Waste Management 29 (2009) 2078–2084

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Impact of nitrate-enhanced leachate recirculation on gaseous releases from a landfill bioreactor cell

G. Tallec [a], C. Bureau [a], P. Peu [b], J.C. Benoist [b], M. Lemunier [c], A. Budka [d], D. Presse [d], T. Bouchez [a,*]

[a] Cemagref, UR HBAN, Parc de Tourvoie, BP44, F-92163 Antony, France
[b] Cemagref, UR GERE, 17 Avenue de Cucillé, CS 64427, F-35044 Rennes, France
[c] Suez-Environnement, CIRADE, 38 Av. Jean Jaurès, 78440 Gargenville, France
[d] SITA France, 132 Rue des 3 Fontanot, 92000 Nanterre Cedex, France

ARTICLE INFO

Article history:
Accepted 27 January 2009
Available online 17 March 2009

ABSTRACT

This study evaluates the impact of nitrate injection on a full scale landfill bioreactor through the monitoring of gaseous releases and particularly N_2O emissions. During several weeks, we monitored gas concentrations in the landfill gas collection system as well as surface gas releases with a series of seven static chambers. These devices were directly connected to a gas chromatograph coupled to a flame ionisation detector and an electron capture detector (GC-FID/ECD) placed directly on the field. Measurements were performed before, during and after recirculation of raw leachate and nitrate-enhanced leachate. Raw leachate recirculation did not have a significant effect on the biogas concentrations (CO_2, CH_4 and N_2O) in the gas extraction network. However, nitrate-enhanced leachate recirculation induced a marked increase of the N_2O concentrations in the gas collected from the recirculation trench (100-fold increase from 0.2 ppm to 23 ppm). In the common gas collection system however, this N_2O increase was no more detectable because of dilution by gas coming from other cells or ambient air intrusion. Surface releases through the temporary cover were characterized by a large spatial and temporal variability. One automated chamber gave limited standard errors over each experimental period for N_2O releases: 8.1 ± 0.16 mg m^{-2} d^{-1} ($n = 384$), 4.2 ± 0.14 mg m^{-2} d^{-1} ($n = 132$) and 1.9 ± 0.10 mg m^{-2} d^{-1} ($n = 49$), during, after raw leachate and nitrate-enhanced leachate recirculation, respectively. No clear correlation between N_2O gaseous surface releases and recirculation events were evidenced. Estimated N_2O fluxes remained in the lower range of what is reported in the literature for landfill covers, even after nitrate injection.

1. Introduction

Landfilling is still a major municipal solid waste (MSW) management technology in France (47% of MSW, Ademe, 2006). In this framework, leachate recirculation has retained increasing attention over the past years as a suitable management strategy for enhancing waste degradation in closed landfill cells (Benson et al., 2007).

MSW has been estimated to contain about 4% protein and, therefore, ammonia (NH_3) is produced during anaerobic decomposition of organic nitrogen (Barlaz et al., 1990; Madigan and Martinko, 1997). Because ammonia is stable under the conditions prevailing within the landfill, it can accumulate in recirculated leachate (Burton and Watson-Craik, 1998), which could cause an inhibition of refuse decomposition (Wens et al., 2001; Vigneron, 2005). To avoid this potential inhibition, it has been proposed to treat leachate before recirculation in order to convert *ex situ* ammonia into nitrate by nitrification (Vigneron, 2005). The landfill could thus be used as an anaerobic bioreactor for denitrification converting nitrate (NO_3^-) into N_2 gas. The use of nitrate instead of carbon dioxide as an electron acceptor for microbial metabolism induces a strong increase in the Gibb's free energy (around six-fold) available for microorganisms' growth. Heterotrophic denitrification could therefore enhance decomposition rates and reduce the aftercare period, a critical issue for the landfill operators. This strategy enables the release of nitrogen outside of the system with concomitant consumption of organic carbon from the waste, thus possibly enhancing MSW stabilization. Nitrate-enhanced leachate recirculation therefore represents a promising strategy for more sustainable landfill management.

However, the issue of possible nitrous oxide (N_2O) emissions during nitrate-enhanced leachate recirculation in bioreactor-landfill should be carefully addressed. N_2O is a strong greenhouse gas. N_2O presents a 310 times stronger 100-year global warming potential than carbon dioxide (CO_2) and a 100–120 years lifetime (IPCC, 2000). The environmental conditions in landfills are favourable to the production of N_2O as they are characterized by the presence

* Corresponding author. Tel.: +33 140 96 60 40; fax: +33 1 40 96 62 70.
E-mail address: theodore.bouchez@cemagref.fr (T. Bouchez).

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doi:10.1016/j.wasman.2009.01.006

of organic waste containing nitrogen and zones under both aerobic and anaerobic conditions. Several biochemical reactions may therefore produce N_2O in landfills. N_2O can be produced by nitrification in the aerobic upper layer of the waste mass, or in the soil top-cover, *via* methanotrophic microorganisms (Mandernack et al., 2000). In these zones, when oxygen becomes limiting, autotrophic ammonium oxidizing bacteria may carry out nitrifier denitrification with N_2O and N_2 as final products (Kuai and Verstraete, 1998; Jiang and Bakken, 1999). Moreover, N_2O may be produced as an intermediate of the heterotrophic denitrification during nitrate-enhanced leachate recirculation (Gaskell et al., 1981; Itokawa et al., 2001; Bonin et al., 2002; Price et al., 2003). It could also be produced during dissimilatory nitrate reduction to ammonia under anoxic conditions (Tiedje et al., 1979; Satoh et al., 1983).

The N_2O emissions, measured from traditional landfills, would be much weaker than those measured in bioreactor-landfill with leachate recirculation, leachate irrigation, sludge- or leachate-amendment on the landfill cover (Rinne et al., 2005; Lee et al., 2002; Mandernack et al., 2000; Bogner et al., 1999; Börjesson and Svensson, 1997). However, only a limited number of studies have been carried out on N_2O emissions from bioreactor-landfill with nitrate-enhanced leachate recirculation (Balslev et al., 2005).

In this study, we evaluated the impact of nitrate injection on a full scale landfill bioreactor, with a particular emphasis on the monitoring of N_2O releases. During several weeks, we monitored gas concentrations in the landfill gas collection system as well as the gaseous releases from the surface with a series of seven static chambers. Measurements were performed before, during and after recirculation of raw and nitrate-enhanced leachate. The biogas concentrations (O_2, CH_4, CO_2 and N_2O), were analyzed *in situ* throughout the recirculation process using gas chromatography coupled with flame ionization (FID) and electron capture detectors (ECD) to allow sensitive detection of all gases.

2. Methods

2.1. Site and experimental cell description

We studied an experimental cell located at a municipal solid waste landfill site near Dijon, France. This landfill site includes a composting plant, an industrial waste stabilization unit as well as a MSW landfilling area with cells in use. Leachate from the MSW landfill is stored in two covered basins (500 m^3 each). A flare allows the combustion of the biogas collected by the whole MSW section.

The filling of the cell studied ended in 2001. It consists of an area of about 5000 m^2 with mean waste thickness of 8 m (45000 m^3 capacity). The cell is closed and covered with temporary compacted clay capping of 0.5 m placed directly over the waste mass. Woven geo-film strips without welding are laid on the top of the cover to avoid scrubbing of clay during rain periods. This geo-film is therefore not tight to gas. Gas diffusing through the temporary clay cover will therefore finally be emitted to the atmosphere.

The cell is equipped with a leachate recirculation network consisting of 5 horizontal trenches located below the cover and filled with gravel. These trenches contain perforated HDPE pipes (about 70 m long, 0.1 m of diameter) placed with a slope of 2% to 4% at a depth of approximately 1.5 m. Leachate is fed by gravity at atmospheric pressure, enabling flow rates ranging between 5 to 10 $m^3\ h^{-1}$ (Fig. 1a, black lines, and b). The biogas collection system is located in the same horizontal trenches, with perforated pipes placed above the ones used for leachate recirculation (Fig. 1a and b). No recirculation had been carried out for at least one year before the experiment started.

This cell is also equipped with a network of electrodes perpendicular to the trenches of recirculation (3 series of 48 electrodes installed at the surface of the waste mass at a depth of approximately 0.15 m), in order to measure resistivity during the recirculation events (Fig. 1a, dotted lines, and b). Temperatures sensors were also installed at four different locations along the A2 resistivity line at different depths under the clay cover (Fig. 1b).

2.2. Leachate recirculation in the experimental cell

All the leachate injections were carried out in October and November 2006 in one trench only (B3, Fig. 1). Recirculated leachate originated from the storage ponds of the landfill site. The average nitrogen concentration was 250 mg $N–NH_4\ L^{-1}$ (without detectable amount of nitrite and nitrate). Raw leachate was first in-

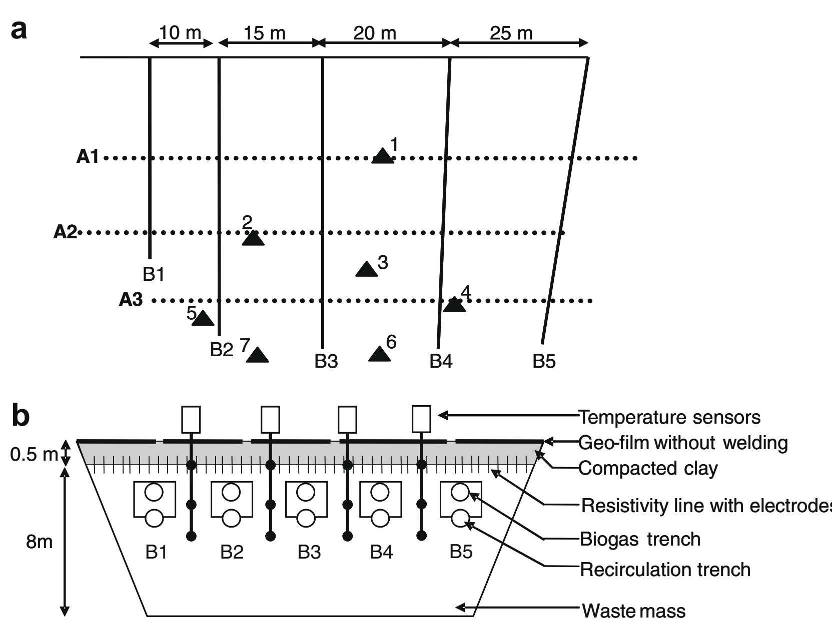

Fig. 1. (a) Plan view of the experimental cell with five horizontal recirculation trenches (B1–B5), the seven static chambers localizations (triangles) and three resistivity lines (A1–A3, horizontal dotted lines), (b) transversal view of the experimental cell.