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Methane and nitrous oxide emissions following anaerobic digestion of sludge in Japanese sewage treatment facilities

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HIGHLIGHTS

- CH_4 and N_2O emissions after anaerobic sludge digestion were investigated.
- CH_4 mission factor was 509 ± 72 mg/m^3-influent and especially high in winter.
- N_2O emission factor was 7.1 ± 2.6 mg/m^3-influent and lower than CH_4 emission.
- The highest CH_4 emissions were during dewatering, followed by continued digestion.
- CH_4 and N_2O emissions after anaerobic digestion are considered to be significant.

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ABSTRACT

Methane (CH_4) and nitrous oxide (N_2O) are potent greenhouse gases with global warming potentials (expressed in terms of CO_2-equivalents) of 28 and 265, respectively. When emitted to the atmosphere, they significantly contribute to climate change. It was previously suggested that in wastewater treatment facilities that apply anaerobic sludge digestion, CH_4 continues to be emitted from digested sludge after leaving the anaerobic digester. This paper studies the CH_4 and N_2O emissions from anaerobically digested sludge in the subsequent sludge treatment steps. Two full-scale treatment plants were monitored over a 1-year period. Average emissions of CH_4 and N_2O were 509 ± 72 mg/m^3-influent (wastewater) and 7.1 ± 2.6 mg/m^3-influent, respectively. These values accounted for 22.4 ± 3.8% of the indirect reduction in CO_2-emissions when electricity was generated using biogas. They are considered to be significant.

1. Introduction

The anaerobic digestion of sewage sludge produces biogas consisting mainly of methane (CH_4), which can be used as an energy source. However, the continuation of CH_4-emissions to the atmosphere from digested sludge during the subsequent sludge treatment steps may significantly contribute to global warming. Indeed, the 100-year global warming potential of CH_4 is 28, meaning that each emitted molecule of CH_4 contributes 28 times more to global warming than an emitted carbon dioxide (CO_2) molecule during this period (IPCC, 2013). CH_4 produced by sludge is, moreover, combustible, hence giving rise to fire and explosion hazards (Astbury, 2008). Whereas CH_4 emissions from composting and wetlands have been reported before in some papers (e.g., Maulini-Duran et al., 2013; Uggetti et al., 2012), little information is available on the emission of CH_4 during the dewatering process after anaerobic digestion (IPCC, 2002). Additionally, nitrous oxide (N_2O) has an even higher 100-year global warming potential of 265 (IPCC, 2013), and is released as a by-product of the nitrification and/or denitrification processes. Here again, only very few studies are available on N_2O emissions following the anaerobic digestion of sludge (Czepiel et al., 1995; Scherson et al., 2014; Toyoda et al., 2011; Tallec et al., 2008).

In Japan, the current emission factor for CH_4 from sewage sludge dewatering amounts 106 mg/m^3-influent (wastewater) (Ministry of the Environment, Japan, 2006). This factor, which is frequently used in estimations for greenhouse gas inventories, was determined from an investigation of treatment plants 20 years ago (Sato et al., 1992; Takeishi et al., 1993) and is based on two assumptions: (i) an anaerobic sludge digestion process is installed before the sludge dewatering, and (ii) all of the dissolved CH_4 in

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digested sludge is emitted during sludge dewatering. Because of these assumptions, all CH_4 generated in the sludge between the anaerobic digestion and the sludge dewatering process was included in the current emission factor. Also, the obtained value of 106 mg/m^3-influent is not applicable for sewage treatment plants in which anaerobic sludge digestion is not employed. A N_2O emission factor for sludge dewatering is not yet available (Ministry of the Environment, Japan, 2006).

Given the contributions of CH_4 and N_2O emissions to global warming, the production of these gases after anaerobic sludge digestion may considerably reduce the environmental benefit of using biogas as an energy source to indirectly reduce CO_2 emissions. A reliable quantification of the CH_4 and N_2O emissions between the anaerobic digestion and sludge dewatering processes is necessary to be able to calculate and take into account these effects. Several recent studies reporting life-cycle assessments on sludge treatment (including anaerobic digestion), did not take into account this aspect and only examined the fugitive biogas emissions by incomplete combustion and failure of combustion (Brown et al., 2010; Cao and Pawłowski, 2013; Patterson et al., 2011; Venkatesh and Elmi, 2013). In Europe, Daelman et al. (2012) concluded that CH_4 generated by the wastewater and sludge treatment processes has a high global warming potential in CO_2 equivalents, and that its impact on global warming reduces the indirect reduction in CO_2 emissions derived from biogas with sewage treatment as a fuel source.

The present study quantified the emissions of CH_4 and N_2O from the anaerobic digestion and sludge dewatering processes and used these data to update the emission factors in Japan. More specific, gaseous CH_4 and N_2O emissions in the unit operations related to the sludge dewatering process were measured (these process steps where these gases are most likely to be emitted). Also the water removed from the sludge during the dewatering process was analyzed for dissolved CH_4 and N_2O. The CH_4 and N_2O emission factors and their influence on the indirect reductions in CO_2 emissions derived from using biogas as a fuel source at two sewage treatment facilities over a 1-year period were also quantified.

2. Methods

2.1. Sewage treatment facilities

Flow diagrams for the sewage treatment facilities and locations of sample collection/measurement points are provided in Fig. 1. Two sewage facilities were selected for this study: one facility employing anaerobic sludge digestion (H) and one without (N). The total amount of wastewater annually treated in the two facilities was very similar: 38,005,000 m^3/year and 38,745,489 m^3/year for facilities H and N, respectively (Japan Sewage Works Association, 2010). Information on the wastewater composition in both facilities throughout the measurement campaign (between April 2011 and March 2014) is provided in the Supplementary information (Table S1). Although, total solids (TS), volatile total solids (VTS), suspended solids (SS) and biochemical oxygen demand (BOD) of the influent in facility H were higher than in facility N, the total nitrogen concentrations and relative composition of the influent in both facilities were comparable. In facility H, the produced sludge is subjected to a mesophilic anaerobic digestion [hydraulic retention time (HRT): 36 days; average temperature: 39 °C; organic matter decomposition ratio: 64.6%] after thickening. Most of the biogas generated during anaerobic digestion is used as auxiliary fuel for sludge incineration. The digested sludge is dewatered using a belt-filter press, producing a sludge cake containing 84% water. The dewatered sludge is then combusted in a fluidized bed incinerator. The off-gas from the dewatering process is deodorized using activated carbon in the exhaust ventilation system. In facility N, the sludge is directly dewatered (without prior anaerobic digestion) using a screw-type dewatering (achieving a sludge cake with a water content of 79%), also followed by fluidized bed incinerator. The off-gas from the dewatering process is biologically deodorized in the exhaust ventilation system.

2.2. Sampling point and dates

Sampling was done at those points in the sewage treatment facility that represent potential sources of CH_4 and N_2O (Fig. 1). Facility H uses an open-type storage tank for the digested sludge, and CH_4 emissions from the surface of the digested sludge were measured using the floating closed-chamber method (Czepiel et al., 1993). CH_4 and N_2O concentrations were also measured in the off-gas and in the water separated from the sludge during the dewatering process. In facility N (not applying anaerobic digestion), CH_4 and N_2O were measured in the off-gas and in the water separated from the sludge during the dewatering process. Odorous off-gas from the storage tank of the sludge cake is used as combustion air in the incineration chamber, hence this gas stream was not included in the measurements as all components are further oxidized during the combustion. Also, samples of digested sludge in facility H and thickened sludge in facility N were collected and their CH_4 and N_2O potentials were compared. On-site sampling and measurements were carried out during two winter and two summer periods: November 2011–March 2012 (winter), August–September 2012 (summer), July–September 2013 (summer), and December 2013–January 2014 (winter).

2.3. Measurements of CH_4 in the digested sludge storage tank

Flux emissions of CH_4 from the storage tank of digested sludge in facility H were measured using the floating closed chamber method (Figs. S2 and S3 in the Supplementary information), consisting of a cylindrical case made of polyvinyl chloride (PVC) and a float around the case composed of polyethylene foam. Two ports were located on the upper side of the case; one of these was used for gas sampling, and the other was attached to a 1-L gas bag inside the case to maintain constant air pressure.

Measurements of CH_4 were carried out during daytime for a total of 8 days in winter and 6 days in summer. The weather was clear on all measurement dates. The floating closed chamber was used at 4–6 points on the sludge surface in the storage tank during 1 day. The temperature inside the chamber was recorded, and gas emitted from the sludge was continuously analyzed for CH_4 concentration using a CH_4 gas detector (XP-3160; New Cosmos Electric Co., Ltd., Osaka, Japan); the gas flow rate and sampling time were 200 mL/min and 5 min, respectively. Measurements of CH_4 were used to calculate rates of CH_4 production (dC/dt), and CH_4 fluxes ($\mu mol/m^2/s$) from the surface of the digested sludge, according to Czepiel et al. (1993). The method of calculation is included in Supplementary information (Section S.3).

N_2O emissions were also measured, but all measurements were below the detection limit and, therefore, will not be discussed further.

2.4. Measurement of CH_4 and N_2O in water separated from sludge during the dewatering process

The water separated from the sludge during the dewatering process in both sewage treatment facilities was analyzed for CH_4 and N_2O. Sampling was done over the course of 6 days in winter and 3 days in summer.

The analytical procedures outlined in Hatamoto et al. (2010) were followed for quantifying CH_4 and N_2O dissolved in water separated from sludge during the dewatering process. Specifically, water samples were collected in 122 mL vials by overflowing the