

RESEARCH ARTICLE

Modeling temperature and moisture dependent emissions of carbon dioxide and methane from drying dairy cow manure

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Abstract Greenhouse gas emissions due to biological degradation processes of animal wastes are significant sources of air pollution from agricultural areas. The major environmental controls on these microbe-induced gas fluxes are temperature and moisture content. The objective of this study was to model the effects of temperature and moisture content on emissions of CO₂ and CH₄ during the ambient drying process of dairy manure under controlled conditions. Gas emissions were continuously recorded over 15 d with paired fully automated closed dynamic chambers coupled with a Fourier Transformed Infrared gas analyzer. Water content and temperature were measured and monitored with capacitance sensors. In addition, on days 0, 3, 6, 9, 12 and 15, pH, moisture content, dissolved organic carbon and total carbon (TC) were determined. An empirical model derived from the Arrhenius equation confirmed high dependency of carbon emissions on temperature and moisture content. Results indicate that for the investigated dairy manure, 6.83% of TC was lost in the form of CO₂ and 0.047% of TC was emitted as CH₄. Neglecting the effect of temperature, the moisture contents associated with maximum gas emissions were estimated as 0.75 and 0.79 g·g⁻¹ for CO₂ and CH₄, respectively.

Keywords carbon dioxide, dairy manure, methane, moisture, temperature

1 Introduction

Annually in the USA, over 9 million dairy cows generate an estimated 226 billion kg of wet manure^[1]. Animal manure and its common use as fertilizer contribute to gaseous emissions, significantly degrading air quality to the detriment of human health and the environment^[2–4].

The challenge in assessing emission rates is complicated by the various factors affecting emissions, including type and number of animals, nutrient inputs and feeding operations, confinement conditions, manure management practices, and environmental conditions^[5]. Temperature and moisture content are the primary environmental variables influencing gas emission rates, through their influence on metabolic activity of microorganisms, manure gas diffusion, nutrient availability and redistribution^[6].

Once manure is excreted, processes of biological decomposition and formation of gaseous compounds continue. The peak emissions occur shortly after deposition and diminish significantly within a few days as manure dries^[7]. During this process, various chemical, rheological and structural changes take place inside manure piles. The organic matter is decomposed by microorganisms and enzymes under anaerobic and/or aerobic conditions, and the end products of CO₂, CH₄ and some other gases, diffuse through the manure surface crust.

Temperature and moisture content significantly impact microbial activity and gas diffusion processes driving emissions from farmyard manure and manure compost^[8–10]. For example, methanogenesis in solid manure increased with increasing temperature^[11]. Greater moisture content induces three kinds of microbial metabolism: inactive, aerobic and fermentative at low, moderate and high moisture contents, respectively, and hence influences gas emission rates^[12]. Relatively high moisture content promotes CH₄ and N₂O emissions from composting dairy manure because of the acceleration of anaerobic conditions arising from reduced oxygen supply inside compost piles^[13]. The reduced oxygen supply is generally limited by low gas diffusion into or out of porous media when the amount of air-filled pore space is below the gas percolation threshold occurring at high moisture levels^[14]. As manure dries, an increase in those anaerobically generated gasses is accompanied by an increase in the number of air-filled pores beyond the gas percolation

Received September 21, 2017; accepted December 3, 2017

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threshold. This results in a commensurate increase in oxygen supply into the manure resulting in a transition from anaerobic to aerobic activity. This optimal aerobic wet condition leads to maximum generation and emission of microbial generated gasses. Continued drying of manure decreases water availability to microbes and diminished water pathways for transport of nutrients, resulting in reduced microbial activity^[15]. In-depth quantitative knowledge about temperature and moisture effects on gas emission rates from dairy manure is still limited. Growing interest in potential sources and mitigation of greenhouse gas emissions motivates interest in understanding and predicting the simultaneous organic matter degradation, moisture content decline and biogas diffusion during the ambient drying process of dairy manure. Recent work in this regard includes characterization of dairy cow manure with respect to the hydraulic and thermal properties needed for modeling of these complex processes^[16]. The objective of this study was to model the combined effects of temperature and moisture content on CO₂ and CH₄ gas fluxes from drying fresh dairy manure over a 15 d period.

2 Materials and methods

2.1 Experimental setup and sampling

Fresh dairy manure was collected from ten lactating dairy cows at the Utah State University Caine Dairy Teaching and Research Center, Wellsville, Utah, USA. After homogenization of the collected samples, about 1 kg of manure was placed into each of 17 PVC cylinders, 203 mm in diameter and 38 mm deep, for ambient drying inside a research greenhouse. The temperature inside the greenhouse was maintained between 5 and 35°C, and fluctuated with outdoor weather conditions. Two of the 17 cylinders were placed under automated closed dynamic chambers (LI-8100-101, LI-COR Biosciences, Lincoln, NE, USA) that were connected to a portable Fourier transformed infrared (FTIR) analyzer (DX-4030, Gasmeter Technology Oy, Helsinki, Finland) for continuous measurement of CO₂ and CH₄ concentrations for flux estimation (F). Air temperature (T_a) and manure moisture content were measured with thermal resistance (PT1000, Thermometrics Corporation, Northridge, CA, USA) and electromagnetic (GS3, Decagon Devices Inc., Pullman, WA, USA) sensors, respectively. The GS3 sensor was embedded in the manure to measure continuous changes in temperature (T) and moisture content. The remaining 15 cylinders were placed on a table and prepared for destructive sampling and periodic chemical analyses.

A preliminary study of gas emissions from fresh manure revealed that CH₄ emissions fell below detection level within two weeks^[7]. This subsequent experiment was

conducted from January 4 to 19, 2013, over a 15 d period. In the beginning of the experiment, a homogenized manure sample was analyzed for initial moisture content and chemical composition. After 3, 6, 9, 12 and 15 d, triplicate samples were collected from below the crust of three manure samples to repeat laboratory analyses.

2.2 Laboratory analyses

The following parameters were analyzed for each manure sample using methods recommended by Peters et al.^[17], (1) wet manure moisture content (M_C , the mass of water relative to total wet mass, g·g⁻¹), (2) total carbon (TC), (3) dissolved organic carbon (DOC), and (4) pH.

The M_C was monitored every 5 min using a capacitance sensor (GS3) that was calibrated based on measurements on oven-dry manure (70°C for 24 h). The TC was analyzed on finely ground (< 250 μm) oven-dried samples with a combustion assay (SKALAR Primacs^{S_LC}, Skalar Analytical BV, Breda, The Netherlands). For DOC analysis, a 3 g subsample of manure was mixed with 30 mL deionized water and agitated for 1 h at room temperature. The solution was then centrifuged at 5000 r·min⁻¹ for 10 min. The supernatant was filtered with Whatman no. 542 filter paper. The extracts were immediately stored in a freezer at -20°C and thawed prior to measurements with a carbon analyzer (Phoenix 8000, Tekmar-Dohrmann, Cincinnati, OH, USA). The pH of the manure was measured with a portable pH meter (Accumet pH meter model 50, Hudson, MA, USA) in a 1:2 manure-water slurry (5 cm³ of manure with 10 mL DI water).

2.3 Gas emission measurements

Two LI-COR surface chambers were used for monitoring gas fluxes. Gas samples were sequentially drawn from the closed chambers through a valve manifold and directed to the FTIR analyzer. The concentrations of CO₂ and CH₄ were recorded in μL·L⁻¹ every 10 s. Each chamber was automatically positioned over the manure sample and sealed for 3 min during measurements of gas concentrations, repeating measurements again at 12 min intervals. During the measurements, air was circulated at a rate of 2 L·min⁻¹ between the closed chamber and the FTIR analyzer and the increase in gas concentration with time was measured. Each 3 min measurement consisted of a 1 min gas line purge prior to 2 min of data collection that was followed by a 3 min break. This cycle was repeated between the two chambers every 6 min. The MATLAB function, *robustfit*, was employed to estimate the gas fluxes from gas concentrations versus time plots. Later the gas flux units were adjusted from μL·L⁻¹·s⁻¹ to μmol·m⁻²·s⁻¹. The gas emission fluxes reported here at 12 min intervals were averaged using one 3-min flux estimate from each of the two chambers.

3 Mathematical model

3.1 Effects of temperature and moisture on gas flux

Husted^[18] found that the Arrhenius equation captured gas production affected by temperature reasonably well,

$$\ln F = -\frac{E_a}{R \cdot (273.15 + T)} + k \quad (1)$$

where F denotes the gas emission flux, E_a designates the energy of activation ($\text{kJ} \cdot \text{mol}^{-1}$), R is the gas constant ($8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$), T is the manure temperature ($^{\circ}\text{C}$) and k is an empirical constant. Maag and Vinther^[19] found that E_a changes with moisture content. Myers et al.^[20] established a second-order polynomial relationship between nitrogen mineralization and soil water content, which is acceptable for general use because it seems to work well for a wide range of soils. Here, a similar relationship was employed to express E_a as a function of moisture content, M_C :

$$E_a = B \cdot M_C^2 + C \cdot M_C + D \quad (2)$$

where B , C and D are model parameters. The combined temperature and moisture dependent gas flux relationship may be expressed as:

$$\ln F = -\frac{B \cdot M_C^2 + C \cdot M_C + D}{R \cdot (273.15 + T)} + k \quad (3)$$

or

$$F = A \cdot \exp\left[-\frac{B \cdot M_C^2 + C \cdot M_C + D}{R \cdot (273.15 + T)}\right] \quad (4)$$

where $A = \exp(k)$.

Two thirds of the gas flux estimates were randomly selected for parameterization and the remaining one third were used for validation of Eq. (4) describing the temperature and moisture effects on gas fluxes.

3.2 Cumulative emissions

The cumulative gas emission (E) is the time integration of the gas flux. Sommer and Ersbøll^[21] fitted the cumulative ammonia loss from manure slurry with the Michaelis-Menten equation:

$$E = \frac{E_{\max} \cdot t}{t + K_m} \quad (5)$$

where t is the time since start of the experiment, E_{\max} is the maximum gas loss when time approaches infinity and K_m is the time when $E = 0.5 E_{\max}$. Misselbrook et al.^[22] showed that for some cases the projected E_{\max} could not be derived with reasonable accuracy from fitting Eq. (5) to measured data. There should also be a significant difference in gas emissions if the manure is exposed to

high evaporative demand (high temperature and low humidity, i.e., higher rate of drying) versus low evaporative demand (lower temperature and higher humidity, i.e., lower rate of drying). In other words, the gas emission rate is a function of the drying rate given as:

$$F = f_F(|dM_C/dt|) \quad (6)$$

where f_F is a time-independent function. Integrating both sides of Eq. (6) with respect to time yields

$$\int_0^t F = \int_0^t f_F(|dM_C/dt|) \quad (7)$$

or equivalently,

$$E = g_F(M_{C0} - M_C) \quad (8)$$

where g_F is the integral function of f_F and M_{C0} is the initial moisture content.

To extend the applicability of Eq. (5) to various potential shapes of the cumulative emission curve, an exponential factor was introduced. Also, to account for the influence of the drying rate, time was replaced with the decrease in water content ($M_{C0} - M_C$):

$$E = E_{\max} \frac{(M_{C0} - M_C)}{(M_{C0} - M_C) + k_m \cdot \exp[-\alpha \cdot (M_{C0} - M_C)]} \quad (9)$$

where k_m and α are positive empirical parameters. Equation (9) maintains the advantages of the Michaelis-Menten equation, where when water is lost, E approaches E_{\max} . By replacing time, which can go to infinity in Eq. (5) with the decrease in water content ($M_{C0} - M_C$), it now approaches a maximum value (M_{C0}).

4 Results and discussion

After 15 d of drying, manure M_C decreased from 0.85 to 0.63 $\text{g} \cdot \text{g}^{-1}$ (Fig. 1a). Diurnal changes of manure temperature during the 15 d drying process are depicted in Fig. 1b. The manure temperature fluctuated with solar radiation and air temperature inside the greenhouse.

Figure 1c and 1d depict fluxes of CO_2 and CH_4 as a function of time and temperature. Over the first 3 d of the drying experiment there was little CO_2 and CH_4 emitted from the manure. The likely causes for these reduced initial emissions include reduced temperature resulting in inhibited microbial activity^[12]. The apparent M_C of manure was above 0.84 $\text{g} \cdot \text{g}^{-1}$ during the first 3 d, which also likely reduced gas (O_2) diffusion due to the low amount of air-filled pore space (i.e., too wet). From days 3 to 6, the emissions of CO_2 and CH_4 increased with the decline in M_C . Then CO_2 emissions remained at a high rate following diurnal temperature fluctuations, while the CH_4 flux gradually decreased until it ceased.

The parameter estimates for gas emission surface plots of CO_2 and CH_4 , with their respective temperature and

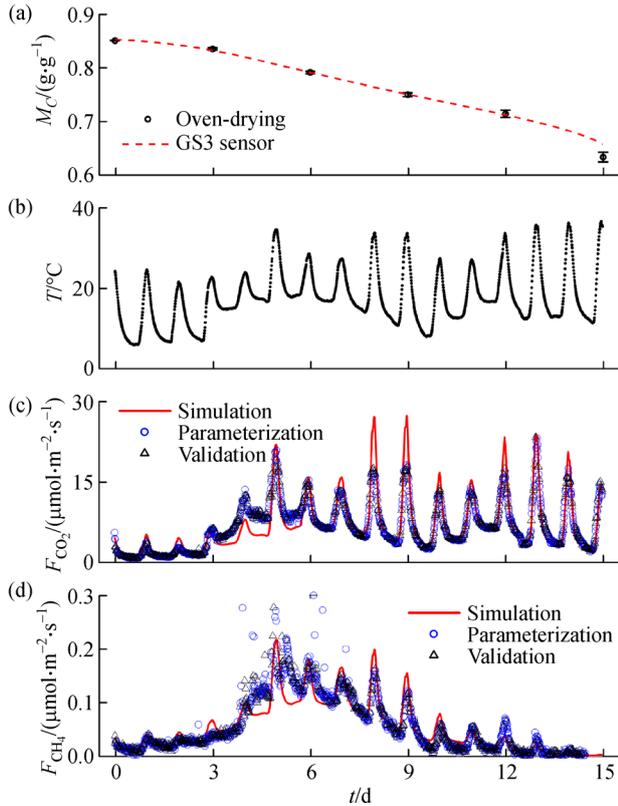


Fig. 1 Sensor-based and gravimetric measurements of wet manure moisture content (M_C) (a), temperature (T) (b), fluxes of CO₂ (c) and CH₄ (d) during the ambient drying process of dairy manure

moisture dependence are shown in Fig. 2a–2b, respectively. The model validation conducted over 15 d resulted in high regression correlation coefficients for CO₂ ($R^2 = 0.862$, $MSE = 2.05$) and CH₄ ($R^2 = 0.717$, $MSE = 0.034$). Determined regression parameters were $A = 3.53 \times 10^{11}$, $B = 2.58 \times 10^5$, $C = -3.87 \times 10^5$, $D = 2.05 \times 10^5$ for CO₂ and $A = 1.09 \times 10^6$, $B = 8.24 \times 10^5$, $C = -1.30 \times 10^6$, $D = 5.49 \times 10^5$ for CH₄.

Neglecting the influence of temperature, the parabolic profiles illustrated in Fig. 2a–2b, reaches peak values when M_C is 0.75 and 0.79 g·g⁻¹ for CO₂ and CH₄, respectively. This peak is expected considering the competing processes of moisture dependent supply of nutrients, which diminishes with reducing water content and oxygen supply, and gas exchange, which increases with reducing water content. In other words, the reduced emissions at higher M_C are associated with reduced gas diffusion through manure with a low number of air-filled pores and the reduction in gas emissions beyond the M_C associated with peak gas emissions results from diminishing resources and mobility. The mechanism for reduced pathways for transport of nutrients and movement of microbes is generally a reduction in cross-sectional area for water transport, resulting in reduced microbial activity^[15].

The net cumulative losses of CO₂ and CH₄ from the

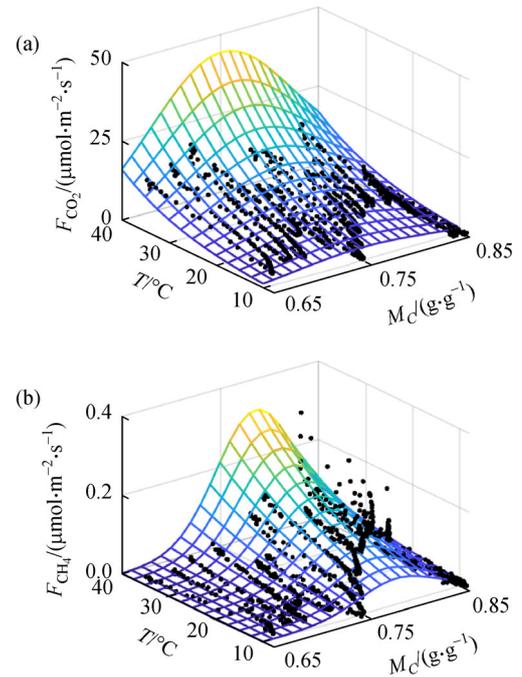


Fig. 2 Two-dimensional fitted surface describing the temperature and moisture dependence of CO₂ (a) and CH₄ (b) gas fluxes from dairy manure

ambient drying process of manure were calculated by integrating the area beneath each gas flux curve (Fig. 1c and 1d) over the total monitoring period (Fig. 3) as used to fit the cumulative flux data of CO₂ and CH₄ versus the loss of moisture content in Fig. 3 using parameters of $E_{\max} = 10.3 \text{ mol} \cdot \text{m}^{-2}$, $k_m = 0.373$, $\alpha = 11.43$ for CO₂; and $E_{\max} =$

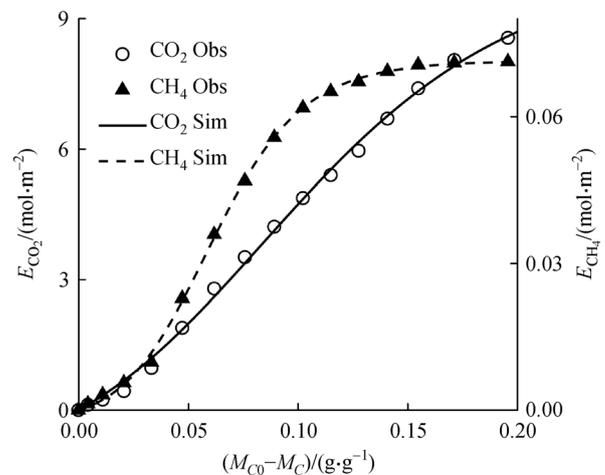


Fig. 3 Cumulative losses of CO₂ and CH₄ as a function of decreasing moisture content during the drying process of dairy manure. Differences were computed based on the initial moisture content, M_{C0} , as reference. Equation (9) fitted E_{\max} values for CO₂ ($10.3 \text{ mol} \cdot \text{m}^{-2}$) and CH₄ ($0.0713 \text{ mol} \cdot \text{m}^{-2}$) represent an estimate of the total gas loss over the manure drying process (i.e., beyond 15 d).

$0.0713 \text{ mol} \cdot \text{m}^{-2}$, $k_m = 0.527$, $\alpha = 34.2$ for CH_4 . The resulting coefficients of determination exhibited high correlation with R^2 values of 0.998 for CO_2 and 0.999 for CH_4 , respectively.

At the beginning of the experiment, there was an average of 1038 g of fresh manure in each cylinder. The initial average moisture content was 85.0% and the initial TC content was 37.8%, yielding an average total C mass of 58.9 g in each cylinder. With the surface area of each cylindrical manure sample of $3.24 \times 10^{-2} \text{ m}^2$ and the estimated E_{max} value from Fig. 3, the predicted maximum cumulative emission of CO_2 would be 4.02 g C, which represents about 6.83% of the total C present in the manure from the beginning of the experiment. This value was slightly lower than the percentage of total C (9.76%) reported by Moral et al.^[23] for a 52 d storage of farm yard manure. Similarly, the predicted maximum cumulative CH_4 emission was 0.028 g C, which accounts for 0.047% of the total initial C content. The cumulative emission of CH_4 here approached the results (0.06%) reported by Yamulki^[24], but was much lower than other literature values that range from 0.6% to 9.7%^[23,25]. Total C loss as CO_2 and CH_4 (6.88%) was much higher than the TC decrease shown in Fig. 5 because the cumulative CO_2 emission estimated using E_{max} extends beyond the 15 d drying process to complete drying (Fig. 3). With the help of the present model, it is feasible to estimate the total C loss within a relatively short experimental period.

The pH value of the manure decreased from 7.25 to 6.77 within the first 3 d, then increased slightly over the following 6 d and rose rapidly from 6.93 to 7.61 over the last 6 d (Fig. 4). Figure 5 illustrates changes in DOC, TC and DOC/TC during the ambient drying process. The DOC content increased initially and reached the peak concentration ($48.9 \text{ mg} \cdot \text{g}^{-1}$) on day 6, then it dropped almost linearly to $32.5 \text{ mg} \cdot \text{g}^{-1}$ measured at the end of the experiment. As the labile organic carbon source is most likely directly utilized by microorganisms, DOC accounts for only a small portion of TC (13.9% at the peak point). The DOC/

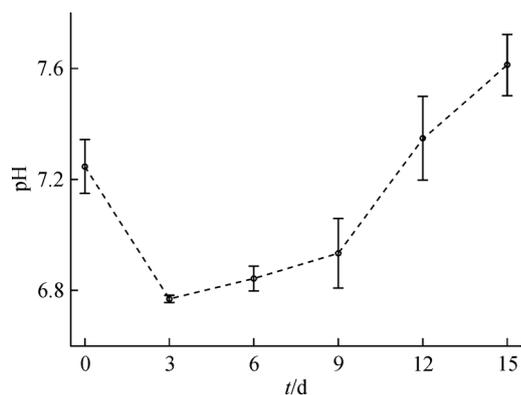


Fig. 4 Changes in pH during the ambient drying process of dairy manure

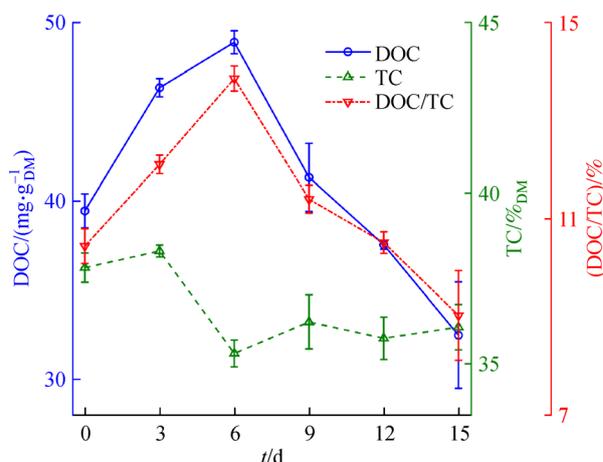


Fig. 5 Changes of DOC, TC and DOC/TC during the ambient drying process of dairy manure. The subscript DM refers to dry matter.

TC ratio follows the same pattern as DOC concentration because the variable range of TC is relatively small. However, there is a slight decline in TC concentration after day 3; then the concentration was relatively low but stable.

The microbial decomposition of organic matter in the manure occurs in two phases^[26]. First, some of the high molecular weight (MW) organic material is degraded to low MW constituents, such as alcohols and organic acids. Then the low MW organic compounds are degraded to CO_2 and CH_4 by microbes. In the beginning of the ambient drying process, manure was nearly saturated with an M_C of $0.85 \text{ g} \cdot \text{g}^{-1}$, which impeded gas (i.e., O_2) diffusion and hence respiration activity. Over the first 3 d of the experiment the production rates of low MW organic compounds were higher than their consumption rates. The TC content was maintained, while the DOC concentration rose gradually. Anaerobic reactions would have been dominant during this stage. The accumulation of organic acids reduced the pH value to acidic conditions. With continued drying, from days 3 to 6, much more of the low MW organic components were decomposed because M_C decreased and temperature increased. CH_4 was generated under anaerobic conditions, while most of the CO_2 was produced aerobically. Emissions of CO_2 and CH_4 showed that the aerobic and anaerobic processes were almost equally important during this stage. However, the reaction rates of the first process were still greater than that of the second. Thus, the content of DOC was still increasing at a slower rate while the TC content slightly decreased. With CO_2 and CH_4 emissions, some organic acids were degraded and pH was slightly increased. Beginning with day 6 until the end of the experiment, most of the high MW organic compounds, which are easier to decompose, were degraded into low MW compounds. With increase in the number of air-filled pores, aerobic reaction rates increased while anaerobic activity decreased and finally ceased.

Aerobic reactions capitalized on this condition consuming abundant DOC. This caused pH to increase to 7.61 by day 15.

5 Conclusions

Gaseous emissions from drying manure are highly dependent on its temperature and moisture content. An empirical surface model derived from the Arrhenius equation was employed to simulate effects of temperature and moisture content on gas fluxes, resulting in excellent agreement with measured CO₂ and CH₄ emissions. The system of equations provides a new, quantitative and more robust tool for characterizing gaseous emissions from drying manure. Ignoring the effect of temperature, the emissions of CO₂ and CH₄ peaked at moisture contents of about 0.75 and 0.79 g·g⁻¹, respectively. The CH₄-C emission during ambient drying of manure was 0.047% of the initial C content. It was mainly generated during the first 10 d. The projected C losses from CO₂ represented 6.83% of the total initial C content. Expansion and universal application of this model to estimate gas emissions from a variety of manure sources across the range of temperature and water content experienced for agricultural applications requires implementation of additional physical, chemical and biological factors, which is part of ongoing research.

Acknowledgements The authors are grateful to Bill Mace, Ricardo Tejada, Jean Almonte and Franyel Silfa for their assistance with technical, hardware and software aspects of this work. This research was supported by funds from the USDA-NIFA AFRI Air Quality Program (2010-85112-50524), the Utah Agricultural Experiment Station, Utah State University, Logan, Utah, USA (approved as journal paper no. 8647), the National Natural Science Foundation of China (41401225), the Fundamental Research Funds for the Central Universities, China (N150204005) and the Natural Science Foundation of Liaoning Province, China (201602250).

Compliance with ethics guidelines Enzhu Hu, Pakorn Sutitarnontr, Markus Tuller, and Scott B. Jones declare that they have no conflicts of interest or financial conflicts to disclose.

All applicable institutional and national guidelines for the care and use of animals were followed.

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