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Methane Emissions from Municipal Wastewater Treatment Processes

Peter M. Czepliel,* Patrick M. Crill, and Robert C. Harriss

Complex Systems Research Center, Morse Hall, University of New Hampshire, Durham, New Hampshire 03824-3525

Methane and carbon dioxide emissions from primary and secondary wastewater treatment processes were measured from mid-winter to summer conditions in Durham, NH. A statistically significant positive relationship between gas flux and wastewater temperature was determined for methane and carbon dioxide in both the aerated and nonaerated areas of the grit tanks. Statistical correlations to temperature measured in a secondary aeration tank were marginal for methane and insignificant for carbon dioxide. Emission factors derived from our measurements were 39 g of CH₄ person⁻¹ year⁻¹ and 35 698 g of CO₂ person⁻¹ year⁻¹ for primary and secondary activated sludge treatment processes.

Introduction

A key to formulating strategies to control and reduce the rate of increase of methane (CH₄) in the atmosphere is the identification and quantification of all CH₄ sources, both natural and anthropogenic. Significant natural sources, including wetlands, natural biomass burning, termites, and oceans and freshwater, are generally not subject to human control. These sources account for approximately 45% of the total global CH₄ emissions which are estimated to range from 440 to 640 Tg of CH₄ year⁻¹ (1). However the anthropogenic sources, especially those in urban areas such as landfills, natural gas systems, fossil fuel combustion, and wastewater treatment facilities, could potentially be regulated once properly resolved and quantified (2). Preliminary studies of whole city methane emissions, scaled to the globe, suggest an urban methane source of approximately 30-60 Tg (3). Relatively small reductions in global methane emissions of 30-50 Tg year⁻¹ would be sufficient to stabilize the global atmospheric concentration, assuming that the rates of destruction by OH and soil uptake remain the same.

The purpose of this study was to quantify CH₄ emissions from a wastewater treatment plant consisting of processes typical of those in use in the industrialized world. The transport and management of residential, commercial, and industrial wastewater is known to produce CH₄ gas (4, 5). CH₄ is produced by bacterial decomposition of organic matter in the absence of oxygen. Aerobic decomposition processes in wastewater demand more oxygen than can be supplied by surface diffusion. Therefore, in the absence of mechanical aeration, methanogenic bacteria activate anaerobic decomposition which produces CH₄ gas as a byproduct. However, few quantitative data exist regarding the generation of CH₄ in wastewater processes other than the anaerobic digestion of sewage sludge. Wastewater treatment plants were estimated to account for approximately 5% of total global emissions or 20-25 Tg of CH₄, year⁻¹ by the IPCC, although the basis for the estimate was not defined (1). The lack of quantitative data describing CH₄ emissions from the specific processes that comprise modern wastewater treatment plays will limit further technical assessments of mitigation options. The field measurements described below were performed to

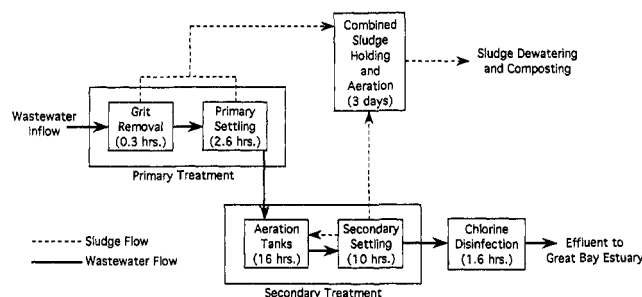


Figure 1. Simplified process diagram for the Durham NH, wastewater treatment plant. Retention times are given in parentheses.

quantify CH₄ emissions from processes in a wastewater treatment plant and to determine the fate of organic carbon in the influent.

CO₂ emissions were measured simultaneously with the CH₄ emission measurements. The CO₂ data are useful as an indication of overall decomposition processes and to a broader goal we have for understanding urban carbon metabolism.

Field Site

The municipal wastewater treatment plant studied is located in Durham, NH. The facility serves the town and the main campus of the University of New Hampshire. The population served during the school year is approximately 12 500 and generates a mean wastewater flow of approximately 4×10^6 L day⁻¹. The population served during the summer is approximately 6200 and generates a mean wastewater flow of approximately 2×10^6 L day⁻¹. During the summer months, the plant processing capacity is normally reduced by one-half through partial shut down of the grit tanks, primary settling tanks, secondary aeration tanks, and secondary settling tanks.

The facility consists of primary and secondary unit operations and processes (Figure 1) that treat a medium-strength municipal wastewater with an average influent BOD₅ at 20 °C of approximately 250 mg/L. Average removal efficiency is 94%. A set of two grit chambers is the first physical unit operation encountered by the influent. Grit, consisting of sand, gravel, and other heavy solids, settles and is removed from the system. The wastewater then enters the primary settling tanks where it is held in a relatively quiescent state which results in the removal of solids with a higher specific gravity than the liquid by settlement and solids with a lower specific gravity by flotation and skimming. The liquid then enters secondary treatment, while the solids are removed from the system as primary sludge. The secondary treatment system consists of four aeration tanks and two clarification tanks. In this process, the wastewater organic matter that enters the aeration tanks is decomposed by suspended bacterial biomass under aerobic conditions. This results in the release of gases to the atmosphere and the growth of microbial biomass. The microbial-rich liquid then passes into the secondary clarification tanks where the microbial biomass is separated from the treated wastewater

by gravity. A portion of the settled microbes is continuously recycled back to aeration tank no. 1 as an inoculum to maintain a proper concentration of microorganisms in the aeration tanks. The remaining fraction of the microbial biomass is treated as sludge. This diversion of sludge is a discontinuous process which occurs periodically depending on load conditions. The treated effluent then moves to the chlorine contact tank for disinfection and is released into the Great Bay Estuary.

The resulting primary and secondary sludge is retained for approximately 3 days in a partially aerated holding tank where the liquid content is reduced by aeration and evaporation. The mixed sludge is then transferred to the sludge processing building where it is dewatered. The dewatered sludge is mineralized by composting with wood chips to maintain aerobic permeability in compost piles that are actively aerated.

Methods

Methane and carbon dioxide fluxes were measured from each tank in the wastewater treatment plant during a preliminary evaluation of plant emissions. The dominant emission sources were determined to be the grit tanks, secondary aeration tanks, and sludge holding tanks. Negligible emissions were measured from the primary settling tanks and secondary clarification tanks. CH₄ and CO₂ fluxes were then measured from the dominant sources at roughly 1-week intervals at approximately the same time of day. The sampling period captured the mid-winter to summer range of operating conditions, especially variations in temperature. Gas flux measurements were collected from aerated and nonaerated liquid surfaces, and dissolved gas content was measured in the secondary sludge. The spatial variability of measured methane concentration was examined early in the study in each source tank. The maximum measured spatial variation was found in the aerated area of the grit tank. This variability was determined to be of negligible impact when compared to the expected temporal variability of emissions. The sample collection procedures employed are described below.

Nonaerated Surfaces. A closed chamber technique was used to measure fluxes from nonaerated liquid surfaces (7). An aluminum enclosure (51 × 51 × 26 cm) was floated on the liquid surface with a thermistor mounted inside the enclosure to measure the enclosed air temperature. Floats were fastened to the enclosure sides, and the chamber was manually held in place during sampling to minimize chamber movement due to surface turbulence. A battery-operated blower circulated the air within the enclosure and through a closed loop of hose from which headspace air samples were withdrawn. Samples were collected in 60-mL polypropylene syringes at 2-min intervals for 10 min. The gas flux, E (mass area⁻¹ time⁻¹), from the liquid surface in the static chamber was calculated by

$$E = (V/A)p(dc/dt) \quad (1)$$

where V is the volume of the chamber, A is the enclosed surface area, p is the density of the gas at the temperature recorded in the chamber, and dc/dt is the linear increase in the gas concentration in the chamber during the sampling period.

Aerated Surfaces. A bag technique was used to measure fluxes from aerated liquid surfaces. A 40-L

polyethylene sample bag, fastened to the inside of a wood support frame, was used to collect gas samples from the agitated surface of the aerated tanks. The open base of the frame allowed gas to be collected from 0.084 m² of the surface. To collect samples, the bag was collapsed and emptied of ambient air and the wood support frame was placed several inches into the liquid. Dissolved gases within the liquid, transferred to the mechanically distributed air by diffusion, then filled the collection bag. Samples were withdrawn from the bag in 60-mL polypropylene syringes. The gas flux, E (mass area⁻¹ time⁻¹), was calculated by

$$E = pcQ/A \quad (2)$$

where p is the density of the gas at the temperature recorded in the sample bag, c is the sample gas concentration, Q is the total diffuse air flow, and A is the total surface area.

Sludge Gases. CH₄ and CO₂ fluxes resulting from sludge storage were determined by two methods. First, gas concentrations in the sludge were determined by sampling the sludge flow, prior to transfer, with a 0.5-L collection bucket. Samples of 30 mL of sludge were collected immediately after withdrawal from the flow in 60-mL polypropylene syringes. An equal volume of laboratory room air was later introduced into the sludge sample syringe, and the syringe was shaken vigorously for 2 min. The gases in the syringe headspace were then transferred to a clean 60-mL polypropylene syringe for analysis. This procedure was repeated three times and stripped greater than 95% of the dissolved CH₄ from the sludge samples. This method estimated the quantities of gases emitted during, and immediately after, transfer of sludge to the holding tanks. Fluxes were calculated under the assumption that the total gas potential of the sludge was realized by aeration stripping during the period of storage prior to final processing.

Second, fluxes resulting from the generation of gases within the sludge holding tanks, and stripped from the liquid during aeration, were determined using the aerated surface collection method described previously. This flux was assumed to be continuously present as a secondary background flux. In addition, emissions from the surface of the nonaerated area of the sludge storage tanks were evaluated using the closed chamber technique described previously.

Gas Analysis. All samples were analyzed as described by Crill (8). Samples were immediately returned to the lab, allowed to equilibrate to laboratory temperature for 1–2 h, and analyzed for CH₄ and CO₂ within 3–4 h after collection. Samples and standards were dried across CaSO₄ as they were injected into the gas chromatograph (GC) injection loop. Analyses for CH₄ were accomplished using a flame ionization detector (FID-GC), while a thermal conductivity detector (TCD-GC) was used to analyze for CO₂ after constituent separation using 2 m × 3.2 mm o.d. columns packed with Poropak Q or HayeSep Q. Carrier gases were helium for TCD-GC and nitrogen for FID-GC. Peaks were quantified with Hewlett Packard recording integrators. CH₄ and CO₂ standard gases were obtained from Scott Specialty Gases and were calibrated with a National Institute of Standards and Technology certified standard. Precision of analysis for calibration gases was 0.3%.

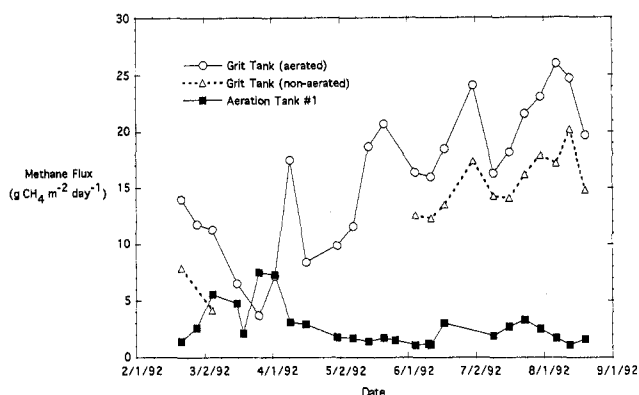


Figure 2. Measured CH_4 fluxes from the aerated and nonaerated areas of the grit tank and secondary aeration tank no. 1.

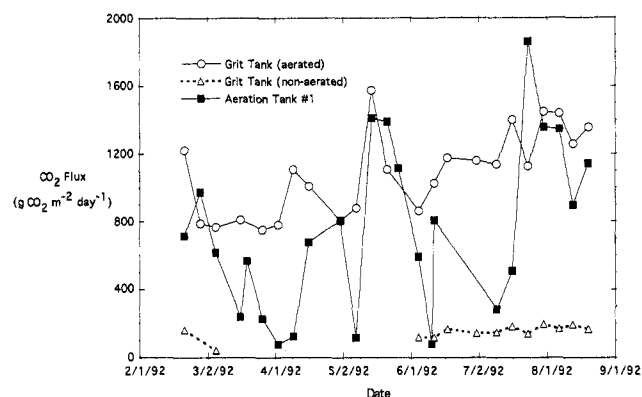


Figure 3. Measured CO_2 fluxes from the aerated and nonaerated areas of the grit tank and secondary aeration tank no. 1.

Statistical Analysis. Emissions data were examined for statistical distribution to determine the appropriate form of statistical analysis. CH_4 flux data from the secondary aeration tank no. 1 was found to be log normally distributed, requiring the application of parametric statistics to the data in log-transformed form. All other sample data were determined to have been drawn from normally distributed populations, permitting application of parametric statistics.

Relationships between emissions and temperature were examined using linear regression techniques (6). A test was also performed to accept or reject the hypothesis that the slopes of the regression lines of emissions as a function of temperature, from aerated and nonaerated areas of the grit tanks, were equal.

Results

Grit Tanks (Aerated). CH_4 and CO_2 fluxes from the aerated area of the grit tanks, measured under normal operating conditions, are presented in Figures 2 and 3. CH_4 fluxes ranged from a minimum of $3.7 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $26 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$. CO_2 emissions ranged from a minimum of $751 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $1577 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$. Fluxes of $39.5 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ and $2281 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ were measured in the aerated area of the grit tanks after the disposal of a septic tank load by a local septage hauler. These septage disposal events occur several times during a typical week at which time gas emissions are elevated. However, these events last only a single detention period and do not contribute significantly to total weekly emissions and were disregarded.

Grit Tanks (Nonaerated). CH_4 and CO_2 fluxes from the nonaerated areas of the grit tanks, measured under normal operating conditions, are presented in Figures 2 and 3. CH_4 fluxes ranged from a minimum of $4.2 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $20.2 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$. CO_2 emissions ranged from a minimum of $42 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $194 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$. As in the aerated areas, higher flux values of $20.6 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ and $284 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ were measured from samples collected during the septage disposal event described previously and are not representative of normal plant operations and were disregarded.

Secondary Treatment Tanks (Aerated). Wastewater from the primary settling tanks and return sludge enter the secondary aeration system in tank no. 1. Preliminary measurements established tank no. 1 as the significant CH_4 source in the four-tank aeration system. Samples were also collected from tank nos. 2–4 to determine emissions for use in organic carbon mass balance calculations. Gases generated and dissolved in the liquid during primary settling, as well as gases generated in the return sludge during settlement in the secondary clarification tanks, are later stripped from the liquid column by aeration. CH_4 fluxes from tank no. 1 ranged from a minimum of $1.1 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $7.5 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$. CO_2 emissions ranged from a minimum of $76 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $1862 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ (Figures 2 and 3). CH_4 fluxes from tank nos. 2–4 ranged from a minimum of $0.2 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $1.2 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$. CO_2 fluxes from tank nos. 2–4 ranged from a minimum of $627 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $2329 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$.

Sludge Holding Tanks. Approximately $2.4 \times 10^6 \text{ L}$ of sludge from the primary and secondary treatment processes are transferred to the sludge holding tanks annually. The sludge is aerated in the holding tanks for several days to reduce the liquid content prior to transfer to the sludge processing facility. Gas samples collected from the aerated surface early in the study indicated highly variable fluxes dependent on the time since sludge transfer. Fluxes from the aerated surface as high as $86 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ and $12749 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ were detected immediately after transfer to the holding tanks, while fluxes of $6.9 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ and $2103 \text{ g of CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ were measured from a batch of sludge after 1 day of aeration.

The flux potential of gases dissolved in the sludge was estimated by determining total dissolved CH_4 and CO_2 per unit volume of sludge and was assumed to be fully realized. CH_4 concentrations ranged from a minimum of $0.0017 \text{ g of CH}_4 (\text{L of sludge})^{-1}$ to a maximum of $0.0035 \text{ g of CH}_4 (\text{L of sludge})^{-1}$. CO_2 concentrations ranged from a minimum of $0.025 \text{ g of CO}_2 (\text{L of sludge})^{-1}$ to a maximum of $0.081 \text{ g of CO}_2 (\text{L of sludge})^{-1}$.

Gas fluxes from stored sludge were measured from the aerated areas of the storage tanks to determine the flux of gases due to production during storage. Only samples collected a minimum of 1 day after sludge transfer were considered appropriate to estimate this secondary flux. A minimum of 1 day of aeration was considered necessary for the depletion of gases dissolved in the sludge during accumulation and transfer. Data collected on seven sampling dates met this criteria. CH_4 fluxes ranged from a minimum of $6.9 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ to a maximum of $10.9 \text{ g of CH}_4 \text{ m}^{-2} \text{ day}^{-1}$. CO_2 fluxes ranged from a minimum

of 954 g of CO₂ m⁻² day⁻¹ to a maximum of 5208 g of CO₂ m⁻² day⁻¹. The nonaerated areas of the storage tanks were also examined as a possible emissions source. Fluxes from the nonaerated areas were found to be less than 10% of those from the aerated areas and were disregarded.

Discussion

Methane is generated in wastewater treatment systems in areas of high biological oxygen demand and low oxygen concentration such as the influent lines, primary settling tanks, secondary clarification tanks, sludge holding tanks, and sludge transfer lines. The full potential of dissolved CH₄ is realized in areas of mechanical aeration where gases are stripped from the liquid by diffusion. In the Durham plant, CH₄ generated in the influent lines was emitted in the aerated areas of the grit tanks by aeration and in the nonaerated areas as the result of turbulent liquid flow initiated by aeration and the subsequent flow of fine bubbles into the nonaerated areas. Some CH₄, generated in the nonaerated liquid, may also have contributed to total grit tank emissions.

CH₄ generated in the primary settling tanks and in the returned sludge was stripped by aeration from the liquid in the secondary aeration tanks. The highest emissions of CH₄ from the secondary aeration system were measured in the first tank where the return sludge and wastewater enter the system. CH₄ emissions decreased as the liquid moved sequentially through the four-tank process and were negligible after transfer of the liquid to the secondary clarifiers.

CH₄ generated in the wasted sludge during storage was emitted during aeration in the holding tanks immediately after transfer. Additional CH₄ was generated in the sludge in the nonaerated areas of these tanks during the 3-day detention period and was stripped from the liquid column as it flowed into the aerated areas.

Annual Emissions. The mean fluxes of CH₄ and CO₂ measured from the aerated area of the grit tanks were 15.9 g of CH₄ m⁻² day⁻¹ and 1087 g of CO₂ m⁻² day⁻¹. During the local university school year, from September through May, two grit tanks are operated in parallel with a total aerated surface area of 11.2 m². During the summer months, from June to August, wastewater flow and organic load are reduced by approximately one-half due to the local population reduction. Primary treatment capacity during the summer low flow period is subsequently reduced by the shutdown of one grit tank, and the total aerated surface area drops to 5.6 m². These reductions preserve the continuity of emissions from the grit tank, requiring only an area weighting in total annual flux calculations. The population weighted area is 9.8 m². The estimated total area-weighted annual emissions from the aerated area of the grit tanks were 5.7 × 10⁴ g of CH₄ year⁻¹ and 3.9 × 10⁶ g of CO₂ year⁻¹.

Mean fluxes measured from the nonaerated area of the grit tanks were 11.1 g of CH₄ m⁻² day⁻¹ and 131 g of CO₂ m⁻² day⁻¹. Total nonaerated surface area was 26 m² from September through May and 13 m² from June through August, yielding a population-weighted surface area of 22.8 m². The estimated total area-weighted annual emissions from the nonaerated area of the grit tanks were 9.2 × 10⁴ g of CH₄ year⁻¹ and 1.1 × 10⁶ g of CO₂ year⁻¹.

Mean CH₄ and CO₂ fluxes from secondary aeration tank no. 1 were 2.7 g of CH₄ m⁻² day⁻¹ and 747 g of CO₂ m⁻²

Table I. Process and Total CH₄ and CO₂ Fluxes (g year⁻¹) from Durham, NH, Wastewater Plant

process	CH ₄	CO ₂
grit tank (aerated)	5.7 × 10 ⁴	3.9 × 10 ⁶
grit tank (nonaerated)	9.2 × 10 ⁴	1.1 × 10 ⁶
aeration tanks	2.2 × 10 ⁵	3.6 × 10 ⁸
sludge storage tank	6.3 × 10 ⁴	2.1 × 10 ⁷
total	4.3 × 10 ⁵	3.9 × 10 ⁸

day⁻¹. All four secondary aeration tanks remained operational throughout the sample collection period and are normally operational throughout the year with single-tank-aerated surface areas of approximately 144 m². The estimated total annual emissions from tank no. 1 were 1.4 × 10⁵ g of CH₄ year⁻¹ and 3.9 × 10⁷ g of CO₂ year⁻¹. The estimated CH₄ emissions from tank nos. 2–4 were 2.5 × 10⁴, 1.4 × 10⁴, and 4.1 × 10⁴ g of CH₄ year⁻¹, respectively. The estimated CO₂ emissions from tank nos. 2–4 were 1.0 × 10⁸, 1.2 × 10⁸, and 1.0 × 10⁸ g of CO₂ year⁻¹, respectively. The total estimated CH₄ emissions from all secondary aeration tanks was 2.2 × 10⁵ g of CH₄ year⁻¹, and the total estimated CO₂ emissions from all secondary aeration tanks was 3.6 × 10⁸ g of CO₂ year⁻¹.

The mean sludge sample gas concentrations were 0.0025 g of CH₄ (L of sludge)⁻¹ and 0.049 g of CO₂ (L of sludge)⁻¹. Approximately 2.4 × 10⁶ L of sludge is transferred annually to the sludge holding tanks for aeration. Assuming complete removal of dissolved gases by aeration yields annual fluxes from sludge transfer of 6.0 × 10³ g of CH₄ year⁻¹ and 1.2 × 10⁶ g of CO₂ year⁻¹.

Mean secondary CH₄ and CO₂ fluxes from the aerated areas of the sludge holding tanks were 8.3 g of CH₄ m⁻² day⁻¹ and 3079 g of CO₂ m⁻² day⁻¹. Two of the three holding tanks were typically utilized for sludge storage on a continuous basis. The total aerated surface area was 18.6 m², yielding annual fluxes from sludge storage due to secondary production of 5.7 × 10⁴ g of CH₄ year⁻¹ and 2.1 × 10⁷ g of CO₂ year⁻¹. Total estimated annual fluxes from sludge transfer and storage were 6.3 × 10⁴ g of CH₄ year⁻¹ and 2.1 × 10⁷ g of CO₂ year⁻¹.

The estimated total process CH₄ and CO₂ fluxes are summarized in Table I. The total estimated annual CH₄ and CO₂ fluxes from the Durham, NH, wastewater treatment plant are 4.3 × 10⁵ g of CH₄ year⁻¹ and 3.9 × 10⁸ g of CO₂ year⁻¹.

Temperature Relationships. A reliable predictive linear relationship between CH₄ and CO₂ fluxes and wastewater temperature was observed in the aerated and nonaerated areas of the grit tanks. CH₄ emissions from the aerated and nonaerated areas of the grit tanks were found to be significantly correlated with liquid temperature (Figure 4). The coefficients of determination for CH₄ flux versus temperature in the aerated and nonaerated areas of the grit tanks were 0.74 and 0.76, respectively, and the associations between flux and temperature were found to be highly significant.

CO₂ emissions from the aerated and nonaerated areas of the grit tanks were also found to be significantly correlated with wastewater temperature. The coefficients of determination for CO₂ flux versus temperature in the aerated and nonaerated areas of the grit tanks were 0.51 and 0.63, respectively, and the associations between flux and temperature were found to be highly significant.

No statistically significant correlation (*r*² = 0.11) was observed in secondary aeration tank no. 1 between CO₂

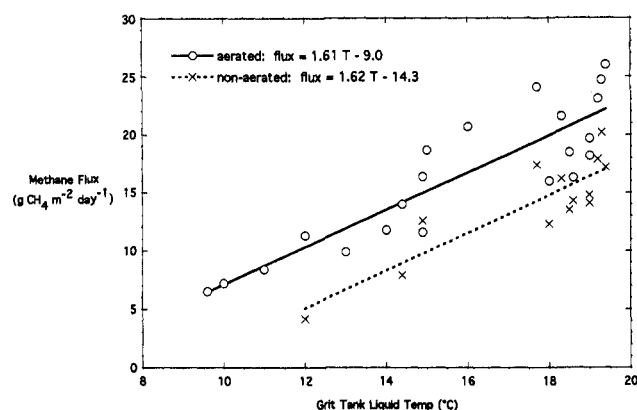


Figure 4. Temperature response of methane flux in the aerated and nonaerated area of the grit tanks; $r^2 = 0.78$ for aerated flux and $r^2 = 0.76$ for nonaerated flux.

Table II. Estimated Emissions to Atmosphere from Significant CH₄-Producing Wastewater Treatment Processes in the U.S. during 1988^a

process	U.S. emissions (10 ⁹ g of CH ₄ year ⁻¹)
primary treatment	5.6
activated sludge treatment	7.9
landfilling of sludge	40*
septic systems	44*

^a Anaerobic digestion is estimated to have generated 837 Gg of CH₄ in digester vessels. The fraction of digester gas that escaped to the atmosphere from the vessels is unknown at this time but may be a significant emission source. *, indicates emission estimates assuming a CH₄ oxidation rate in cover soils of 70%.

emissions and wastewater temperature, while a very marginally significant correlation ($r^2 = 0.23$) was observed between CH₄ emissions and wastewater temperature.

Emission Factors. A weighting must be applied to calculate the population served by the wastewater treatment plant due to annual variations in the number of town residents. Approximately 12 500 people from September through May and approximately 6200 from June through August generate wastes that are treated at this facility. The time-weighted average of this population is 10 925 people, which yields per capita direct gas fluxes of 39 g of CH₄ person⁻¹ year⁻¹ and 35 698 g of CO₂ person⁻¹ year⁻¹ from wastewater treatment plants with typical primary and activated sludge treatment processes.

Flow-based emission rates can also be determined. Based on an annual wastewater flow of 1.1×10^9 L year⁻¹, annual gas fluxes are estimated to be 1.4×10^{-4} g of CH₄ (L of wastewater)⁻¹ and 4.5×10^{-3} g of CO₂ (L of wastewater)⁻¹ for the primary treatment system and 2.0×10^{-4} g of CH₄ (L of wastewater)⁻¹ and 0.33 g of CO₂ (L of wastewater)⁻¹ for the activated sludge secondary treatment system.

U.S. Emissions. Total annual CH₄ emissions from wastewater treatment in the United States can be estimated using our emission factors and national wastewater and sludge handling statistics. Table II shows estimated national CH₄ production and emission values for significant CH₄-producing wastewater treatment and sludge disposal processes. Wastewater flow through primary treatment in the United States in 1988 was estimated to be 4.1×10^{13} L year⁻¹ (9). This study also found return-activated sludge to be the most widely used method of secondary treatment with a flow of 3.9×10^{13} L year⁻¹.

Total CH₄ emissions from these systems, based on the Durham wastewater treatment plant emission factors, are estimated to be 5.6×10^9 g of CH₄ year⁻¹ from primary treatment and 7.9×10^9 g of CH₄ year⁻¹ from secondary activated sludge treatment.

The treatment and disposal of sludge presents a significant potential for CH₄ emissions nationally. The treatment of sludge by anaerobic digestion utilizes microorganisms to mineralize a portion of the organic fraction of a sludge. The fraction of the sludge volatile solids (VS) converted to digester gas varies, depending on the digester detention time and type of digester, and typically ranges from 50 to 70% (10). The gas generated is composed of approximately 65% CH₄, 30% CO₂, and other trace gases. A total of 3.1×10^{13} L of sludge is estimated to have been treated by anaerobic digestion in 1988, while 638.4 g of CH₄ (kg of volatile solids destroyed)⁻¹ is estimated to be generated during the anaerobic digestion of sludge (4, 9). The total quantity of digester gas generated annually in the U.S. can be estimated assuming a typical sludge dry solids generation rate of 0.12 g of dry solids (L of sludge)⁻¹, a volatile solids content of 70% of total solids, and a typical rate of volatile solids destroyed during digestion of 50% (4, 10). This yields an estimate of 0.84 Tg of CH₄ produced per year by anaerobic digestion. Digester gas is normally utilized on site as fuel for space or process heating or it is flared. However, digester system leaks, collection system leaks, and flaring system upsets can release uncombusted CH₄. The fraction of CH₄ produced nationally that escapes to the atmosphere is unknown at this time, but could be a significant emission source.

The method of ultimate sludge disposal will also have an impact on the total CH₄ potential from sludge. The most widely used sludge disposal methods include landfill burial, incineration, and land application. Landfill burial is the only method that provides the anaerobic environment necessary for the large-scale conversion of sludge organic matter to CH₄. An estimated 4.5×10^{13} L of sludge was disposed of in landfills in 1988 (9). Assuming a dry solids generation rate of 0.12 g of dry solids (Liter of sludge)⁻¹ yields total dry solids landfilled of 4.2×10^{12} g year⁻¹. A significant, but unknown, fraction of that sludge will have been digested prior to landfilling and will have contributed a reduced quantity of volatile solids to the total sludge content. If a total volatile solids content of 50% of total solids is presumed, and carbon comprises 40% of the volatile solids, 8.5×10^{11} g of carbon as sludge is buried in landfills annually. Current landfill gas models assume an average time constant for the conversion of readily decomposable organic matter to gas of 3–4 years (11, 12). If this conversion is presumed constant over the period of degradation, and CH₄ content is typical of landfill gas (55% CH₄), 1.3×10^{11} g of CH₄ are produced annually from sludge. This quantity of CH₄ will be produced each year, typically for 3–4 years, from a single annual batch of landfilled sludge. The actual steady-state annual emissions from landfills will be the sum of emissions from each annual sludge batch. Assuming a constant rate of sludge disposal, 4.5×10^{11} g of CH₄ is produced from sludge in landfills annually. The quantity of CH₄ produced that actually reaches the atmosphere is currently a point of conjecture. Much of the methane produced in landfills may never reach the atmosphere due to oxidation by methanotrophs that may be as high as 70% (13). At an oxidation rate of 70% of CH₄ produced, 4.0×10^{10} of CH₄

would be emitted from U.S. landfills each year, for 3–4 years, from the sludge landfilled in 1988. Prior and subsequent annual batches would have an additive effect on total yearly CH₄ emissions.

The conversion of solids in individual septic systems must also be considered in the estimation of CH₄ emissions from wastewater treatment systems. In 1987, an estimated population of 58 675 800 were served by septic systems in the United States (14). Assuming a typical volatile solids loading rate of 10.6 g VS person⁻¹ day⁻¹ yields a mass of volatile solids collected annually of 2.3×10^{11} g (4). If the previously stated rate of CH₄ production due to anaerobic digestion is assumed and 70% of methane was oxidized in septic tank soil covers, 4.4×10^{10} g of CH₄ was emitted by septic systems in 1988.

Our results are a first step to a quantitative national inventory of CH₄ emissions from wastewater treatment and should be viewed as an approximation of the order of magnitude of these emissions. Sampling a wide variety of treatment technologies and operating conditions will be necessary to better refine these estimates. Sludge handling and processing appear to hold the greatest potential for uncertainty in emission estimates. Large quantities of methane are known to be produced as a result of the anaerobic digestion of sludge, and anecdotal evidence suggests that some of this methane escapes to the atmosphere. However, the magnitude of fugitive emissions is unknown. A large-scale assessment of digester integrity will be required to quantitatively assess the fraction of CH₄ lost.

Also, wastewater and sludge lagoons are a known source of methane emission that have not been quantitatively evaluated. Lagoons are used worldwide for wastewater treatment, in many different configurations, and will require a detailed evaluation of emissions to accurately estimate their impact on national and global methane budgets.

Acknowledgments

We would like to thank Antonio Nobre, Tim Finnegan, and Paul Carroll for their invaluable assistance in the field. We also thank the staff of the Durham, NH wastewater treatment facility for their helpful cooperation. This

research was supported by the National Aeronautics and Space Administration Interdisciplinary Research Program.

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Received for review February 12, 1993. Revised manuscript received July 15, 1993. Accepted August 3, 1993.*

* Abstract published in *Advance ACS Abstracts*, September 15, 1993.