Hydrology and Earth System Sciences, 3(3), *429–438* (1999) © EGS

Hydrology & Earth System Sciences

An experimental evaluation of the solute transport volume in biodegraded municipal solid waste

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Abstract

A large undisturbed sample (3.5 m^3) of 22-year-old, biodegraded solid waste was set up to estimate the volume fraction participating in the transport of solutes through the waste material. Altogether, five tracer tests were performed under ponding and sprinkling conditions, and under steady-state and transient conditions. The experimental break through curves (BTCs), which indicated a non-equilibrium transport of the solute by early peaks and long right-hand tails, were used to parameterize log-normal solute travel time probability density functions. The expected solute travel times (i.e. the median solute travel times) were assessed and the corresponding fraction of the experimental volumes active in the transport of solutes was estimated. The solute transport volume fractions defined by the median solute travel times were estimated to vary between 5 and 10% of the total experimental volume. Further, the magnitudes of the solute transport volume fractions defined by the modal (peak) solute travel times were estimated to vary between 1 and 2% of the total experimental volume. In addition, possible boundary effects in terms of rapid flow along the wall of the experimental column were investigated.

Introduction

Most of the solid waste generated world-wide is disposed of in landfills and uncontrolled landfilling, known as open dumping, is the world's most common disposal method (Meadows *et al.*, 1997; Rushbrook, 1997). Open dumping is characterised by low standards of environmental protection, e.g. no or ineffective liner systems, landfill gas and leachate collection and management. Hence the leachate from an open dump site may affect directly the groundwater and/or the surface water.

In high-income countries, over the last 4 decades, controlled landfilling (known as sanitary landfilling) has developed. The definition of sanitary landfilling requires the isolation of the waste from the environment until the emissions can be considered to be harmless. Minimising the time over which collection and treatment of leachate and landfill gas is required has therefore become a key issue in developing sanitary landfill technologies and practices.

The physical structure of the solid waste material composing a landfill is highly heterogeneous. Consequently, the presence and mobility of water in landfills is highly non-uniform and the degradation processes, which depend on the moisture content and the transport of substances within the landfill, are accordingly non-uniform. Since the water flow in a landfill is highly non-uniform, the leachate quality reflects only the parts of the landfill where water flow occurs. Sudden changes in the physical structure of the landfill (e.g. settlements due to biodegradation) may change the water flow pattern and, thus, severely influence the quality of the leachate (El-Fadel *et al.*, 1997; Åkesson and Nilsson, 1997).

The non-uniform water flow pattern through a landfill makes the typical sanitary landfill operate as an unmixed water deficient biological reactor (Bogner and Spokas, 1993), and the role of water in biodegradation and landfill gas production has been pointed out (Klink and Ham, 1982; Ehrig, 1983; Barlaz *et al.*, 1990; Augenstein and Pacey, 1991; Ehrig, 1991; Bogner and Spokas, 1993; El-Fadel *et al.*, 1996; Christensen *et al.*, 1996). An increase in moisture content in a landfill enhances the anaerobic degradation processes by facilitating the redistribution of substrates and nutrients and the spreading of micro-organisms between the micro-environments in the landfill, leading to an increase in the methane production rate (Barlaz *et al.*, 1990; Augenstein and Pacey, 1991; Christensen et al. 1996; El-Fadel *et al.*, 1996).

Due to the highly heterogeneous structure of the solid waste material composing a landfill, parameters such as

hydraulic conductivity and field capacity vary greatly. As has been pointed out by Zeiss and Major (1993) and Bendz *et al.* (1998), the material geometry of a landfill facilitates the flow of water in restricted channels and voids. The spatial variation in moisture content in landfills is large and it varies from saturated conditions to complete dryness (Harris, 1979). Field capacity is defined as the maximum moisture content that a porous medium can retain against gravity before continuous downward flow occurs. In landfills, leachate is often released prior to reaching the field capacity (El-Fadel *et al.*, 1997). Bengtsson *et al.* (1994) suggested that water may be flowing in locally saturated regions even though most of the landfill may be well below field capacity. Zeiss and co-workers (Zeiss and Major, 1993; Zeiss and Uguccioni, 1995; Uguccioni and Zeiss, 1997; Zeiss and Uguccioni, 1997) report the practical field capacity, defined as the moisture content at which free drainage just starts, to be considerably lower than the values of the conventional field capacity. The existence of rapid flow in favoured flow paths in solid waste media has been reported in studies on a field scale (e.g. Harris, 1979; Ehrig, 1983; Blight *et al.*, 1992; Bendz *et al.*, 1997b; Rosqvist *et al.*, 1997) and on a laboratory scale (e.g. Blakey, 1982; Uguccioni and Zeiss, 1997).

Numerous column experiments with the aim of measuring water flow or solute transport in solid waste have been reported. In these experiments, fresh municipal solid waste was packed into the experimental column (e.g. Korfiatis *et al.*, 1984; Colin *et al.*, 1991; Lee *et al.*, 1991). No experiments are known to the authors in which hydraulic experiments were carried out on an undisturbed, solid waste sample. In recent studies, with the aim of estimating the fractions of the cross sectional area through which downward water flow occurred, flow sensor plates were placed in experimental columns filled with fresh municipal solid waste (Zeiss and Major, 1993; Uguccioni and Zeiss, 1997). The fraction of the cross sectional area through which the downward water flow occurred was estimated to be about ⅓ of the total cross sectional area of the experimental columns.

With a few exceptions (e.g. Rosqvist *et al.*, 1997; Uguccioni and Zeiss, 1997; Bendz *et al.*, 1998), little effort has been made to investigate the non-uniform flow field in solid waste material. However, a great body of research has been carried out emphasising non-uniform solute transport in structured soils. Several studies have reported that the transport of solutes in structured soils is significantly affected by fast flow in favoured flow paths bypassing other parts of the medium (Brusseau and Rao, 1990). To describe this process, preferential flow is a frequently used generic term (Luxmoore, 1991). As a consequence of preferential flow, the water in the remaining parts of the medium must flow considerably slower, or not at all. Fast advective solute transport in the preferential flow paths is accompanied by diffusive mass transfer of solutes between the preferential flow domain and the domains with slow flow, or stagnant water. The latter domains then behave as solute sink/source components. This phenomenon has been termed physical or transport nonequilibrium (Brusseau and Rao, 1990), and is believed to limit severely the feasibility of predicting flow and transport processes in porous media. To model solute transport in soils which are in a state of transport non-equilibrium, two-domain or multi-domain concepts are often employed (Gerke and van Genuchten, 1993).

As shown in reviews by Bendz *et al.* (1997a) and El-Fadel *et al.* (1997), the prevailing approach for modelling water flow and solute transport in solid waste media relies on the assumption of an homogenous, porous medium (e.g. Ahmed *et al.*, 1992; Demetracopoulus *et al.*, 1986; Korfiatis, 1984; Lee *et al.*, 1991; Straub and Lynch, 1982; Vincent *et al.*, 1991). However, due to the heterogeneous nature of the solid waste medium, this assumption may be questioned. According to Ehrig (1983) and Uguccioni and Zeiss (1997), rapid flow in favoured flow paths is believed to be the reason why existing models are not in agreement with actual field observations. In recent work (Uguccioni and Zeiss, 1997; Bendz *et al.*, 1998), the non-uniform flow has been taken into consideration. Uguccioni and Zeiss (1997) compared a one-dimensional water balance model with a two-domain fractured-porous media flow model. Bendz *et al.* (1998) presented a water flow model taking channel flow into consideration, based on data from the experiments described in this paper.

Successful prediction of leachate quantity and quality is a highly complex and difficult task, and understanding of the spatial and temporal distribution of moisture is not complete (El-Fadel *et al.*, 1997). Zeiss and co-workers (Zeiss and Major, 1993; Zeiss and Uguccioni, 1995; Uguccioni and Zeiss, 1997; Zeiss and Uguccioni, 1997) have pointed out the need to revise and develop existing models to predict the leachate formation better.

In this study, a 3.5 m^3, undisturbed, 20-year-old, solid waste sample was installed to investigate parameters influencing water flow (e.g. porosity, field capacity and hydraulic conductivity) and to measure the distribution of the solute travel time. Altogether, five tracer tests were performed, two under constant water head and steady-state conditions. Three tracer tests were performed under sprinkling conditions, two under steady-state and one under transient conditions. The data were analysed using the transfer function approach which provides a simple theory for estimating the behaviour of solute movement through a porous medium, addressing the distribution of travel times through the porous medium as the principal entity of the solute transport process (Jury, 1982). In the present study, break through curves originating from the tracer tests performed under sprinkling conditions were used to parameterize the solute travel time probability density functions. The expected solute travel times (i.e. the median solute travel times) were assessed and the corresponding fraction of the experimental volumes active in

the transport of solutes (the solute transport volume fractions), was estimated. Processes governing the solute travel times for non-reactive tracers, e.g. slow water flow in the matrix and diffusional mass exchange between flow domains, were discussed. Estimated parameters influencing the water flow are discussed and compared with values reported in the literature. The estimation of the solute transport volume fraction is regarded as the main objective of this study.

Material and methods

THE SOLID WASTE MATERIAL

The experimental landfill from which the large-scale sample was taken was built in 1973 in an area with a high groundwater table. Since the late 1970s, when the deposit area was closed and abandoned, the landfill has, due to seasonal fluctuations, periodically been below the groundwater. The landfill has an area of 300 m^2 and an original depth of 1.8 m, it had a plastic liner at its base and was covered with a soil layer. In 1995, when the sample was taken, the depth was 1.2 m, excluding the soil cover. The waste material consisted of shredded household waste, originally mixed with 35 weight percentage sludge, and it was compacted to a wet density of 0.91 at a water content of 0.53 (Persson and Rylander, 1977). This equals a dry density of 0.38 (Table 1). In Flyhammar *et al.* (1998) the waste characteristics of the sample were investigated. It was found that the amount of paper had decreased by more than 40 percent and the easily degradable materials were almost completely degraded. The largest fractions, sheets, threads and irregular structures from materials such as plastic, textiles, leather, rubber, metal and wood, were found to range in size from 0.2 to 10 cm, the cellulose fibres ranged from 0.2 to 1 cm and the sizes of the fine residuals were less than 0.2 cm (Table 1). The increase in the dry density, compared to the original value, is attributed to settlement due to biodegradation.

SAMPLING PROCEDURE

The experimental column was a steel cylinder 2.0 m high and 1.93 m in diameter, and it acted both as a sampling device and as an experimental column. The depth of the sample was 1.2 m and the diameter 1.93 m. Thus, the diameter of the sample was almost 20 times larger than the largest fractions of the waste, and the depth of the sample was more than 10 times larger. The sample was therefore regarded as being sufficiently large to cover the heterogeneities in the solid waste medium.

Table 1. Comparison of waste characteristics (partly after Flyhammar *et al.*, 1998)

	1973	1995
Depth (m)	1.8	1.2
Wet density (10^3kg m^{-3})	0.91	1.0
Dry density (10^3kg m^{-3})	0.38	0.59
Water content (m^3 m^{-3})	0.53	0.41

Waste fractions	Dry substances (tonnes)	Dry substances (tonnes)
Metal	13.7	11.6
Plastic, textile/rubber/leather	13.4	15.3
Paper	66.5 (59.9[1])	40.3 (25.5[1])
Inert	12.9	10.7
Animal	3.4	0.8
Vegetable	19.3	2.8
Residual	6.2 (< 10 mm)	19.4 (< 2 mm)
Sludge	4.5	—
TOTAL	144.6	101.1

[1] volatile solids (VS) = (dry weight—ash weight)

Table 2. The tracer tests in the large-scale, undisturbed, solid waste sample.

Test No.	Water input	Q (l/min)	Area of applied tracer (m^2)	Tracer	C_{pulse} (mg/l)	m_{pulse} (mg)	C_0 (mg/l)
1	ponding	7.6	1.46	Brill. Blue FCF	1770	17700	0
			1.46	lithium	52	520	
2	ponding	7.6	2.92	lithium	200	4000	0.08
3	sprinkling	2.8	2.92	lithium	200	4000	0.71
4	sprinkling	1.72	2.92	lithium	428	4280	0.99
5	sprinkling	1.72(6.9)[1]	2.92	lithium	405	4050	0.89

[1] True inflow rate during sprinkling in pulses

Before the sample could be taken, the soil covering the landfill was removed. The column was then carefully driven into the waste by alternately excavating the waste material surrounding the column with a mechanical excavator and applying pressure to the top of the column. Cutting around the column by hand using a spade was necessary. When the bottom liner of the landfill was reached, two 10 mm steel sheets were placed flat on the bottom of the pit beside the column and the column was slid on to them. The steel sheets were then welded to the bottom of the column. The weights of the column and the steel sheets were known and, before it was moved to the laboratory, the whole assembly was reweighed to determine the weight of the waste sample. The moisture content of the sample, when it was taken in the landfill, was 0.41 m^3m^{-3} (Table 1), which was the same as the field capacity determined in a smaller undisturbed sample, measuring 0.3 m in diameter and 0.44 m in height (Table 3).

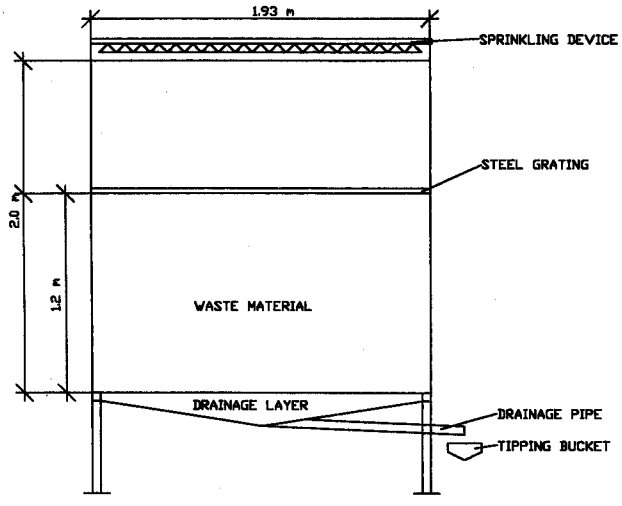

Fig. 1. *A section of the experimental column.*

THE LABORATORY SET-UP

In the laboratory, the steel sheets at the bottom of the experimental column were removed and the column was provided with a drainage layer of coarse gravel and a 75 mm drainage pipe. The bottom edge of the column was sealed to a stand with a silicone sealant. To register the discharge during the experiments, a tipping-bucket device connected to a data-logger was arranged at the outlet of the drainage pipe. Each bucket held 0.489 litre.

The inflow of water to the surface of the experimental volume was arranged in two ways, constant water head and sprinkling. The constant water head (referred to below as ponding conditions) was achieved by adjusting the inflow of water so that a constant head of water was maintained during the test. To sprinkle the water (referred to below as sprinkling conditions), an irrigation system of 19 microsprinklers was set up, mounted on a circular bar which was kept rotating to ensure that the water was applied evenly. The irrigation flux was measured with an electronic flowmeter. To prevent the waste material from expanding when water was added, a steel grating was fixed on top of the waste material. Figure 1 shows a section of the experimental column.

PROPERTIES OF THE SOLID WASTE MATERIAL

The field capacity (FC) was determined by drying one of the small-scale solid waste samples, which was first saturated and then left to drain until the drainage ceased. In this paper, the porosity (n) is defined as the total pore space which water can occupy. Consequently, the porosity corresponds to the saturated moisture content. Thus, possible closed, air-filled pores are not included in the porosity. The porosity in the large-scale sample was calculated as the sum of the field capacity and the water that was drained by gravity after saturation. To estimate the moisture content during tracer tests, the discharge volume was measured after the inflow of water was turned off at the end of the experiment. The moisture content during the

tracer tests was then calculated as the sum of the field capacity and the drained water volume. The moisture content under ponding conditions was measured in tracer test No. 2 and under sprinkling conditions in tracer tests 4 and 5. The calculated results for the moisture content under ponding and sprinkling conditions were 46 and 43 volumetric percent respectively. The saturated hydraulic conductivity (K_{sat}) in the large-scale sample was determined by using a falling head permeameter as described by Jury *et al.* (1991). Dry density was determined by drying the small undisturbed sample until there were no further changes in weight with time.

TRACER TESTS

Altogether, five tracer tests were carried out. The tracers, lithium (LiBr) in experiment Nos. 1 to 5 and Brilliant Blue FCF in experiment No. 1 were regarded as being conservative. Lithium has been used as a conservative tracer in solid waste material by several researchers (e.g., Harris, 1979, Blakey *et al.*, 1997 and Blight, 1996). Brilliant Blue FCF was chosen for the simplicity it provides in analysing the concentrations in the water samples. Although the adsorption of Brilliant Blue FCF could not be neglected, it may be assumed to be minor for tracer tests performed using high water velocities, in soils (Flury and Flühler, 1995).

The experiments started with a period during which water was supplied until a steady discharge rate was attained, after which the water was turned off. Immediately after the water had infiltrated, a pulse of tracer solution was applied with the concentration (C_{pulse}) listed in Table 2. The tracer solutions were applied by being evenly sprinkled by hand over the surface of the experimental volume from four sprinkling cans. As soon as the tracer solution had been infiltrated, the inflow of water was re-established. The sampling procedure started immediately after the tracer solution had been applied. Samples (100 ml) were taken every minute for the first twenty minutes and then at gradually increasing time intervals.

Tracer test No. 1 was carried out to investigate whether any boundary effects due to water flow along the wall of the steel column could be detected. The surface of the experimental volume was therefore divided into two equal areas using a metal ring that was forced 5 cm into the solid waste. Brilliant Blue FCF was applied in the outer area and lithium in the inner circle. It was assumed that high inflow rates would enhance any boundary effects, therefore tracer test No. 1 was performed under ponding conditions. Since the first experiment indicated boundary effects, a slurry of very fine quartz sand was firmly packed at the edge of the steel cylinder to avoid water flow along the wall of the steel column in the following experiments. Tracer test No. 2 was then performed to investigate if the measures taken to prevent boundary effects along the wall of the steel cylinder were effective. Tracer test No. 2 was performed under the same flow rate as in the first tracer test, but this time the lithium pulse was applied over the whole surface area of the experimental column.

Tracer tests Nos. 3 to 5 were performed to investigate solute transport under unsaturated conditions. In these tracer tests, lithium was applied over the whole surface area of the experimental column. Tracer tests Nos. 3 and 4 were performed under steady-state sprinkling conditions at different flow rates of the inlet water (Table 2). In the fifth tracer test, the water inflow was sprinkled over the surface of the experimental volume in pulses at intervals of 24 minutes. Each sprinkling period lasted for 6 minutes. The average inflow rate in experiments 4 and 5 was the same (Table 2).

Since lithium was used as tracer in experiments following upon each other, the experimental volume was contaminated and thus, the background concentration of lithium increased with the tracer tests. In Table 2 the measured background concentrations (C_0) are compiled. To avoid clogging in the bottom of the experimental volume or in the drainage layer of coarse gravel, each tracer test was kept of as short duration as possible. The risk of clogging was also the reason for not performing two experiments under identical conditions. In Table 2, information about the five tracer tests is summarised. Q is the inflow and m_{pulse} is the amount of tracer applied.

Results

PROPERTIES OF THE SOLID WASTE MATERIAL

In Table 3, estimated parameters of the undisturbed sample are compared with values reported in the literature. The parameters reported by others were estimated in fresh solid waste that was packed into experimental columns. Compared with the results reported by others, the porosity (n) of the undisturbed sample was in the same range (Zeiss and Major, 1993) or lower (Colin *et al.*, 1991). The high porosity reported by Colin *et al.* (1991) is consistent with the low dry density. Despite the differences in experimental set-ups, the field capacity (FC) reported in this study was of the same order as the value reported by Colin *et al.* (1991). The practical field capacity given by Zeiss and Major (1993) is defined as the moisture content at which free drainage just starts, and it is thus estimated differently from the conventional field capacity reported here. The dry density (ρ_{dry}) reported in this study was, due to settlement following the degradation of the material, expected to be higher than the value obtained in fresh waste. It is suggested that the high dry density value stated in Korfiatis *et al.* (1984) was due to the differences in experimental set-ups and it also illustrates the variability in the nature of the waste material. The saturated hydraulic conductivity (K_{sat}) in this study was in the same range ($10^{-5} - 10^{-4}$ ms^{-1}) as the values reported by Korfiatis *et al.* (1984). The values reported by Colin *et al.* (1991)

Table 3. Parameters of the large-scale, undisturbed, solid waste sample compared to results reported in the literature.

	n ($m^3\ m^{-3}$)	FC ($m^3\ m^{-3}$)	ρ_{dry} ($10^3\ Kgm^{-3}$)	K_{sat} ($10^{-5}\ ms^{-1}$)
Korfiatis *et al.* (1984)			0.62	8–13
Colin *et al.* (1991)	0.65	0.40	0.40	10–50
Zeiss and Major (1993)	0.53	0.136*		
Large-scale undisturbed sample	0.53	0.41	0.59	4

* Practical FC

were up to 10 times higher, possibly due partially to the high porosity.

INVESTIGATION OF BOUNDARY EFFECTS

Tracer test No. 1 was designed to investigate whether any boundary effects in terms of rapid flow along the wall of the steel cylinder could be detected (see Materials and Methods section). The peak concentration of Brilliant Blue FCF applied in the outer area of the surface of the experimental column appeared ahead of that of lithium, after 5 and 11 minutes respectively (Fig. 2). Thus, boundary effects in terms of rapid flow along the wall of the steel cylinder were indicated. The next tracer test (No. 2) was performed to investigate whether the measures to prevent the boundary flow along the wall of the steel cylinder were effective. In Fig. 3 the lithium BTCs in tracer tests Nos. 1 and 2 are compared. The peak concentration of tracer test 2 appears slightly before that of tracer test No. 1 (after 9 and 11 minutes respectively). This indicated that the boundary effects may not have been completely eliminated. However, since the following tracer tests were performed under sprinkling conditions and thus at much lower flow rates, boundary effects due to rapid flow along the wall of the steel cylinder were assumed to be negligible.

ESTIMATION OF THE SOLUTE TRANSPORT VOLUME FRACTION

Transfer function analysis was carried out to estimate the expected solute travel time (i.e. the median solute travel time) and the corresponding solute transport volume fraction. For a narrow-pulse input of a tracer through the surface of the experimental volume at time zero, and on the assumptions that the water flow is steady and that the solute is non-reactive, the solute travel time probability density function (pdf) ($g(t)$), can be expressed as the normalised mass flux (White *et al.*, 1986)

$$g(t) = \frac{q \cdot C_{ex}(t)}{\rho_A} \quad (1)$$

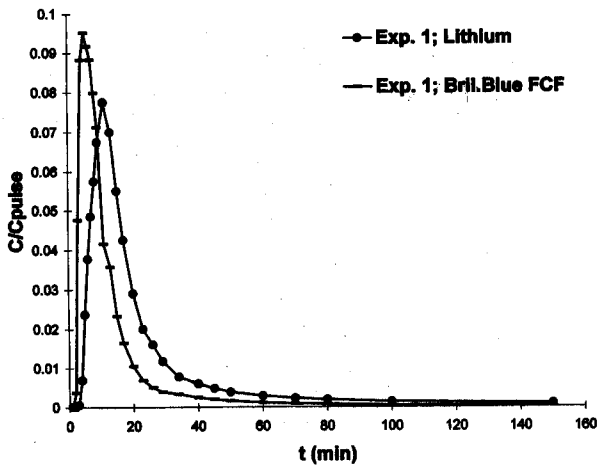

Fig. 2. *BTCs of lithium and Brilliant Blue FCF of tracer test No. 1 in terms of normalised concentrations.*

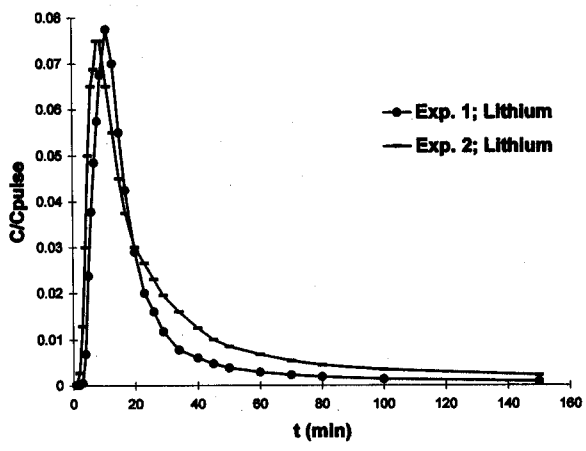

Fig. 3. *BTCs of lithium of tracer tests Nos. 1 and 2 in terms of normalised concentrations.*

Table 4. Results of the tracer tests performed under sprinkling conditions (Nos. 3 to 5).

Test No.	3	4	5
Inflow conditions	Steady-state	Steady–state	Transient
q (10^{-5} ms^{-1})	1.60	0.98	0.98 (3.92)[1]
μ (–)	4.86	4.70	5.37
σ (–)	1.65	1.30	1.50
r^2 (–)	0.86	0.89	0.88
$\tilde{t}$(s)	7740	6600	12840
$\tilde{\theta}$ (m^{3}m^{-3})	0.10	0.05	0.10
t^{mode} (s)	780	1920	2640
Θ_{st}^{mode} (m^{3}m^{-3})	0.01	0.02	0.02

[1] True flux rate during sprinkling in pulses.

In Eqn. (1), q is the steady-state Darcian water flux, $C_{ex}(t)$ is the solute concentration in the output flow, ρ_A is the mass per unit area of the applied solute, and Eqn. (1) yields

$$\int_0^\infty g(t)dt = 1. \qquad (2)$$

Positively skewed BTCs have successfully been described by a two-parameter log-normal pdf $g(t)$ (Jury, 1982; Jury and Sposito, 1985; White *et al.*, 1986; White, 1987),

$$g(t) = \frac{1}{\sqrt{2 \cdot \pi} \cdot \sigma \cdot t} \cdot e^{\frac{-(\ln(t)-\mu)^2}{2 \cdot \sigma^2}} \qquad (3)$$

where μ and σ are fitting parameters and t is the time variable. The parameters μ and σ^2 represent the mean and the variance of ln(t), respectively. In these experiments, it was possible to achieve good fits to all the BTCs using Eqn. (3) (Figs. 4 to 6). The fitting parameters μ and σ and the r^2 values for tracer tests Nos. 3 to 5 are listed in Table 4.

The solute transport volume ($\tilde{\theta}_{st}$) is defined as the volume fraction of the wetted pore space which is active in the advective movement of solute (White *et al.*, 1986; Butters *et al.*, 1989) and it corresponds roughly to the mobile water content in the mobile-immobile water model described by van Genuchten and Wierenga (1976), (Jury *et al.*, 1991). $\tilde{\theta}_{st}$ can be estimated from the median of the

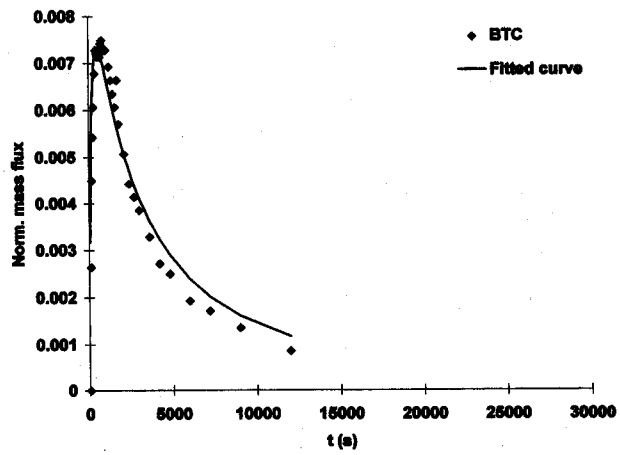

Fig. 4. *BTC in terms of normalised mass flux and fitted log-normal distribution of tracer test No. 3.*

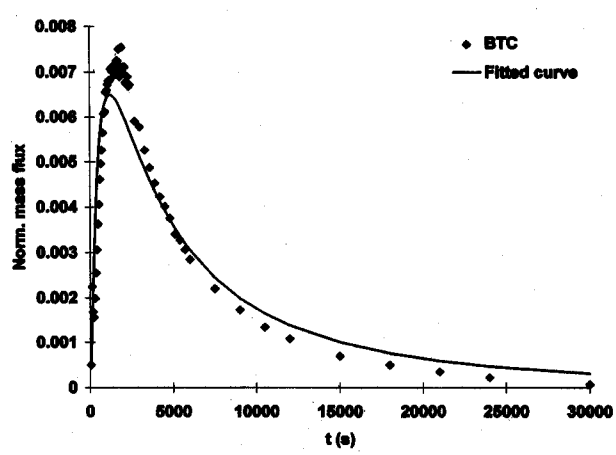

Fig. 5. *BTC in terms of normalised mass flux and fitted log-normal distribution of tracer test No. 4.*

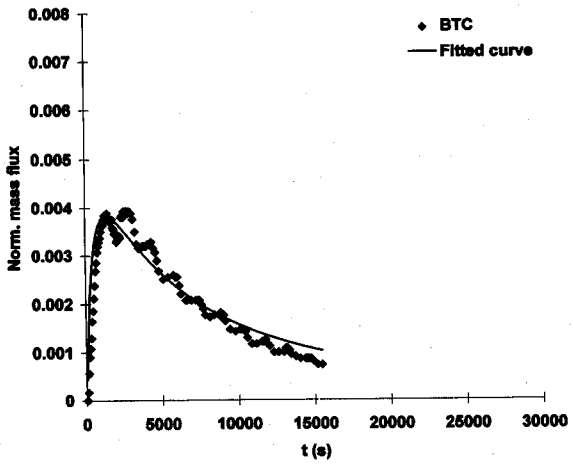

Fig. 6. *BTC in terms of normalised mass flux and fitted log-normal distribution of tracer test No. 5.*

solute travel time distribution ($\tilde{t}$) (White *et al.*, 1986; Butters *et al.*, 1989)

$$\tilde{\theta}_{st} = \frac{q \cdot \tilde{t}}{L} \tag{4}$$

where

$$\tilde{t} = \exp(\mu) \tag{5}$$

The high water flux rate during pulses in tracer test No. 5 resulted in a high expected solute travel time (Table 4). It is suggested that, during transient conditions, water is forced into less mobile domains during the pulses and then, between the pulses, these domains are drained. This process, resulting in a reversible advective solute transport between the domains, is believed to enhance the process in which the domains with less mobile water are acting as a sink/source component in the solute transport process. Moreover, the low peak concentration of tracer test No. 5 (Fig. 6) indicated the processes whereby the less mobile domains are acting as solute sink/source components. The high $\tilde{\theta}_{st}$ values found in tracer tests Nos. 3 and 5 (0.10 m^3 m^{-3}, Table 4) indicate that a high flow rate, steady-state (No.3) and in pulses (No. 5), makes a larger fraction of the experimental volume participate in the transport of the solutes.

The modal travel time (t^{mode}) defines the arrival of the peak or maximum average concentration. The modal travel time and a fraction of the transport volume defined relative to the modal travel time ($\tilde{\theta}_{st}^{mode}$) may be important in characterising certain environmental problems since this travel time indicates the earliest arrival of significant concentrations of solute at the lower boundary of the transport volume (White *et al.*, 1986; Butters *et al.*, 1989). The solute transport volume relative to t^{mode} is defined by replacing $\tilde{t}$ with t^{mode} in Eqn. (4). As expected, the high steady-state water flux rate in tracer test No. 3, compared to that of No. 4, resulted in an earlier arrival of the peak concentration (t^{mode}, Table 4). When the tracer test performed under transient conditions (No.5) was compared to a steady-state tracer test performed with the same average flow rate (No.4), high water flux rates in the pulses resulted in a late arrival of the peak concentration. The late arrival of the peak concentration is believed to be a result of the processes whereby a high water flux during the pulses forces the water to enter domains with less mobile (or stagnant) water. However, the estimated $\tilde{\theta}_{st}^{mode}$ values were of the same magnitude, 0.01 for test No.3 and 0.02 for tests Nos. 4 and 5 (Table 4). It is therefore suggested that the fast solute transport represented by t^{mode} is dominated by fast advective flow and that it takes place in a small fraction of the total volume.

Discussion and conclusions

During high water flux rates, steady-state and transient, larger $\tilde{\theta}_{st}$ were estimated. It is suggested that the high flux rates forced water to enter less mobile domains resulting in slower displacement of the solute, which then may enhance the mass exchange due to diffusion. The large $\tilde{\theta}_{st}$ during transient inflow conditions is suggested to be due partially to a reversible advective transport between domains with rapid flow and domains with less mobile (or immobile) water. This phenomenon has been discussed in structured soils (Reedy *et al.*, 1996; Hutson and Wagenet, 1995), and shown in a modelling study by Hutson and Wagenet (1995) to enhance the exchange of solutes between domains. The magnitude of the water flux rates appeared to have a minor influence on $\tilde{\theta}_{st}^{mode}$, which was estimated to be small and dominated by the fast advective transport. It seems physically reasonable that the highly non-uniform solute transport in the waste mass is due to the structure of the large fractions of the waste material, leading to continuous voids enhancing the preferential flow.

The results show that, in order to improve long term prediction of the leachate quality and quantity, the magnitude of the true volume participating in the water flow through a landfill must be taken into account. As a consequence of the water flow in favoured flow paths, bypassing other parts of the medium, the leachate quality reflects only the flow paths and their surroundings. Sudden changes in the physical structure of the landfill (e.g. settlements due to biodegradation) may change the water flow pattern and thus new parts of the landfill may be exposed to moving water. As a consequence, the quality of the leachate may be changed dramatically.

In this study, high water flux rates resulted in larger $\tilde{\theta}_{st}$ participating in the transport of solutes. Therefore, techniques enhancing the moisture distribution in solid waste landfills, such as recirculation of leachate and the flushing bioreactor concept, in order to speed up the biodegradation and the leaking of pollutants, are supported. However, to force larger parts of the landfill volume to participate in the biodegradation processes and the leachate formation, existing techniques need to be developed further.

Acknowledgement

The financial support for this study was provided by the Swedish Waste Research Council (AFN).

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