MOVEMENT OF POLLUTANTS THROUGH CLAYEY SOIL

by

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ABSTRACT

This paper examines a number of factors which should be considered when attempting to predict the impact of landfill sites on groundwater contamination. The relative importance of transport mechanisms such as diffusion, dispersion and advection are discussed as well as the significance of attenuation mechanisms. Techniques for determining relevant parameters are outlined and the applicability of laboratory techniques for determining diffusion and distribution coefficients is discussed with respect to the observed migration of contaminants beneath the Sarnia Landfill.

Simple but effective models for calculating the migration of contaminant from landfills are discussed and their application illustrated by a number of examples. Finally, factors such as the impact of the leachate collection system and the migration of contaminant from landfills designed to have an inward gradient is examined.

INTRODUCTION

The movement of potential contaminants through clayey soils is usually by the processes of advection (seepage), diffusion and dispersion. This paper sets out to (1) briefly review these transport processes; (2) comment on their

relative importance, (3) examine how relatively simple techniques can be used to determine a number of relevant parameters, and (4) discuss some relatively simple techniques which can be used to make an engineering assessment of potential contaminant impact of a waste disposal site.

Attention will be focused on contaminant migration through relatively thin clayey barriers which may be underlain by a man-made collection system or a natural aquifer. The clayey barriers are assumed to be saturated (or nearly saturated). This assumption is likely to be applicable to many natural clayey deposits which are located below the watertable and to well constructed compacted clay liners (i.e. liners which are compacted wet of optimum moisture content and are not allowed to dry out).

Much of the current state-of-the-art with respect to Geotechnical Practice for Waste Disposal was summarized in the proceedings of an ASCE specialty conference held in 1987 (Woods, 1987). This present paper does not attempt to review those proceedings but, rather, attempts to highlight and summarize some of the issues raised by the author in a paper at that conference (Rowe, 1987) as well as in a number of more recent papers by the author and/or his co-workers (Quigley et al., 1987; Rowe, 1988; Rowe et al., 1988; Barone et al., 1988).

CONTAMINANT TRANSPORT PROCESSES

The primary mechanisms for contaminant transport through saturated (or nearly saturated) clayey barriers are advection and diffusion. Geotechnical engineers are generally quite familiar with advective transport which involves the movement of contaminant with flowing water. In the absence of diffusion, contaminant would be transported out of a landfill at the groundwater (seepage) velocity. Again, in the absence of diffusion, it is obvious that there would be

no outward contaminant transport through a landfill barrier if the advective flow were into the landfill. However, diffusion can not be neglected. Diffusion is a process whereby chemical species move from the location of high chemical concentration to points of low chemical concentration. If the direction of diffusive transport is the same as the direction of advective flow then it will increase the amount of contaminant transported and decrease the time it takes for contaminant to move to a given point away from the source. Diffusion can also occur in the direction opposite to advective transport and so, as will be demonstrated later, it is possible for contaminant to escape from a landfill even though the groundwater flow is directed into the landfill. The level of impact can be calculated and often the landfill can be designed to maintain an acceptable level of impact (which may be no significant impact).

The detailed processes associated with diffusion of contaminants through clayey soils may be quite complicated (e.g. see Quigley et al., 1987). Nevertheless, geotechnical engineers can obtain parameters and calculate impacts for use in design using relatively simple techniques. Both the determination of parameters and the assessment of potential impact involve the use of mathematical modelling. For the simplest case of one-dimensional advective-diffusive transport, the governing differential equation can be readily obtained from consideration of conservation of mass within any small region, and can be written as

$$n \frac{\partial c}{\partial t} = (nD \frac{\partial^2 c}{\partial z^2} - nv \frac{\partial c}{\partial z}) - \rho K \frac{\partial c}{\partial t} \qquad (1)$$

where n = effective porosity of the soil

c = concentration at depth z and time t

D = D_e = diffusion coefficient of the species of interest for this soil

v = seepage (average linearized groundwater) velocity

ρ = soil dry density

K = distribution or partitioning coefficient

v_a = nv is the Darcy or discharge velocity

This equation simply states that the increase in contaminant concentration within a small volume of soil is equal to the increase in mass due to advective-diffusive transport minus the mass of contaminant removed from solution by what are loosely referred to as "sorption processes". Here, it is assumed that the sorption processes are linear and can be represented in terms of partitioning or distribution coefficient K (see Freeze and Cherry, 1979, for more details regarding "sorption processes"). The product ρK (where ρ is the soil density) is a dimensionless measure of the amount of sorption which is likely to occur. A contaminant species is said to be "conservative" if there is no sorption (i.e. $\rho K = 0$). A typical example of a conservative contaminant would be the ion chloride (Cl^-). Typical examples of contaminants whose migration and impact may be retarded by sorption processes are the metals Na^+, K^+, Pb^{++}, Cd^{++}, Fe^{++}, Cu^{++}.

Diffusion Coefficients

Table 1 summarizes the diffusion coefficients of a number of ionic species. As is evident from Table 1, the diffusion coefficient may vary depending on the species of contaminant being considered and the nature of the porous media through which diffusion is occurring. Thus, for example, the diffusion coefficient through the sand-bentonite mixtures examined by Gillham et al. (1984) varied depending on the proportions of sand or bentonite. Also, it is evident that the diffusion of chloride through a clayey soil (porosity $\approx$ 0.4) and shale (porosity = 0.1) are different. What is not evident from Table 1 is the fact

TABLE 1 SOME DIFFUSION COEFFICIENTS

Silica Sand-Bentonite Mixture	Diffusion Coefficients		Reference
	Cl^- cm^2/s	^3H(Tritium) cm^2/s	
100% - 0%	10×10^{-6}	17×10^{-6}	1
90% - 10%	7×10^{-6}	9×10^{-6}	
85% - 15%	8×10^{-6}	10×10^{-6}	
50% - 50%	9×10^{-6}	11×10^{-6}	
0% - 100%	10×10^{-6}	12×10^{-6}	

	Cl^- (cm^2/s)	Na^+ (cm^2/s)	Ca^{++} (cm^2/s)	K^+ (cm^2/s)	
Sarnia Silty Clay	6×10^{-6}	5×10^{-6}	4×10^{-6}	7×10^{-6}	2
Queenston Shale	1.5×10^{-6}				3

[1] Gillham et al., 1984

[2] Rowe et al., 1988

[3] Barone and Rowe, 1987

that diffusion coefficients (and sorption parameters) for a given species can also depend on the chemical composition of the source leachate (e.g. see Barone et al., 1988). This can be true even for conservative ionic species such as chloride. Thus the diffusion (and sorption) coefficients should be determined for the proposed soil using a leachate as near as practicable to that expected in the field situation. Techniques for determining parameters in this way will be outlined in a later section. Fortunately, despite the potential variation in diffusion coefficients due to soil and leachate composition, the range of variation is relatively small compared to that of many other parameters (e.g. hydraulic conductivity). In engineering terms, diffusion is in fact a very predictable process and natural diffusion processes established over thousands of years have been shown to be consistent with very simple theoretical predictions based on Equation 1 (e.g. see Desaulniers et al., 1981).

Dispersion Coefficients

In the foregoing discussion, the coefficient D was taken to be the diffusion coefficient. This tends to be appropriate when contaminant is moving at very low Darcy velocities through clayey soils. For the higher flows often associated with aquifers, an additional mechanism called "mechanical dispersion" (e.g. see Freeze and Cherry, 1979) can give rise to a spreading of the contaminant plume. Although this mechanism is totally different from the diffusion process, for most practical purposes, it can be mathematically modelled in the same way and hence the two processes are often lumped together as a composite parameter D called the "coefficient of hydrodynamic dispersion" viz.

$$D = D_e + D_m \tag{2}$$

TABLE 2   REGIONAL DISPERSIVITIES (after Anderson, 1979)

Aquifer	Location	Porosity	Longitudinal Dispersivity (m)	α_T/α_ℓ
Alluvial	Rocky Mountain	0.30	30.5	1.0
	Colorado	0.20	30.5	0.3
	California	NR	30.5	0.3
	Lyons, France	0.2	12	0.33
	Barstow CA	0.40	61	0.3
	Sutter Basin CA	0.05-0.2	80-200	0.1
Glacial	Long Island, NY	0.35	21.3	0.2
Limestone	Brunswick, GA	0.35	61	0.3
Fractured	Idaho	0.10	91	1.5
Basalt		0.10	91	1.0
	Hanford Site, WA	NR	30.5	0.6
Alluvial	Barstow, CA	0.40	61	1/330
	Alsace, France	NR	15	0.067
Glacial Till		0.001 and	3.0 and	
Over Shale	Alberta, Canada	0.053	6.1	0.2
Limestone	Cutler Area, FL	0.25	6.7	0.1

Note: α_ℓ = longitudinal dispersivity; α_T = transverse dispersivity

neglecting diffusion; and (c) considering both advection and diffusion for a
range of advective velocities. The results are replotted in Figure 1. The esti-
mated flux was based on the maximum of the flux obtained from either case (a) or
(b) and the actual flux was that deduced from case (c) for a given Darcy velocity
v_a. The neglect of either diffusion or advection gives rise to an underesti-
mate (i.e. unconservative estimate) of the contaminant flux through the liner.
Examining Fig. 1, it is evident that diffusion is the dominant mechanism at low
Darcy velocities (i.e. less than 2×10^{-4} m/a). The error associated with not
considering both advective and diffusive transport is greatest for a Darcy velo-
city of 0.006 m/a. This Darcy velocity is typical of what might be encountered
for a liner with a field hydraulic conductivity of 10^{-7} cm/s and an outward
gradient of 0.2. In many practical situations, the hydraulic conductivity and
gradient will be such that the Darcy velocity is less than (or equal to) 0.006
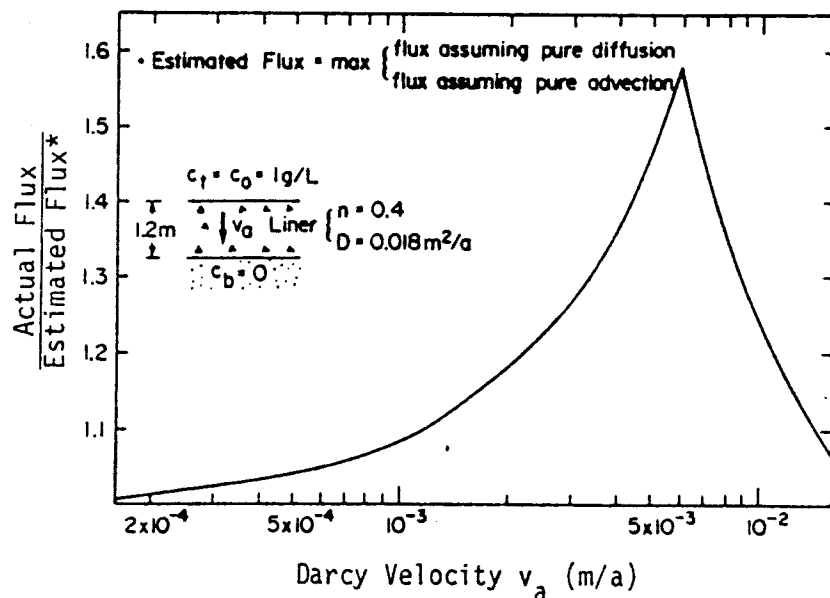m/a. Diffusive transport must be considered for these cases.

FIGURE 1   ERROR IN THE CALCULATED PEAK FLUX THROUGH A 1.2 m THICK CLAY LINER
WHICH CAN ARISE FROM NOT CONSIDERING BOTH DIFFUSION AND ADVECTION
(After Rowe, American Society of Civil Engineers, 1987)

DETERMINATION OF PARAMETERS

The Darcy velocity depends on both the hydraulic gradient and the hydraulic conductivity of the soil. The gradient may depend upon the initial hydrogeologic conditions or may be totally engineered. The gradient will often depend on the design of the leachate collection system and the height of leachate mounding. The construction and quality assurance associated with the hydraulic conductivity of compacted clay liners has been discussed by Lahti et al. (1987). For a discussion of the effect of interaction between leachate and clay upon hydraulic conductivity, see Mitchell and Madsen (1987), Fernandez and Quigley (1985; 1988), Bowders and Daniel (1987).

The author's experience has been that the effective porosity, n, of saturated (or near saturated) clayey barriers is often reasonably estimated based on water content determined according to usual geotechnical practice. However, situations can be envisaged where the effective porosity could be less. The effective porosity can be determined using the procedure described by Rowe et al. (1988). The soil density ρ can be determined according to normal geotechnical practice.

The parameters least familiar to the geotechnical engineer are the diffusion coefficient $D = D_e$ and the partitioning coefficient K. Both parameters can be estimated from a single test as outlined below and described in detail by Rowe et al. (1988).

In the proposed test, an undisturbed sample of soil is placed in a column and the leachate of interest is placed above the soil. Contaminant is then permitted to migrate through the specimen under the prescribed head (which may be zero). The volume of leachate above the soil will normally be selected to be sufficiently small to allow a significant drop in concentration of contaminant

within the source solution (typically the height of leachate in the column above the clay will range from 0.05 to 0.3 m). This drop in concentration with time should be monitored.

A number of possible boundary conditions at the base of the sample may be considered. If the test is to be conducted with advective transport through the specimen, then a porous collection plate can be placed beneath the sample and the effluent collected and monitored. If there is no advective flow, then two other base boundary conditions may be considered. Firstly, the base could be an impermeable plate (see Fig. 2a). The second alternative is to have a closed collection chamber (reservoir) similar to that for the leachate but initially having only a background concentration of the contaminant of interest (see Fig. 2b). Thus, as contaminant passes through the soil, it accumulates (and can be monitored) in this collection chamber.

Height of Leachate H_f

Source leachate has initial concentration C_0. Concentration will decrease as contaminant moves into the soil.

Soil

H

Zero Outflow

(a)

H_f

Soil

H

Collection chamber. Concentration will increase as contaminant moves through the soil. Concentration variation with time can be monitored.
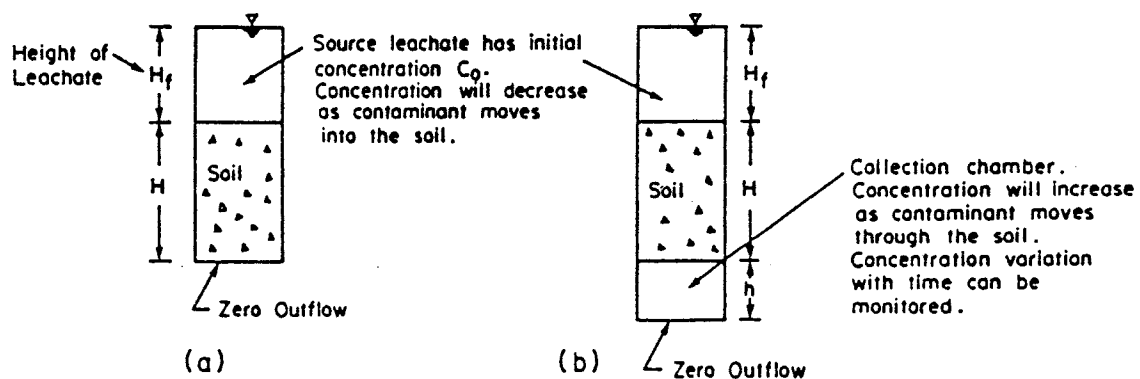
h

(b)

Zero Outflow

FIGURE 2   SCHEMATIC OF PURE DIFFUSION TESTS (a) ZERO FLUX AT BASE OF THE SOIL, (b) MIGRATION INTO A COLLECTION CHAMBER (After Rowe, Caers and Barone, Canadian Geotechnical Journal, 1988)

Suppose that the volume of source solution (leachate) is equal to $A \cdot H_f$ where A is the plan area of the column and H_f is the "height of the leachate" in the column (e.g. see Fig. 3). Then at any time t, the mass of any contaminant species of interest in the source solution is equal to the concentration $c_t(t)$ in the solution multiplied by the volume of solution (assuming here that the solution is stirred so that $c_t(t)$ is uniform throughout the solution). The principle of conservation of mass then requires that at this time t, the mass of contaminant in the source solution is equal to the initial mass of the contaminant minus the mass which has been transported into the soil up to this time t. This can be written algebraically as

$$c_t(t) = c_0 - \frac{1}{H_f} \int_0^t f_t(\tau) d\tau \tag{4}$$

where $c_t(t)$ is the concentration in the source solution at time t_i

c_0 is the initial concentration in the source solution (t=0)

A   is the plan area of the column

H_f is the height of leachate (ie. the volume of leachate per unit area)

$f_t(\tau)$ is the mass flux of this contaminant into the soil at time τ

Contaminant is allowed to migrate from the source chamber through the soil and, if present, into the collection chamber. If no additional contaminant is added to the source chamber, then the concentration of contaminant will decrease with time as mass of contaminant diffuses into the soil (see Fig. 4). The rate of decrease can be controlled by the choice of the height of leachate, H_f. Conversely, as contaminant diffuses into the collection chamber (Fig. 2), the increase in mass gives rise to an increase in contaminant concentration in this reservoir (see Fig. 4). The rate of decrease in concentration in the source and increase in the collection chamber should be monitored with time. At some time t_f, the test is terminated and the concentration profile through the soil

CONCEPT
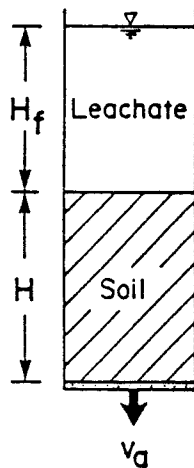
Concentration in leachate varies with time as contaminant moves into the soil

Concentration at time t = Initial Concentration − Total mass flux into the soil per unit volume of leachate

$$c(t) = c_0 - \frac{1}{H_f} \int_0^t f(T)\, dT$$

Where flux $f = nvc - nD \dfrac{\partial c}{\partial z}$

FIGURE 3 SCHEMATIC SHOWING HOW THE CONCENTRATION OF CONTAMINANT IN THE SOURCE VARIES AS CONTAMINANT IS TRANSPORTED INTO THE SOIL

PROCEDURE

- Monitor Source Leachate Concentration With Time

- Monitor Effluent Concentration With Time

- Determine Concentration Profile Through Sample at end of Test (Time t_f)

- Calculate D & ρK by Fitting Theoretical Solution to the Experimental Curves
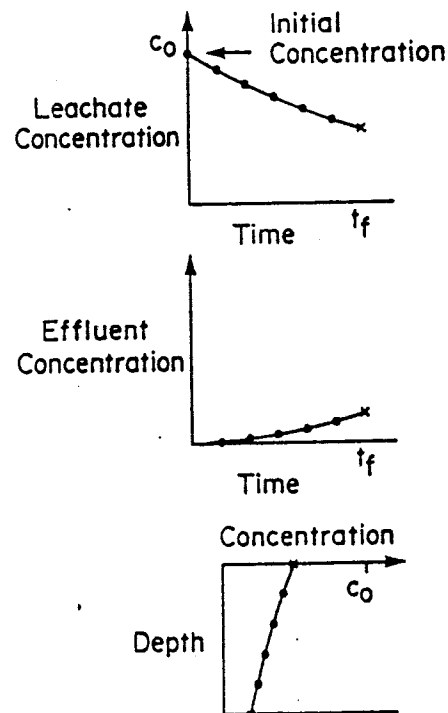
FIGURE 4 EXPERIMENTAL PROCEDURE USED TO DETERMINE THE DIFFUSION COEFFICIENT D AND DISTRIBUTION COEFFICIENT K (After Rowe, Canadian Geotechnical Journal, 1988)

sample may be determined (see Fig. 4). Assuming linear sorption, theoretical models can then be used to estimate the parameters n, D and ρK. This theoretical analysis has been described in detail by Rowe and Booker (1985a, 1987) and has been implemented in the computer program POLLUTE (Rowe et al., 1984). This approach permits very accurate calculation of concentration in only a few seconds on a microcomputer and hence is well suited for use in interpretation of the results of the column tests.

To illustrate the application of the procedure, Fig. 5 shows the chloride concentration profile through a sample of clay till from Sarnia, Ontario (see Rowe et al., 1988 for details). Also shown is the calculated concentration profile for D = 0.019 m^2/a (6x10^{-6} cm^2/s) and it can be seen that a good match can be obtained between the observed and experimental profiles.

Despite the fact that this test had only been run for four days (without any advection), some chloride had migrated to the bottom of the sample (i.e. 4 cm). When performing these tests using an impermeable base, it is desirable to terminate the test at about the time chloride reaches the bottom of the deposit. If concentration is allowed to build up at the base (as seen in Fig. 5) then accuracy may be lost; the greater the buildup the greater the potential loss in accuracy. For example, in the worst case the test could be run long enough for steady state conditions to develop (i.e. a uniform concentration throughout). In this case, the concentration profile is independent of the diffusion coefficient and hence a match could be obtained for any number of different diffusion coefficients. On the other hand, providing the test is terminated while there is significant diffusion still occurring (e.g. when contaminant is just reaching the base) then the parameter can be determined uniquely (to within the experimental accuracy of the concentration determination). To achieve this, an initial
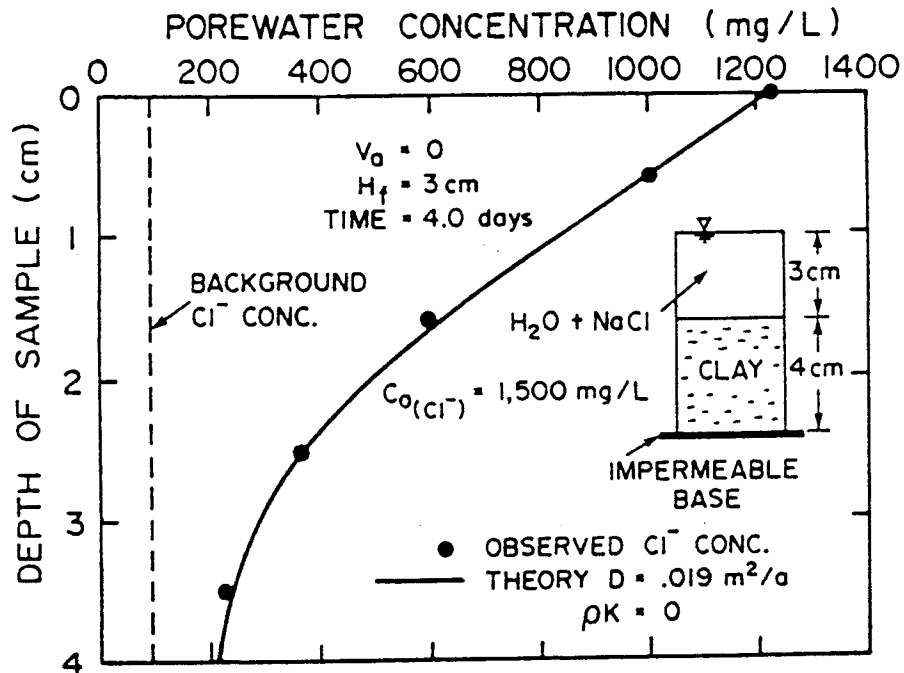
FIGURE 5   CHLORIDE CONCENTRATION VS DEPTH OF SAMPLE FOR PURE DIFFUSION MODEL
(After Rowe, Caers and Barone, Canadian Geotechnical Journal, 1988)

estimate of the time of termination can be made by estimating the diffusion coefficient and using POLLUTE to simulate the migration. This, of course, assumes that the diffusion coefficient can be reasonably estimated prior to the test. Nevertheless, even if a good estimate cannot be made initially and a buildup in concentration does occur, the results of this test will usually give a fairly good estimate of the diffusion parameter. This parameter can be used to determine a better termination time for a second test, and the test can be repeated as a check. For the case shown in Fig. 5, the diffusion coefficient was not affected by the buildup of concentration at the base.

COMPARISON BETWEEN OBSERVED AND CALCULATED FIELD PROFILES

The Confederation Road landfill near Sarnia, Ontario is one of the best documented case histories where the migration of contaminants has been monitored in an insitu natural clay barrier. The migration has been carefully monitored over a 14 year period by researchers at The University of Western Ontario (Goodall and Quigley, 1977; Crooks and Quigley, 1984; Quigley and Rowe, 1986; Quigley et al., 1987a,b; Yanful et al., 1988). Figure 6 shows the observed concentration profiles for chloride (Cl^-), sodium (Na^+) and potassium (K^+) which were obtained in 1984 (after about 16 years migration). It is evident from Fig. 6 that over 16 years chloride (a conservative species) has migrated about 1.3 m below the waste. The cations Na^+ and K^+ are being retarded by interaction (cation exchange) with the clay with the more highly sorbed K^+
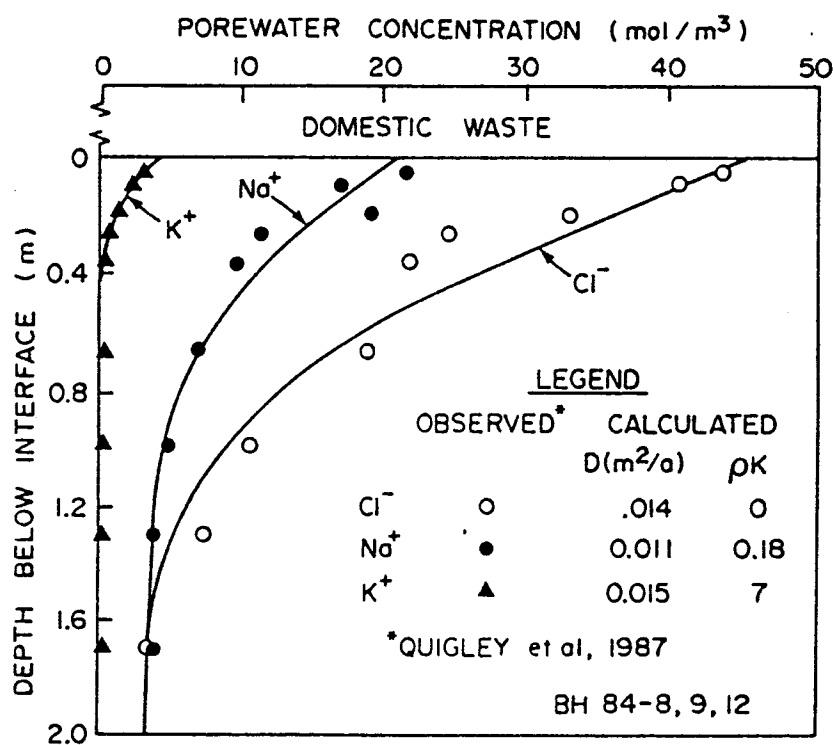

FIGURE 6   COMPARISON OF OBSERVED AND CALCULATED MIGRATION PROFILES BENEATH THE
CONFEDERATION ROAD LANDFILL AFTER 16 YEARS MIGRATION

having migrated less than 0.4 m. An investigation of the migration of the heavy metals copper, lead, zinc and iron (Quigley et al., 1987; Yanful et al., 1988) has shown that they have been restricted to the upper 0.1 to 0.2 m of the clay in this same time period.

Laboratory tests were performed to determine diffusion and partitioning coefficients for the three species Cl^-, Na^+ and K^+ (Rowe et al., 1988). The diffusion coefficients which were obtained at laboratory temperature were adjusted for field temperature (e.g. see Crooks and Quigley, 1984; Quigley et al., 1987) and the concentration profiles were calculated and plotted in Fig. 6. Allowing for inevitable experimental scatter, it can be seen that the agreement between observed and calculated profiles is quite encouraging. (A more detailed discussion of modelling at this landfill is given in Quigley and Rowe, 1986).

MASS OF CONTAMINANT AND LEACHATE COLLECTION

For waste disposal sites such as municipal landfills, the mass of any potential contaminant within the landfill is finite. The process of collecting and treating leachate involves the removal of mass from the landfill and hence a decrease in the amount of contaminant which is available for transport through the liner and into the general groundwater system. Similarly, the migration of contaminant through the barrier also results in a decrease in the mass available within the landfill. For a situation where leachate is continually being generated (e.g. due to exfiltration through the landfill cover), the removal of mass by either leachate collection and/or contaminant migration will result in a decrease in leachate strength with time (i.e. there will be a decrease in concentration similar to that observed in the laboratory test described in a previous section).

On the simplest level, suppose that the infiltration into the landfill was q_0, the exfiltration through the liner was q_a and the leachate collected (per unit area) q_c (see Fig. 7) then, assuming the landfill is at field capacity, continuity of flow requires that

$$q_0 = q_c + q_a \qquad (5)$$

If each of these quantities is a representative average value, then the proportion of contaminant which can pass into the soil is q_a/q_0 (similarly, the proportion of contaminant collected would be q_c/q_0). Thus if the initial mass m_{TC} of a contaminant species (e.g. Cl^-) can be estimated, then the mass available for transport into the soil m_0, is given by

$$m_0 = m_{TC} \cdot q_a/q_0 \qquad (6)$$

If the peak concentration of this contaminant species is c_0, then the mass of contaminant m_0 can be represented as an equivalent volume of leachate V_0 viz.

$$V_0 = m_0/c_0 \qquad . \qquad (7)$$

In general, this volume will not correspond to the actual volume of leachate because (i) it only represents the portion of the total mass which is available for transport into the hydrogeologic system and (ii) it is based on the conservative assumption that all this available mass can be quickly leached from the solid waste. It is convenient for both mathematical and physical reasons to express the volume V_0 in terms of an "equivalent height of leachate" H_f, where H_f is defined as the volume of leachate divided by the area, A_0, through which contaminant passes into the primary "barrier" i.e.,

$$H_f = V_0/A_0 \qquad (8a)$$

EQUIVALENT HEIGHT OF LEACHATE H_f

Landfill with a leachate collection system — "Solid Waste"
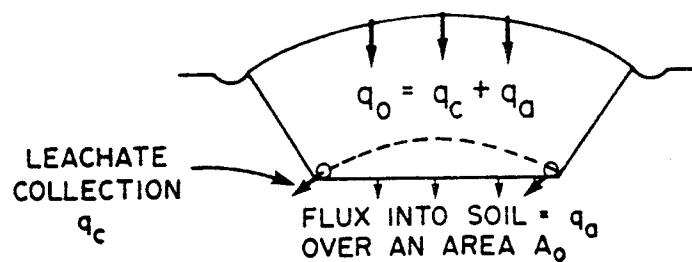
Proportion of contaminant which can pass into the soil $= q_a/q_o$

Maximum "Initial" Concentration of Contaminant Species $= c_o$

Total mass of contaminant species in the waste $= m_{TC}$

Mass of contaminant likely to be transported into the soil: $m_o = m_{TC} \cdot q_a/q_o$

Equivalent volume of leachate $=$ mass/Initial Concentration: $V_o = m_o/c_o$

Equivalent height of leachate $=$ (volume)/(area $\perp$ flow): $H_f = V_o/A_o$

$$\text{i.e.} \quad H_f = \frac{m_{TC}}{c_o A_o} \cdot \frac{q_a}{q_o}$$

FIGURE 7   DERIVATION OF THE "EQUIVALENT HEIGHT OF LEACHATE" TO REPRESENT THE MASS OF CONTAMINANT AVAILABLE FOR TRANSPORT INTO THE SOIL

or

$$H_f = \frac{M_{TC}}{c_o A_o} \cdot \frac{q_a}{q_o} \tag{8b}$$

As noted above, q_o represents the volume of leachate generated within the landfill (per unit area) and can usually be taken to be equal to the infiltration into the landfill. The quantity q_a may be defined as the average flux into the

barrier normalized (divided) by the average concentration within the landfill and referred to here as the normalized average flux. For situations where advection is the dominant transport mechanism, q_a is approximately equal to the Darcy velocity, v_a, (i.e. $v_a = nv = ki$ where k is the hydraulic conductivity of the barrier and i is the outward hydraulic gradient in the barrier). However, for situations where diffusion is a significant transport mechanism, the determination of the normalized average flux q_a is a little more complicated but can be readily estimated as outlined below.

Using mathematical modelling (e.g. Program POLLUTE), the contaminant flux into the clay barrier can be determined for any combination of advection and diffusion as outlined by Rowe (1988). The average mass flux, f_a, into the liner can be determined and the normalized average flux, q_a, can then be calculated by dividing by the initial concentration c_0, viz.

$$q_a = \frac{f_a}{c_0} \tag{9}$$

For the purposes of determining q_a, it is conservative to assume that (i) the concentration in the landfill remains constant; and (ii) the concentration in the aquifer is zero. The results obtained for this case are shown in Fig. 8 for a barrier of thickness greater than or equal to 1 m. Here, the normalized average flux (q_a) is plotted against the Darcy velocity through the liner for a range of values of the product nD where n is the porosity of the barrier and D is the effective diffusion coefficient of the contaminant being considered. Thus, for example, if the porosity of the soil were $n = 0.4$, the diffusion coefficient $D = 0.02$ m^2/a and the Darcy velocity $v_a = 0.002$ m/a then $nD = 0.4 \times 0.02 = 0.008$ and hence, from Fig. 8, $q_a \simeq 0.013$ (noting the logarithmic scale).
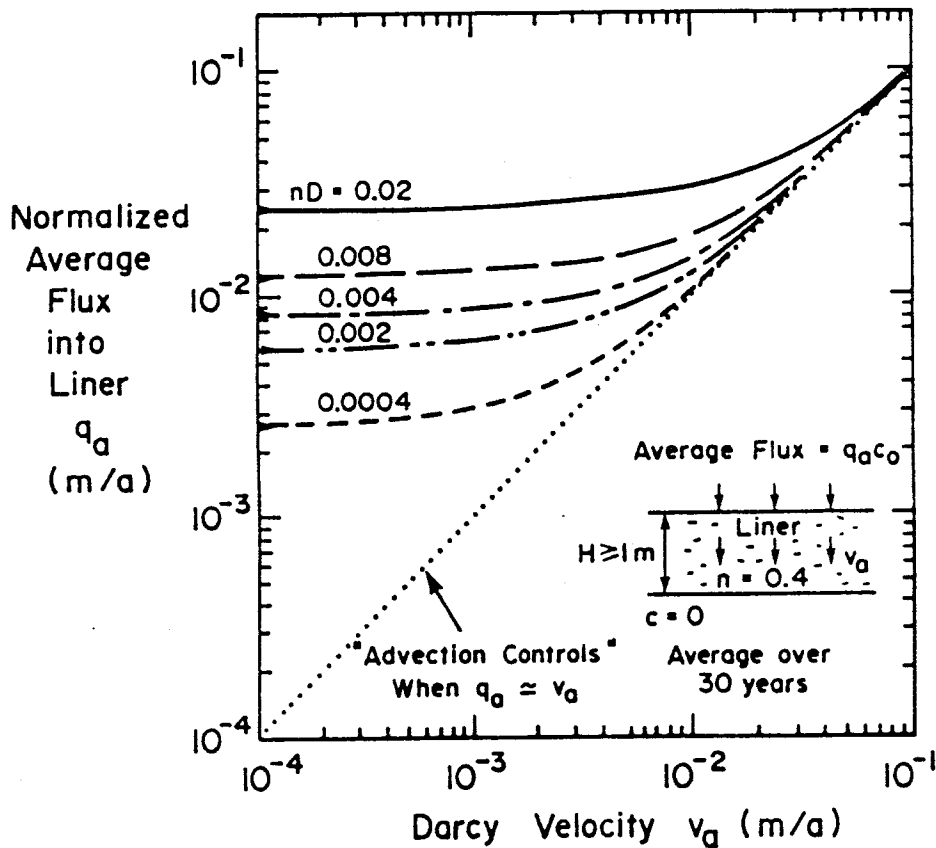
FIGURE 8   RELATIONSHIP BETWEEN NORMALIZED AVERAGE FLUX INTO A LINER AND THE
DARCY VELOCITY FOR A RANGE OF DIFFUSION COEFFICIENTS (After Rowe,
Canadian Geotechnical Journal, 1988).

The results presented in Fig. 8 may be conservatively used for situations
where the concentration in the leachate decreases with time and/or the concentra-
tion beneath the barrier is greater than zero.

Advection controls contaminant transport when the normalized average flux
(q_a) is approximately equal to the Darcy velocity (v_a). For typical situa-
tions involving clayey barriers this will be the case for Darcy velocities
greater than 0.03 m/a. For velocities less than this, diffusion may noticeably
increase the normalized average flux and by inspection of Fig. 8 it can be seen

that for a given diffusion coefficient there is a minimum value of q_a. Thus, even if there were no flow into the soil (i.e. zero hydraulic gradient), contaminant would still pass into the barrier.

Having determined q_a from an analysis program such as POLLUTE, or from Fig. 8, the mass of contaminant available for transport can be estimated. For example, based on limited data, a reasonable estimate for the mass of chloride may be 0.1% of the total mass of waste. Thus, for example, if the total mass of waste in a proposed landfill of area A_o = 25 ha were 2 Mt then the total mass of chloride in the waste would be 0.1% of 2 Mt i.e. m_{TC} = $0.001 \times 2 \text{-} 10^6$ t = 2000 t. If the estimate peak chloride concentration in the landfill c_o = 2000 mg/L, then the equivalent height of leachate can be determined (see Fig. 7) to be

$$H_f = \frac{m_{TC}}{c_o A_o} \frac{q_a}{q_o} = \frac{2000 \times 10^6}{2000 \times 25 \times 10^4} \frac{q_a}{q_o} = 4 \frac{q_a}{q_o} \text{ (m)}$$

The value of q_a may be determined as described above. The infiltration q_o through the landfill cover must be estimated. In Ontario, a value of q_o commonly used is 300 mm/a. It should be noted that when estimating contaminant impact it is important to be realistic in the estimation of the value of q_o. For example, it is not conservative to use a design value of q_o = 0.3 m/a if the realistic infiltration is, say, 0.15 m/a.

ANALYSIS OF CONTAMINANT IMPACT

There are a wide variety of techniques available for the analysis of contaminant impact, including finite element, finite difference, finite layer, boundary element and analytic solutions. Of these, the finite element programs are most general. For many practical problems, however, techniques such as the

finite layer method provide a very attractive alternative for use in practical design situations. Finite layer techniques have been described by Rowe and Booker (1985a,b; 1987) and are available as computer programs for 1½ D (POLLUTE: Rowe et al., 1983) and 2D (MIGRATE: Rowe and Booker, 1988) conditions.

For situations involving a clayey liner overlying a drainage layer which can be pumped or overlying a thin natural aquifer (e.g. see Figs. 9-12), a reasonable initial estimate of contaminant impact can be obtained in seconds using a micro-computer and 1½ D finite layer programs such as POLLUTE (e.g. see Rowe and Booker, 1985a). The designation of these programs as 1½ D is intended to indi-cate that they consider one-dimensional transport down into the upper aquifer (or drainage layer) and also approximately take account of lateral migration within the aquifer. These techniques will often provide a reasonable estimate of both the magnitude of the peak impact beneath the landfill and the time at which this occurs.

A more rigorous solution of this problem can be obtained using a full 2D analysis (e.g. program MIGRATE). A comparison of the two approaches has been given by Rowe and Booker (1985b). The 2D approach allows one to consider mul-tiple layers (e.g. see Fig. 9) and impact at points outside the landfill. The full 2D analysis can be readily performed on a microcomputer but it does involve substantially more computation than the 1½ D analysis. Before performing 2D analyses, a 1½ D analysis should <u>always</u> be performed to estimate the likely magnitude of impact and its time of occurrence beneath the landfill; the 2D pro-gram can then run for appropriate times using the 1½ D results as a reference. One of the advantages of finite layer techniques over conventional finite element methods is that it is unnecessary to determine solutions at times prior to the

time period of interest (which is usually the time period when peak impact will occur). Thus it is unnecessarily wasteful of the engineer's (and computer's) time to evaluate solutions at times well before (or after) the peak impact will occur. As noted above, this waste can be avoided by using a 1½ D program to determine the time period where attention should be focused.

To illustrate the application of some of the concepts discussed in the previous sections, consider a landfill with dimensions 200 m x 1250 m (A_o = 25 ha) separated from a thin underlying aquifer (1 m thick) by a 2 m thick clayey till liner. The groundwater flow is in the direction of the shorter side (i.e. 200 m). The thin aquifer is assumed here to be underlain by an additional 10 m of clay till and a second 2 m thick aquifer, however the primary impact will clearly be on the upper thin aquifer and attention will be focused on this. Figure 9 shows concentration at the downgradient monitoring point "x" for an infinite mass of contaminant (H_f = $\infty q_a/q_o$) and a finite mass of contaminant (H_f = 4 q_a/q_o (m)) for assumed downward Darcy velocities of 0.03 m/a and 0.003 m/a. If one assumes that the concentration in the source remains constant for all time (i.e. H_f = ∞) then a ten-fold decrease in Darcy velocity v_a only reduces the peak concentration by about 35% from 0.46 c_o to 0.3 c_o. However, when one considers the finite mass of contaminant (specifically H_f = 4 q_a/q_o) then this ten-fold decrease in Darcy velocity gives rise to a more than ten-fold decrease in peak concentration from 0.11 c_o to 0.01 c_o. The corresponding increase in the time required to reach this peak was from a little over 60 years to about 700 years. For a Darcy velocity of 0.003 m/a as considered here, assuming a constant source concentration would result in an overestimate of the peak concentration by a factor of thirty if the mass of contaminant corresponds to H_f = 4 q_a/q_o m (e.g. 2000 tonnes over a site of area 25 ha at an initial source concentration of 2000 mg/L).
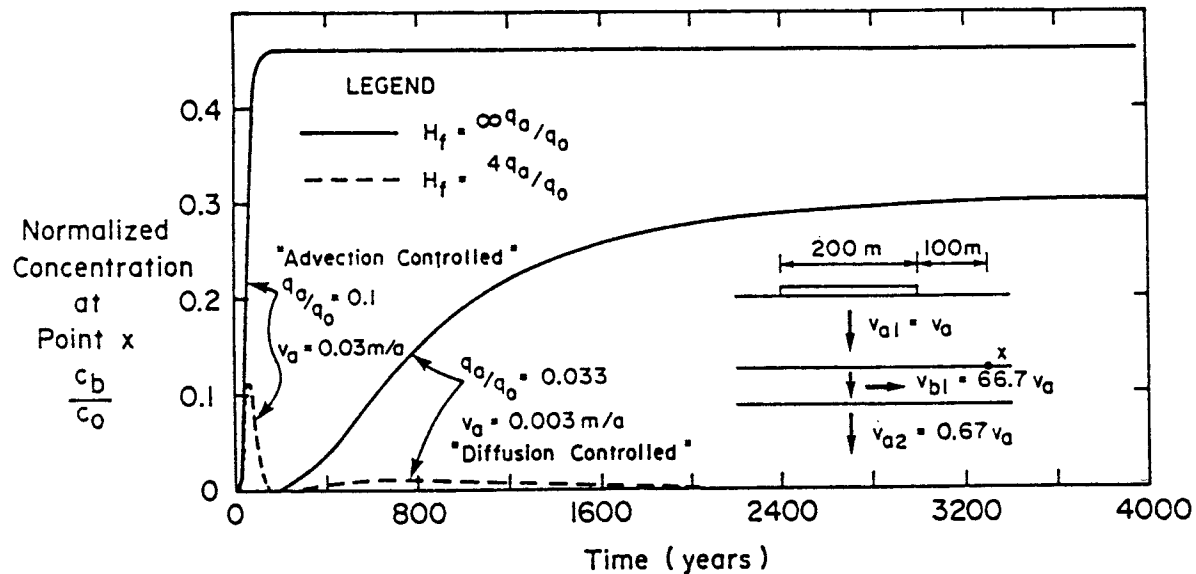
FIGURE 9 EFFECT OF EQUIVALENT HEIGHT OF LEACHATE (H_f) ON THE VARIATION IN CONCENTRATION WITH TIME AND CONTAMINANT IMPACT AT A POINT IN THE AQUIFER BENEATH THE SITE BOUNDARY (After Rowe, Canadian Geotechnical Journal, 1988).

Attenuation

Figure 9 shows results for a conservative contaminant species ($\rho K = 0$) which does not interact with the soil. Many contaminant species will interact and will be removed from solution by processes such as cation exchange (e.g. metals) or by partitioning with the organic matter in the soil (e.g. organic contaminants such as benzene, toluene etc.). Figure 10 shows the normalized variation in concentration in an aquifer with time at two locations (i.e. at the downgradient edge of the landfill: x = 100 m and 300 m downgradient of the landfill: x = 400 m) for both a conservative species ($\rho K = 0$) and a moderately sorbed contaminant species ($\rho K = 10$). These results are for a finite mass of contaminant ($H_f = 1$ m) and hence the concentration at any point increases to a peak value and then subsequently decreases. As might be expected, the peak value is reached at the

downgradient edge of the landfill (x = 100 m) before it is reached at the point x = 400 m. This delay is largely due to diffusion of contaminant from the sand layer into the clay. This reduces the mass of contaminant in the aquifer and results in a decrease in concentration as can be appreciated by comparing the peak impact at the points x = 100 m and 400 m.

Effect of Base Velocity

The results presented in Fig. 10 were obtained for a specific value of the advective velocity within the aquifer (v_b = 1 m/a). This parameter is important; it is also difficult to determine in practice. What can be determined is a reasonable estimate of the range in which the velocity is expected to lie. Under these circumstances, finite layer techniques can be easily used to determine the effect of this uncertainty upon the expected impact. For example, Fig. 11 shows the peak concentrations obtained at x = 100 and 400 m for analyses performed for a range of base velocities v_b.

Beneath the edge of the landfill (x = 100 m), the maximum concentration decreases monotonically with increasing base velocity due to the consequent increased dilution of the contaminant in high volumes of water. However, at points outside the landfill area, there is a critical velocity which gives rise to the greatest "maximum" concentration. As indicated by Rowe and Booker (1985b), this situation arises because of the interplay of two different attenuation mechanisms. The first of these, diffusion into the surrounding clayey soil, is dependent on the time required to reach the monitoring point. Generally, the lower the velocity v_b, the more time there is for contaminant to diffuse away and hence the lower the maximum concentration. The second mechanism, dilution, involves decreasing contaminant concentration due to higher volumes of water (i.e. higher v_b).
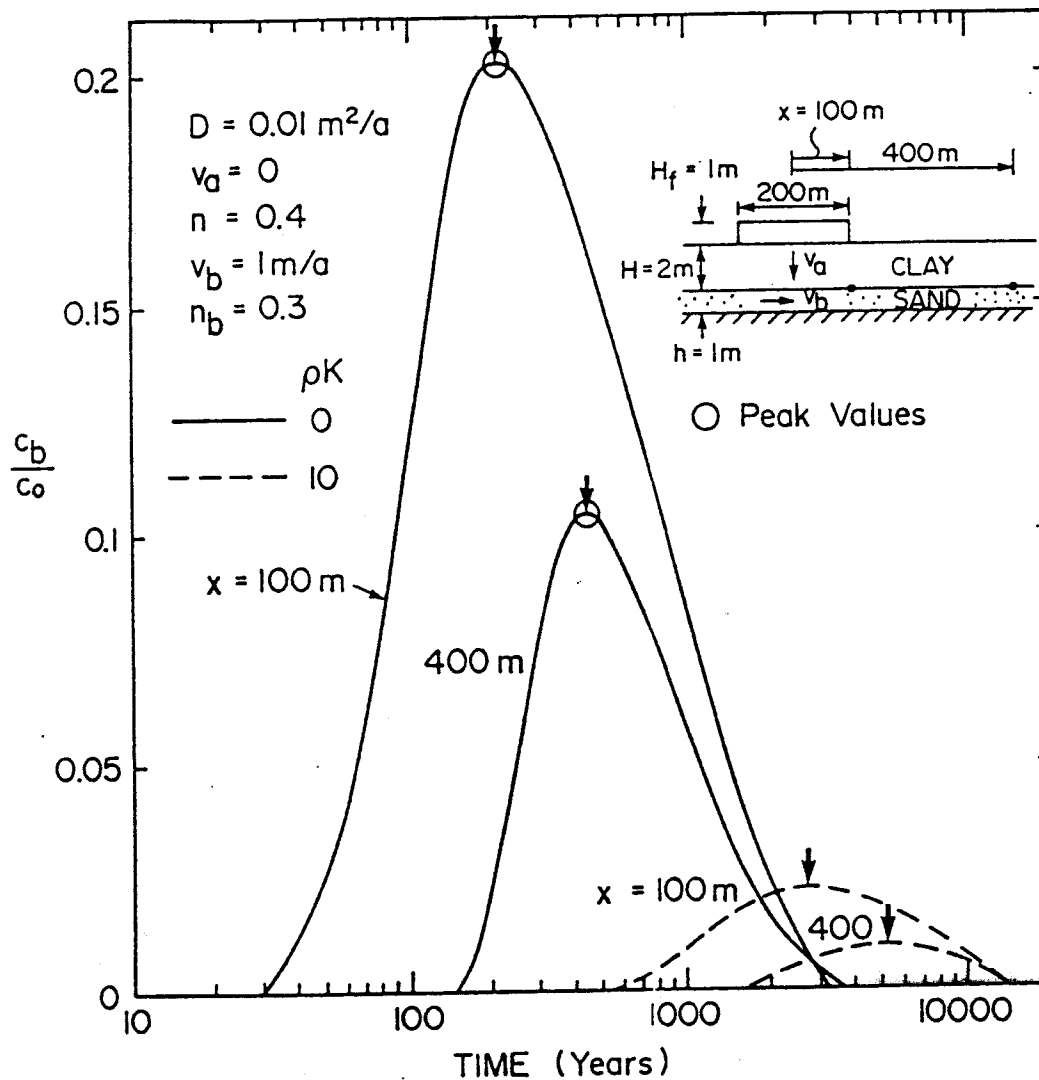
FIGURE 10   ATTENUATION OF THE CONTAMINANT DUE TO DIFFUSION INTO THE CLAY
ADJACENT TO THE AQUIFER AND DUE TO SORPTION (After Rowe and Booker,
Canadian Geotechnical Journal, 1985b).

An important practical consequence of the foregoing is that it is not neces-
sarily conservative to design only for the maximum and minimum expected veloci-
ties in the aquifer. In performing sensitivity studies, sufficient analyses
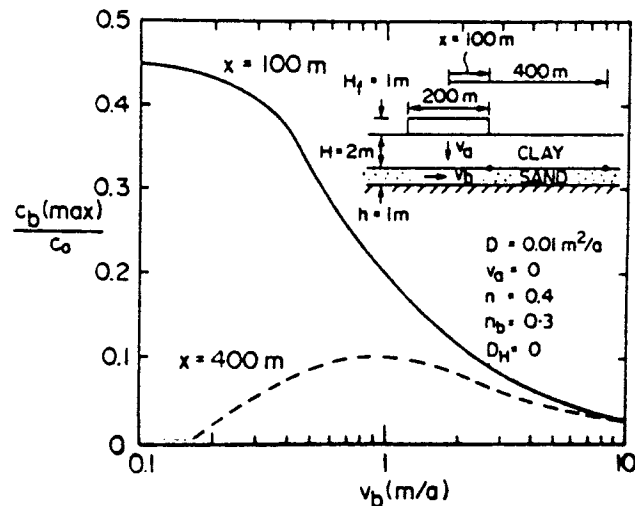should be performed to either determine the critical velocity or, alternatively,

FIGURE 11   VARIATION IN PEAK CONCENTRATION AT TWO POINTS (x=100 m, 400 m) AS A
FUNCTION OF THE DARCY VELOCITIES IN THE AQUIFER, v_b (After Rowe
and Booker, Canadian Geotechnical Journal, 1985b).

to show that the critical velocity does not lie within the practical range of
velocities for the case being considered.

Effect of Landfill Size

The foregoing results have all been for a landfill with a width (parallel to
the direction of flow in the aquifer) of 200 m. Figure 12 shows the variation in
the peak cocentration at the edge of the landfill (point E) and 300 m downgra-
dient (point x) for a range of landfill widths L. For the problem considered, it
can be seen that increasing the width of the landfill increases the concentration
at both points of interest, although the maximum concentration tends to become
asymptotic to a constant value for L approaching 1000 m. As indicated by Rowe
and Booker (1986), this increase in concentration with L arises because of the
increased mass loading of the aquifer which arises from a large total mass of
contaminant within the landfill. The tendency of the asymptote to reach a con-
stant value for very large L arises because significant diffusion can occur into
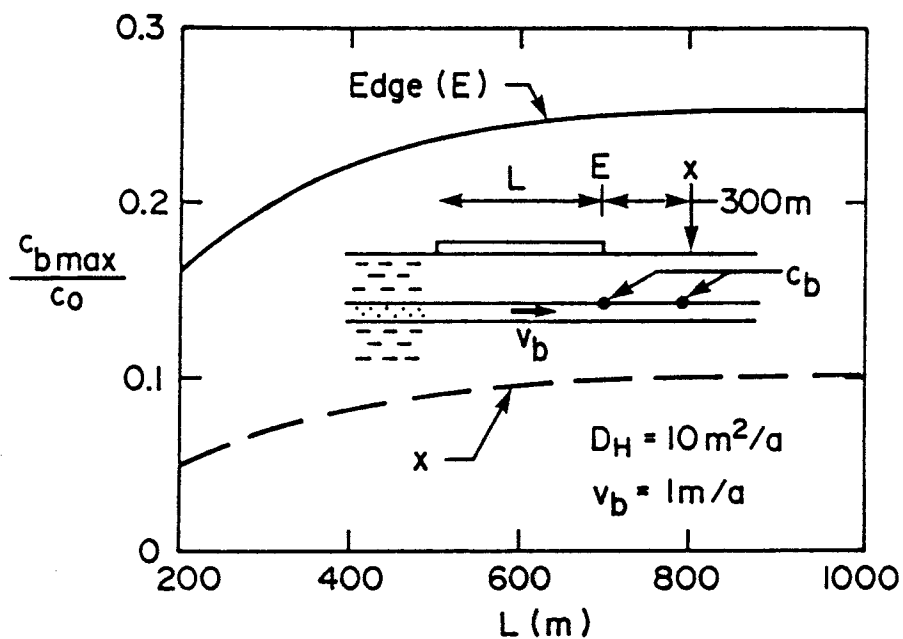
FIGURE 12   EFFECT OF LANDFILL SIZE ON PEAK CONCENTRATION (After Rowe and Booker, Geotechnique, 1986).

the underlying clay between the time that the contaminant enters the aquifer near the upstream edge and the time that it approaches the downstream edge when the width of the landfill, L, is large. (The value of L at which this occurs will depend on the specific parameters for each case.)

"Hydraulic Trap"

As noted earlier, contaminant migration from a landfill can be minimized if the landfill is designed so that groundwater flow is into the landfill. However, the fact that there is inward flow does not necessarily mean that there will be no contaminant migration out of the barrier. The assessment of potential impact on groundwater gradiation involves two stages: viz.

(i)   determine the Darcy velocity into the landfill.

(ii) determine the diffusive movement out of the barrier.

Figure 13 shows the calculated steady-state contaminant flux passing into an aquifer beneath a 1 m thick clayey barrier for a range of inward Darcy velocities and assumed diffusion coefficients.

The flux f has been divided by the initial source concentration and the resulting normalized flux f/c_0 has units of velocity. (This may, in fact, be thought of as the equivalent outward velocity of contaminant migration which occurs due to the outward diffusive transport which is in opposition to inward flow). The effective diffusion coefficient for many contaminants lies in the range $D = 0.01$ to 0.02 m^2/a. For these conditions, Fig. 13 shows that the inward Darcy velocity would have to exceed 0.025 and 0.05 m/a for $D = 0.01$ and 0.02 m^2/a respectively before the outward flux was reduced to negligible levels for a 1 m thick liner.
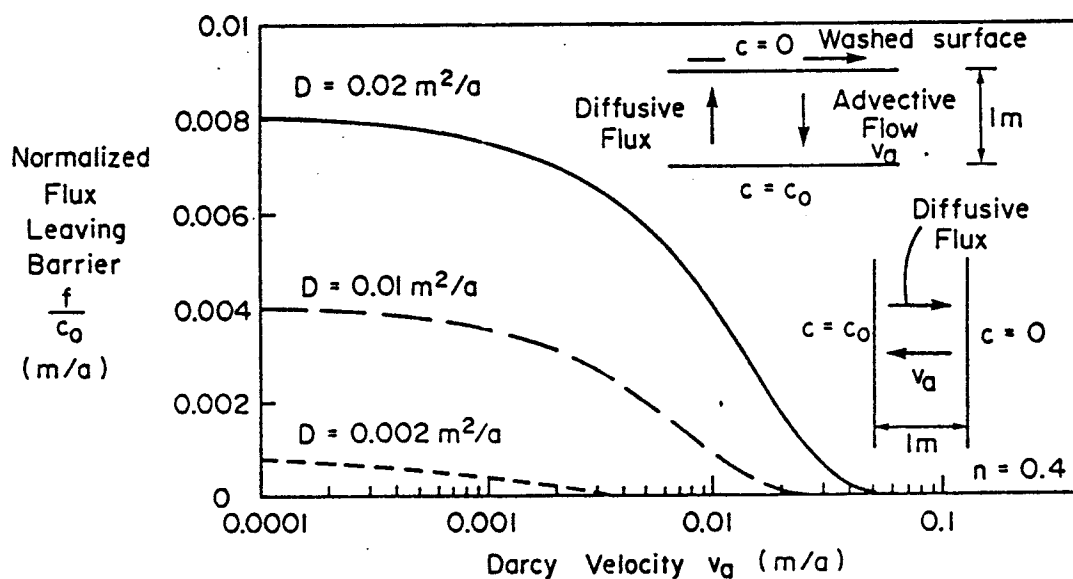

FIGURE 13   DIFFUSIVE FLUX OF CONTAMINANT INTO AN AQUIFER AGAINST AN INWARD FLOW AT A DARCY VELOCITY v_a FOR STEADY STATE CONDITIONS (After Rowe, Canadian Geotechnical Journal, 1988).

CONCLUSION

This paper has reviewed transport process and has concluded that diffusion is an important (often critical) mechanism controlling contaminant migration in well designed modern landfills with clayey barriers. Advective-diffusive transport can be readily modelled on a microcomputer using techniques such as the finite layer method. Parameters such as the diffusion coefficient and distribution coefficient can also be determined using these modelling techniques combined with laboratory column tests. These tests should be performed using the soil to be used in the liner and with a leachate which is considered to be representative of that expected in the field.

Examination of actual diffusion profiles beneath existing landfills shows that while diffusion does occur it is slow and predictable. Furthermore, considerable attenuation of heavy metals and organic chemicals can occur due to interaction with the clay.

Using modern laboratory and computer modelling techniques safe landfills can be designed using clayey liners.

ACKNOWLEDGEMENT

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