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Multiple-tracer tests for contaminant transport process identification in saturated municipal solid waste

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ABSTRACT

Two column tests were performed in conditions emulating vertical flow beneath the leachate table in a biologically active landfill to determine dominant transport mechanisms occurring in landfills. An improved understanding of contaminant transport process in wastes is required for developing better predictions about potential length of the long term aftercare of landfills, currently measured in timescales of centuries. Three tracers (lithium, bromide and deuterium) were used. Lithium did not behave conservatively. Given that lithium has been used extensively for tracing in landfill wastes, the tracer itself and the findings of previous tests which assume that it has behaved conservatively may need revisiting. The smaller column test could not be fitted with continuum models, probably because the volume of waste was below a representative elemental volume. Modelling compared advection-dispersion (AD), dual porosity (DP) and hybrid AD–DP models. Of these models, the DP model was found to be the most suitable. Although there is good evidence to suggest that diffusion is an important transport mechanism, the breakthrough curves of the different tracers did not differ from each other as would be predicted based on the free-water diffusion coefficients. This suggested that solute diffusion in wastes requires further study.

1. Introduction

There is considerable uncertainty over the mechanisms of flow and transport through biologically active municipal solid waste (MSW). This uncertainty translates into a poor ability to predict the efficiency of contaminant mass removal from a waste body under different flushing conditions. The importance of this relates to the need to understand and predict how the water polluting potential of landfills will reduce over time and hence for how long active leachate control and management measures will be required at a site. Current estimates indicate that management timescale will run into many centuries after a site has closed (Hall et al., 2004). A difficulty in predicting the transport behaviour of degrading, gassing waste is that state variables such as water flow, gas production rate and water content change over time. 'Landfill simulators' therefore typically need to model several coupled processes, combining a large number of assumptions and parameters in a manner which makes it difficult to infer much about the constitutive relationships or to obtain unique parameter values. The purpose of this study is to extricate from this complexity an understanding of the dominant mass-transport processes in waste. Our approach is to conduct an experiment under highly controlled conditions, such that we can decouple physical transport mechanisms (including advection, mechanical dispersion and diffusion) from thermal, biological, and mechanical processes and multi-phase flow effects.

In a landfill experiencing downward infiltration of water, two distinct regimes of flow are normally considered to exist. At the top of the landfill there is two phase-flow, comprised of downward liquid flow and predominantly upwards landfill gas flow. In this zone it is normally assumed that due to capillarity the liquid pressure will be below that of the gas phase, although localised perching is possible. Several authors (Straub and Lynch, 1982; Demetracopoulos et al., 1986; McDougall and Silver, 2005; Kazimoglu et al., 2006) have attempted to address this regime. McCreanor and Reinhart (2000) modelled this zone using the Richards equation and White et al. (2004) modelled both liquid and gaseous phases.

Vertical transport in the zone beneath the leachate table has received less attention in the literature. In this zone (which is frequently called the 'saturated zone', although may not be due to the presence of landfill gas), leachate and gas are under positive hydrostatic pressure.

Solutes that are measured at the base of a landfill will have arisen from and travelled through either the saturated zone or both

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the saturated and unsaturated zones. Contaminant flushing models for a landfill under these conditions will need to simulate both above and below the leachate-table. Concern over the potential for leakage of contaminants through landfill bases means that the leachate table is often maintained at a low level within the waste, making the saturated zone the minority proportion of the landfill volume. Despite the regulatory barriers to raised leachate tables, an advantage of increased water content would be that it would potentially encourage greater microbial activity and a higher flushing efficiency (Beaven et al., 2004). There is therefore a strong motivation for addressing transport in the saturated zone.

There are particular problems for experimentation in the saturated zone. Firstly, it is difficult to obtain representative *in situ* samples. Secondly, the high levels of compressive stress and compaction are difficult to reproduce in the laboratory. Thirdly, the control of liquid flow during gas production is difficult to achieve.

Even with perfect control, single tracer tests can provide insufficient information to unambiguously identify the underlying transport processes (Jury and Roth, 1990). Flow interruption is a standard method for identifying diffusive non-equilibrium during a tracer test (Brusseau et al., 1997; Fortin, 1997), since the diffusion process continues even though advection has stopped during an interruption in flow. Wehrer and Totsche (2008) applied flow-interruption to MSW incinerator ash. When applied to MSW (Beaven et al., 2002) this technique has provided evidence suggestive of the presence of diffusive exchange with immobile (or less mobile) zones. However, during active degradation changes to flow rates cause changes in the amount of gas within the waste, and therefore also the volume of liquid within the waste. Flow-interruption is therefore not ideally-suited to gassing wastes as a tool for diagnosing transport processes.

One further way to test the importance of diffusion is to inject tracers with different diffusion coefficients into the waste. This method has been used with success in soils and groundwaters (Garnier et al., 1985; Maloszewski and Zuber, 1992; Becker and Shapiro, 2000). Here the method is evaluated with application to waste. Previous studies have used simultaneous application of multiple tracers in MSW (Woodman et al., 2013; Woodman, 2014), however the primary focus in these studies was on the effect of operating conditions rather than on process identification.

There have been a few previous column tracer tests on saturated methanogenic MSW wastes that were suggestive of dual-porosity processes (Beaven et al., 2003; Rosqvist and Bendz, 1999; Fellner and Brunner, 2010). However, difficulties in hydraulic control (specifically, flow rate and saturation level) meant that they were not entirely conclusive (Woodman, 2007).

The primary aim of the experiment described here is to test the hypothesis that dual-porosity exchange is the predominant mechanism accounting for solute mixing. Implicit to this hypothesis is the assumption that flow is restricted to a fraction of the porosity (a 'mobile' zone) and that dispersion will be affected by the diffusion coefficient of the solute in question.

2. Method

2.1. Description of experiments

Two column tests ('Test 0A' and 'Test 0B') were designed to emulate contaminant transport in the complex sub-water table environment of biologically active MSW by tracking the flushing of methanogenic high-bromide leachate with water dosed with lithium chloride and deuterium oxide tracers (i.e. indigenous bromide was flushed from the waste whist lithium and deuterium were passed through the waste as a tracer 'pulse'). Test 0A was performed in a 2 m diameter column, in contrast to Test 0B which was in a smaller (0.26 m) diameter column. Prior to the flushing, a number of separately reported hydraulic tests were performed on the larger column. Subsequently, leachate was recirculated over five weeks to achieve dynamically stable hydraulic conditions and the equilibration of the high bromide leachate throughout the waste mass. Test 0B was packed and manually compressed to the same bulk density as that in Test 0A. Recirculation was also carried out in Test 0B for approximately five weeks (34 days) prior to the test.

Over the course of the tests samples collected from the outputs of the columns recorded flushing (thereby emulating the flushing of a real landfill) as well as the simultaneous recovery of the two introduced tracers. Bromide was selected as a key flushed species. Aside from differing in scale, both tests were performed under very similar conditions. Test 0A, a large-scale test (5.2 m^3), was performed first. It will be shown that for this test the estimates of lithium (Li) tracer mass recovery were low. Because this result was of concern a second tracer test (Test 0B) was performed in a smaller-scale laboratory column (0.04 m^3).

2.2. Experiment

Test 0A was performed in the Pitsea compression cell (shown schematically in Fig. 1). The compression cell is a 2 m diameter steel cell capable of delivering uniaxial loads representative of those within full-scale landfills, whilst allowing detailed logging of compression, temperature, piezometric pressures through the column, total weight, gas production and leachate electroconductivity (EC).

The cell was loaded in September 2004 with 3563 kg (dried mass) of fresh MSW that was shredded with a hammer mill and passed through an 80 mm screen.

Sub-samples of the waste were characterised for material content and particle size (see Supplementary Information Table S1). Layers of gravel were placed at the top and bottom of the waste to encourage a uniform distribution of flow in and out of the waste (amounting to depths of 0.08 m above and 0.095 m of gravel above and below the waste respectively).

The residual (i.e. drained) water content of the waste in the cell was established from an oven-dried (at 80 °C) 10 kg sub-sample obtained at the time of loading. Thereafter the total water content in the waste was estimated based on the cell weight (measured using load cells) and a water balance. This and other key details of both tests are given in Table 1.

The waste was subject to long-term monitoring and a number of hydraulic tests and was then compressed using an upper platen at 87 kPa (thereafter the platen was locked in position to maintain the waste at a fixed volume). Methanogenic leachate with a high bromide content (Table 2) was obtained from Pitsea Landfill and then introduced and recirculated for two months to allow physical and chemical equilibration.

The design of Test 0A was optimised based on parameters previously established from a pilot tracer test in the Pitsea compression cell (Beaven et al., 2002; Woodman, 2007). The leachate re-circulation was achieved using a positive-displacement pump (LMI Milton Roy C785- 139 35T) drawing from a 400 litre supply tank, giving a stable flow of 3.7 L/h. Manual readings from a gas-meter connected to an outlet manifold at the top of the cell indicated stable gas generation rates before the start of the test and monitoring of cell weight during the test indicated no major change to the water content of the waste over the duration of the test (see data for Phase 1 in Woodman et al., 2009). Consequently, a dynamic hydraulic equilibrium is assumed within the cell for the duration of the test (i.e. gas production rate equal to gas release rate).