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Research article

Nitrous oxide and methane emissions during storage of dewatered digested sewage sludge

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ABSTRACT

This study investigated the effect on greenhouse gas emissions during storage of digested sewage sludge by using a cover during storage or applying sanitisation measures such as thermophilic digestion or ammonia addition. In a pilot-scale storage facility, nitrous oxide and methane emissions were measured on average twice monthly for a year, using a closed chamber technique. The thermophilically digested sewage sludge (TC) had the highest cumulative emissions of nitrous oxide (1.30% of initial total N) followed by mesophilically digested sewage sludge stored without a cover (M) (0.34%) and mesophilically digested sewage sludge stored with a cover (MC) (0.19%). The mesophilically digested sewage sludge sanitised with ammonia and stored with a cover (MAC) showed negligible cumulative emissions of nitrous oxide. Emissions of methane were much lower from TC and MAC than from M and MC. These results indicate that sanitisation by ammonia treatment eliminates the production of nitrous oxide and reduces methane emissions from stored sewage sludge, and that thermophilic digestion has the potential to reduce the production of methane during storage compared with mesophilic digestion. The results also indicate a tendency for lower emissions of nitrous oxide and higher emissions of methane from covered sewage sludge compared with non-covered.

1. Introduction

Sewage sludge (SS) from municipal wastewater treatment contains plant nutrients and organic matter that can be beneficial in agriculture. Approximately 12,970,000 Mg dry matter (DM) of SS are produced in the EU28 annually, approximately 38% of which is applied to arable land (Eurostat, 2012). In Sweden, 200,000 Mg (DM) are produced annually, of which approximately 23% is applied to arable land (Eurostat, 2012). Most sewage sludge produced in Sweden is anaerobically digested, and this is also the most common stabilization process in EU (Smith, 2009). A goal of recycling 40% of the P and 10% of the N in SS to arable land has been proposed by Swedish Environmental Protection Agency (SEPA), but not yet agreed by the government. Owing to the risk of occurrence of human pathogens in SS, SEPA has previously suggested that SS should be sanitised before being used on land and that storage for one year would meet the minimum requirement for sanitisation (class C) (Swedish Environmental Protection Agency, 2002). In a proposal in 2013, storage for one year was no longer considered sufficient for sanitisation of SS prior to use on arable land and specific sanitisation measures, such as digestion under thermophilic temperatures (45–65 °C) or treatment with ammonia (NH_3), were suggested instead (Swedish Environmental Protection Agency, 2013). The latter can be achieved with addition of urea ($(NH_2)_2CO$).

The waste and wastewater industry contributes approximately 3% of global anthropogenic emissions of greenhouse gases, according to the Intergovernmental Panel on Climate Change (IPCC, 2007). Storage of SS may lead to high emissions of nitrous oxide (N_2O) and methane (CH_4) (Flodman, 2002;). The global warming potential for CH_4 and N_2O corresponds to 34 and 298 carbon dioxide (CO_2)-equivalents, respectively, in a 100-year perspective (IPCC, 2013). According to Flodman (2002), annual emissions of N_2O and CH_4 from sludge storage would comprise around 5% and 0.1% of total Swedish anthropogenic emissions of N_2O and CH_4, respectively, if all the SS in Sweden were stored for one year.

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In Sweden, the dominant process for stabilising SS is by digestion at 25–45 °C (mesophilic digestion), with a hydraulic retention time (HRT) in the digester of 15–30 days. However, digestion can also be conducted at other temperatures, *e.g.* 45–65 °C (thermophilic digestion) or <20 °C (psychrophilic digestion). During digestion, various groups of anaerobic microorganisms convert organic carbon (C) to CH_4 and CO_2 and there is a risk of emissions of these gases during subsequent storage of the digested SS. Previous studies on the storage of similar organic fertilisers, such as solids separated from pig slurry and deep litter pig manure, show that CH_4 emissions are positively related to temperature, with higher temperatures during storage leading to increased emissions (Hansen et al., 2006). This is evident in practice as *e.g.* seasonal variations in emissions from stored cattle slurry (Rodhe et al., 2009). N_2O emissions can also be positively correlated to temperature, as demonstrated in a study on SS storage by Majumder et al. (2014).

Covering organic fertilisers such as SS during storage has been shown to reduce emissions of N_2O, most probably because the cover reduces aeration and prevents the material from drying, as shown *e.g.* for solid manure (Hansen et al., 2006). However, since covering organic fertiliser heaps has also been shown to decrease (Chadwick, 2005; Hansen et al., 2006) and occasionally also to increase CH_4 emissions (Chadwick, 2005), further studies are required. Anaerobic conditions due to the SS being covered promote CH_4 production, but the cover could also lead to lower temperatures and hence a reduction in emissions (Chadwick, 2005).

In high concentrations, NH_3 has a sanitising effect as the NH_3 molecule can diffuse across microbial cell membranes, causing the pH to increase inside the cell, which affects the ion balance and produces a toxic effect (Schneider et al., 1996). Ammonia treatment can be achieved by the addition of urea which is hydrolysed to form ammonium (NH_4^+), which in turn is protolysed to form NH_3. If the SS is covered in an efficient manner so that NH_3 is not lost, the sanitising effect is expected to continue throughout the storage period and hence pathogenic microorganisms will not be able to regrow. However, high pH also increases the risk of NH_3 being emitted to the atmosphere (Brady and Weil, 2008).

Nitrifying bacteria rely on mineralised nitrogen (N) as their principal energy source. The major genera of nitrifying bacteria in wastewater treatment processes are *Nitrosomonas, Nitrobacter* and *Nitrospira* (Siripong and Rittmann, 2007). These strictly aerobic bacteria are sensitive to high temperatures (Jiang and Bakken, 1999; Grunditz and Dalhammar, 2001) and also to high concentrations of NH_3 (Anthonisen et al., 1976). Their activity, and thus N_2O emissions, could therefore be reduced by anaerobic digestion at high temperatures (thermophilic digestion) and by NH_3 sanitisation. Methanogens are also sensitive to high levels of NH_3 (Hansen et al., 1998), and thus NH_3 sanitisation should decrease CH_4 emissions.

Very little experimental data are available on N_2O and CH_4 emissions from storage of digested SS, and therefore the contribution from this source cannot be estimated with any accuracy. The overall aim of this study was to characterise emissions of N_2O and CH_4 during storage of dewatered digested SS and to test the hypotheses that: (1) covering SS reduces N_2O emissions during storage and (2) sanitisation of SS with NH_3 (by addition of urea) or by thermophilic digestion reduces emissions of N_2O and CH_4 during storage.

2. Materials and methods

2.1. Site description and experimental design

A pilot storage facility consisting of 12 high-density polyethylene cylinders, 2 m high and 1.6 m in diameter with a horizontal surface area of 2.1 m^2 and a volume of 4.2 m^3, were set up 3 km north-east of Uppsala (59°50′N, 17°39′E), Sweden. To mimic the thermal conditions in full-scale storage, the cylinders were surrounded by mesophilically digested and dewatered SS about 1.5 m high. The experiment was organised in a completely randomised block design with three replicates (blocks) and four treatments: mesophilically digested SS stored with no cover (M) or with a cover (MC), ammonia-treated (by addition of 1.5% urea by weight) mesophilically digested SS stored with a cover (MAC) and thermophilically digested SS stored with a cover (TC).

The mesophilically digested SS used in treatments M, MC and MAC originated from the municipal wastewater treatment plant in Uppsala, while the thermophilically digested SS used for treatment TC originated from the municipal wastewater treatment plant in Sunne (59°50′N, 13°8′E). Both SS types were mixtures of SS from primary (mechanical), secondary (biological) and tertiary (phosphorus removal) treatment steps. Phosphorus at Uppsala is removed with ferric chloride (PEX111, Kemira Oyj, Helsinki, Finland), while phosphorus at Sunne is precipitated with aluminium chloride (Ekoflock 90, Feralco Nordic AB, Helsingborg, Sweden). The SS is then dewatered by addition of polymers. For pre-dewatering, Zetag 7557 (BASF, Ludwigshafen am Rhein, Germany) is used at Uppsala. For final dewatering, Superfloc C-498 and Sedifloc 1060c (Kemira Oyj, Helsinki, Finland) are used at Uppsala and Sunne, respectively. Other sludge properties are presented in Table 1.

The cover used was 0.47–0.57 mm thick polyester fabric covered with PVC, with a raised peripheral edge to prevent precipitation on the cover from entering the stored SS. The cover rested directly on the surface of the sludge. Excess water was removed with a wet vacuum cleaner when necessary.

The MAC treatment involved mixing urea (AB Hanson och Möring, Halmstad, Sweden) into mesophilically digested SS just before filling the cylinders. All SS was weighed and the cylinders were filled using a tractor loader to approximately 1.3 m height on 14–15 September 2011, after which the first gas sampling for determination of CH_4 and N_2O emissions was conducted. In total, emissions of CH_4 and N_2O were measured 21 times over the 352-day study period (20 September 2011–6 September 2012).

The temperature of the SS in each of the 12 cylinders was continuously recorded every hour at a point approximately 0.2 m from the cylinder base by sensors (Tiny tag Aquatic, Intab Interface Teknik AB, Stenkullen, Sweden) and every 15 min at 0.2 m below

Table 1
Background data on the sewage sludge used in storage treatments.

Digestion	Digestion temperature (°C)	Hydraulic retention time (days)	DM to Reactor (%)	DM from Reactor (%)	Degree of degradation (% of DM)	Load (PE)
Mesophilic	37.5	15[a]	3.6	2.6	28	130,000
Thermophilic	53	15–17[b]	6	4	33	20,000

DM = dry matter.
PE = population equivalents, one PE corresponds to a load of 70 g biochemical oxygen demand day^{-1}.
[a] No guaranteed retention time.
[b] Guaranteed retention time 3 h.