



Biosolid stockpiles are a significant point source for greenhouse gas emissions



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ABSTRACT

The wastewater treatment process generates large amounts of sewage sludge that are dried and then often stored in biosolid stockpiles in treatment plants. Because the biosolids are rich in decomposable organic matter they could be a significant source for greenhouse gas (GHG) emissions, yet there are no direct measurements of GHG from stockpiles. We therefore measured the direct emissions of methane (CH₄), nitrous oxide (N₂O) and carbon dioxide (CO₂) on a monthly basis from three different age classes of biosolid stockpiles at the Western Treatment Plant (WTP), Melbourne, Australia, from December 2009 to November 2011 using manual static chambers. All biosolid stockpiles were a significant point source for CH₄ and N₂O emissions. The youngest biosolids (<1 year old) had the greatest CH₄ and N₂O emissions of 60.2 kg of CO₂-e per Mg of biosolid per year. Stockpiles that were between 1 and 3 years old emitted less overall GHG (~29 kg CO₂-e Mg⁻¹ yr⁻¹) and the oldest stockpiles emitted the least GHG (~10 kg CO₂-e Mg⁻¹ yr⁻¹). Methane emissions were negligible in all stockpiles but the relative contribution of N₂O and CO₂ changed with stockpile age. The youngest stockpile emitted two thirds of the GHG emission as N₂O, while the 1–3 year old stockpile emitted an equal amount of N₂O and CO₂ and in the oldest stockpile CO₂ emissions dominated. We did not detect any seasonal variability of GHG emissions and did not observe a correlation between GHG flux and environmental variables such as biosolid temperature, moisture content or nitrate and ammonium concentration. We also modeled CH₄ emissions based on a first order decay model and the model based estimated annual CH₄ emissions were higher as compared to the direct field based estimated annual CH₄ emissions. Our results indicate that labile organic material in stockpiles is decomposed over time and that nitrogen decomposition processes lead to significant N₂O emissions. Carbon decomposition favors CO₂ over CH₄ production probably because of aerobic stockpile conditions or CH₄ oxidation in the outer stockpile layers. Although the GHG emission rate decreased with biosolid age, managers of biosolid stockpiles should assess alternate storage or uses for biosolids to avoid nutrient losses and GHG emissions.

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1. Introduction

Greenhouse gas (GHG) concentrations of methane (CH₄), nitrous oxide (N₂O) and carbon dioxide (CO₂) have increased in the atmosphere due to direct and indirect human activities (Dalal et al., 2003). The waste and wastewater industry sectors contribute approximately 3% to the global anthropogenic emissions of GHG (IPCC, 2007), which has led to increased public awareness and the need to better estimate or measure GHG emissions from the sector

(Ishigaki et al., 2005). The wastewater sector emits CH₄, CO₂ and N₂O directly through nutrient transformations during the wastewater treatment process and degradation of organic matter (Boldrin et al., 2009; Czepiel et al., 1996) and indirectly through fossil fuel consumption for pumping, transport, operations and processing (Monteith et al., 2005). In all wastewater treatment plants large quantities of sewage sludge are produced (Brown et al., 2008). The dried sludge is called biosolids and still contains significant quantities of organic matter, macro-nutrients and trace elements (Bright and Healey, 2003). The quantity of biosolid produced in Australia is around 360,000 dry Mg per year (Pritchard et al., 2010) and in the state of Victoria it is approximately 68,250 dry Mg (AWA, 2013). These biosolids are often stored in large

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stockpiles for many years within the wastewater treatment site, before they are moved off-site and used as a resource.

Currently, GHG emissions from waste and wastewater treatment plants are often based on simple emission factors or mathematical models developed from kinetic relationships of mass and energy balances (Yerushalmi et al., 2013). One of the main uncertainties in GHG accounting is associated with the method of calculation, which is frequently not based on, or validated by, direct GHG flux measurements. For example, waste industries use minimum data to predict the GHG emissions based on population, waste generation rate, solid waste composition and methods of solid waste disposal (El-Fadel and Massoud, 2000). Another uncertainty confronting GHG accounting in the wastewater industry is access to long term monitoring of GHG emissions and under a wide range of climatic and management conditions (Cowie et al., 2012; Bogner et al., 2008; Czepiel et al., 1993). Hence, to reduce these uncertainties it is important to obtain reliable long-term data to better understand the magnitude of direct GHG emissions and the variation over seasonal or inter-annual time frames. There are many studies investigating GHG emissions from landfill waste (Bastian et al., 2013; Stanisavljevic et al., 2012; Goldsmith et al., 2012), stockpile composting (Chan et al., 2011; Ahn et al., 2011; Boldrin et al., 2009) and stockpile manure management (Petersen et al., 2013; Wood et al., 2012). However, GHG emissions from stockpiled biosolids have not yet been measured in detail.

The overall objective of this study was therefore to investigate the direct GHG emissions from various ages of biosolids stockpiles at a wastewater treatment plant. The specific objectives were to:

- (1) measure the flux of CH₄, CO₂ and N₂O from different ages of biosolid stockpiles across different seasons and investigate GHG concentrations (CH₄, CO₂) at two different biosolid stockpile depths,
- (2) investigate the relationship between GHG emissions from biosolid stockpiles and environmental variables,
- (3) calculate the cumulative magnitude of annual CH₄, CO₂ and N₂O fluxes from different aged biosolid stockpiles and compare this against modeled emissions.

2. Material and methods

2.1. Site description and experiment set up

The Western Treatment Plant (WTP) is located 35 km to the south-west Melbourne and services wastewater from a population of about 1.6 million people from the western and northern suburbs of Melbourne (38°1'52''S, 144°34'82''E). Climate in this area is temperate with warm dry summer and cool winters with maximum rainfall occurring during spring. The long-term average rainfall is 542 mm (Stickland et al., 2013) and mean evaporation is generally highest between December and February (Parameswaran, 1999). The WTP was established more than 100 years ago and currently processes 52% of Melbourne sewage. The sewage is transported from domestic (70%) and industrial (30%) sources to the WTP via pipes and pumping stations. The plant receives an average 500 ML of wastewater per day and has a maximum capacity of 2000 ML per day. Wastewater in the WTP is distributed to lagoons for processing in two stages. The first stage is anaerobic treatment in an initial deeper lagoon, which is covered to allow capture of CH₄ gas produced for biogas energy. The second stage are aerobic lagoons to provide an active process for nitrogen removal. The treatment processes produces sludge that settles to the floor of the lagoons that therefore require removal. The sludge from the base of the aerobic and anaerobic settling ponds is

collected using a floating pump and then transported to large open air drying pans to produce biosolids. The length of air-drying depends upon the potential evaporation and rainfall conditions, but generally ranges between 10 and 20 weeks. The dried biosolid product is then stored onsite in a stockpile format. The composition of each stockpile is likely to vary quite significantly depending on the relative contribution of sludge that has been collected from aerobic or anaerobic settling ponds and the time it has taken to dry the sludge in the drying pans. The anaerobic settling ponds had not been dredged for a long period of time and continuous dredging started in 2009. Hence, biosolids from anaerobic ponds are of varying age and consistency, depending where they were collected.

The total annual biosolid production in WTP is around ~24,000 dry Mg (Stickland et al., 2013). There are currently 15 stockpiles onsite in the WTP. Three stockpiles of different age ranges were selected to assess the magnitude of GHG flux emissions over a two-year period and to better understand the biosolid characteristics and environmental conditions that may influence emissions. We selected three different age ranges to assess if GHG emissions change during the ageing of biosolids. The main sources of biosolids in the oldest stockpile (>3 years old, yo) were from the sludge of various drying pans collected before 1995. The 1–3 yo biosolids and <1 yo biosolids were sourced from the ongoing sludge drying pan collections. The dimensions of the three different biosolid stockpiles were: >3 yo (130–150 m long, 40–50 m wide and 8–10 m high), 1–3 yo (130–150 m long, 50–60 m wide and 10–12 m high) and <1 yo (100–120 m long, 30–40 m wide and 6–8 m high).

2.2. Measuring GHG flux from biosolid stockpiles

The closed static chamber technique (Hutchinson and Mosier, 1981) was used to quantify the GHG flux rates of CH₄, CO₂ and N₂O from the top surface of each stockpile. Gas samples were collected every month from December 2009 to November 2011 from the >3 yo and 1–3 yo stockpiles, and from March 2010 to November 2011 for the <1 yo stockpile. Eight chambers were placed in a row about 10–12 m apart on the top surface of each stockpile. Chambers were made from non-transparent PVC pipe (diameter 25 cm, height 24.5 cm, volume 12.0 L, basal area 0.045 m²) with a twist-lid incorporating a butyl-rubber septum and a rubber O-ring to form a gas tight seal. Chambers were inserted to a depth of 3–4 cm at least 15–30 min before closing the lid. Once closed, 20 mL headspace gas samples were taken at 0, 4, 8 and 12 min after closure and stored in 12 mL pre-evacuated Exetainer™ (Labco, UK) gas vials. Gas samples were collected between 10:30 am and 14:30 pm. After collecting gas samples, the chamber lids were removed to measure the internal chamber height at four points to calculate the internal headspace volume of each chamber. Gas samples stored in Exetainers™ were transferred to the laboratory and analysed for CH₄, CO₂ and N₂O concentration through gas chromatography (Shimadzu GC17A with N₂ carrier gas). Methane and CO₂ concentrations were determined using a flame ionization detector (FID), and N₂O concentration determined using an electron capture detector (ECD). Gas concentrations (%) of CH₄, CO₂ and O₂ were also measured at two different depths (0.5 m and 1 m depth) from August 2010 to November 2011 from the >3 yo and 1–3 yo biosolids stockpiles using GA 2000 landfill gas analyser.

2.3. Measuring biosolid properties

The temperature of biosolid (BT) was measured at a depth of 10 cm using a short temperature probe (Cole–Parmer, USA) for the entire measurement period and at a depth of 50 cm using a longer Digi-Sense™ Type K thermocouple thermometer from August 2010 onwards. Biosolid samples were collected from 0 to 10 cm using a

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