

The 14-year performance of a compacted clay liner used as part of a composite liner system for a leachate lagoon

CRAIG B. LAKE^{1,*} and R. KERRY ROWE²

¹*Department of Civil Engineering, Dalhousie University, Halifax, Nova Scotia, Canada B3J 1Z1*

²*GeoEngineering Centre at Queen's-RMC, Department of Civil Engineering, Queen's University, Kingston, Ontario K7L 3N6*

(Received 9 January 2004; revised 9 January 2004; accepted 26 April 2004)

Abstract. The migration of contaminants through a 2.9 m thick compacted clay liner (CCL) for a landfill leachate lagoon is examined 14 years after construction. The clay liner formed the lower portion of the composite liner system but the geomembrane (GM) was found to have defects that had allowed leachate to migrate between the GM and CCL. Chloride, sodium, potassium, calcium and magnesium pore water profiles through the CCL are examined. It is shown that chloride migrated approximately 1.7 m into the CCL during the 14 years of the lagoon operation, sodium approximately 1.2 m, and potassium 0.7 m. Diffusion and sorption data from laboratory diffusion testing are utilized in combination with a finite layer contaminant transport model to predict field contaminant migration profiles through the composite liner system and to establish the time of 'failure' of the geomembrane at sometime between 0 and 6 years after installation. Relatively high sorptive uptake of potassium by the CCL soil is observed from batch testing and diffusion testing with field data suggesting an even larger amount of sorption. It is hypothesized that organic sludge matter at the base of the lagoon is responsible for potassium uptake from the leachate. This field case highlights the importance of the compacted clay liner as part of the composite liner system in acting as a diffusion barrier during the lifetime of the lagoon as well as using relatively non-conservative contaminants such as chloride and sodium to estimate geomembrane 'failure' times.

Key words. compacted clay, composite liner, contaminant transport, diffusion, lagoon, leachate.

1. Introduction

The migration of contaminants from solid waste landfills through both natural and compacted clay has been examined through several well documented case histories (Goodall and Quigley, 1977; Barone et al., 1991; King et al., 1993). In most cases, these low hydraulic conductivity natural clay deposits or compacted clay liners minimize the advective flux of contaminants through the clay resulting in diffusion becoming the dominant contaminant transport mechanism (for an exception, see Munro et al., 1997). Diffusion, the movement of contaminants from areas of high

*Corresponding author: Department of Civil Engineering (Dalhousie University, Halifax, Nova Scotia, Canada B3J 1Z1 (e-mail: craig.lake@dal.ca)

concentration to areas of low concentration, has been researched extensively for low hydraulic conductivity barriers such as compacted clay liners (CCLs) (Rowe et al., 1988, 2004), geosynthetic clay liners (GCLs) (Lake and Rowe, 2000), and geomembranes (Park and Nibras, 1993; Rowe et al., 1995, 2004). In particular, it is known that intact HDPE geomembranes provide an excellent diffusive barrier to ions (e.g. Cl^- , Na^+ , and K^+) and negligible diffusion of these contaminants would be expected through an intact geomembrane over a period of 14 years (Rowe, 1998; Rowe et al., 2004).

Modern landfills and lagoons widely utilize composite liners (e.g. a geomembrane over compacted clay) but unfortunately there is a paucity of published field investigations that have examined contaminant transport through the complete geosynthetic composite liner system. The understanding of factors associated with design for contaminant transport mitigation, selection of materials and long term performance of composite liners has improved considerably over the last two decades. The importance of some of these factors can be best illustrated with reference to case records. The objective of this paper is to examine the 14 year performance of the compacted clay liner portion of the composite liner for a leachate lagoon and provide information related to contaminant transport of several ionic constituents found in the leachate through this liner system. In particular, the retardation of K^+ during diffusive and advective migration through the CCL is examined based on field observations and a laboratory study. Possible mechanisms of the high amounts of sorption observed in the field are also discussed in the paper. This paper complements Rowe et al. (1998, 2003) and which focused on the physical characteristics of the geomembrane portion of the composite liner system discussed herein as well as chloride migration.

2. Background

2.1. LANDFILL

The lagoon under study was situated in a landfill located in Ontario, Canada which encompassed 28 hectares at the time of this study. Non-hazardous industrial, municipal, and commercial wastes were being placed in this multi-cell landfill that is lined with a compacted clay liner and leachate collection system (Gartner Lee Ltd., unpublished report, 1995). Below the landfill lies the Rochester Shale Formation, consisting of 'thin beds of aphanitic shales and shaley dolostones' (Gartner Lee Ltd., unpublished report, 1995). Horizontally bedded dolostone of the Lockport Formation surround the landfill above the Rochester Shale. Leachate generated within the landfill is collected by a leachate collection system and transferred to the aerated leachate lagoon studied in this paper (constructed with an unprotected geomembrane over a compacted clay liner). The original location of the lagoon was temporary and served the landfill for 14 years at which time a new lagoon was constructed and the original lagoon decommissioned.

Table 1 shows 1995 leachate results for selected constituents of the leachate monitoring program sampled from a manhole directly before the leachate passed into

Table 1. Landfill Leachate Characteristics (1994/1995 Data: Gartner Lee Ltd., unpublished report, (1995))

Parameter	Ontario drinking water objective (mg/L)	Arithmetic mean (mg/L)
Lab pH	6.5–8.5	7.9
Conductivity		7985 us/cm
TDS	500	5153
Alkalinity	30–500	2978
Hardness	100	1074
COD		505
BOD		90
<i>Major Ions</i>		
Cl ⁻	250	472
Na ⁺	200	1528
K ⁺		143
Ca ²⁺		154
Mg ²⁺		167
NH ₃ -N		148
<i>Metals</i>		
Al	0.1	0.47
Fe	0.3	2.84
Pb	0.01	0.03
Mn	0.05	0.30

the aerated leachate lagoons. Precipitation events, the addition of new landfill cells and some changes in proportions of waste types has caused variations in leachate concentrations with time as shown for chloride, sodium, and potassium in Figure 1.

Lysimeters form part of the landfill monitoring program and have been installed under the clay liner of the lagoon and landfill in an attempt to monitor leachate constituents migrating through the liners. At the time of this study, lysimeter water underneath the lagoon was a dilute mixture of sodium (31 mg/L), potassium (28 mg/L), and chloride (25 mg/L) with concentrations of calcium (573 mg/L) and magnesium (308 mg/L) elevated relative to that in the leachate (Gartner Lee Ltd., unpublished report, 1995). None of the leachate indicator parameters were detected in the lysimeter at the time of this study.

2.2. LAGOON

Lagoon storage capacity was approximately 2500 m³ (side slopes of 3:1) and were lined with a 1.5 mm high density polyethylene (HDPE) geomembrane over a compacted clay liner (2.9 m thick) situated above the quarry's shale floor (Figure 2). Leachate heights in the lagoon averaged 3 m above the bottom of the lagoon. A leveling surface of broken shale and trap rock from quarrying operations was placed on the highly weathered quarry floor prior to construction of the compacted clay liner of the lagoon. The water table in the shale was estimated at approximately 3 m below the bottom of the compacted clay. The bottom of the quarry floor was exposed at the base of the west slope of the lagoon.

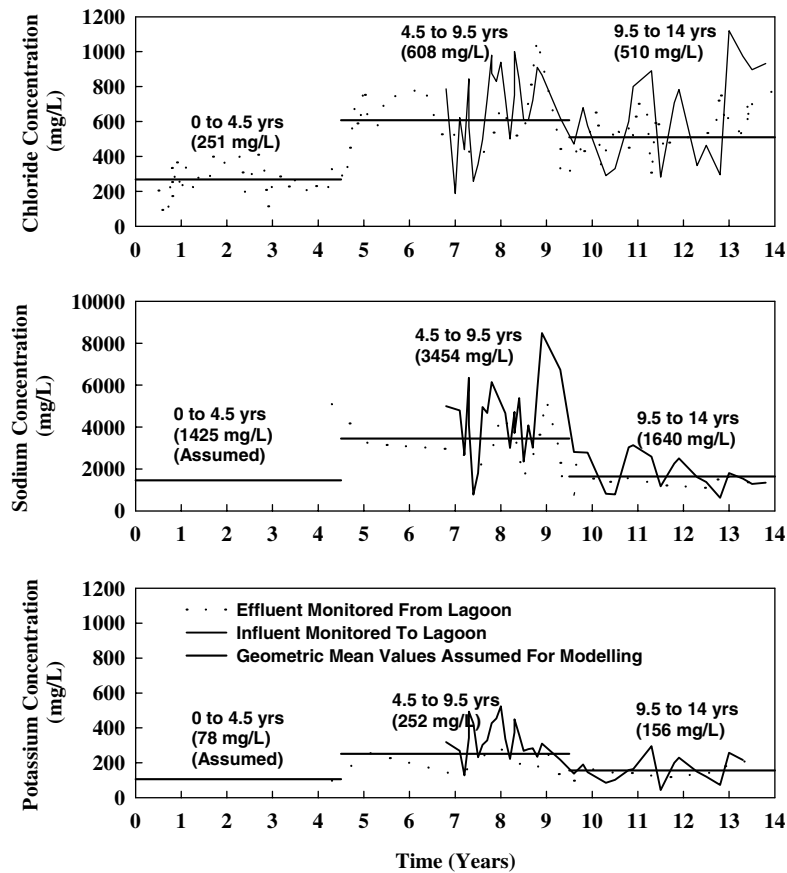


Figure 1. Temporal leachate lagoon Cl, Na, and K concentrations and corresponding geometric means for time periods 0 to 4.5 year, 4.5 to 9.5 year, and 9.5 to 14 year.

According to Chapman and Putnam (1973) varying amounts of lacustrine clays and glacial tills formed during the Pleistocene glaciation period are present in this area (Quigley, 1971). Construction records (Proctor and Redfern, unpublished report, 1982) show the compacted clay liner was constructed from 'silty clay' borrow material present near the landfill. Table 2 summarizes mineralogical and chemical data obtained from a sample of the compacted clay liner approximately 1.8 m below the top of the compacted clay below the lagoon. Illite and chlorite were found to be the predominant clay minerals with some vermiculite also present in the sample tested. This is similar to the mineralogy reported by Quigley (1971) for a clay in the area.

3. Field Exhumation of Composite Liner System

To assess the performance of the geomembrane/compacted clay composite liner system for the lagoon at the landfill, the lagoon was drained of leachate and

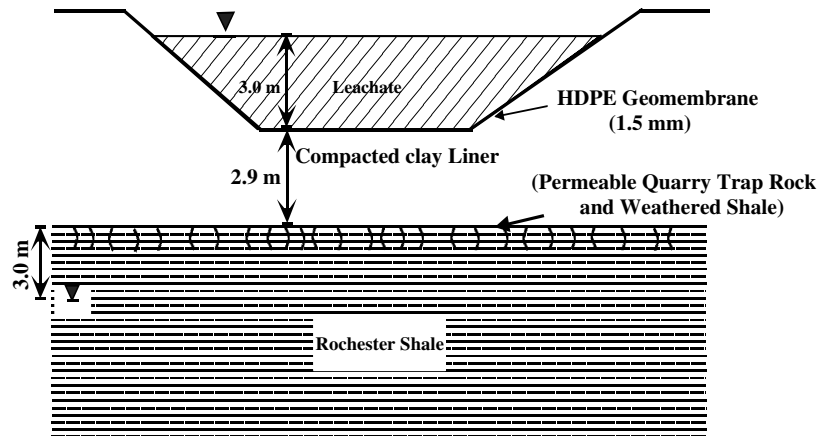


Figure 2. Schematic cross-section of leachate lagoon.

subsequently decommissioned. At first appearance, the bottom of the lagoon looked like a 'waterbed' with a significant amount of liquid trapped between the geomembrane and the compacted clay. A similar observation had been made by landfill staff during previous maintenance operations of the lagoons. According to the operators of the landfill, the lagoon had a history of problems and had to be periodically drained and defects present on the geomembrane patched and repaired during their lifespan. The geomembrane portion of the composite liner was visually examined as described by Rowe et al. (2003). In summary, investigation of the geomembrane revealed 82 cracks, holes, and patches over a total area of 1350 m² (600 defects per hectare for the 14 year period of operation of the lagoon). Even though the majority of the holes, cracks and patches were above the leachate levels in the lagoon, the defects below the leachate level appear to have been sufficient to allow leachate to

Table 2. Mineralogical and chemical data for compacted clay* (modified from Rowe et al. 2003)

% Sand	Trace
% Silt	63%
% Clay	37%
Quartz	~ 36%
K Feldspar	~ 4%
Na Feldspar	~ 5%
Calcite	~ 18%
Dolomite	~ 4%
Illite	~ 20%
Chlorite	~ 10%
Vermiculite	~ 3%
Liquid limit	35%
Plasticity index	12%
Activity	0.3
Total organic carbon	0.2%
pH	8.1
CEC	12.0 meq/100 g

*Results of one sample (1.8 m below top of CCL, BH 4).

migrate between the geomembrane and clay under low effective stress conditions. This is the probable explanation of the observed geomembrane 'waterbed' effect at the bottom of the lagoon.

After the completion of geomembrane inspection and sampling, portions of the compacted clay liner underneath the geomembrane were examined. The clay liner above the leachate level in the lagoon was noticeably desiccated. Desiccation of clay liners underneath geomembranes has also been observed by others (Corser et al., 1992; Basnett and Brungard, 1992). Below the leachate level of the lagoon, the clay appeared saturated with a thin layer of black sludge (~25 mm) found in the top portion of the clay under the geomembrane on both the base and side slope. In some areas a 20–80 mm thick mixture of sludge and particulate matter was observed at the base of the lagoon, under the geomembrane.

Continuous, 75 mm diameter soil samples were taken of the compacted clay liner at the base of the lagoon at five locations with sampling depths ranging from 1.1–2.1 m. The soil was visually classified as a light brown clay of low plasticity during sampling with no indication of fractures present. Samples obtained from each borehole were transferred to PVC casings that were subsequently capped, taped and placed into a styrofoam box packed with ice. Samples were transferred to the geotechnical laboratory at The University of Western Ontario, taken out of the PVC casings, sectioned into various lengths (ranging from approximately 2 to 18 cm), coated with wax and stored in an environmentally controlled room at 5 °C ambient temperature. Soil samples were taken during sectioning for mass determination of water content. Soil samples were stored for geotechnical index tests, flexible wall hydraulic conductivity tests, diffusion tests, batch tests, and chemical mineralogical testing. Pore-water samples for each borehole (at various depths) were obtained using a pneumatic pore squeeze apparatus by applying a 25 MPa pressure to selected soil samples. Pore-water obtained from the samples was analyzed for sodium, potassium, magnesium, and calcium using an atomic absorption spectrometer and chloride using a specific ion electrode.

4. Laboratory Investigation

4.1. BATCH AND DIFFUSION TESTS: COMPACTED CLAY

To obtain the diffusion and sorption coefficients required for contaminant transport modelling of the composite liner system, batch and diffusion tests were conducted as described below.

Two batch tests were performed with a sample of the compacted clay liner soil (Borehole 1, 1.8 m below lagoon); one with the municipal solid waste leachate (MSWL) shown in Table 3 and the second with potassium chloride (KCl) solutions (maximum K^+ concentration of 250 mg/L). Batch testing involved mixing the leachate at various dilutions (total volume 100 mL) or KCl solutions with 10 g of water wet soil. Details of all these testing procedures can be found in Lake (2000).

Table 3. Chemical characteristics of the municipal solid waste leachate used for diffusion testing (sampled from landfill lagoon)

Cl ⁻ (mg/L)	1500
SO ₄ ²⁻ (mg/L)	589
Na ⁺ (mg/L)	1503
K ⁺ (mg/L)	208
Ca ²⁺ (mg/L)	103
Mg ²⁺ (mg/L)	205
pH	7.2
Temperature at sampling (°C)	18 (8°C air temperature)

Where practical, batch test results (plotted as mass of cation sorbed per unit mass of dry soil vs. the equilibrium solution concentration) were fitted by a linear partitioning coefficient, K_d . However, for potassium, there was some non-linearity and the data was also fitted by a Langmuir isotherm (e.g. see Yong et al., 1992 or Rowe et al., 2004):

$$S = \frac{S_m b \cdot c}{1 + b \cdot c}, \quad (1)$$

where S = mass of concentration removed from solution per mass of dry soil [-]; c = equilibrium concentration of solute in the soil pore fluid [ML⁻³]; S_m = maximum solid phase concentration, obtained experimentally [-]; and b = an experimental parameter [L³M⁻¹].

A diffusion test was performed with the MSWL solution (Table 3) and an intact sample of the compacted clay using a procedure similar to that described by Rowe et al. (1988) and Barone et al. (1989). Diffusion coefficients for Cl⁻, Na⁺, and K⁺ were obtained from diffusion tests by modelling the experimental diffusion test results with the contaminant transport program POLLUTE (Rowe and Booker, 1999). At the end of the diffusion test, adsorbed cation profiles for Na⁺, K⁺, Ca²⁺ and Mg²⁺ were measured at sample depths of 0.5, 1.5, 3.5 and 5.5 cm. X-ray analyses were also performed for a sample from the leachate/soil interface (0.5 cm depth) and at the bottom of the sample (8.7 cm depth). The wet soil was pressure oriented (~160 MPa) on a round metal disk using a hydraulic jack to form a 'pellet' for X-ray analysis, (Yanful et al., 1990). X-ray analyses were performed with a Rigaku diffractometer, generating cobalt radiation from a rotating anode source (Department of Earth Sciences, The University of Western Ontario). The diffractometer was operated at 45 kV and 160 mA with a scan rate of 5°/min.

5. Results

5.1. FIELD COMPACTED CLAY LINER GEOTECHNICAL CHARACTERISTICS

At the time of the field investigation, liner moisture contents ranged from a maximum of 38% at the top of the compacted clay liner (i.e. just below the geomembrane) to approximately 20–24 % below a depth of 0.8 m. The liquid observed to be trapped between the geomembrane and compacted clay during the field

investigation, along with low effective stresses present in the lagoon are probably responsible for swelling of the clay (i.e. increased moisture content) at the surface of the compacted clay liner. Based on records of the original investigations of the clay borrow source and lagoon liner construction reports (Proctor and Redfern, unpublished report, 1982), the majority of the clay liner was compacted in the range of 1–4% above the standard proctor optimum moisture content that ranged from 15% to 21%. Hydrometer analyses of soil randomly selected throughout the liner (7 samples) obtained from the current investigation showed the proportion of clay size particles in the liner varied between approximately 30% to 50% which is consistent with records of the initial investigation into the borrow material (Proctor and Redfern, unpublished report, 1982). Three flexible wall hydraulic conductivity tests performed on the compacted clay yielded hydraulic conductivities of 1.8×10^{-10} , 2.2×10^{-10} , and 3.0×10^{-10} m/s, with a mean of 2.3×10^{-10} m/s.

5.1.1. K^+ and Na^+ Laboratory Batch Tests – KCl and MSWL

An initial series of batch tests was performed using a KCl solution and then a subsequent series (described later) was performed with municipal solid waste leachate. There were two reasons for starting with a single cation component solution. First, the KCl batch test provides an upper bound value for the sorption of K^+ to the compacted clay soil compared to the multi-component cation municipal solid waste leachate. If a portion of the sorption process is dependant on ion exchange processes then a single component KCl solution would give a higher K^+ exchange with the clay, without competition from other ions. Second, the KCl is a relatively stable ionic solution. Any mass retention of K^+ during batch tests is most likely due to sorption processes and not losses due to K^+ precipitation from solution. These KCl results were not used for modelling of diffusion testing but provide some insight regarding the sorptive capacity of the clay with regards to K^+ .

Figure 3 shows that K^+ exhibited a non-linear sorption isotherm with the CCL for the range of concentrations examined using the KCl solution. The pH of the KCl solution ranged from 8.0 to 8.4 after testing compared to the pH of the control bottle of 7.1. This higher pH in the soil suspensions was most likely due to alkalinity of the soil used for batch testing. This was also observed by Yanful (1984) for batch tests performed with a carbonate rich soil. It was decided not to adjust pH values closer to neutral values since such an increase in pH also occurs as the leachate enters the soil. This non-linear sorption data for potassium was fit to the Langmuir sorption model with the Langmuir parameters shown on Figure 3. For comparison, a linear partitioning coefficient K_d , of 7 mL/g is also shown on Figure 3 over the range of equilibrium concentrations.

To properly quantify sorption of K^+ to the compacted clay soil for contaminant transport modelling, it was deemed important to use a sample of leachate from the landfill. Figure 4 shows batch test results with the municipal solid waste leachate.

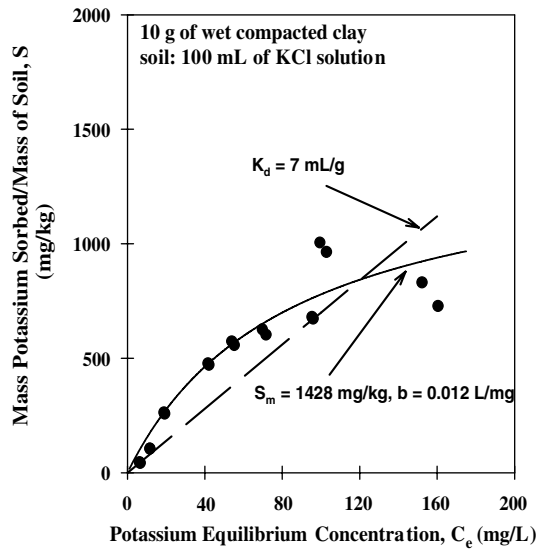


Figure 3. Batch test sorption isotherm of wet compacted clay and KCl solution.

Again, potassium (K^+) exhibited non-linear isotherm batch test results over the range of concentrations tested. The pH of soil/leachate solutions at the end of testing ranged from 8.0 to 8.5 compared to a leachate pH prior to testing of 7.4. Figure 4 shows the isotherm for K^+ batch test results as well as the Langmuir sorption parameters found from a regression analysis. A linear partitioning coefficient, K_d , of 3 mL/g is also shown for comparison. As the ionic concentration in the leachate increases, not only does the concentration of K^+ increase, but other cations in the

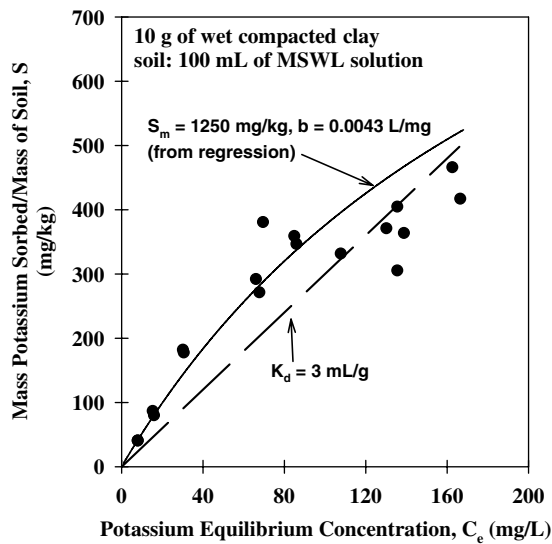


Figure 4. Batch test sorption isotherm of wet compacted clay and municipal solid waste leachate.

leachate as well, which increases the competition for exchange sites between K^+ and these other cations. Evidence supporting this hypothesis is found in a comparison of the KCL and MSWL batch test data. The sorptive capacity of the clay is higher for the KCL solution than that of the MSWL solution. As will be seen in the next section, the Langmuir model also provides a reasonable fit to the experimental data of the diffusion test.

Only slight sorption of Na^+ was observed over the range of concentrations examined and sorption was approximated by the linear partitioning coefficient, K_d of 0.2 mL/g.

5.2. DIFFUSION TEST ON LABORATORY SAMPLES

Table 4 summarizes the diffusion coefficients obtained from diffusion testing. For illustrative purposes, Figure 5 shows diffusion test results for potassium. Both Langmuir and linear sorption parameters were utilized for modelling diffusion test K^+ concentrations with very little difference being observed. This is most likely due to equilibrium concentrations in the diffusion test sample being in the range of the initial linear portion of the batch test results. As shown in Figure 5, K^+ concentrations decrease significantly in the leachate due to migration into the clay sample, resulting in an increase in pore fluid concentrations in the top 1 cm of the sample. This migration distance of K^+ was approximately 7 cm less than that of chloride for the test duration of 8 days (this attenuation was also observed in K^+ field porewater concentrations as will be discussed later in the paper).

The diffusion test results indicated elevated levels of K^+ adsorbed concentrations at the soil-leachate interface (Figure 6). Mass balance calculations for potassium indicated that only 63% of the original mass of potassium was accounted for based on the concentration in pore water and that which could be recovered by cation exchange for K^+ associated with the clay after the tests. This difference between K^+ response in sorption and desorption is considered to be due to K^+ fixation to vermiculite clay minerals present in the clay. K^+ associated with fixation is essentially unrecoverable, resulting in an apparent mass loss of K^+ . Support for K^+ fixation to the vermiculite clay minerals arises first from the change in CEC values for the top section of soil in the diffusion test which dropped from 17.6 meq/100 g to 16.4 meq/100 g. The 37% of K^+ that could not be recovered for this soil section can be explained by a decrease in CEC of 0.5 meq/100 g due to

Table 4. Parameters obtained from laboratory diffusion and batch test results

Parameter	Effective diffusion coefficient, D_e (m^2/s)	Sorption
Cl^-	7.0×10^{-10}	$K_d = 0$ mL/g
Na^+	4.0×10^{-10}	$K_d = 0.2$ mL/g
K^+	7.0×10^{-10}	<i>Langmuir coefficients</i> $S_m = 1250$ mg/kg $b = 0.0043$ L/mg

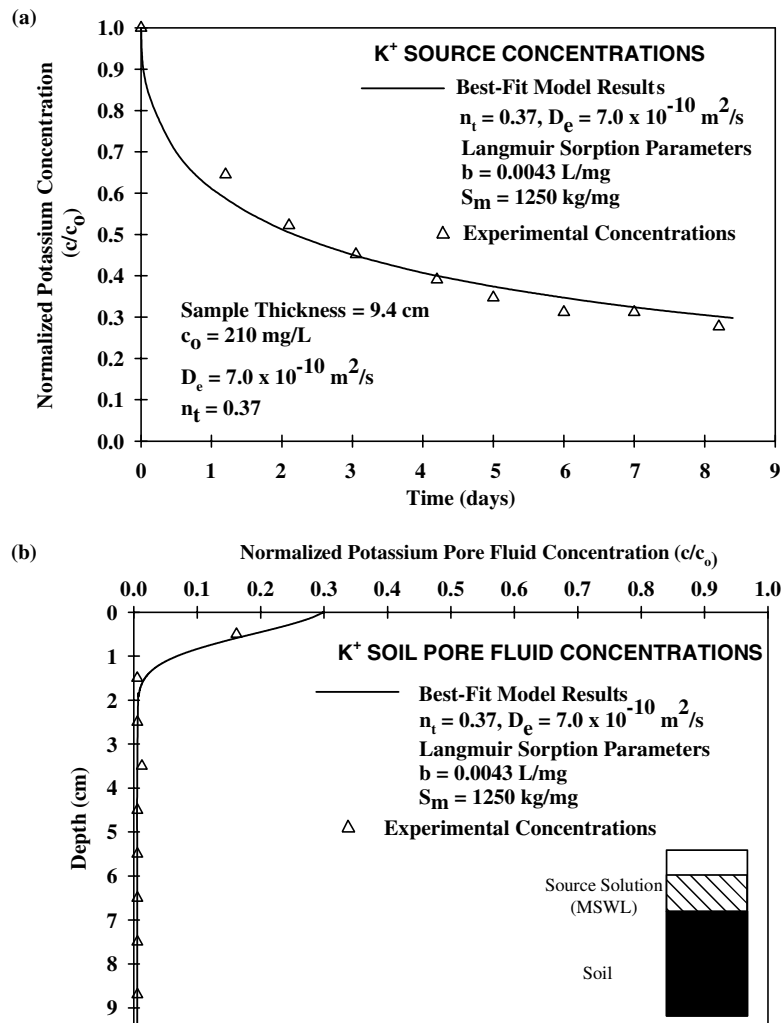


Figure 5. Observed and calculated potassium concentrations (a) in the source solution and (b) with depth at the end of diffusion testing.

K^+ fixation. This represents about a half of the change in CEC that was observed. Although it is acknowledged that experimental error also may be responsible for some of the difference in CEC, the remaining change in CEC is probably due to NH_4^+ fixation. X-ray analysis of the soil at the leachate interface (0.5 cm depth) showed the ratio of the 1.0 nm peak to the 1.4 nm peak is higher relative to the soil at the base of the sample (depth 8.7 cm). This is consistent with (a) the collapse of vermiculite minerals at a d -spacing of 1.4 nm to illite at a d -spacing of 1.0 nm as the result of K^+ fixation between the tetrahedral sheets of the vermiculite minerals (Rowe et al., 2004) and (b) the decrease in CEC due to K^+ satisfying some of charge deficiency in the collapsed vermiculite mineral.

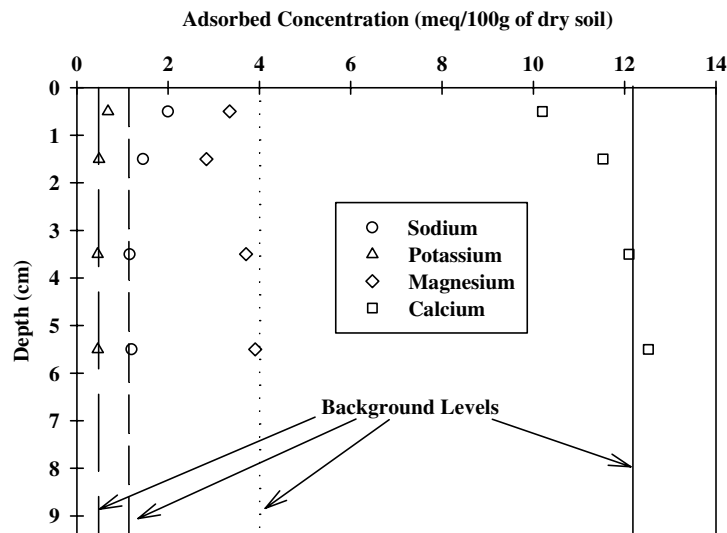


Figure 6. Adsorbed cation concentrations with depth at the end of diffusion testing.

Quigley et al. (1989) observed significant collapse of 'vermiculite/smectite' and 'interlayered illite/smectite' present in a sample of water-wet Leda clay after permeation with leachate. This was attributed to K^+ fixation of soil vermiculite to illite. It is also noted that when modelling the K^+ diffusion test results with POLLUTE, batch test sorption parameters provided a good fit to potassium diffusion data providing independent verification of the sorption of K^+ at the levels expected based on sorption tests.

The results obtained from laboratory diffusion test modelling were used to provide theoretical comparisons to field CCL pore fluid concentrations.

5.3. COMPACTED CLAY LINER IONIC PORE FLUID DISTRIBUTION WITH DEPTH – FIELD RESULTS

Figures 7–11 present pore-water concentration profiles for calcium, magnesium, chloride, sodium, and potassium through the compacted clay liner at each borehole. When comparing boreholes, there is some scatter, possibly the result of non-uniform distribution of defects found on the geomembrane liner (holes, tears, wrinkles) occurring at different time periods, producing differences in ionic pore-water concentrations between boreholes. Also of note is the apparent back diffusion of chloride and sodium and possibly calcium, potassium and magnesium from the compacted clay into the lagoon. The leachate lagoon had been decommissioned for a period of approximately one month before the field investigation was performed. Rain water that accumulated in the bottom of the lagoon would have caused dilution of leachate present at the bottom of the lagoon, causing back-diffusion of each contaminant into the lagoon.

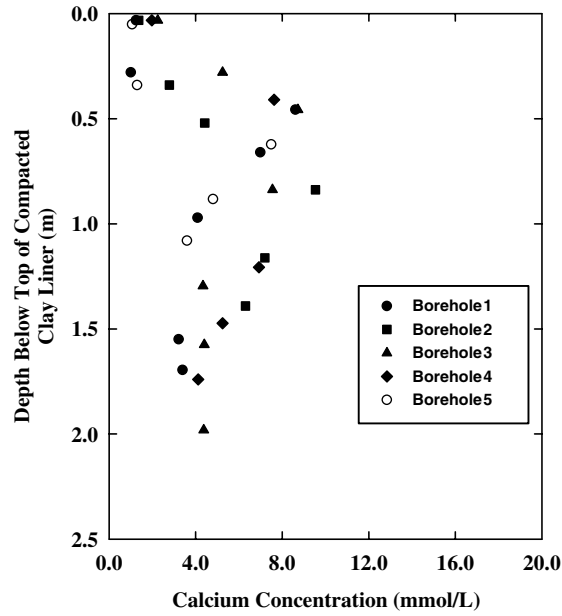


Figure 7. Calcium pore fluid profile through compacted clay liner component of composite liner system.

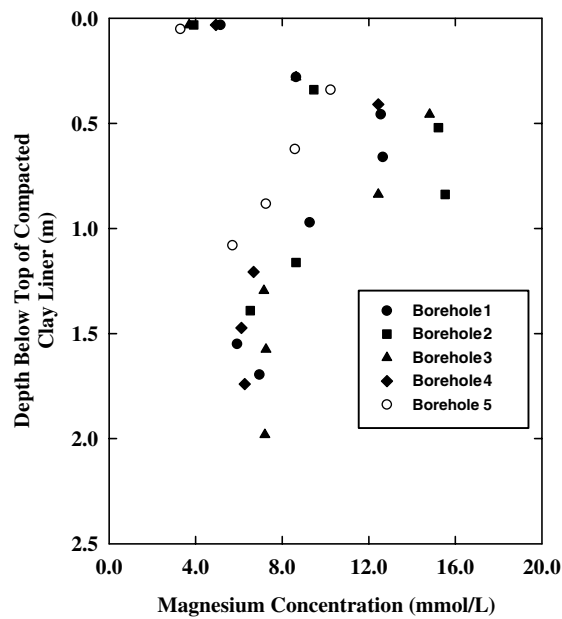


Figure 8. Magnesium profile through compacted clay liner component of composite liner system.

After 14 years of lagoon operation, chloride had migrated approximately 1.7 m into the compacted clay liner (Figure 9). Sodium, which was slightly retarded with respect to chloride, had migrated approximately 1.2 m (Figure 10). Chloride is often

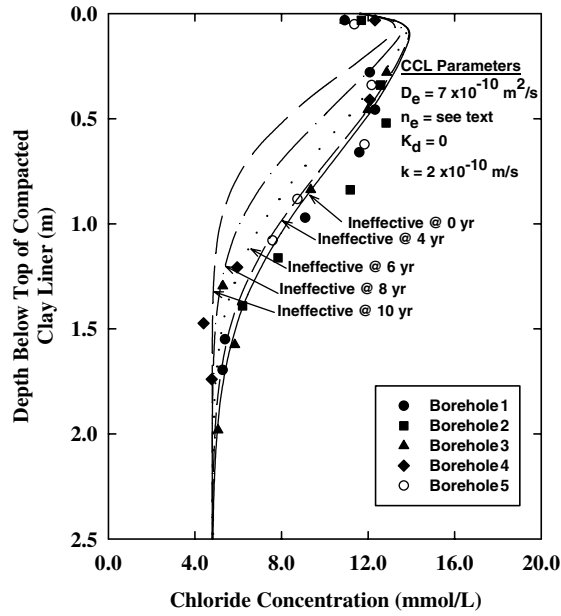


Figure 9. Prediction of chloride pore fluid concentrations through compacted clay liner component of composite liner for different assumed lifespans of the geomembrane (Modified from Rowe et al., 2003).

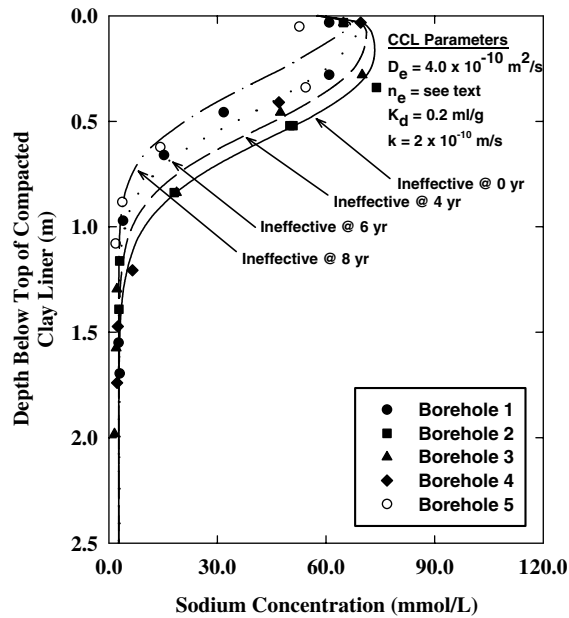


Figure 10. Prediction of sodium pore fluid concentrations through compacted clay liner component of composite liner for different assumed lifespans of the geomembrane.

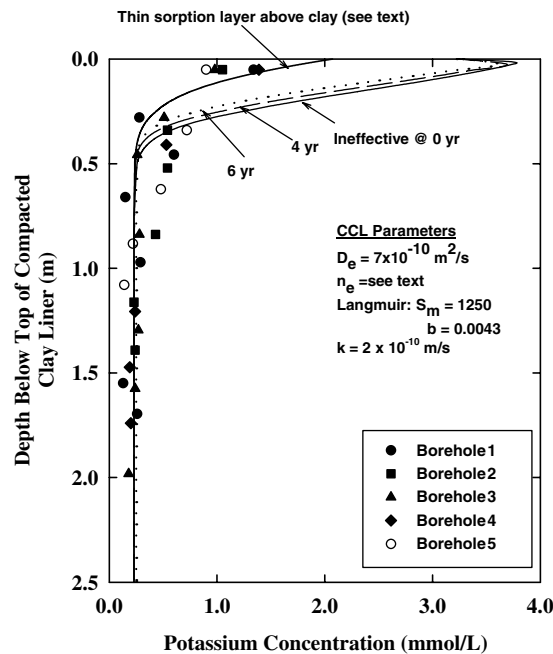


Figure 11. Prediction of potassium pore fluid concentrations through compacted clay liner component of composite liner for different assumed lifespans of the geomembrane.

considered a conservative contaminant that does not participate in sorption processes under typical alkaline groundwater conditions. The slight retardation of sodium relative to chloride indicates that some exchange reactions are occurring between the high concentrations of sodium in the migrating leachate and the adsorbed calcium and magnesium cations present on the clay exchange sites. The desorbed magnesium and calcium ions arising from this exchange process provide the charge balance for anions, such as chloride, as they migrate through the liner; this is exhibited as 'hardness halos' for calcium and magnesium (Figures 7 and 8).

Further confirmation of exchange of calcium and magnesium for sodium as well as potassium is evident by examining a plot of adsorbed cations with depth in the compacted clay from borehole 4 (Figure 12). It should be noted that adsorbed cation concentrations are normalized by the percentage of clay sized particles present for the soil sample tested. A relative decrease in Ca^{2+} and Mg^{2+} adsorbed cation concentrations near the surface of the compacted clay is apparent when compared to a relative increase in Na^{+} and K^{+} adsorbed cations. Typically Na^{+} is not easily exchanged for Mg^{2+} and Ca^{2+} on clay exchange sites (Rowe et al., 2004). However, the mass loading of Na^{+} in the leachate is high relative to Ca^{2+} and Mg^{2+} (see Table 1) and is probably causing the desorption of Ca^{2+} and Mg^{2+} on the clay exchange sites (Mitchell, 1993). The migration of potassium is highly retarded with respect to chloride and sodium.

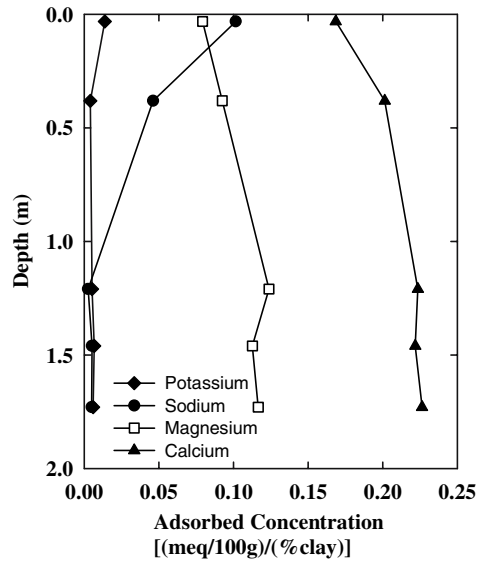


Figure 12. Adsorbed cation concentration with depth (borehole 4).

Even though chloride from the leachate has migrated approximately 1.7 m through the compacted clay, it is noted that there was still over 1 m of compacted clay and approximately 3 m of quarry rock separating this diffusion front from the water table in the Rochester Shale. It should also be noted that the water quality in the Rochester Shale is very saline and unpotable. Hence, while the geomembrane component of the composite liner system had apparently not performed well, the compacted clay had performed its desired function of preventing leachate migration into the underlying ground water during its 14 year lifespan. Contaminant transport modelling of the composite liner system confirms this effectiveness of the CCL as discussed below.

5.4. CONTAMINANT TRANSPORT MODELLING OF COMPOSITE LINER SYSTEM

The differential equation to predict one dimensional contaminant transport for a single reactive solute (no degradation) through a saturated porous medium (i.e. CCL) is given by Rowe et al. (2004) as:

$$n_e \frac{\partial c}{\partial t} = \left(n_e D_e \frac{\partial^2 c}{\partial z^2} - n_e \bar{v} \frac{\partial c}{\partial z} \right) - \rho \frac{\partial S}{\partial t}, \quad (2)$$

where c = concentration at depth z and time t [ML^{-3}]; n_e = effective porosity [-]; D_e = effective diffusion coefficient [L^2T^{-1}]; \bar{v} = average linearized groundwater velocity [LT^{-1}]; ρ = dry density [ML^{-3}]; S = mass of solute removed from solution per unit mass of solute [-]. The contaminant transport program POLLUTE (Rowe and Booker, 1999) solves this equation numerically by imposing the appropriate boundary conditions for the particular problem. For contaminant migration

through the compacted clay liner, the bottom boundary condition was considered infinitely thick for modelling purposes. This is a reasonable assumption since the chloride diffusion front had only migrated 1.7 m through the 2.9 m compacted clay liner and hence the underlying shale did not directly participate in the contaminant migration process. The top boundary condition of the leachate in the lagoon was assumed to be a constant concentration over each of three different time periods as shown in Figure 1.

As summarized in Figure 1, geometric means were calculated for both influent and effluent leachate lagoon concentration data for three time periods (0–4.5 years; 4.5–9.5 years; and 9.5–14 years). Sodium and potassium were not monitored from 0 to 4.5 years and therefore it was assumed that the ratio proportional decrease of Cl from 608 to 510 mg/L (81%) and Na from 3454 to 1640 mg/L (47%) followed the same proportion increase from 0–4.5 to 4.5–9.5 years (41% Cl to 23% Na). The same logic was followed for K^+ to give concentrations in years of 0–4.5 as shown in Figure 1. Although calcium and magnesium porewater concentrations were obtained from the field program, they were not considered in the modelling exercise since varying degrees of desorption processes are occurring for both ions. A model that considers contaminant transport of a single parameter is unable to predict the field profiles for ions in which a major contributor to the pore-water profile is desorption.

To evaluate the effectiveness of the geomembrane, contaminant transport modelling was performed assuming different times at which the geomembrane ceased to be effective and allowed leachate into direct contact with the clay liner. The parameters given in Tables 4 and 5 were used to calculate the Cl^- , Na^+ , and K^+ pore fluid concentrations in the compacted clay. To account for back-diffusion of ions for the one month prior to the field investigation, it was assumed that dilution of the small amount of liquid at the bottom of the lagoon during this time caused a linear decrease in leachate concentrations of 20% over this time period. These theoretical curves were then compared with the pore fluid concentrations obtained from the field investigation.

As discussed by Rowe et al. (2003) the geomembrane component of the composite liner system had a large number of defects (600 defects per hectare) and any attempt to use small leakage rates typical of those used in landfill design (e.g. 2.5 holes per hectare) through the geomembrane resulted in no fit to the field data. This was consistent with observations made during the field investigation, confirmed by discussions with maintenance staff, that there was a significant amount of leachate trapped under the geomembrane and was in direct contact with the compacted clay

Table 5. Soil parameters used for contaminant transport modelling of compacted clay liner

Depth range of layer used in modelling (<i>m</i>)	Darcy velocity (<i>m/a</i>)	Porosity of layer (-)	Dry density of layer (g/cm^3)
0–0.25	0.13	0.48	1.4
0.25–0.50	0.13	0.42	1.5
0.5–bottom	0.13	0.38	1.7

for much of its period of service. Thus at some point during the lifetime of the lagoon, the geomembrane stopped functioning as designed and started to allow leachate to come into contact with the compacted clay.

Figure 9 shows contaminant profiles for chloride assuming the geomembrane was ineffective (i.e. the advective flux through the clay liner was based on the gradient through, and hydraulic conductivity of, the compacted clay assuming that the leachate was in direct contact with the clay). Concentrations at various times (4, 6, 8, and 10 years) relative to the time of first use of the lagoon (time = 0 year) were calculated. As shown in Figure 9, theoretical chloride pore fluid profiles assuming geomembrane failure at 8 and 10 years provide a poor fit to experimental chloride pore fluid concentrations. However, assuming the geomembrane functioned effectively for elapsed times of 0, 4 and 6 years after construction provides a reasonable fit to the majority of the data. Pore fluid concentrations at depths 0.5–1.2 m seem to be slightly under-predicted using a chloride diffusion coefficient of $7 \times 10^{-10} \text{ m}^2/\text{s}$ and a CCL hydraulic conductivity of $2 \times 10^{-10} \text{ m/s}$. To examine the effect of hydraulic conductivity of the compacted clay on modelling results, approximate maximum and minimum values obtained from laboratory testing were utilized (1.5×10^{-10} and $3.0 \times 10^{-10} \text{ m/s}$) as shown in Figure 13. Reasonable fits to field data are obtained. Increasing the hydraulic conductivity by an order of magnitude produced poor fits to chloride field results. These same general conclusions were reached when modelling Na^+ results (see below).

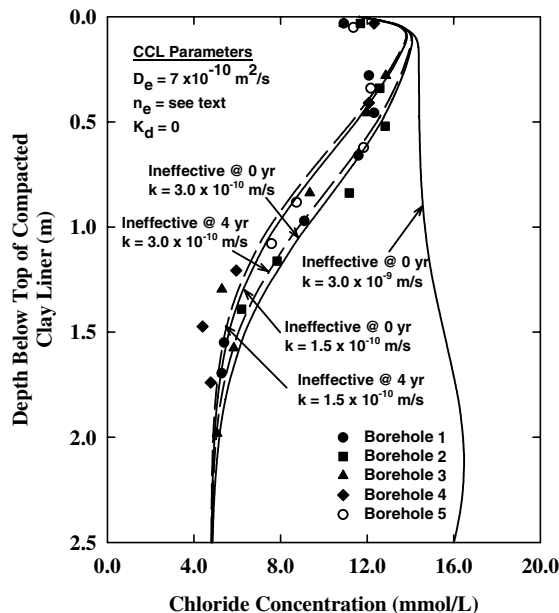


Figure 13. Prediction of chloride pore fluid concentrations through compacted clay liner component of composite liner for different assumed lifespans of the geomembrane (Influence of Hydraulic Conductivity).

The same variation in parameters was examined (time of GM failure and hydraulic conductivity) for Na^+ and K^+ . Figure 10 shows that Na^+ pore fluid concentrations are predicted best when the GM is assumed to fail either at between 4 and 6 years. However, there is not as good a theoretical fit for all the boreholes using the range of parameters previously described for chloride. This was mainly because Na^+ pore fluid profiles (Figure 10) exhibited more scatter than those of chloride (Figure 9). Examining chloride leachate concentrations in Figure 1, it can be seen that for the first 4.5 years of the lagoon, chloride concentration was approximately 250 mg/L or 7 mmol/L which is only slightly higher than that of the Cl^- background pore fluid in the compacted clay (approximately 4 mmol/L). Since there is very little chemical gradient of chloride into the clay, there is not much difference between predicted Cl^- curves assuming failure at 0 or 4 years (Figure 9). However when considering Na^+ concentrations for the first 4.5 years of the lagoon, the assumed concentrations in the leachate (~ 62 mmol/L) are much higher relative to Na^+ background pore fluid values in the compacted clay (~ 3 mmol/L). Thus the sodium profiles would be more sensitive to when leachate came into contact with the clay at different locations in the liner during the early period of operation (0 and 6 years – Figure 10) than chloride profiles (Figure 9). Based on this observation, it is hypothesized that the range in data (which is internally consistent to each bore hole and does not appear to be the result of experimental error) is a result of different times at which leachate came into contact with the geomembrane at the locations of the different bore holes during the first 6 years of operation, with almost immediate contact (i.e. in less than 4 years) with leachate being inferred at Borehole 2 to as much as about 6 years at Boreholes 1 and 5.

Theoretical K^+ profiles generated by modelling the field conditions (Figure 11) both over predict (at leachate interface) and under-predict (between 0.3 and 0.6 m) K^+ concentrations through the clay. A sensitivity study using a lower diffusion coefficient than that obtained from diffusion testing or different hydraulic conductivities did not improve the fit for assumed geomembrane failure times of 0, 4 or 6 years. This small dependency of K^+ concentrations on the time of failure, diffusion coefficient and hydraulic conductivity is a result of the high sorption onto the clay which dominates the transport of K^+ . Thus K^+ is not a good indicator of GM failure in this instance.

As shown on Figure 11, there is a poor match between theoretical and experimental lagoon/compacted clay interface K^+ concentrations which may be due to geochemical process occurring between the leachate and the black sludge observed on top of the compacted clay. Also, between depths of 0.3 and 0.7 m, K^+ concentrations are under predicted using the modelling parameters previously discussed. Since other parameters such as time of failure, diffusion coefficient, and hydraulic conductivity have very little influence on predicted concentrations, the discrepancy may be due to sorption parameters used for modelling.

As discussed above, sorption of K^+ appears to be dominating the transport process and therefore any reasonable variation in sorption parameters could potentially

have a large influence on predicted results. To estimate the influence of uncertainty the dry density and sorption parameters had on modelling, the high and low values of dry density used for each layer (from the field investigation) in combination with corresponding upper and lower bound sorption values from batch tests were used for modelling. However, prediction of K^+ field concentrations did not improve with these upper and lower bound parameters (Lake 2000).

It is believed that the reason for these discrepancies at the leachate interface may be due to geochemical processes occurring between the leachate and the black sludge in the top surface of the clay, as well as, above the clay, under the geomembrane. As discussed by vanLoon and Duffy (2000), organic matter can have a 'CEC' range of 150–500 meq/100 g when pH ranges from approximately 6–9. For the samples near the leachate interface of the lagoon, organic matter contents from 0.03 to 0.6 m were approximately 0.6% or more than three times that of samples at depth, (batch tests and diffusion tests were performed on a sample 1.8 m below this interface, with a minimal organic matter content of 0.2%). This increase in organic matter could cause an increase in sorption of K^+ . Even more importantly, a layer of black sludge that accumulated at the bottom of the lagoon (the reason for periodic cleaning of the lagoon) would have substantial organic matter content. In order to match leachate interface concentrations in the clay, a thin (30 mm) layer of organic matter with appreciable sorption of 300 mL/g (100 times that of clay based on organic matter's CEC of 325 meq/100 g and 50% organic matter) was considered in the modelling as shown in Figure 11. A reasonable fit is obtained at the leachate interface, but at the expense of data at depth (0.4–1.0 m). It is probable that a combination of K^+ interaction with organic matter in the clay and sludge in the base of the lagoon resulted in lower concentrations of K^+ available for contaminant transport into the clay. This time dependant generation of sludge would therefore have an influence on the resulting K^+ profile, possibly causing increased transport of K in early times. However, at later times, as the sludge accumulated, higher sorption would result. Any attempt to quantify this would be speculative based on the available information. Variation in clay sized particles and hence the amount of clay minerals throughout the compacted clay may also be influencing results.

The leachate interface modelling data did predict reasonably well the depth of K^+ assuming a geomembrane failure of some time between 0 and 4 years. From an engineering standpoint, the results are reasonable consistent with the findings deduced for Cl^- and Na^+ .

6. Summary and Conclusions

This paper has examined the performance of a geomembrane/compacted clay composite liner at the end of its 14 year operational lifespan. Observations of defects present on the HDPE geomembrane as well as the presence of leachate trapped between the geomembrane and compacted clay suggests that the unprotected geomembrane was ineffective at containing the leachate in the lagoon. This was

confirmed by the presence of ionic contaminant profiles through a portion of the clay which would not have been present if the geomembrane had been intact throughout the lifetime of the lagoon. Modelling of the contaminant migration through the compacted clay suggests that the geomembrane probably was effective for less than 4–6 years after initial operation of the lagoon.

Results of diffusion, and sorption tests, as well as, field results indicated high sorptive uptake of K^+ . Batch testing indicated non-linear sorption of the K^+ with the soil which was fitted to the Langmuir sorption model. However these parameters could not predict the low concentrations in the clay near the interface without invoking sorption in the organic matter (black sludge) observed above the clay liner. Thus it is inferred that there was geochemical interaction of K^+ with the organic matter present at the base of the lagoon. Modelling of a layer of organic matter at the bottom of the lagoon improved the fit to field data near the leachate interface but at the expense of data at depth. Due to the high sorption of K^+ , it was found that Cl^- and Na^+ were better indicators of geomembrane failure times than K^+ .

This field case also highlights the importance of the compacted clay liner component of the composite liner system for this lagoon. The clay provided a low hydraulic conductivity ($2-3 \times 10^{-10}$ m/s) barrier which controlled both diffusive and advective contaminant transport over the 14 year period of operation. Within this time chloride had migrated (primarily by diffusion) to a depth of 1.7 m into the 2.9 m thick clay liner. Thus the clay liner prevented the ionic contaminants studied from reaching the groundwater system during the lifetime of the lagoon.

Acknowledgements

The study was financially supported by the Natural Science and Engineering Research Council of Canada (NSERC) and the Centre for Research in Earth and Space Technology (CRESTech). The authors are indebted to Mr. Steve Usher of Gartner Lee Ltd. for his initiation of this project as well as acting as a valuable information resource. The authors appreciate the help from Dr. Henri Sangam and Dr. Paul Dittrich with the field work, and Ms. Cristina Orsini with some of the chemical analyses of porewater samples.

References

- Barone, F.S., Yanful, E.K., Quigley, R.M. and Rowe, R.K. (1989) Effect of multiple contaminant migration on diffusion and adsorption of some domestic waste contaminants in a natural clayey soil, *Canadian Geotechnical Journal*, **26**, pp. 189–198.
- Barone, F.S., Mucklow, J.P., Quigley, R.M. and Rowe, R.K. (1991) Contaminant transport by diffusion below an industrial landfill site, in *Proceedings of the 1st Canadian Conference on Environmental Geotechnics*, Montreal, May 1991, pp. 81–90.
- Basnett, C. and Brungard, M. (1992) The clay desiccation of a landfill composite lining system, *Geotechnical Fabrics Report*, January/February, p. 38.
- Chapman, L.J. and Putnam, D.F. (1973) *The Physiography of Southern Ontario*, Ontario Research Foundation, University of Toronto Press, 386 pp.

- Corser, P., Pellicer, J. and Cranston, M. (1992) Observations on the long term performance of composite clay liners and covers, *Geotechnical Fabrics Report*, November, pp. 6–16.
- Goodall, D.C. and Quigley, R.M. (1977) Pollutant migration from two sanitary landfill sites near Sarnia, Ontario, *Canadian Geotechnical Journal*, **14**, pp. 223–236.
- King, K.S., Quigley, R.M., Fernandez, F., Reades, D.W. and Bacopoulos, A. (1993) Hydraulic conductivity and diffusion monitoring of the Keele Valley Landfill liner, Maple, Ontario, *Canadian Geotechnical Journal*, **30**, pp. 124–134.
- Lake, C.B. (2000) Contaminant transport through geosynthetic clay liners and a composite liner system, PhD dissertation, The University of Western Ontario, 412 pp.
- Lake, C.B. and Rowe, R.K. (2000) Diffusion of sodium and chloride through geosynthetic clay liners, *Geotextiles and Geomembranes*, **18**, pp. 103–131.
- Mitchell, J.K. (1993) *Fundamentals of Soil Behavior (2nd edn.)*, John Wiley and Sons, N.Y., 437 pp.
- Munro, I.R.P., MacQuarrie, K.T.B., Valsangkar, A.J. and Kan, K.T. (1997) Migration of landfill leachate into a shallow clayey till in southern New Brunswick: A field and modelling investigation, *Canadian Geotechnical Journal*, **34**, pp. 204–219.
- Park, J.K. and Nibras, M. (1993) Mass flux of organic chemicals through polyethylene geomembranes, *Water Environment Research*, **65**(3), pp. 227–237.
- Quigley, R.M. (1971) The geotechnical design of the Townline Road-Rail Tunnel: Discussion, *Canadian Geotechnical Journal*, **8**, pp. 609–610.
- Quigley, R.M., Fernandez, F. and Ohikere, C. (1989) Compatibility of Leda clay with municipal solid waste leachate, in *Proceedings Technology Transfer Conference 1989*, Ontario Ministry of the Environment, Toronto, Vol II- Liquid and Solid Waste: 25–47.
- Rowe, R.K. (1998) Geosynthetics and the minimization of contaminant migration through barrier systems beneath solid waste – Keynote Lecture, in *Proceedings, 6th International Conference on Geosynthetics*, Atlanta, Vol. 1, pp. 27–102.
- Rowe, R.K. and Booker, J.R. (1999) *POLLUTE v.6.3, 1-D Pollutant Migration Through a Non homogeneous Soil, 1983, 1990, 1994, 1997, 1999*. Distributed by GAEA Environmental Engineering Ltd., Ontario, Canada.
- Rowe, R.K., Caers, C.J. and Barone, F. (1988) Laboratory determination of diffusion and distribution coefficients of contaminants through undisturbed soils, *Canadian Geotechnical Journal*, **25**, pp. 108–118.
- Rowe, R.K., Hrapovic, L. and Kosaric, N. (1995) Diffusion of chloride and dichloromethane through an HDPE geomembrane, *Geosynthetics International*, **2**, pp. 507–536.
- Rowe, R.K., Quigley, R.M., Brachman, R.W.I. and Booker, J.R. (2004) *Barrier Systems for Waste Disposal Facilities*, E & FN Spon, London, 579 pp.
- Rowe, R.K., Sangam, H.P. and Lake, C.B. (2003) Evaluation of an HDPE geomembrane after 14 years as a leachate lagoon liner, *Canadian Geotechnical Journal*, **40**(3), pp. 536–550.
- Rowe, R.K., Hsuan, Y.G., Lake, C.B., Sangam, P. and Usher, S. (1998) Evaluation of a composite (geomembrane/clay) liner for a lagoon after 14 years of use. in *Proceedings of the 6th International Conference on Geosynthetics*, Atlanta, Georgia, March 1998, pp. 233–238.
- vanLoon, G.W. and Duffy, S.J. (2000) *Environmental Chemistry – A Global Perspective*. Oxford University Press, 492 pp.
- Yanful, E.K. (1984) Heavy metal migration through clay. *PhD Dissertation*, The University of Western Ontario, 357 pp.
- Yanful, E.K., Haug, M.D. and Wong, L.C. (1990) The impact of synthetic leachate on the hydraulic conductivity of a smectite till underlying a landfill near Saskatoon, Saskatchewan, *Canadian Geotechnical Journal*, **27**(4), pp. 507–519.
- Yong, R.N., Mohamed, A.M.O. and Warkentin, B.P. (1992) *Principals of Contaminant Transport in Soils, Developments in Geotechnical Engineering*, Elsevier, Vol. 73, 327 pp.