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# Pollutant migration through clay soils

## Migration des polluants à travers les sols argileux

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**SYNOPSIS** A new and very simple method of calculating contaminant migration profiles through soils is illustrated by comparing observed and predicted migration profiles both in laboratory model tests and in the field. This technique permits consideration of a changing concentration in the leachate due to mass transport into the soil, diffusion, advection and sorption within the soil, as well as the effects of the presence of a permeable strata beneath the clay. Using this method of analysis, both the diffusion/dispersion coefficient and the distribution coefficient can be deduced for a particular contaminant from a single laboratory model test as described in the paper.

### INTRODUCTION

The movement of contaminants from waste disposal sites into the general groundwater system may be inhibited by a liner which can take the form of a natural insitu clay soil or a compacted clay borrow. Since the liner will often be underlain by a relatively permeable soil, it should be designed to limit the concentration of contaminant in the pore fluid of this underlying strata. However, design of an appropriate liner will usually require some calculation of the contaminant concentration profile and in particular, an estimate of the maximum contaminant concentration in the groundwater will be needed.

In this paper, the use of a new and very simple method for calculating concentration profiles will be illustrated for situations where the clay liner and underlying strata are of finite thickness. Consideration will first be given to the use of this technique for backfiguring soil parameters from laboratory column tests. Potential problems arising from the use of an effective diffusion coefficient will then be illustrated and a procedure for determining both the dispersion and sorption parameters from one test will be described. Finally, the use of the technique in the back analysis of field results will be demonstrated.

### THE ANALYSIS AND PROGRAM POLLUTE

The movement of contaminants from waste disposal sites (e.g. landfills) into the surrounding soil is often governed by the one-dimensional dispersion-advection equation viz.

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial z^2} - v \frac{\partial c}{\partial z} - \frac{Kc}{n} \frac{\partial c}{\partial t} \quad (1)$$

where  $c$  = concentration of contaminant at depth  $z$  at time  $t$ ;

$D$  = coefficient of hydrodynamic dispersion for the contaminant;  
 $v$  = average linearized seepage velocity;  
 $n$  = effective porosity of the soil;  
 $K$  = distribution (sorption) coefficient;  
 $\rho$  = bulk density of soil  
 $t$  = real time.

The dispersion coefficient  $D$  is usually determined from back-calculations of earlier field tests or from laboratory model tests and includes the effect of tortuosity. The interpretation of field migration and laboratory model tests involves finding the solution to equation 1. The parameter  $K$  is usually determined from separate batch tests (e.g. Hajek and Ames, 1968).

For most cases involving mass transport through clay liners, the advective (superficial, Darcy) velocity  $v_a = nv$  will be relatively small and the coefficient  $D$  will primarily reflect diffusion rather than dispersion effects. The adsorption term is assumed to be linear. This is the approach adopted by many previous workers (e.g. see Gillham and Cherry, 1982) and although sorption may not be precisely linear, the use of a suitable linear relationship determined over the appropriate concentration range may be expected to provide reasonable and conservative results.

For many landfills with a clay liner, it can be assumed that the pollutant species of interest has a maximum concentration,  $c_0$ , shortly after construction (time zero) and that this concentration will then decrease with time as material is transported into the soil. The total mass of any particular species of pollutant available for migration from the landfill is directly related to the concentration of that species within the leachate and to the total volume of leachate. The concentration can be measured. The volume of contaminant is more difficult to determine but may be estimated from the porosity of the landfill material and the time average height of the water table within the landfill. The height of

leachate  $H_f$  represents the volume of leachate divided by the area of the landfill. An examination of a limited number of landfills would suggest that  $H_f$  is likely to range from 0.5 - 10 m, with values of 1-5 m being most probable.

If the clayey liner (which may be natural or manmade) is underlain at some finite depth  $H$  by a far more permeable stratum with water flow in the horizontal direction with a superficial (discharge, Darcy) velocity  $v_b$ , then solute will be transported away from the landfill at a rate dependent on the velocity, porosity and geometric dimensions of this layer. The concentration in this permeable layer will tend to zero as the base velocity tends to infinity, however in many cases the velocity will be relatively small and there will be a change in concentration with time in this stratum. An efficient technique for solving equation 1 for these boundary conditions has been described by Rowe and Booker (1984a,b).

The strategy of this solution consists of taking the Laplace Transform of the governing equation and boundary conditions, finding an analytical solution in transformed space and then inverting the transform numerically using an algorithm developed by Talbot (1979). This solution has been implemented in the computer program POLLUTE (Rowe and Booker, 1983) which solves the one-dimensional dispersion-advection equation for a layered deposit of finite depth. The program has the following capabilities and features:

- the deposit may be subdivided into individual layers where each layer may have different parameters (e.g.,  $D$ ,  $v$ ,  $n$ ,  $\rho$  and  $K$ );
- the effect of geochemical reactions on a non-conservative ion may be considered (i.e., sorption or desorption of the contaminant from the clay surface);
- provision is made for depletion of the contaminant in the landfill with time (i.e., the concentration of contaminant in the landfill can be automatically reduced as contaminant is transported into the soil. The case where the landfill concentration remains constant represents a limiting situation);
- the deposit may be underlain by either a relatively permeable or an impermeable base strata (e.g., a thin sand layer which is permeable relative to the clayey deposit or intact sound rock which is impermeable relative to the clayey deposit);
- provision is made for horizontal flow within any permeable stratum beneath the less permeable barrier;
- the maximum concentration of contaminant in a permeable base stratum can be automatically determined;
- the program requires minimal data preparation and generally provides accurate results for relatively little computational effort.

Unlike finite element and finite difference formulations, POLLUTE does not involve a "time marching" procedure. Thus, the concentration of contaminant can be directly determined at any specified time without calculating the concentration of earlier times.

#### BACKFIGURING A DISPERSION COEFFICIENT FROM MODEL TESTS

One of the most commonly reported techniques for estimating the dispersion coefficient for any particular contaminant consists of the use of laboratory models in which the contaminant migrates through the soil of interest under controlled conditions. Contaminant concentration profiles in the soil are measured and a dispersion coefficient is obtained by using various values of  $D$  and fitting the theoretical profile to the observed results. This approach provides the most practical solution to determining a value of  $D$ , particularly when an analysis is available (e.g. POLLUTE) which allows accurate modelling of the boundary conditions in the experiment. To illustrate the suitability of POLLUTE for modelling even quite complicated boundary conditions, the program was used to re-analyze the laboratory model test results performed at The University of Western Ontario (Crooks and Quigley, 1984).

Crooks and Quigley reported the results of two model tests involving the migration of sodium chloride ( $\text{NaCl}$ ) through a silty clay. The clay was compacted in the models to a thickness of 15 cm at a moisture content of 21%. The compacted unit weight of the soil was  $19 \text{ kN/m}^3$  at a porosity of 0.38. The soil was maintained at the laboratory ambient temperature (19-23°C). In model I, a head of 3.7 m at the top of the soil column gave rise to a superficial velocity of 0.022 m/a. Model II was similar except that the head of 1.0 m gives a superficial velocity of 0.0056 m/a. An important detail in the model design was the 0.6 cm thick porous (porosity 0.43) polyethylene plate at the bottom of the column which served to collect fluid exiting from the soil and allowed this fluid to be collected from an outlet pipe (see Fig. 1). Soil was taken from the models at different depths and times (see Crooks and Quigley, 1984 for details) and was analyzed to determine the concentrations of  $\text{Na}^+$  and  $\text{Cl}^-$  in the pore fluid.

Typical results for Chloride and Sodium migration through the silty clay in Model I are presented in Figs. 1 and 2. A preliminary analysis of this data using the analytical solution of Ogata (1970) for a halfspace with a uniform surface concentration was reported by Crooks and Quigley (1984) and gave values for the diffusion coefficient  $D$  of  $0.031 \text{ m}^2/\text{a}$  for Chloride and  $0.011 \text{ m}^2/\text{a}$  for Sodium. It was recognized that the Ogata solution did not accurately simulate the actual test conditions and considerable judgement was required to obtain reasonable values of  $D$ . Consequently, the test data was re-analyzed using the far more general solution to 1-D migration problems (Rowe and Booker, 1984) together with some additional data concerning the actual test conditions which could be used in conjunction with program POLLUTE.

POLLUTE permitted the modelling of the 0.6 cm thick polyethylene plate as a permeable base stratum ( $n_b = 0.43$ ) with an outflow velocity dictated by flow continuity requirements. It also permitted a consideration of changes in leachate concentration in the source reservoirs due to

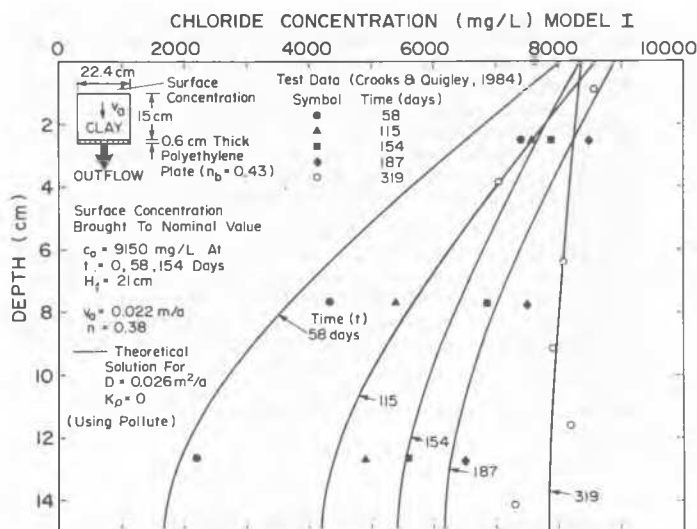


Fig. 1 Backfiguring the Dispersion Coefficient From the Results of a Laboratory Model Test

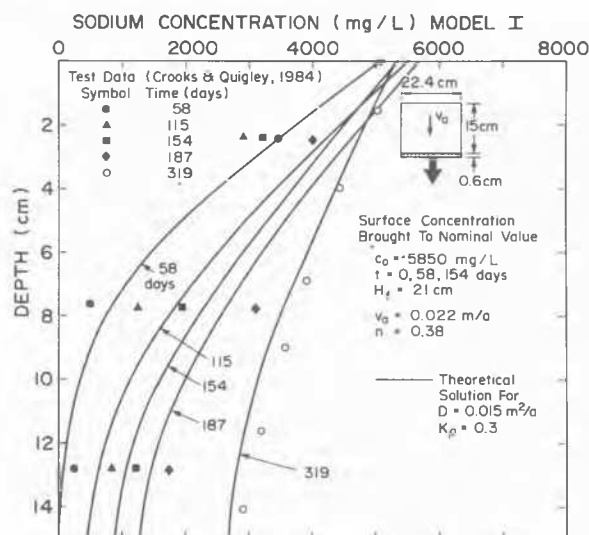


Fig. 2 Backfiguring D and  $K_p$  From the Results of a Laboratory Model Test

salt migration into the clay. It should be noted that maintaining a constant surface concentration (as required if the data is analyzed using Ogata's solution) involves practical difficulties at the model scale when the volume of leachate is relatively small. These difficulties arise from the fact that significant mass transport into the soil will occur and consequently the source concentration will drop. Maintaining a uniform concentration would require frequent replacement of the leachate however this results in undesirable fluctuations in the flow through the soil. In practice, a compromise must be met between allowing some drop in concentration and avoiding too frequent replacement of leachate.

In the model tests under discussion, the initial Sodium and Chloride concentrations were 5850 mg/L and 9150 mg/L respectively (Stage 1). After 58 days, the concentrations of Sodium and Chloride in the reservoir were measured to be 15% ( $Na^+$ ) and 17% ( $Cl^-$ ) lower than the initial leachate values (presumably due to significant mass transport into the clay). At this point, the model reservoir was drained and refilled with leachate at the initial concentration (Stage 2). This procedure was repeated again at 154 days after the start of the test (Stage 3).

The three stage sequence described above was directly simulated using the program POLLUTE. The dispersion coefficient  $D$  (and in the case of Sodium, the dimensionless sorption parameter  $K_p$ ) were adjusted to provide the "best fit" to the experimental data. The parameters giving the "best fit" are summarized in Table I and the theoretical concentration profiles are compared with the observed data for model I in Figs. 1 and 2 for Chloride and Sodium respectively. The agreement between the observed and predicted profiles is quite good. It should be noted that the Ogata solution will not provide a good fit to the data over the entire depth for any of these times and, furthermore, any value of  $D$  which gives a fair fit at any time will give a poor fit at the other times. This is simply because the

TABLE I

Parameters From Model Tests  
( $T \approx 20^\circ C$ )

Species	Model Test	$v_a$ (m/a)	$D$ (m <sup>2</sup> /a)	$K_p$	$D$ ( $7^\circ C$ ) * (m <sup>2</sup> /a)
$Cl^-$	I	0.022	0.026	0.0	.019
	II	0.0056	0.018	0.0	.013
$Na^+$	I	0.022	0.015	0.3	.011
	II	0.0056	0.013	0.3	.0095

\* Corrected for estimated field temperature (i.e.  $D_{7^\circ C} = 0.73D_{20^\circ C}$ ; see Crooks and Quigley, 1984).

boundary conditions in the model test are too far removed from those assumed in developing the Ogata solution.

These results (and comparable agreement obtained for model II tests) serve to demonstrate that the proposed theory, and the program POLLUTE, can adequately model dispersive (diffusive)-advective transport in clay subject to quite complex boundary conditions.

USE OF AN EFFECTIVE DIFFUSION COEFFICIENT

Equation 1 may be rewritten as

$$\frac{\partial c}{\partial t} = D^* \frac{\partial^2 c}{\partial z^2} - v^* \frac{\partial c}{\partial z} \quad (2)$$

where  $D^* = \frac{D}{1 + \frac{K_p}{n}}$  is an effective diffusion coefficient; and

$$v^* = \frac{v}{1 + \frac{K\rho}{n}}$$

is an effective velocity.

This representation of the diffusion/dispersion-advective process in terms of only two effective parameters ( $D^*$ ,  $v^*$ ) may be quite useful for situations where the advective velocity is small and the boundary concentrations are maintained at specified values. However, even in the laboratory, it is not always easy to maintain these boundary concentrations while in the field many situations will arise where the concentration in the leachate and any underlying aquifer will vary with time as mass is transported into or out of the clay liner. These cases where the boundary concentrations are flux controlled can be modelled using the theory proposed by Rowe and Booker (1984) and program POLLUTE however in doing so care is needed to avoid potential errors which can arise from the inappropriate use of an effective diffusion coefficient and effective velocity.

To illustrate the effect of the boundary conditions on the results obtained using actual and effective parameters, consideration was given to a hypothetical "laboratory test" as indicated in Fig. 3. Supposing that the soil has  $D = 0.01 \text{ m}^2/\text{a}$ ,  $K\rho = 10$ ,  $n = 0.4$  and  $v_a = 0.02 \text{ m/a}$ , an analysis was performed assuming a height of leachate  $H_f$  of 15 cm. The results shown in Fig. 3 indicate that there is a substantial difference between the results obtained using the actual and the corresponding "effective" parameters. This difference arises because the use of only the effective parameters gives incorrect fluxes at the boundaries. (The same conclusion would be reached even if  $v_a$  were zero.)

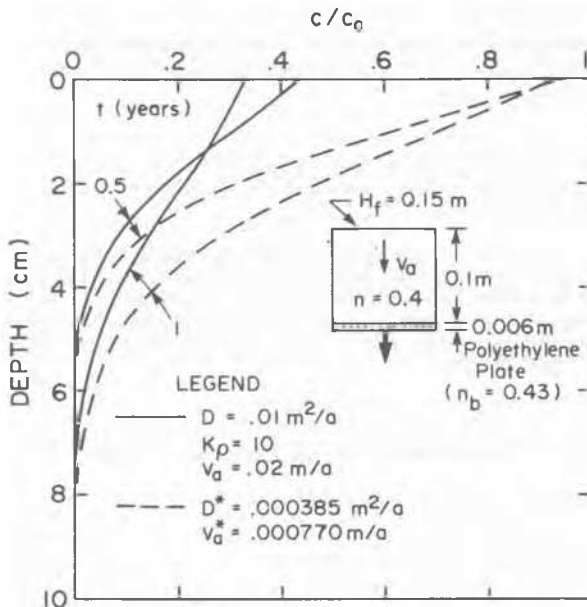


Fig. 3 Effect of Using "Effective" Parameters  $D^*$ ,  $v^*$  Upon Predicted Concentration Profiles in a Hypothetical Model Test

To illustrate the practical implications of the use of "effective" parameters  $D^*$  and  $v^*$ , consider a 200 m long landfill resting on a 2 m thick clay barrier which in turn is underlain by a 1 m thick sand layer. Assuming that the advective velocity in the clay and sand are 0.02 m/a and 5 m/a respectively, Fig. 4 shows the variation in the base concentrations with time predicted using "actual" parameters  $D = 0.01 \text{ m}^2/\text{a}$ ,  $n = 0.4$ ,  $K\rho = 10$  and the corresponding effective parameters  $D^*$  and  $v^*$ . It is apparent from Fig. 4 that the analysis performed using only the effective parameters  $D^*$  and  $v^*$  is both incorrect and unconservative. Because of potential problems inherent in the use of effective parameters, it is recommended that the use of these parameters be avoided in any analysis where the boundary conditions are flux controlled.

SUGGESTED PROCEDURE FOR DETERMINATION OF PARAMETERS FROM LABORATORY MODEL TESTS

As previously noted, there are some practical difficulties associated with maintaining a constant concentration of contaminant within a source fluid in model tests. This difficulty can be avoided by adopting a simple test where the source concentration is initially specified and then allowed to drop due to mass transport into the soil. This situation is readily analyzed using POLLUTE and in fact the marked different effects of  $D$  and  $K\rho$  which are apparent under these circumstances can be used to permit the determination of both  $D$  and  $K\rho$  from the one test. For example, Fig. 5 shows a single column which consists of 0.1 m of source fluid (leach-

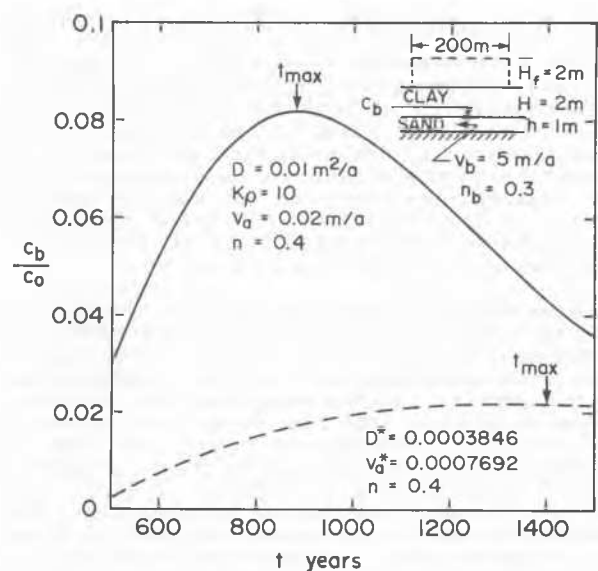


Fig. 4 Effect of Using "Effective" Parameters  $D^*$ ,  $v^*$  Upon Base Concentration  $c_b$

ate), overlying 0.1 m of clay overlying 0.006 m of polyethylene porous plate. At the start of the test, the species of interest in the leachate has a concentration  $c_0$ . Diffusion/advection is then allowed to reduce the concentration within the leachate (and this reduction with time should be monitored). The effluent exiting from the base plate should also be collected and the concentration of species in this effluent monitored. The test should be terminated at some time  $t$  (the actual time is not critical provided that there has been time for significant diffusion into the clay sample) and the concentration profile in the soil determined. Knowing the advective velocity  $v_a$ , the initial concentration  $c_0$ , the height of leachate  $H_f$  and the time  $t$ , theoretical profiles can be determined for different values of  $D$  and  $K\rho$ . The values of  $D$  and  $K\rho$  are adjusted until a reasonable fit of the experimental profile is obtained.\* The validity of these best fit parameters can then be checked by comparing the predicted and observed variation in concentration within the leachate and effluent. To illustrate the sensitivity of the procedure, Fig. 5 shows the concentration profiles obtained at three times for  $D = 0.01 \text{ m}^2/\text{a}$  and  $K\rho = 0$  and  $0.25$  and it can be seen that even a relatively low level of sorption ( $K\rho = 0.25$ ) can have an appreciable effect on the concentration profile and hence can be readily detected. The results shown in Fig. 5 were obtained for a relatively small height of leachate  $H_f$  (0.1 m) and advection velocity (0.003 m/a). Even for much larger values of  $H_f$  and  $v_a$ , the effect of small values of  $K\rho$  is readily apparent and consequently both  $D$  and  $K\rho$  may be deduced.

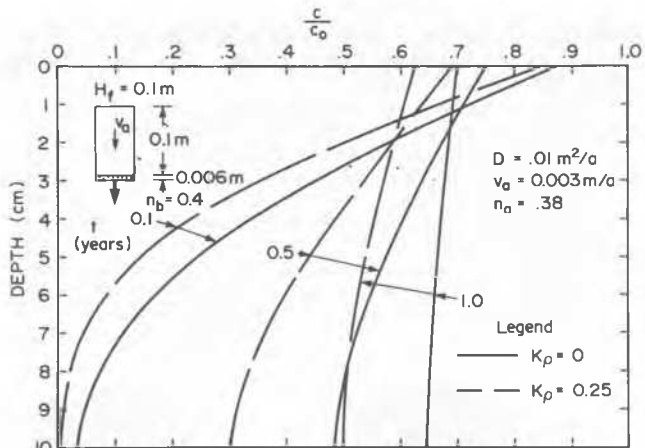


Fig. 5 Effect of  $K\rho$  on a Concentration Profile for  $H_f = 0.1 \text{ m}$

\* An initial estimate of  $D$  can be obtained by first determining the value of  $D$  for a non-reactive reference species (e.g. Chloride) and then multiplying this value by the ratio of the diffusion coefficient of the species of interest in pure water to the diffusion coefficient of the reference species in pure water.

APPLICATION TO FIELD SITUATIONS

The concentration profile beneath the Confederation Road landfill near Sarnia, Ontario has been extensively studied in recent years (Goodall and Quigley, 1977; Crooks and Quigley, 1984). Fig. 6 shows the observed Chloride profile at two locations beneath the Confederation Road landfill 12 years after completion. The concentration of Chloride in the leachate at these two locations is 1700 mg/L and 3000 mg/L at Boreholes 101 and 102 respectively. An inspection of the measured concentrations in the silty clay beneath the landfill indicates a substantial drop in concentration compared to the leachate values over a very small distance. These results would suggest that something special is happening at the interface between leachate and the general mass of clay (similar results were obtained for the Sodium ions). There are several ways of explaining this behaviour (e.g. see Crooks and Quigley, 1984; Quigley et al., 1984) and the various mechanisms will be discussed in detail in a forthcoming publication. In this paper, we wish to show that the large drop in concentration from the leachate values to those measured a short distance below the refuse-clay interface can be modelled by postulating a thin interface layer of thickness  $H_I$  with an apparent dispersion coefficient  $D_I$ . Analyses were performed varying the ratio  $D_I/H_I$  and the dispersion coefficient in the clay to obtain a fit with the data at BH102. The best fit was obtained for a value of  $D_I/H_I = 0.02 \text{ m/a}$  and  $D = 0.018 \text{ m}^2/\text{a}$ . The dispersion coefficient in the clay is consistent with that obtained from model tests on the same clay once an adjustment is made for the difference in temperature in the laboratory and field (see Table I).

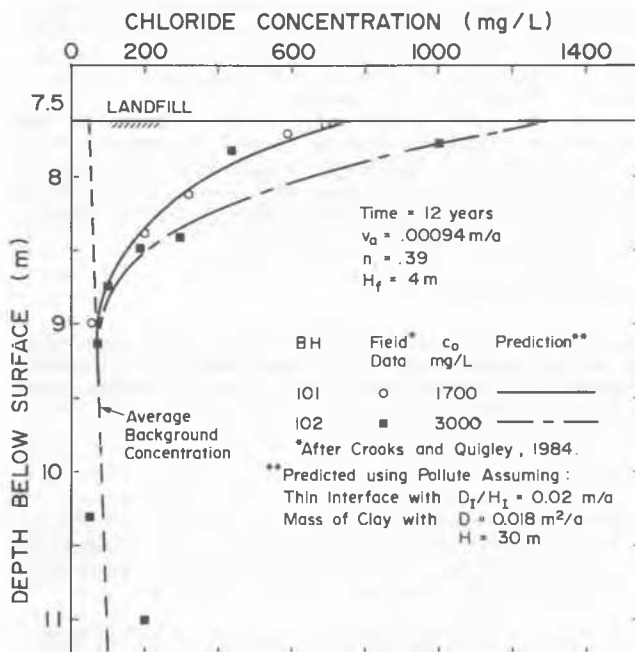


Fig. 6 Measured Porewater Chloride Concentration and Calculated Profiles - Sarnia Landfill

These parameters were then used to predict the concentration profile at BH101 and the resulting predictions shown in Fig. 6 are in very good agreement with the observed concentration profile. The consistency of the results at two locations suggests that some form of interface effect is occurring at the landfill which was not evident in the model tests on the same clay (see Figs. 1 and 2\*). The mechanisms responsible for this interface are currently under investigation at The University of Western Ontario and will be discussed in detail in a subsequent publication.

In the design of landfills, it would be conservative to neglect the effects of any interface layer which may be formed. However, the possible presence of such a layer should be considered when attempting to backfigure parameters from laboratory tests. The "leachate" in laboratory tests (by Crooks and Quigley) described earlier, did not contain any metals suitable for forming precipitates at the interface or bacteria which could multiply and cause clogging, however it is possible that a true leachate which is chemically very complex and possibly bacterially active may well result in an interface effect. If present, this interface effect should be modelled when backfiguring  $D$  and  $K_p$  from the model tests and only the values determined away from the interface should be used in design of landfills. Failure to allow for possible interface effects in model tests could give rise to an artificially low dispersion coefficient and/or an artificially high value of sorption coefficient  $K_p$ .

#### CONCLUSION

A new and simple method of calculating concentration profiles in laboratory column tests and in field applications has been shown to provide good agreement with observed behaviour. This technique has then been used to demonstrate the potential dangers which may arise from the use of an effective diffusion (dispersion) coefficient. Finally, a procedure for determining parameters from laboratory column tests has been proposed. This procedure readily permits the determination of both the dispersion (diffusion) coefficient and the sorption potential (in terms of the distribution coefficient  $K$ ) for a given species of contaminant.

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#### REFERENCES

Crooks, V.E. and Quigley, R.M. (1984). Saline Leachate Migration Through Clay: A Comparative Laboratory and Field Investigation. Canadian Geotechnical Journal, 21(2), 349-362.

\* As previously noted, the modest drop in concentration in the model tests is the result of a drop in concentration within the leachate. This mechanism cannot explain the large concentration drop observed in the field.

- Gillham, R.W. and Cherry, J.A. (1982). Predictability of Solute Transport in Diffusion-Controlled Hydrogeologic Regimes. Proc. Symp. on Low Level Waste Disposal: Facility Design, Construction and Operating Practices, Nuclear Regulatory Commission, Washington, D.C.
- Goodall, D.E. and Quigley, R.M. (1977). Pollutant Migration From Two Sanitary Landfill Sites Near Sarnia, Ontario. Canadian Geotechnical Journal, 14, 223-236.
- Hajek, B.F. and Ames, L.L. (1968). Trace Strontium and Cesium Equilibrium Distribution Coefficients: Batch and Column Determinations. Battelle Pacific Northwest Laboratories, Richland WA.
- Ogata, A. (1970). Theory of Dispersion in a Granular Medium. U.S. Geological Survey Professional Paper 411-1.
- Quigley, R.M., Crooks, V.E. and Yanful, E. (1984). Contaminant Migration Through Clay Below a Domestic Waste Landfill Site, Sarnia, Ontario, Canada. Int. Symp. on Groundwater Resources, Utilization and Contaminant Hydrogeology, Montreal, May.
- Rowe, R.K. and Booker, J.R. (1983). Program POLLUTE - 1D Pollutant Migration Analysis Program. SACDA, Faculty of Engineering Science, The University of Western Ontario, London, Ont.
- Rowe, R.K. and Booker, J.R. (1984a). 1-D Pollutant Migration in Soils of Finite Depth. Journal of Geotechnical Engineering, ASCE (in press).
- Rowe, R.K. and Booker, J.R. (1984b). The Analysis of Pollutant Migration in a Non-Homogeneous Soil. Geotechnique (in press).
- Talbot, A. (1979). The Accurate Numerical Integration of Laplace Transforms. J. Inst. Maths. Applics. 23, 97-120.