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A new tracer experiment to estimate the methane emissions from a dairy cow shed using sulfur hexafluoride (SF_6)

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Abstract. Methane emission from livestock and agricultural wastes contribute globally more than 30% to the anthropogenic atmospheric methane source. Estimates of this number have been derived from respiration chamber experiments. We determined methane emission rates from a tracer experiment in a modern cow shed hosting 43 dairy cows in their accustomed environment. During a 24-hour period the concentrations of CH_4, CO_2, and SF_6, a trace gas which has been released at a constant rate into the stable air, have been measured. The ratio between SF_6 release rate and measured SF_6 concentration was then used to estimate the ventilation rate of the stable air during the course of the experiment. The respective ratio between CH_4 or CO_2 and SF_6 concentration together with the known SF_6 release rate allows us to calculate the CH_4 (and CO_2) emissions in the stable. From our experiment we derive a total daily mean CH_4 emission of 441 L_{STP} per cow (9 cows nonlactating), which is about 15% higher than previous estimates for German cows with comparable milk production obtained during respiration chamber experiments. The higher emission in our stable experiment is attributed to the contribution of CH_4 release from about 50 m^3 of liquid manure present in the cow shed in underground channels. Also, considering measurements we made directly on a liquid manure tank, we obtained an estimate of the total CH_4 production from manure: The normalized contribution of methane from manure amounts to 12-30% of the direct methane release of a dairy cow during rumination. The total CH_4 release per dairy cow, including manure, is 521-530 L_{STP} CH_4 per day.

1. Introduction

Methane is an important anthropogenically produced greenhouse gas and plays a key role in atmospheric chemistry, especially in the stratosphere. The atmospheric mixing ratio of methane has increased by more than a factor of 2 since preindustrial times to a present global mean concentration of about 1750 ppb mainly because of man's activity [*Dlugokencky et al.*, 1994]. The rate of increase has slowed in recent years, however, because of causes yet unknown. One of the most important man-made sources of atmospheric methane is its production and emission by domestic animals. The amount of methane globally emitted by vertebrates (including animal waste) is estimated to be 110 Tg CH_4 per year (85-130 Tg per year (1 Tg = 10^{12} g)) [*Prather et al.*, 1995]. This corresponds to about 20% of the total methane emission from all sources (about 535 Tg per year) or to more than 30% of all anthropogenic sources.

The method generally applied to obtain values of the global CH_4 emission rate by cows is to use respiration chamber measurements. From these respiration chamber experiments, normalized emission rates for different animal classes (beef, dairy cows, sheep, etc.) are then calculated, also taking into account parameters like age distribution, fractional diet, energy loss, milk production, etc. [*Johnson et al.*, 1993]. Using Food and Agriculture Organization estimates of animal numbers then leads to global methane emission rates [*Crutzen et al.*, 1986; *Johnson et al.*, 1993]. The experiments with respiration chambers were mainly designed to measure the convertible energy in food for animals. In this sense the loss of energy from domestic animals in developed countries through methane emission is a quite remarkable share of about 5-7% of the input energy through the diet [*Blaxter and Clapperton*, 1965; *Johnson et al.*, 1993].

We present here an experiment specifically designed to measure the emission of methane from dairy cows in their accustomed environment, namely, in a normal cow shed. In this way we are able to measure the methane emission of many cows simultaneously and, moreover, can avoid any bias caused by stress situations possibly occurring in respiration chambers. One shortcoming of this kind of

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experiment is, however, that we cannot directly separate the methane contribution from manure also present in the stable. In our experiment the methane emission rate from dairy cows is determined via a tracer (SF_6) which is continuously released at a constant rate throughout the duration of our measurements (24 hours). The technique to determine the ventilation rate using SF_6 has been previously reported. *Leonard et al.* [1984] used SF_6 to study the heat and moisture production of animals housed under commercial conditions. The concentration trend of the SF_6 tracer, released at a constant rate, was found to be a direct measure of the air ventilation rate in the stable. From this ventilation rate and the parallel concentration trend of CH_4 in the stable air the methane emission rate could be determined.

2. The Experiment

2.1. Description of the Experiment

The tracer experiment was made in a modern cow shed of a medium size German farm, suitable to host about 50 adult animals. The cow shed has an outline of 16 m by 32 m and a height of 2.5 to 4.5 m due to a roof slope of about 8 degrees (see Figure 1). The milking device is located in the northeast corner of the shed. The stable is actively ventilated by three fans located at the highest point of the roof. When all doors are closed, the air inlet is solely through an air shaft located directly under the roof along the north side of the stable. During the experiment, 43 dairy cows were hosted in the shed, 9 of them nonlactating. The total milk production of all cows in the stable was about 650 L per day. The diet per cow consisted of 2 kg hay, 40 kg silage (2/3 maize and 1/3 turnip leaves), 3.2 kg concentrated feed, and 1.7 kg grain. The liquid manure from the cows drained under the slotted floor to a container outside. During the experiment, about 50 m^3 of liquid manure were sitting in the drain.

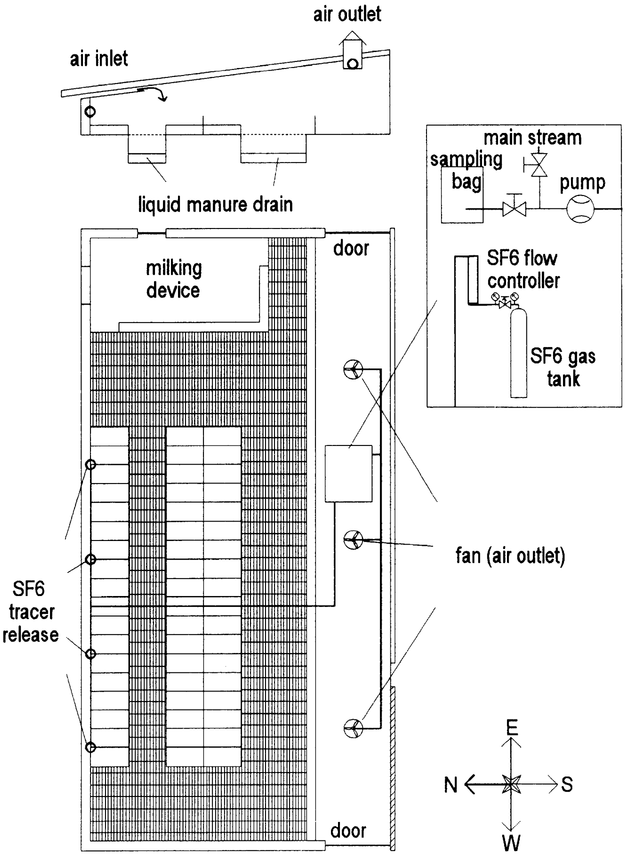

Figure 1. Sketch of the dairy cow shed. The areas to which the cows have access are checkered.

2.2. Sampling Technique

Integrated air samples were collected continuously from 10 min to 1 hour in polyethylene (PE) coated aluminum bags (Tesseraux, Germany). Before final filling, the bags were flushed with stable air for "conditioning" of the PE surface. The sample air was taken simultaneously from the shafts of the three fans through three polyvinyl chloride tubes. The tubes were continuously flushed at a flow rate of more than 100 L per hour. The bags were filled at a constant low flow rate through a bypass of the main stream behind the membrane pump (Sauer, Germany). In addition to the stable air, spot samples were taken every 2 hours from outside the stable to determine the background concentration of the three trace gases. The tracer gas SF_6 was released at four points close to the fresh air inlet shaft on the north side of the stable (see Figure 1). A 10-L-high pressure tank with synthetic air and SF_6 at a concentration of 9.7 ppm ± 5% (DEUSTE Steininger, Germany) was used for the experiment. This gas was released with a constant flow rate of 34.9 L_{STP} per hour throughout the experiment.

CH_4 and CO_2 concentration was measured by gas chromatography with flame ionization detector (Siemens Sichromat 3, Germany), using a nickel catalyst for CO_2 conversion to CH_4. Ultrapure nitrogen was used as carrier gas; the column temperature was at 90°C [*Born et al.,* 1990]. SF_6 concentration was measured by gas chromatography with electron capture detector [*Maiss et al.,* 1996]. The reproducibility of sampling and analysis for all three components, CH_4, CO_2, and SF_6, was of the order of $\pm 1\%$ (1σ). The release rate of the tracer gas was also measured to better than ±1%. The main error of the absolute methane emission rates derived in this experiment is therefore determined by the absolute accuracy of the SF_6 tracer gas, namely, ±5%.

2.3. Schedule of the Experiment

The experiment started with tracer release and air sampling on March 27, 1992, at 0727 local time during feeding and milking of the cows from 0630 to 0810. During this day the wind was light, coming from different directions. Around 1900 the wind increased to about 5 m s^{-1} for about 2 hours. During the next day, around 0200 and 0600, higher wind speeds of about 6 m s^{-1} were observed. During the course of the experiment all doors were kept closed as continuously as possible. On the evening of March 27 from 1817 until 1915 and on the following morning from 0650 to 0841 the door in the south west was open during milking and feeding periods. The tracer release was stopped on March 28, 1992, at 0744; sampling of stable air stopped at 0918.

3. The Model

If we consider the volume of air V in the stable being constant with time (constant temperature and pressure), the time dependent source strength $Q(t)$ for any trace gas emitted in the ventilated stable air is given by

$$Q(t) = \frac{d}{dt} M(t) + (c_f(t) - c_i(t))\, \dot{V}(t) =$$

$$\frac{d}{dt}\int_V c(x,y,z,t)\,dV + (c_f(t) - c_i(t))\dot{V}(t) \qquad (1)$$

$M(t)$	amount of a trace gas in the stable [L$_{STP}$ of tracer];
$c(t)$	volume mixing ratio of a trace gas [L$_{STP}$ tracer per L$_{STP}$ air];
$c_f(t)$	volume mixing ratio of a trace gas at the air outlet [L$_{STP}$ tracer per L$_{STP}$ air];
$c_i(t)$	volume mixing ratio of a trace gas at the air inlet [L$_{STP}$ tracer per L$_{STP}$ air];
V	air volume of the stable [L$_{STP}$ air];
$\dot{V}(t)$	ventilation rate [L$_{STP}$ air per hour];
$Q(t)$	tracer emission rate [L$_{STP}$ of tracer per hour];
$c(x,y,z,t)dV$	amount of trace gas in volume dV [L$_{STP}$ of tracer].

If we consider that the air at the outlet is representative of the mean stable air concentration ($c_f(t) = \bar{c}(t)$) and the stable is considered as a box with one way in and one way out, (1) can be simplified to

$$Q(t) = \frac{d}{dt} c_f(t) V + (c_f(t) - c_i(t))\dot{V}(t) \qquad (2)$$

The first term in (2) describes the change of concentration with time in the stable; the second term describes the tracer transport by ventilation of the stable air. The contribution to the source strength from the first term is determined from the difference of concentrations from one sampling period to the next times the volume of the stable (about 1700 m^3). This term is small, as the temporal concentration variations during the experiment are small. In our experiment the main contribution to the source strength is from the second term because of the high ventilation rate of the stable (about 10,000 m^3 h^{-1}) and the large concentration differences between outside and inside air. Under these conditions the emission rate $Q(t)$ of each trace gas emitted in the stable can be approximated by

$$Q(t) = (c_f(t) - c_i(t))\dot{V}(t) \qquad (3)$$

The emission rate Q_{SF6} of the tracer gas SF$_6$ in the stable is known and constant throughout the experiment. From this and the measured SF$_6$ concentration record inside and outside the stable the ventilation rate $\dot{V}(t)$ in (3) can be determined. The emission rates of CH$_4$ and CO$_2$ can then be calculated from the measured concen-tration records according to (4)

$$Q_{CH4,CO2}(t) = \frac{(c_f(t) - c_i(t))_{CH4,CO2}}{(c_f(t) - c_i(t))_{SF6}} Q_{SF6}(t). \qquad (4)$$

4. Results and Discussion

4.1. Concentration Records

Figure 2 shows the concentration records of SF$_6$, CH$_4$, and CO$_2$ plotted versus time. The CO$_2$ and CH$_4$

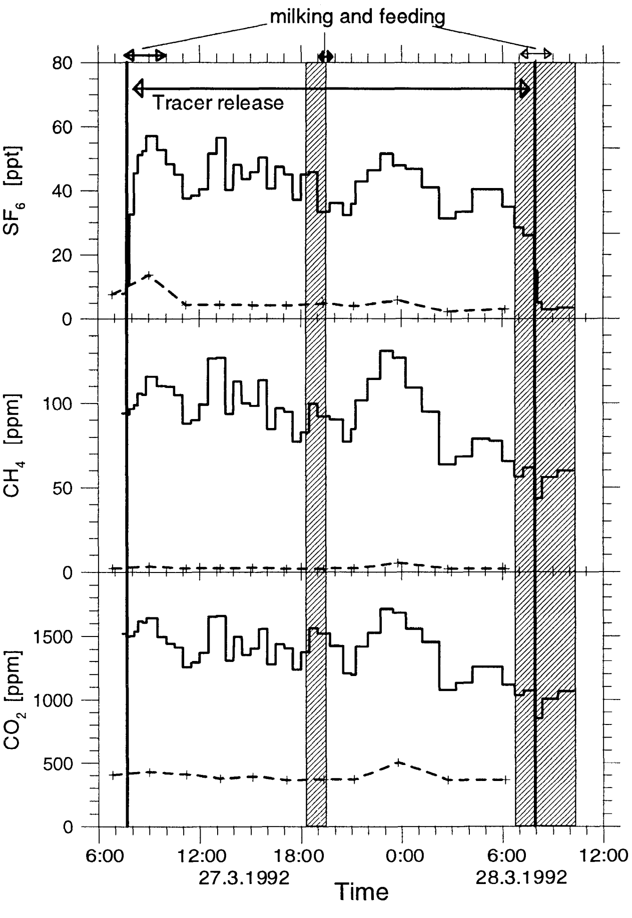

Figure 2. Concentration record of the trace gasses SF$_6$, CH$_4$, and CO$_2$ at the air outlet of the stable (solid lines) and outside the stable (dashed lines). The thick vertical lines mark the start and stop of tracer release. The hatched areas mark the time periods when the door in the south-west was open; the periods marked by arrows indicate feeding and milking times.

concentration records show a very similar behavior with a general decrease of concentration during the course of the experiment. Also, the concentration of SF$_6$ follows the general time trend about 1 hour after the start of tracer release. This indicates that the ventilation rate of the stable was not constant during the experiment in spite of the fans running at a constant speed. The very fast increase of SF$_6$ close to steady state concentrations within less than 1 hour shows that the exchange rate of the stable air was very high. The concentration of all measured trace gases outside the stable was not significantly higher than the concentration for these gases in the free troposphere, except for SF$_6$ at the beginning of the experiment. For emission estimates the concentrations c_i have been interpolated between the measured points as shown in Figure 2.

At the beginning of the experiment when the emission of SF$_6$ began, the gas was not well mixed within the stable air. In this nonsteady state situation the calculated CH$_4$ and CO$_2$ emission rates were estimated too high. After closure of stable doors, when cow feeding and milking were finished, the tracer SF$_6$ was well mixed within the stable air, and the outlet air was representative of the air in the stable. This holds for the tracer as well as for the gases CH$_4$ and CO$_2$. In the evening feeding and milking phase (1817-1915) the southwest door was opened, and the circulation pattern in the stable was disturbed. The simple model of a box with one way out and one well-defined way into the stable is no longer valid. The southwest door of the stable was also a way in for outside air, and the normal air inlet shaft at the north wall of the stable could work as an outlet for stable air. This effect caused a decrease of the SF$_6$ concentration and thus a seemingly higher ventilation rate. In addition, during feeding and milking a large number of cows were eating at the trough, and the air collected at the fans was enriched in CH$_4$ and CO$_2$ with respect to the tracer SF$_6$, which was relatively depleted. After the doors were closed and the fans were working again as the only way out for stable air, a second steady state phase started. Release of SF$_6$ was stopped on March 28 at 0715 and the SF$_6$ dilution phase began. The concentration of the tracer SF$_6$ declined exponentially to the background level during this period.

4.2. Emission rates

For the course of the experiment, $Q_{CH4}(t)$ and $Q_{CO2}(t)$ have been calculated according to (4); the emission rates are displayed in Figure 3. Only the two "steady state phases" outside the feeding and milking periods could be used to calculate reliable emission rates for CH$_4$ and CO$_2$ using (4). To calculate the CH$_4$ emission during feeding and milking phases, CO$_2$ was used as a tracer. This is appropriate because the emission of CO$_2$ by cows in the steady state phases was very constant. Using CO$_2$ is also advantageous in that it is emitted at the same place as the CH$_4$. The SF$_6$-derived emission rates for CO$_2$ in the steady state phases were used to extrapolate the CO$_2$ emission in the other phases (dashed line in Figure 3). Then the extrapolated CO$_2$ emission rate was used instead of Q_{SF6} to calculate the CH$_4$ source according to (4). The results for the methane emission rates are plotted in Figure 4.

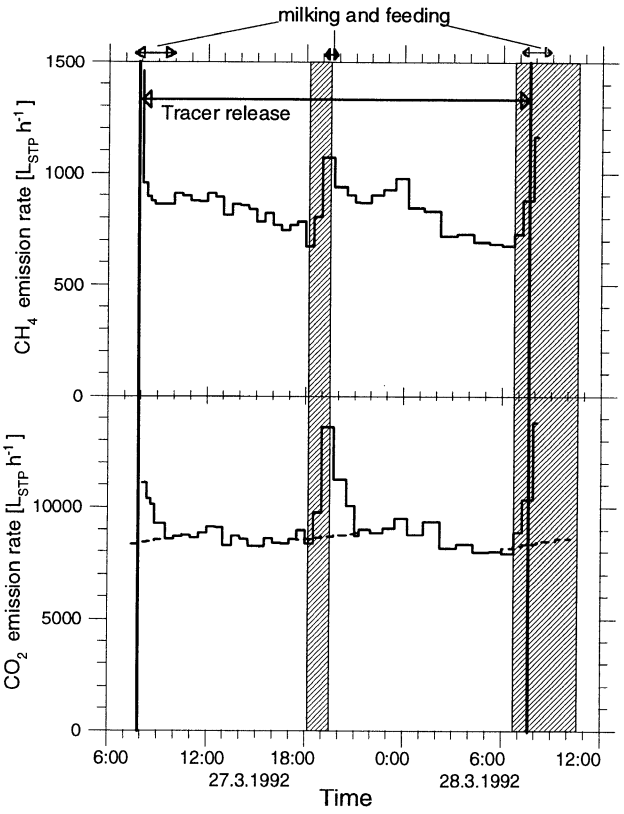

Figure 3. SF$_6$-derived CH$_4$ and CO$_2$ emission rates (solid lines), extrapolated CO$_2$ (dashed line). The thick vertical lines mark the time of start and stop of tracer release. The hatched areas mark the time periods when the southwest door was open; the periods marked by arrows indicate feeding and milking times.

4.3. Discussion

CH$_4$ emission rates were relatively constant during the experiment and show only a small 12-hour cycle: about 1 hour after the beginning of feeding, methane emissions increase for about 4 hours up to values about 20% higher than the daily mean. After the maximum, during the day, CH$_4$ emissions drop slowly for about 8 hours to a minimum value shortly before the next feeding. The methane emission drop during the night is sharper, and a constant low value is reached 6 hours after feeding.

The mean CH$_4$ emission rate during our experiment was 791±40 L$_{STP}$ per hour. This corresponds to 441 L$_{STP}$ CH$_4$ per cow per day. Respiration chamber experiments at the Agricultural Research Center in Braunschweig-Völkenrode (Germany) result in mean values of about 400 L$_{STP}$ CH$_4$ per cow per day and about half that value for nonlactating cows (D. Gädeken, personal communication, 1992). From these numbers a total CH$_4$

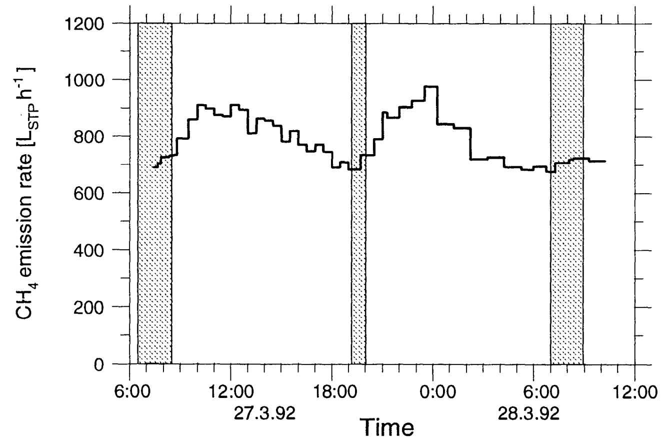

Figure 4. SF_6- and CO_2-derived CH_4 emission rate during the experiment. The hatched areas mark the feeding periods. A significant 12-hour cycle is observed with minima during feeding periods and maxima about 4 hours after feeding.

emission rate of about 642 L_{STP} per hour would be expected in our experiment. This value is about 19% lower than the release rate we actually observed.

In order to compare our results directly with those reported by *Blaxter and Clapperton* [1965], *Crutzen et al.* [1986], and *Johnson et al.* [1993], we would have to calculate the digestible energy content of the diet. These data, unfortunately, are not available for our experiment. We therefore have to use mean CH_4 emission rates for comparable cows also reported by these authors. *Crutzen et al.* [1986] calculated a mean emission of 95 kg CH_4 per dairy cow per year (364 L_{STP} CH_4 per cow per day), and *Johnson et al.* [1993] calculated for an American dairy cow 492 L_{STP} CH_4 per day. The high number of *Johnson et al.* is very close to our result (493 L_{STP} per dairy cow per day) if we assume that nonlactating cows (9 out of 43) produce half the methane of dairy cows.

A nonnegligible contribution of methane emission in our experiment originates from the liquid manure in the drain under the slotted floor. Very little information is available on methane emissions from animal wastes, which makes it difficult to estimate the possible CH_4 contribution from the manure during our experiment. In their review, *Johnson et al.* [1993, Table 7] report potential CH_4 emissions (under optimal conditions) per kilogram dry organic matter (OM) of manure from dairy cows of 240 L_{STP} CH_4 per kilogram OM. They assumed that 13% of this potential production is finally converted into CH_4. Taking these values and an exudation of 4.8 kilogram OM per dairy cow per day ($\hat{=}$ 0.05 m^3 liquid manure) [*Maurer and Winkler,* 1982] would lead to a methane production from manure of 240 L_{STP} CH_4 per kilogram OM $\times$ 4.8 kilogram OM $\times$ 0.13 = 150 L_{STP} CH_4 per dairy cow per day.

In our stable the floor space of the manure drain is 125 m^2, and the filling height is 0.4 m. The liquid manure in the stable thus amounted to 50 m^3. This amount of manure corresponds to the production from 43 animals during the last 23 days, when assuming a daily exudation of 0.05 m^3 manure per cow and no dilution with water. If the emission data for rumination from the respiration chamber experiments in Germany are valid also for our experiment, namely, 400 L_{STP} CH_4 per dairy cow per day, a CH_4 emission rate from the manure of about 3600 L_{STP} CH_4 per day or 3 L_{STP} per hour per cubic meter of liquid manure can be estimated.

We measured CH_4 emissions from liquid manure in a storage tank of a German farm during the course of a whole year [*Marik,* 1993]. These results showed a large variation with an exponential dependence of methane emissions on temperature (see Figure 5). The amount of methane possibly originating from the liquid manure in our stable experiment (3 L_{STP} CH_4 m^{-3} h^{-1}) lies in the upper range of fluxes for the storage tank at 12°C. The mean temperature of 12°C observed during our experiment was measured only in the stable air, not directly in the manure. It is possible that the temperature of the liquid manure was higher than the temperature in the stable air, partly explaining the relatively high methane production from manure during our experiment. However, from Figure 5 we conclude that a CH_4 production of 3 L_{STP} CH_4 m^{-3} h^{-1} from liquid manure is clearly an upper limit for this number.

As a lower limit, we may use the production rate we measured at the manure tank at 12°C, namely, 0.47 L_{STP} CH_4 m^{-3} h^{-1} or 23 L_{STP} CH_4 per 50 m^3 per hour. Subtraction of this manure fraction from the mean total

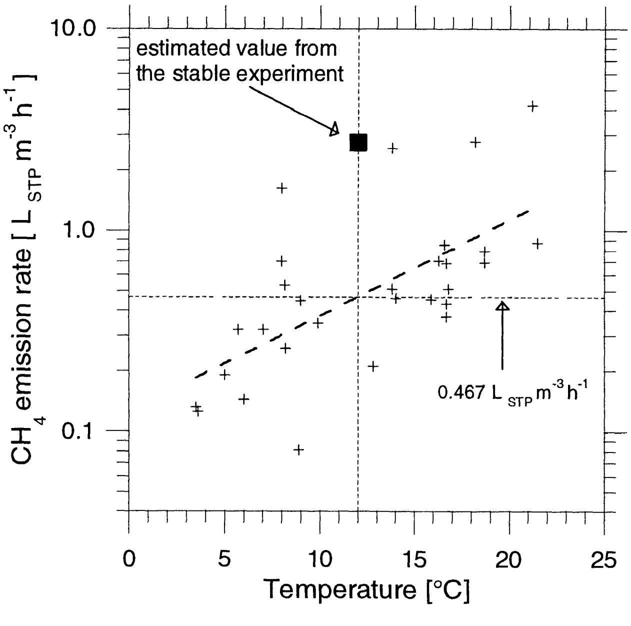

Figure 5. Temperature dependence of the CH_4 emission from liquid manure in a storage tank. An exponential function is obtained: $Q_{CH4} = Q_0 \exp(T/T_e)$ with $Q_0 = 0.125$ L_{STP} per m^3 per hour and $T_e = 9.1$ °C (T in °C).

CH_4 emission during the stable experiment (791 L_{STP} CH_4 h^{-1}) leads to a contribution directly from the ruminating cows of 479 L_{STP} CH_4 per dairy cow per day (again assuming 50% of this emission for a nonlactating cow). With these estimates we obtained a range for daily CH_4 production per dairy cow of 400-479 L_{STP} CH_4 and a range for CH_4 production from manure in the stable of 0.47-3 L_{STP} m^{-3} h^{-1}. In order to calculate the cow-related methane production from manure, we have to take into account not only the residence time of the manure in the stable (in our experiment 23 days) but also the storage time outside the stable in manure tanks. From our manure tank measurements over a whole year [*Marik,* 1993] we can calculate a mean methane emission of 0.742 L_{STP} CH_4 m^{-3} h^{-1}. The mean residence time of the manure in the storage tank was 43±5 days. After that period the manure was distributed on the fields and anaerobic methane production stopped. The total lifetime of the manure as a methane emitter can therefore be estimated to be 66±5 days. The mean storage time of manure is very similar for dairy farms with this kind of manure handling.

A mean methane emission of 17.8 L_{STP} CH_4 m^{-3} d^{-1} for outside manure storage (43 days) and the emission range cited above, 11-72 L_{STP} $m^{-3}day^{-1}$, for the residence time inside the stable (23 days) yields a total CH_4 emission rate of manure of 1018-2421 L_{STP} CH_4 per cubic meter of manure. The daily manure exudation per cow is about 0.05 m^3; the total CH_4 emission from this daily exudation thus amounts to 51-121 L_{STP} per day. In this calculation the lower value, mathematically, is associated with a higher CH_4 emission during rumination (479 L_{STP} CH_4 per day), whereas the higher manure emission, mathematically, is associated with our lower limit of CH_4 production during rumination. The total methane emission per dairy cow, including manure, can be determined as the sums of the respective values, namely, 400+121=521 to 479+51 =530 L_{STP} CH_4 per dairy cow per day.

Conclusion

Our tracer experiment turned out to be an easy and reliable way to obtain estimates of the total CH_4 emission from a modern dairy cow shed. With this experimental design it was possible to obtain mean emission rates from a whole dairy cow herd in its natural environment. One difficulty in our experiment to obtain emission rates was the presence of liquid manure in underground channels. Methane emissions from the manure turned out to be 3-16% of the total methane production rate in the stable (441 L_{STP} CH_4 per cow per day). With the emissions out of the storage tank outside the stable the best estimate for the total emission of methane in this type of dairy farming is 521-530 L_{STP} CH_4 per dairy cow per day.

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