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Temperature response of methane production in liquid manures and co-digestates

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HIGHLIGHTS

- Temperature dependence of methane emission in liquid slurry materials was quantified
- Arrhenius parameters were derived including 95% confidence limits
- Different slurry materials had similar temperature sensitivity of methane emission
- Temperature sensitivity of methane emission from slurry aligned with other ecosystems

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ABSTRACT

Intensification of livestock production makes correct estimation of methanogenesis in liquid manure increasingly important for inventories of CH_4 emissions. Such inventories currently rely on fixed methane conversion factors as knowledge gaps remain with respect to detailed temperature responses of CH_4 emissions from liquid manure. Here, we describe the temperature response of CH_4 production in liquid cattle slurry, pig slurry, and fresh and stored co-digested slurry from a thermophilic biogas plant. Subsamples of slurry were anoxically incubated at 20 temperatures from 5–52 °C in a temperature gradient incubator and CH_4 production was measured by gas chromatographic analysis of headspace gas after a 17-h incubation period. Methane production potentials at 5–37 °C were described by the Arrhenius equation (modelling efficiencies, 79.2–98.1%), and the four materials showed a consistent activation energy (E_a) which averaged 81.0 kJ mol^{-1} (95% confidence interval, 74.9–87.1 kJ mol^{-1}) corresponding to a temperature sensitivity (Q_{10}) of 3.4. In contrast, the frequency factor (A) differed among the slurry materials ($30.1 < \ln A < 33.3$; mean, 31.3) reflecting that origin, age and composition of the manure affect this parameter. The E_a estimate, based on individual slurry materials, was intermediate when compared to published values of 63 and 112.7 kJ mol^{-1} derived from composite data, but was similar to E_a estimated for CH_4 production at microbial community level across aquatic ecosystems, wetlands and rice paddies (89.3 kJ mol^{-1}). This supports that the derived temperature sensitivity parameters may be applicable to dynamic modelling of CH_4 emissions from livestock manure.

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1. Introduction

Methane (CH_4) is an atmospheric greenhouse gas (GHG) produced by methanogenic Archaea (methanogens) in diverse anaerobic environments, such as waterlogged soil and the digestive tract of animals (Le Mer and Roger, 2001). Large quantities of CH_4 are also produced and released from man-made ecosystems such as landfills and rice paddies, and from confined animal feeding operations where both livestock and manure are sources of atmospheric CH_4 (Knapp et al., 2014). Thus, manure management was recently estimated to account for about 10% of the total CH_4 emissions from agriculture (Serrano-Silva et al., 2014). Intensification of livestock production can be observed in many regions of the world (Bouwman et al., 2013) and, especially where manure is handled in liquid form (slurry), the emission of CH_4 during storage can be significant (MacLeod et al., 2013; Opio et al., 2013). Accordingly, the correct estimation of methanogenesis in liquid manure becomes increasingly important for inventories of CH_4 emissions.

Methane production in manure depends on storage temperature; CH_4 emissions from storages have been observed at temperatures of <5 °C, but typically attains a maximum in the mesophilic temperature range, for example at 30–37 °C (Cullimore et al., 1985; Safley and Westerman, 1990). Other controlling factors include manure composition (e.g., organic matter degradability, ammonia concentration and pH) and size and composition of the methanogenic community as modified by storage conditions and pre-treatment (Zeeman, 1994; Chen et al., 2008; Witarsa and Lansing, 2015). Most national inventories of CH_4 emissions from manure management are based on guidelines

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developed by the Intergovernmental Panel on Climate Change (IPCC, 1997, 2006) where the temperature dependency of CH_4 production is taken into account via fixed methane conversion factors (MCFs), defined for a range of average annual temperatures. However, local circumstances with respect to pre-treatment, storage conditions and residence time may significantly influence annual CH_4 emissions (Sommer et al., 2009), and proper accounting for management effects therefore may require a more dynamic approach.

Models with different levels of complexity have been presented to describe CH_4 emission from liquid manure storage with a daily to monthly time resolution (Mangino et al., 2001; Sommer et al., 2004; Chianese et al., 2009). Yet, significant knowledge gaps remain with respect to CH_4 production potentials for specific storage conditions, including the detailed effect of slurry temperature. Generally, the temperature response of microbial activity below the optimum temperature can be described by the Arrhenius equation (Elsgaard and Jørgensen, 2002; Davidson and Janssens, 2006), i.e., rate $= A \exp(-E_a/RT)$, where A is the frequency factor, E_a is the activation energy (J mol^{-1}), R is the gas constant (8.314 J mol^{-1} K^{-1}) and T is temperature (K). Previous attempts to estimate methanogenesis in livestock manure have relied on compilations of data from dissimilar studies to derive an exploratory temperature relationship, with no possibility to estimate slurry-specific variation or parameter uncertainties (Safley and Westerman, 1990; Sommer et al., 2004).

The objective of this study was to determine the temperature dependency of CH_4 production in separate liquid manure materials, including cattle and pig slurry, and co-digested slurry from a thermophilic biogas plant. To ensure a superior data coverage we used a temperature-gradient incubator (TGI) to allow for simultaneous slurry incubation at 20 different temperatures ranging from 5 to 52 °C (Elsgaard and Jørgensen, 2002). We expected this methodology to allow for invention of robust temperature relationships for methane production (i.e., with low parameter uncertainties) and to allow for tests of potential differences among the slurry types.

2. Materials and methods

2.1. Slurry materials

Cattle slurry was collected from the storage tank of a beef cattle farm (Nedergaard, Tjele) in April 2013. The animals were fed grass-clover and whole-crop silage, with only a minor group of calves receiving concentrates. The slurry had been collected during six months, and was mixed on the day of sampling. Pig slurry originated from different production facilities at the Research Centre AU-Foulum and represented both finishing pigs and farrowing sows. Pig slurry was collected from a mixed storage tank in January 2014; age of the slurry at the time of sampling was at least six months. Livestock slurry co-digested with other organic substrates was collected in May 2014 from a biogas plant at Research Centre AU-Foulum with an 1100-m^3 reactor operated at 52 °C (hydraulic retention time, 13–14 d). Various organic materials, including maize silage and glycerol/fish silage that together constituted *c.* 20% by volume, were co-digested with cattle and pig slurry (Dr. Alastair Ward, pers. comm.). At the biogas plant, digestate is first stored in a post-digestion storage tank (from where gas is collected during the cooling phase), and then transferred at monthly intervals to a secondary tank for final storage. For this study, fresh digestate was collected directly from the outlet of the reactor, while stored digestate (>1 month) was collected from the secondary tank.

Before use, the collected slurry materials were sieved (<2 mm) to enable reproducible incubation in test tubes (see below) — it was thus assumed that the methanogenic community of the sieved fraction had the same temperature response as that of bulk slurry. The sieved slurry materials were stored in (almost filled) stoppered 300-mL infusion bottles at 2 °C for a maximum of 14 d before determination of temperature responses.

2.2. Temperature gradient incubator

The TGI used was described in detail by Elsgaard and Jørgensen (2002). Briefly, the TGI consists of an insulated aluminum bar (240 × 79 × 65 cm) with 30 rows of six replicate sample wells for incubation of 28-mL test tubes. The incubator is heated at one end by an electric plate and cooled at the other end by thermoelectric Peltier elements; this produces a linear thermal gradient over the 30 sample rows. Temperatures are monitored continuously and controlled by three automated PC-operated control loops. In the present study temperature gradients ranging from 5 to 52 °C were produced, corresponding to increments of ~1.6 °C between adjacent incubation temperatures (20 of the 30 incubation temperatures were used for slurry incubations). During operation the standard deviation around mean temperatures was 0.2–0.4 °C, as calculated from temperatures logged at 5-min intervals at 15 sample rows along the thermal gradient.

2.3. Incubation procedure

For determination of the temperature response of a slurry material, a stoppered 300-mL infusion bottle with the slurry was first pre-incubated at 20–22 °C for 4 h to activate methanogenesis. During this time, a flow of N_2 was passed through the headspace of the infusion bottle to ensure that oxygen was excluded. Then, through a second gas line, the slurry was gently bubbled for 10 min with N_2 to remove CH_4 from the liquid phase; this was done to reduce the background of dissolved CH_4 in the aliquots subsequently conditioned for incubation in the TGI. While continuously flushing slurry and headspace with N_2, subsamples of ~3-mL were transferred to 28-mL test tubes ($n = 70$) using a 5-mL pipette with a cut-off plastic tip while also gas flushing the recipient test tube to avoid oxygen contamination (Macy et al., 1972). Following slurry addition, each test tube was immediately closed (under N_2 headspace) with a butyl rubber stopper (1 cm thick) and placed on ice to temporarily arrest methanogenesis. Stoppers of the test tubes were secured with crimp seals, and the tubes were evacuated and refilled with He three times; they were then left at atmospheric pressure on ice until all samples were ready for incubation (within 1–2 h). A total of 60 test tubes were placed in the TGI according to a randomization scheme, with triplicate samples for each of 20 different temperatures covering the range from 5 to 52 °C. The last ten test tubes were used for determination of background CH_4 concentrations and were processed for CH_4 measurements at the time of starting the TGI incubation.

Three different incubation periods were evaluated, i.e., 3 h (short-term), 17 h (over-night) and 41 h (over-night + 24 h). Methane production rates after 3 and 17 h were compared for the cattle slurry, and CH_4 production rates after 17 and 41 h were compared for fresh digested slurry. By the end of an incubation period, gas samples were taken from the headspace of each test tube in the TGI. The pressure inside test tubes was expected to vary, partly because test tubes were all at room temperature when closed, but at different temperatures when sampled, and partly because of temperature effects on gas production during incubation. In order to avoid pressure deficits at sampling, the test tubes were all pressurized by injecting between 2 and 5 mL He (5 mL at the lowest temperatures); this was done 0.5 h prior to gas sampling to allow the gas phase temperature to re-adjust to the specific incubation temperature. A 10-mL glass syringe was then used to determine gas volumes at atmospheric pressure; this was done by inserting the glass syringe (with a hypodermic needle) through the stoppers while the test tubes were still in the incubator. After reading the gas volume, a 3-mL sample of the headspace gas was transferred to a 6-mL Exetainer (Labco Inc., Lampeter, UK) previously equilibrated to atmospheric pressure with He.

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