remote sensing method

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Improved characterization of distributed emission sources of greenhouse gases such as methane from concentrated animal feeding operations require more accurate methods. One promising method is recently used by the USEPA. It employs a vertical radial plume mapping (VRPM) algorithm using optical remote sensing techniques. We evaluated this method to estimate emission rates from simulated distributed methane sources. A scanning open-path tunable diode laser was used to collect path-integrated concentrations (PICs) along different optical paths on a vertical plane downwind of controlled methane releases. Each cycle consists of 3 ground-level PICs and 2 above ground PICs. Three- to 10-cycle moving averages were used to reconstruct mass equivalent concentration plum maps on the vertical plane. The VRPM algorithm estimated emission rates of methane along with meteorological and PIC data collected concomitantly under different atmospheric stability conditions. The derived emission rates compared well with actual released rates irrespective of atmospheric stability conditions. The maximum error was 22 percent when 3-cycle moving average PICs were used; however, it decreased to 11% when 10-cycle moving average PICs were used. Our validation results suggest that this new VRPM method may be used for improved estimations of greenhouse gas emission from a variety of agricultural sources.

Keywords: Open-path tunable diode laser (OP-TDLAS), vertical radial plume mapping (VRPM), agricultural sites, greenhouse gas, methane.

Introduction

Agricultural sites such as rice paddies, feedlots and treatment lagoons are significant emission sources of the greenhouse gas methane.[1–3] Although accurate assessment of gas emission from agricultural sites is very important, the accuracy depends strongly on the methods employed and the surrounding environments.[4] Greenhouse gas emissions from a point source such as an animal house with mechanical ventilation could be adequately estimated by multiplying ventilation rates and gas concentrations at the fan outlet. However, estimation of greenhouse gas emission from distributed sources such as treatment lagoons, treatment wetlands, land spread of manure, and feedlots requires more complicated methods such as chamber and various micrometeorological methods. Furthermore, these more sophisticated methods do not provide higher reproducibility of measurements. For example, different measurement methods taken in over-lapping days for a treatment lagoon produced widely different volatilization rates.[5] Harper[6] reported more than an order of magnitude difference in ammonia, methane and nitrous oxide emissions from swine lagoons using these methods. Clearly, new reliable methods to measure emission of these trace gases are urgently needed.

Among various micrometeorological methods, Wilson et al.[7] concluded that the integrated horizontal flux method (IHF) proved to be the most satisfactory, followed by the backward Lagrangian stochastic method (BLS). Laubach and Kelliher[8] also preferred the IHF technique because it does not rely on the similarity assumptions. Although the IHF technique estimates the emission rate by simply determining the difference of the integrated mass fluxes from up- and down-wind sides, successful application of the IHF technique requires accurate concentration profile information. Unfortunately, these values continuously changes in both time and height. Venkatram[9] suggested the use of a dispersion model to better estimate the concentration profile for the IHF.

Instead of using the dispersion model, the new USEPA's path-integrated optical remote sensing (PI-ORS) method

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utilizes open-path Fourier transform infrared spectroscopy (OP-FTIR) or open-path tunable diode absorption spectroscopy (OP-TDLAS) to obtain multiple downwind path-integrated concentrations (PIC) at different heights.[10–12] A bivariate Gaussian smooth basis function minimization (SBFM) approach is used to reconstruct a crosswind-smoothed mass-equivalent concentration map in a vertical plane from the downwind PIC data. Once the mass-equivalent gas concentrations are mapped in the vertical plane perpendicular to wind direction, the IHF method is employed to calculate the total mass rate of the gas passing the vertical plane This method is currently listed under the USEPA Technology Transfer Network Emission Measurement Center Category C, which may be considered for use in federally enforceable State and local programs once approved by an EPA Regional State Implementation Plans (SIP) process.[19] The vertical radial plume mapping (VRPM) software developed by Arcadis Inc. (Research Triangle Park, NC) automatically calculates the emission rates based on PIC data, wind speed and direction information using above PI-ORS method.

The objective of this research is to investigate the accuracy of the new PI-ORS method as a potential method for directly measuring methane emissions from agricultural sites.

Materials and methods

VRPM methodology for ground-level emission

When the emission source exists at the ground level such as treatment lagoons, spray field, and animal houses, the following bivariate Gaussian SBFM approach is used to reconstruct a crosswind-smoothed mass-equivalent concentration map in a vertical plane from the downwind PIC data.[13] First, using the ground-level PIC data, the values of σ_y and m_y of Equation 1 are estimated by fitting the data into a univariate Gaussian function via minimization of the sum of squared errors (SSE).

$$SSE(B, m_y, \sigma_y) = \sum_i \left\{ PIC_i - \frac{B}{\sqrt{2\pi\sigma_y^2}} \int_0^{r_i} \exp\left[-\frac{1}{2}\left(\frac{r - m_y}{\sigma_y} \right)^2 \right] dr \right\}^2 \quad (1)$$

where

B = area under the one-dimensional Gaussian distribution (integrated concentration),
r_i = path length of the i beam (m),
m_y = peak location in y direction, i.e., right angle to the wind direction (m),
PIC_i = measured PIC value of i beam (ppmm),
σ_y = standard deviation in horizontal direction (m).

With the values of σ_y and m_y estimated from the ground-level PIC data, the values of A and σ_z of Equation 2 are then estimated from fitting the above-ground PIC data into the bivariate Gaussian function.

$$SSE(A, \sigma_z) = \sum \left(PIC_i - \frac{A}{2\pi\sigma_y\sigma_z} \times \int_0^{r_i} \exp\left\{ -\frac{1}{2}\left[\frac{(r\cos\theta_i - m_y)^2}{\sigma_y^2} + \frac{(r\sin\theta_i)^2}{\sigma_z^2} \right] \right\} dr \right)^2 \quad (2)$$

where

A = normalizing coefficient adjusting for the peak value of the bivariate surface,
θ_i = vertical angle of beam i from the ground,
σ_z = standard deviation in vertical direction (m).

Once all the parameters for the bivariate Gaussian function are found for a specific run, the VRPM procedure calculates the mass-equivalent concentration values for every square elementary unit (4 × 4 m) in a vertical plane. Then, the VRPM procedure computes and integrates the elementary unit flux over the entire vertical plane with corresponding wind speed data.

USDA-ARS OP-TDLAS system setup

The path integrated optical remote sensing (PI-ORS) system employed at the USDA-ARS Coastal Plains Research Center (Florence, SC) consisted of a OP-TDLAS (GasFinder2.0 for CH_4, Boreal Laser Inc., Spruce Grove, Canada) mounted on an automatic positioning device (Model 20 Servo, Sagebrush Technology, Inc.), five retroreflectors (Boreal Laser), two cup anemometers (CS800-L Climatronics Wind Speed and Direction Sensor, Campbell Scientific, Logan, UT) mounted on a 10-m weather station mast (at two heights, 2 m and 10 m), and a laptop computer (IBM Thinkpad R60). Three retroreflectors were placed 1 m above the ground using tripods (RR1, RR2, and RR3) and two were mounted on a tower at elevations of 5 (RR4) and 10 m (RR5), respectively (Fig. 1 and Table 1).

The distance from the GasFinder2.0 to each retroreflector was measured with a 100-m tape. These 5 retroreflectors were positioned in such a way that the vertical plane containing the reflectors was approximately perpendicular to the mean wind direction. The automatic positioning device directed the infrared beam of the GasFinder2.0 to each retroreflector sequentially; the GasFinder2.0 collected about 12–15 downwind PIC data at each retroreflector before moving to next position. Each cycle consists of 3 ground-level PICs and 2 above ground PICs. The coordinates of the retroreflectors were entered into the VRPM software (Arcadis Inc., NC), and the PIC data for each retroreflector was saved for reconstructing plane-integrated concentration maps.

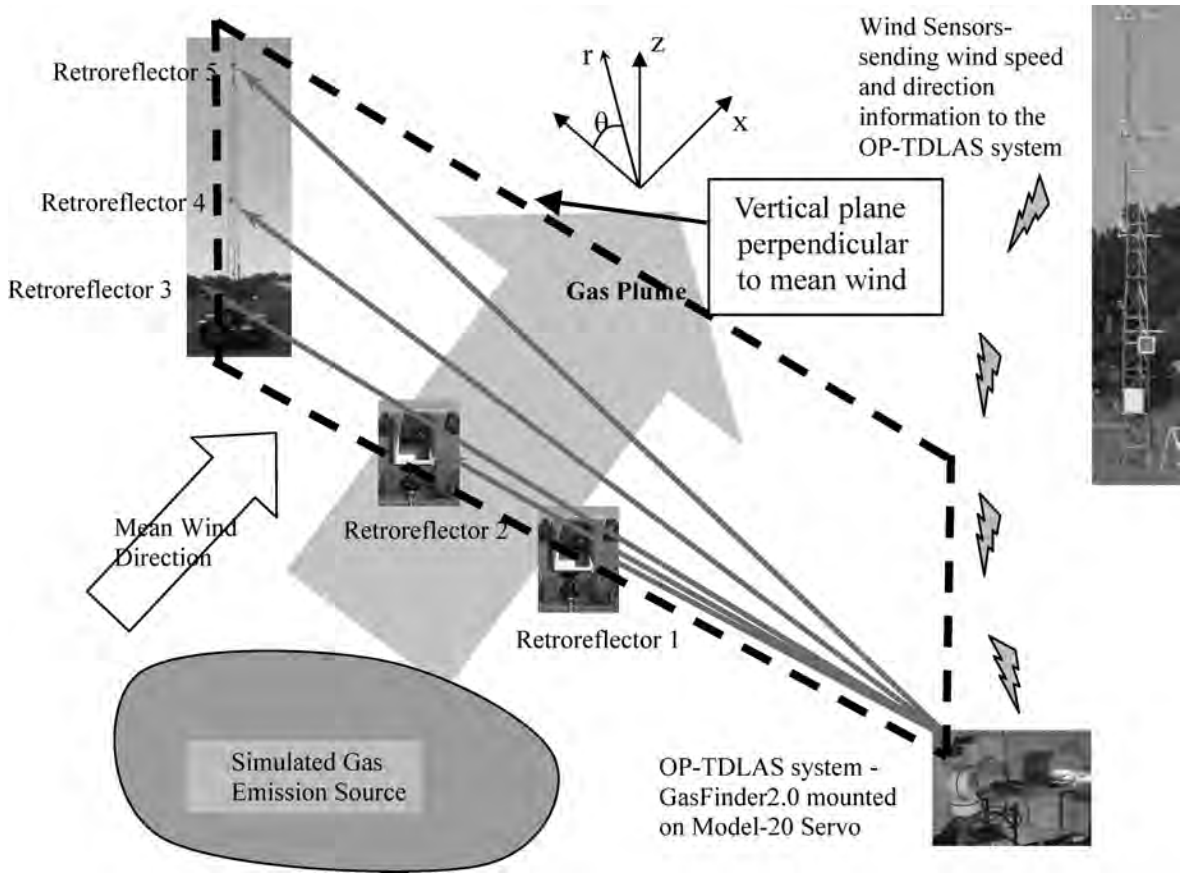

Fig. 1. Simulated field configuration and OP-TDLAS beam geometry.

The automatic positioning device eliminates the need for multiple OP-TDLAS systems and substantially reduces the total costs. The communication between the wind sensors and the base computer was achieved through the RF401 spread spectrum data radio/modems (Campbell Scientific, Logan, UT). The VRPM software used 3- to 10-cycle moving average PIC data to reconstruct the plane-integrated concentration maps on the vertical plane; it calculated the mass rates of gas passing through the vertical plane using the wind information.

Study site and simulated gas emission source

The validation study was conducted on a grass plain at the USDA-ARS Coastal Plains Soil, Water & Plant Research Center at Florence, SC (N 34°14.741′ and W 79° 48.605').

Table 1. Retroreflector positions.

	Ground distance from GasFinder 2.0 (m)					
Test Date	*RR1 (H = 1 m)**	*RR2 (H = 1 m)*	*RR3 (H = 1 m)*	*RR4 (H = 5 m)*	*RR5 (H = 10 m)*	*Simulated emission sources*
9/22/06	77	172	244	200	200	3.1 m × 3.1 m Square (Perforated PVC pipe network)
10/12/06	100	200	309	284	284	3.1 m × 3.1 m Square (Perforated PVC pipe network)
10/31/06	64	138	219	200	200	3.1 m × 3.1 m Square (Perforated PVC pipe network)
6/4/07	43	86	132	134	134	20 m Diameter Circle (Soaker hose)

*H: Height above ground.

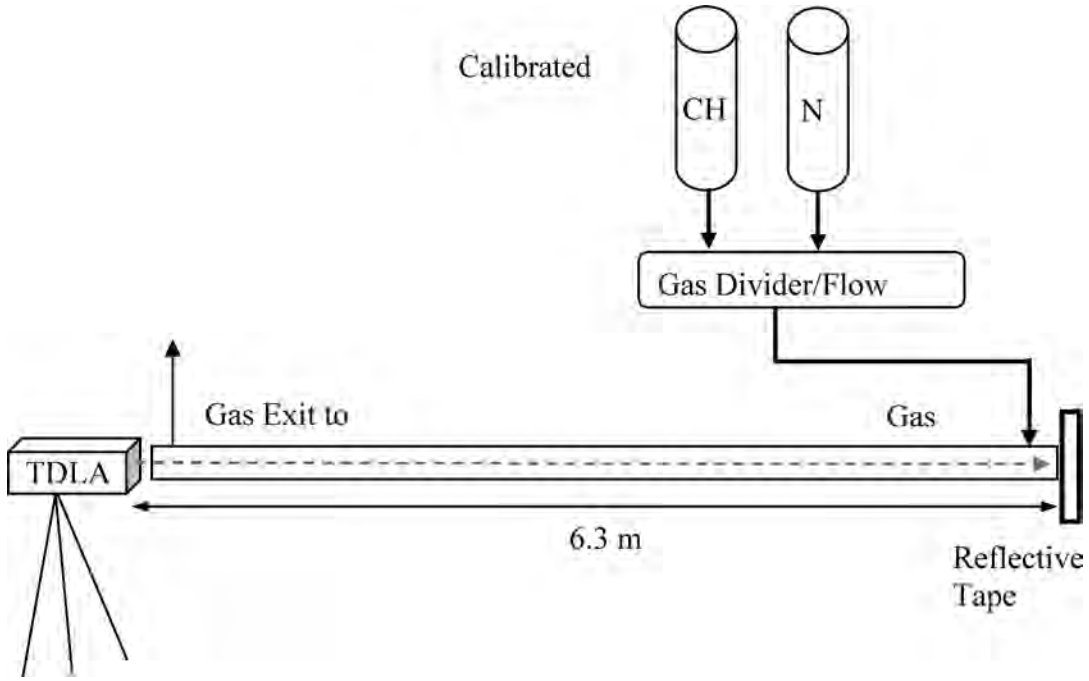

Fig. 2. TDLAS calibration system.

The study site was covered with short grass (typically less than 0.1 m). At the time of the validation study, most of soybeans and peanuts planted under two center pivot irrigation systems (270 m in diameter) had been harvested. The predominant wind direction was NE-SW, which was along the length of the grass plain (approximately 700 m in length). The atmospheric stability conditions during the validation studies were characterized with the stability ratio, a simplified approximation of the Richardson number.[14–17] A micrometeorological station constructed at the validation study site provided wind speed and temperature information for calculating the stability ratio.[18]

Three different distributed emission sources were simulated using 20 cm perforated PVC pipe (3.1 m × 3.1 m square) and six to seven soaker hoses stretching out from a gas cylinder to make star-shaped circular emission sources with a diameter of 20 m. These simulated emission sources were placed about 30 m upwind from the center of the VRPM vertical plane. The background CH_4 around the study site was about 1.7 ppm. Volumetric flow rates (30 – 52 L min^{-1}) of the CH_4 gas (Linde Gas, 93% purity) were released during the validation studies through two air flow meters (Gilmore Instruments). The masses of CH_4 gas released were measured with a floor scale (CW11-2EO model, Ohaus) periodically.

Accuracy of TDLAS

The GasFinder2.0 has an internal reference cell with a known concentration of CH_4 that is traceable to the NIST standards. It is programmed to recalibrate itself automatically every 60 measurements (i.e., about 60 sec). However,

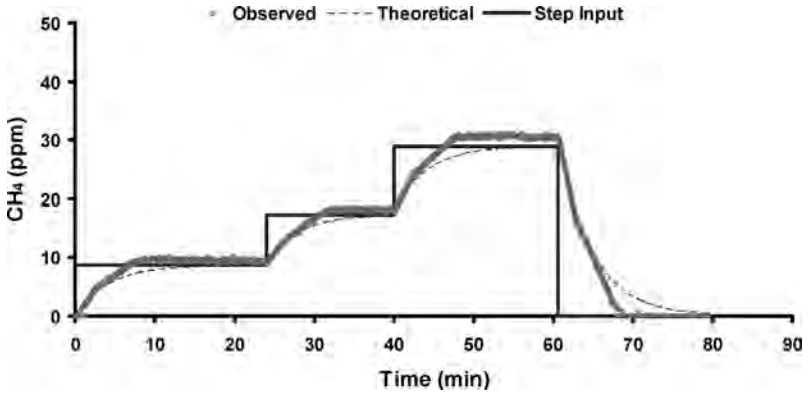

Fig. 3. Comparison of theoretical and observed CH_4 concentration in the PVC pipe.

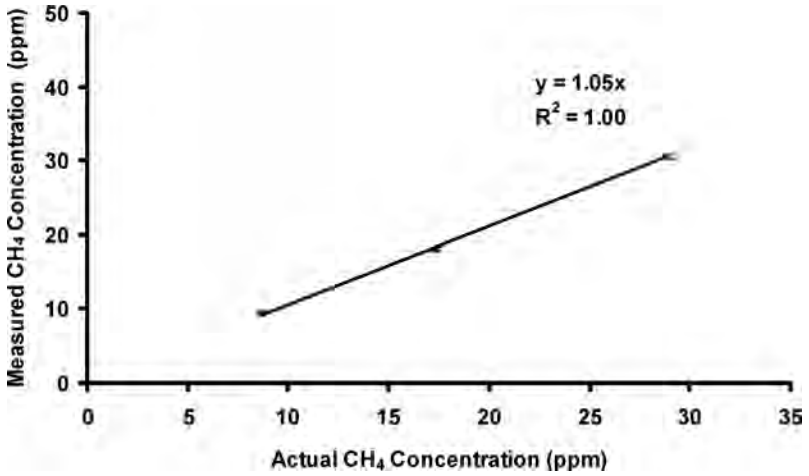

Fig. 4. Accuracy of the GasFInder2.0.

the physical integrity of the reference cell is not known. The accuracy of the PIC may be adversely affected if there is any leak in the reference cell. Because the PI-ORS method depends strongly on accurate measurements of PICs, we decided to verify the accuracy of the GasFinder2.0 by measuring PIC through a 6.3-m PVC pipe (5.1 cm diameter) in which 3 l min^{-1} of calibrated gas with known concentration of CH_4 (28.9 ppm, National Welders Supply Co., Inc., NC) was entered and exited at the both ends of the PVC pipe via Teflon tubing (ID 0.32 cm) as shown in Figure 2.

The two ends of the PVC pipe were sealed with transparent plastic film. Initially the PVC was filled with ultra pure nitrogen gas (National Welders Supply Co., Inc.). At time zero, a gas mixture of 30% CH_4+ 70% N_2 was introduced at one end of the PVC tube. The composition of the gas mixture was changed to 60% CH_4+ 40% N_2, 100 % CH_4, and 100 % N_2 at t = 24 min, 40 min, and 61 min, respectively. Although the actual CH_4 concentration inside the PVC pipe varies with position along the pipe, the system can be treated as a completely mixed reactor with a uniform concentration having the path length-averaged CH_4 concentration measured by the GasFinder2.0. The concentration profile of a completely mixed reactor subjected to step input of CH_4 can be derived by performing a mass balance around the PVC pipe.

$$\frac{dC}{dt}V = QC_{in} - QC \tag{3}$$

where

C = length-averaged concentration in the PVC pipe (ppm),
C_{in} = influent concentration (ppm),
Q = volumetric flow rate of gas (l min^{-1}),
t = time (min),
V = volume of the PVC pipe (l).

The length-averaged concentration in the PVC pipe can be calculated by integrating Equation 3 with an initial concentration (C_o).

$$C = C_o \cdot e^{-t/\tau} + C_{in}(1 - e^{-t/\tau}) \tag{4}$$

where

C_o = initial concentration in the PVC pipe (ppm),
τ = gas residence time (min)
$= V/Q$

Table 2. Validation study results.

Test date	*Actual release rate (g/s)*	*Total No. VRPM estimates*	*No. of VRPM estimates with CCF > 0.8 –10 < WD < 25*	*Avg. WD from normal (deg)*	*Average Wind speed (m/s)*	*VRPM* emission estimates (g/s)*	*Error #*
9/22/06	1.09	38	25	9 ± 16	2.9 ± 0.5	1.33 ± 0.52	22 %
10/12//06	0.96	41	41	11 ± 7	3.9 ± 0.8	1.09 ± 0.41	14 %
10/31/06	0.92	39	22	24 ± 10	3.8 ± 0.5	1.06 ± 0.28	16 %
6/4/07	0.70	12	12	15 ± 5	4.6 ± 0.4	0.61 ± 0.22	−13 %

*3 cycle moving average.
(VRPM estimate – actual rate) × 100/actual rate.

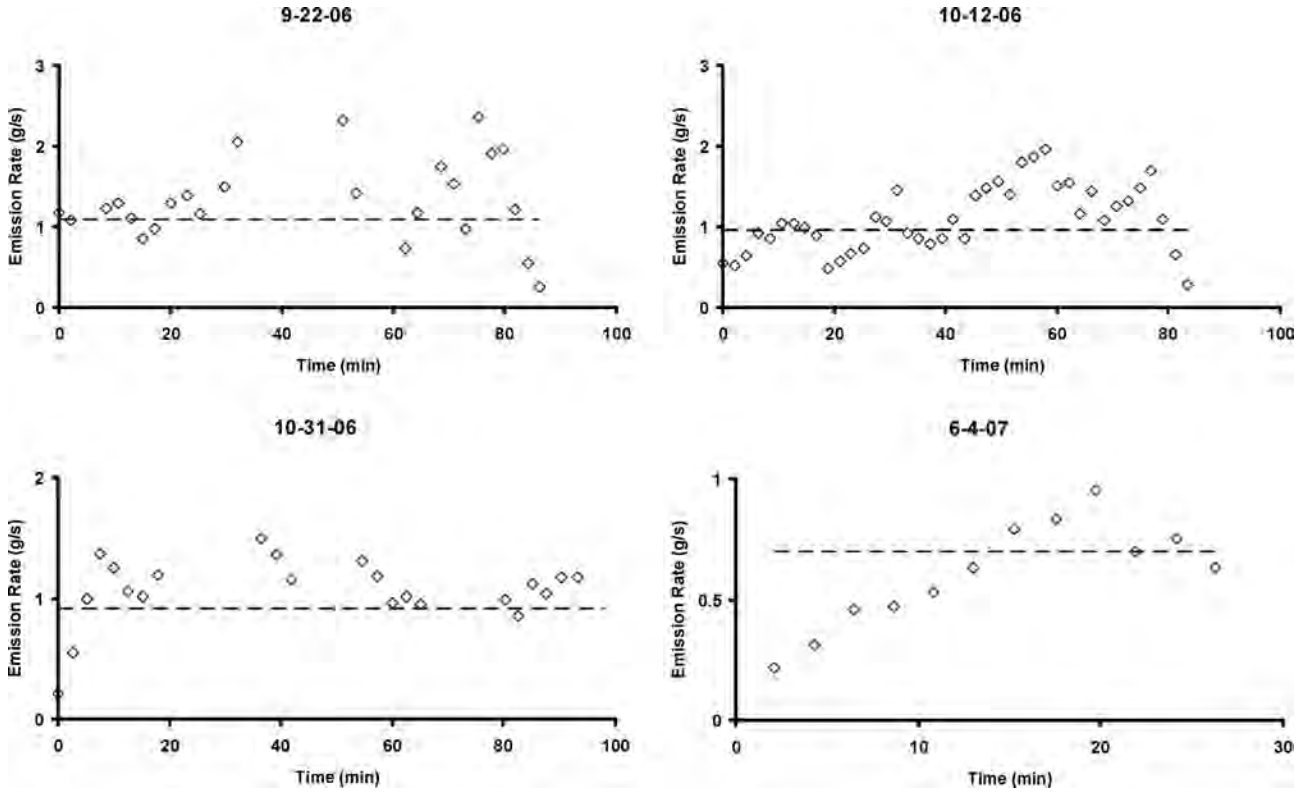

Fig. 5. VRPM estimates and average CH_4 release rates for the 4 validation studies (◇ VRPM estimates, —- Average release rate).

Results and discussion

The GasFinder2.0 slightly over estimated the CH_4 concentration flowing inside the PVC pipe. As shown in Figure 3, the GasFinder2.0 measured concentration followed closely to the following theoretical curve obtained from performing mass balance around the PVC pipe (Equation 4). Assuming the CH_4 concentration in the PVC pipe became practically the same as the influent concentration after 3 gas residence times (i.e., 13 min), the measured CH_4 concentrations were compared with the actual influent concentrations (Fig. 4). The concentration measured by the GasFinder2.0 was about 5% higher than the expected concentration. The calibrated gas from the National Welders Supply Co., Inc. has the relative error of 2% traceable to the NIST standards. The interference caused by the scattered light from the transparent plastic film to which the GasFinder2.0 was closely placed might caused this small deviation of concentration. In this paper, it was assumed that the small deviation of the measured concentration would not contribute significant errors in estimating emission rates using the VRPM algorithm.

The VRPM estimates of the downwind CH_4 mass rate crossing the VRPM optical plane and the corresponding CH_4 release rates based on mass difference before and after each validation studies are shown in Figure 4. The VRPM estimates were based on 3-cycle moving averages of PICs of each retroreflectors. For the plume reconstruction by the VRPM algorithm to be valid, the statistical parameter concordance correlation factor (CCF) must be greater than 0.8 as recommended by the U.S. EPA.[19] The CCF was used to represent the level of fit in reconstructing the mass-equivalent plume map based on the measured PIC data.[19] The VRPM software automatically calculates the values of CCF for each cycle. The CCF values for most of our VRPM data were higher than 0.97. However, the USEPA PI-ORS protocol also recommends only the use of PIC data during those cycles with wind direction -10° to $+25^\circ$ from perpendicular to the optical plane. When these criteria for CCF especially for the narrow wind directions were used, the number of valid VRPM estimates of CH_4 emission rates reduced (Table 2). The average wind speeds during the validation studies were from 2.9 to 4.6 m/s.

The typical VRPM estimates were slightly higher than the actual average CH_4 release rate for the duration of the study period as shown in Figure 5 and Table 2. Nonetheless, the accuracies for all four validation tests were better

Table 3. Atmospheric stability during validation studies.

Test date	*Atmospheric stability*
9/22/06	5% neutral and 95% unstable
10/12/06	100% unstable
10/31/06	100% unstable
6/4/07	25% neutral and 75% unstable

Table 4. VRPM estimates of CH_4 emission rate with different number of cycle moving averages.

Test date	*Actual release rate (g/s)*	*VRPM estimates with 3-cycle moving average PICs (g/s)*	*Error 3-cycle*	*VRPM estimates with 5-cycle moving average PICs (g/s)*	*Error 5-cycle*	*VRPM estimates with10-cycle moving average PICs (g/s)*	*Error 10-cycle*
9/22/06	1.09	1.33 ± 0.52	22 %	1.29 ± 0.42	18 %	1.17 ± 0.35	7 %
10/12//06	0.96	1.09 ± 0.41	14 %	1.12 ± 0.40	16 %	1.06 ± 0.42	11 %
10/31/06	0.92	1.086 ± 0.28	16 %	1.03 ± 0.22	11 %	0.90 ± 0.15	−2 %
6/4/07	0.70	0.61 ± 0.22	−13 %	0.62 ± 0.18	−12 %	0.63± 0.07	−10 %

than the accuracy of 25% suggested by the US EPA ORS Protocol.[19] Previously, we presented preliminary results of the first three validation tests at the 2007 ASABE national meeting. [18] The results were somewhat enigmatic. While the RPM study conducted on 9/22/06 produced a very high error of 91% for the validation, the other two study dates yielded less than 20% errors. After careful reexamination of each individual PIC data, we discovered that the VRPM software made errors in filtering quality PIC data. We located and filtered out bad data with incomplete strings, reran the VRPM calculations, and the accuracy improved significantly. The accuracies of the VRPM estimates for emission rates of three other validation testes conducted on 10/12/06, 10/31/06, and 6/4/07 were all less than 20% errors. Thoma et al.[12] suggested that the VRPM method would significantly underestimate emission rate under unstable atmospheric conditions. However, our validation results suggested that the atmospheric stability conditions did not play an important role on the accuracy of VRPM as shown in Table 3.

Real time VRPM estimates of CH_4 emission were compared with actual release rates measured frequently during the validation study conducted on 10/31/06 as shown in Figure 6. Except for the first 2 data points, it appeared that the VRPM generally overestimated the actual release rate for the first 60 min. However, the accuracy improved for the remainder of the study. We also investigated increasing the number of cycles included in the PIC moving averages as a way to improve the VRPM accuracy. This gave an improvement. Except for the data obtained on 10/12/06, the VRPM accuracy improved with the increased numbers of cycles in the PIC moving averages (Table 4). The maximum error was reduced from 22% using 3-cycle moving average PICs to 11% using 10-cycle moving average PICs.

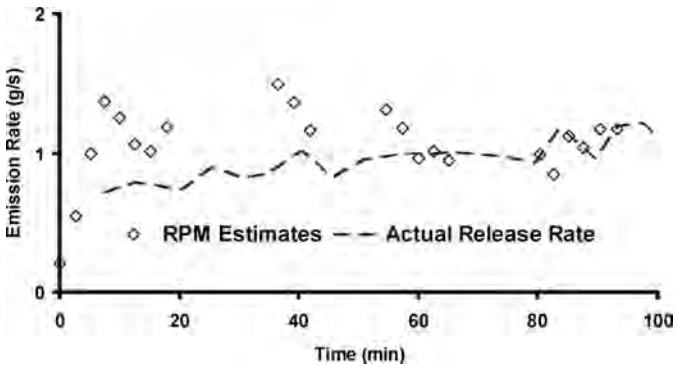

Fig. 6. Real time VRPM estimates of CH_4 emissions.

Conclusion

This paper presents the validation results of the new US EPA PI-ORS method using the OP-TDLAS system and the VRPM software. Methane was used as a target compound because of low atmospheric background concentrations and non-interference from absorption. The accuracy of the VRPM estimates of CH_4 emission rate based on 3-cycle moving averages of PIC data ranged from -13 to 22% errors. The atmospheric stability conditions did not play a significant role on the accuracy. The VRPM method was able to estimate the simulated CH_4 emission rates with less than 13% error. Increasing the number of cycles from 3 to 10 in the moving averages of PIC data improved estimate accuracy. The real-time monitoring of emission rates may be possible, but it needs further verification. The OP-TDLAS system coupled with the VRPM software could become a convenient technology for directly measuring gas emission rates from livestock operations with significantly improved accuracies relative to existing micrometeorological methods.

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