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Storage management influences greenhouse gas emissions from biosolids

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ABSTRACT

Biosolids produced by wastewater treatment plants are often stored in stockpiles and can be a significant source of greenhouse gases (GHG). Growing trees in shallow stockpiled biosolids may remove nutrients, keep the biosolids drier and offset GHG emissions through C sequestration. We directly measured methane (CH_4), carbon dioxide (CO_2) and nitrous oxide (N_2O) flux from a large biosolid stockpile and two shallow stockpiles, one planted with *Salix reichardtii* (willow) trees, from December 2009 to January 2011. All stockpiles emitted large annual amounts of GHG ranging from 38 kg CO_2-e Mg^{-1} dry biosolid for the large stockpile, to 65 kg CO_2-e Mg^{-1} for the unplanted shallow stockpile, probably due to the greater surface area to volume ratio. GHG emissions were dominated by N_2O and CO_2 whilst CH_4 emissions were negligible (<2%) from the large stockpile and the shallow stockpiles were actually a CH_4 sink. Annual willow tree growth was 12 Mg dry biomass ha^{-1}, but this only offset 8% of the GHG emissions from the shallow planted stockpile. Our data highlight that biosolid stockpiles are significant sources for GHG emissions but alternate management options such as shallow stockpiles or planting for biomass production will not lead to GHG emission reductions.

1. Introduction

Biosolids are an end product of the sewage treatment processes and their production gradually increases every year due to sewage production from an increasing human population (Wang et al., 2008). For example, in Australia there is an approximate 3% increase in biosolid production from wastewater treatment plants (WTPs) annually (Australian Water Association, 2014The storage of biosolids within WTPs is necessary either temporarily, or long-term, depending upon whether an ultimate end-use is available. Desirable end uses have a low environmental impact or even an environmental and economic benefit, such as biosolid application to agricultural or production forestry systems (Pritchard et al., 2010). Biosolids are often stored in large stockpiles to minimize the use of space, but this can present a fire risk (spontaneous combustion), pollution risks (leachate and particulate) and increased GHG emission risks as they are rich in organic matter and nutrients (Fernandes et al., 2005). In fact, biosolid stockpiles can emit large amounts of greenhouse gases, especially in young stockpiles (Majumder et al., 2014).

A potential alternative end use, or long term storage option for biosolids are shallow stockpiles (e.g. 0.5 m deep) over larger areas and within which woody vegetation can be planted for carbon offset gains, biosolid stabilization and pollutant/nutrient removal (Laidlaw et al., 2012). In such a system, the high labile carbon and nitrogen content of the biosolid, in combination with rainfall and/or supplementary irrigation, could lead to high plant biomass production and therefore a value adding product for bioenergy or biochar production and/or carbon offset potential of related GHG emissions from the WTP. However, microbial decomposition and transformations of labile carbon and nitrogen in these stockpiles may still lead to significant production of methane (CH_4), carbon dioxide (CO_2) and nitrous oxide (N_2O) under aerobic and anaerobic conditions. In WTPs, GHG emissions are generally estimated using emission factors based on the initial chemical properties of the wastewater or sewage sludge (Brown et al., 2010) and as such there is substantial uncertainty in these GHG emissions estimates (Bogner et al., 2008). This also relates to the storage and management of dried biosolids as direct measurements of GHG emissions from biosolids in stockpiles or other interim storage options are

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lacking.

The main aim of this study was to assess the magnitude of GHG emissions from three different biosolid storage and management systems: i) large stockpiles (LS), ii) unplanted shallow stockpiles (USS) and iii) planted shallow stockpiles (PSS) with willow (*Salix x reichardtii* A.Kern).

The specific objectives of this study were to:

(1) measure the seasonal variation and magnitude of CH_4, CO_2, and N_2O emissions from the three different biosolid management options;
(2) investigate the relationship between greenhouse gas fluxes and environmental variables in these storage management systems;
(3) calculate the annual CH_4, CO_2, and N_2O flux balance from the three different biosolid management systems and evaluate the carbon offset potential of woody biomass growth in biosolids.

2. Materials and method

2.1. Experimental set-up and site description

The study site was located at the Western Treatment Plant (WTP) of Melbourne Water, which is located in Weribbee, 35 km south-west of Melbourne (37°59′35″S, 144°36′58″E). The WTP treats wastewater of around 1.8 M people of the western and northern suburbs of Melbourne. The temperate climate in the region is characterized by warm and dry summers and a relative even rainfall distribution with a maximum in spring. Historical average annual rainfall (1941–2012) is 514.4 mm (BOM, 2013). Currently, there are approximately 1,573,000 Mg of air-dried biosolids stockpiled at the WTP (Melbourne Water, 2010). For this study, three different management practices of biosolids storage and use were selected to quantify their GHG emissions. These were: i) large stockpiles (LS), which are commonly used (~9.0 m high), ii) unplanted shallow stockpiles (USS = 0.5 m high) and iii) planted shallow stockpiles (PSS = 0.5 m high) with willow (*Salix x reichardtii* A.Kern). The same biosolids were used for all three management options. The biosolids were collected before 1995 from the mixed sludge of a number of drying pans and were stored in biosolid stockpiles prior to the experiment commencing. The experimental area of shallow stockpiles was set up in 2005 and covered an area of approximately 7000 m^2, with 2000 m^2 of this being planted with willows at a stocking rate of 40,000 plants ha^{-1} (or 4 plants m^{-2}). The willow planting was irrigated with Class C wastewater during the summer months, see Laidlaw et al. (2012) for more details.

2.2. Measurement of GHG flux from three different management practices of biosolid

The GHG flux rates of CH_4, CO_2 and N_2O were measured using the closed static chamber technique (Hutchinson and Mosier, 1981) at the surface of each stockpile in the management practices LS, USS and PSS. We collected gas samples once a month from December 2009 to January 2011. On the top of each stockpile we placed eight chambers in a row about 1–2 m apart. The manual chambers consisted of non-transparent PVC pipe (diameter 25 cm, height 24.5 cm, volume 12.0 L) and had a twist-lid incorporating a butyl-rubber septum and a rubber O-ring to form a gas tight seal. The basal area of each chamber was 0.045 m^2. Manual chambers were inserted to a depth of 3–4 cm depth at least 15–30 min before closing the lid. After closing the chamber lids, we collected 20 mL headspace gas samples with a syringe at intervals of 0, 4, 8 and 12 min for the large stockpile and at intervals of 0, 10, 20 and 30 min for the shallow stockpile because of initial differences from trial gas flux measures. The gas samples were collected between 10:30 and 14:30 in pre-evacuated 12 mL vials (Exetainers™, Labco Pty Ltd, UK). After collecting gas samples, chamber height was measured at four positions to calculate headspace volume of each chamber individually. Gas samples were analyzed using gas chromatography (GC) (Shimadzu GC17A, with N_2 carrier gas) to determine CH_4 concentrations using a flame ionization detector (FID) and CO_2 concentrations through the addition of a Methaniser (SRI Instruments, USA) before the FID. N_2O concentrations were determined using an electron capture detector (ECD) in the same GC run.

2.3. Measurement of biosolid environmental properties

We measured biosolid temperature (BT) with short temperature probes (Cole-Parmer, USA) at a depth of 10 cm at the same time as gas samples were collected. We collected biosolid samples at each measurement from the upper 0.1 m with a stainless steel bulk density ring near each chamber to estimate biosolid moisture content (MC), bulk density and to provide samples for NO_3^- and NH_4^+ concentration analysis. We dried subsamples at 105 °C for 48 h and determined biosolid MC gravimetrically by weighing samples before and after drying. The NO_3^- and NH_4^+ concentration of biosolids was measured in 1 M KCl extracts (1:4, biosolid:KCl) on a Technicon™ auto-analyser. The bulk density of biosolids was calculated dividing the mass of oven dried biosolids (g) by the volume of stainless steel ring (cm^3) (Gifford and Roderick, 2003).

2.4. GHG flux calculation

Curvilinear regressions best described the CH_4, CO_2 and N_2O fluxes from biosolid stockpile because the gas concentration in the chamber headspace decreased gradually with time of chamber closure (Matson and Harriss, 1995). The following equation was used to measure the flux

$$f_o = V^*(C_1 - C_0)^2 \Big/ [A^*t_1^*(2^*C_1 - C_2 - C_0)]^*\ln[(C_1 - C_0) \times /(C_2 - C_1)] \quad (1)$$

where, f_o is the flux at time 0, V is the chamber headspace volume (L), A is the soil surface area (m^2), C_0, C_1, and C_2 are the chamber headspace gas concentrations ppm(v) at different times after closure (0, 4, 8 and 12 min for LS and 0, 10, 20, 30 min for USS and PSS), respectively, and t_1 is the time between gas sampling points (Minato et al., 2013). The unit of f_o is μL trace gas m^{-2} min^{-1}.

This flux (f_o) was then transformed to μmol CH_4, CO_2 and N_2O m^{-2} h^{-1} by accounting for pressure, temperature and volume based on the ideal gas law by applying Eq. (2)

$$F_{\mu mol} = \frac{F_{\mu L} \times P}{R \times T} \quad (2)$$

where $F_{\mu mol}$ is the flux in μmol CH_4, CO_2 and N_2O m^{-2} h^{-1}, $F_{\mu L}$ is the flux in μL CH_4, CO_2 and N_2O m^{-2} h^{-1}, P is the atmospheric pressure in kPa at the site depending on the altitude, T is air temperature in K (273+ °C), R is the ideal gas constant = 8.3144 L kPa mol^{-1} K^{-1}). The fluxes of μmol CH_4, CO_2 and N_2O m^{-2} h^{-1} were converted to μg CH_4–C, CO_2–C and N_2O–N m^{-2} h^{-1} depending on the molecular mass of each gas.

We measured fluxes prior to the start of the campaigns at different times of the day and we observed only small diurnal variation of flux for all GHG. We therefore decided to measure the