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Migration of Landfill Leachate nrough Glacial Tills

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ABSTRACT

To evaluate the potential of natural clay minerals for attenuating and preventing the pollution of water resources by landfill leachates, leachate was collected by anaerobic techniques from the 15-year old Du Page County sanitary 'fill near Chicago, Illinois, and was passed through 44 ›ratory columns that contained various mixtures of calcium-saturated clays and washed quartz sand. The columns were constructed to simulate slow, saturated, anaerobic flow, and manometers were placed at five locations in each column to measure any changes in hydraulic conductivity. Leachate was run through the columns for periods ranging from 6 to 10 months, during which time effluents were collected periodically and analyzed for 21 chemical constituents. The columns were then sectioned and the clays analyzed to determine the vertical distribution of the chemical constituents in each column. Sterilized landfill leachate was used in a duplicate set of columns to determine the effects of gross biological activity.

Chloride and certain organic compounds were relatively unattenuated by passage through the clay columns; monovalent cations, such as Na, K, and NH_4, were moderately attenuated; and heavy metals, such as Pb, Cd, and Zn, were attenuated by even small amounts of clay. Concentrations of Ca and Fe were much higher in the column effluents than they were in the original leachate. The increase in Ca in the column effluents was due to cation exchange with ions in the refuse leachate. The Fe increase probably resulted from reduction of the oxidized Fe on clay surfaces by anaerobic refuse leachate to more soluble reduced Fe.

Both biologically active and sterilized leachate reduced the hydraulic conductivity of the clay-mixture columns during the experiment. The active leachate reduced the hydraulic conductivity to a much greater degree than the sterile leachate.

Results of the laboratory data were compared witl field data from the Du Page County sanitary landfill and from other existing landfills where detailed data are available. The field data show a "hardness halo" corresponding to the Ca release in the columns. The relative attenuation rates of some of the ions were also confirmed by the field data. The change in hydraulic conductivity was not as clearly shown.

Ion exchange capacity, hydraulic conductivity, and buffering capacity of the earth materials were all shown to be important in assessing the potential of landfills for water pollution.

INTRODUCTION

Land disposal of solid wastes, both domestic and industrial, has been practiced for many years, and the open burning dump has gradually been replaced by the sanitary landfill over the past 30 years. Garland and Mosher (1975) have estimated that there are about 14,000 landfills in the United States; Clark (1975) estimated that there are more than 2,000 known disposal sites in Illinois, although only about 240 are legally operating at this time. A reduction in the number of operating landfills is typical throughout the United States. This reduction in the number of disposal sites has effectively concentrated increasing volumes of refuse at fewer and fewer sites, especially near the urban centers.

There are only three media into which man can place his ever-increasing volume of waste—air, water, and land. The increasingly strict regulations governing the discharge of pollutants to the air and water

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have placed new emphasis on land disposal. This restriction of waste discharge to one medium only serves to increase the stress upon that medium. Since there are interchanges between all the media, land disposal of waste ultimately discharges some of the waste products back to the water or air.

The environmental effects of land disposal are difficult to determine, inasmuch as the subsurface is complex and commonly insufficiently understood to monitor properly. The success of land disposal is judged primarily by the rate of return of the pollutants to the air or water; agricultural and biological scientists are just now beginning to study the uptake of some pollutants through biological systems.

There have been sufficient studies to show that leachate will form in sanitary landfills where there is enough moisture. This condition is met in all but the more arid regions of the nation. The pollutants thus leached from the refuse leave the site as a gas discharged to air or as a leachate discharged to the ground water or surface water. The migration of the pollutants in the subsurface is governed by geologic and hydraulic principles that have been delineated in a number of studies (for example, see Hughes *et al.*, 1971).

Most regulations in force today require either that refuse be placed in materials of sufficiently low hydraulic conductivity to prevent the leachate from entering a ground-water resource in quantities large enough to significantly degrade water quality, or that the disposal site be hydrologically controlled to prevent indiscriminant discharges of leachate. The low-conductivity material is usually a clayey material that either occurs there naturally or is placed there to prevent the escape of leachate that could cause ground-water pollution.

This paper reports some of the results of a laboratory study conducted at the Illinois State Geological Survey and supported in part by U.S. Environmental Protection Agency Contract 68-03-0211 (Cincinnati, Ohio). The purpose of the study was (a) to investigate and evaluate the attenuating properties of clay and the use of clay minerals as liners (natural or artificial) for sanitary landfills, and (b) to relate this laboratory work with the Geological Survey's ongoing program of field evaluation and research at sanitary landfill sites.

This paper relates two significant phenomena noted in the laboratory to field observations around sanitary landfills: The elution of large amounts of the calcium ion from the study columns (Griffin and Shimp, 1976) and the reductions in hydraulic conductivities that resulted from the introduction of leachate to the clay-sand mixtures. The applicability of laboratory studies to the complex field situation is necessary in order that the data may be used in designing the sanitary landfill. On the basis of these laboratory results, one can estimate the amounts of clay required to attenuate leachate as it passes through the clay liner. The results presented in this paper apply only to the leachate of the Old Du Page County landfill; the extent that these results may be generalized to other sites with leachate of different chemical composition is problematic.

EXPERIMENTAL

Laboratory Studies

The clays used in this study were kaolinite (1:1 lattice), montmorillonite (1:1 expanding lattice), and illite (2:1 nonexpanding lattice, mica type). These clay minerals were chosen for study because they are available in commercial quantities and it was felt that their compositions are more representative of natural materials used at landfills. The details of the clay mineralogy and chemistry and of the methods of study have been reported previously (Griffin and Shimp, 1976). The predominantly Ca-saturated <2 μm particle-size fraction of the three clays was then used in the column-leaching studies.

The leachate used in this study was collected from well MM63 at the Du Page County sanitary landfill near Chicago, Illinois. The site description and well location are given by Hughes *et al.* (1971). Details of the leachate collection, processing, and storage are described by Griffin and Shimp (1976). One drum was taken to the Argonne National Laboratories and sterilized by gamma-ray irradiation using a cobalt source that gave a dose of 3.36×10^6 rad at the center of the drum. The second drum was allowed to remain biologically active. Chemical analyses of both the sterilized and natural leachate used in the column-leaching study are given in Table 1.

The laboratory apparatus used in the study consisted of laboratory columns containing mixtures of clay minerals and washed quartz sand (Ottawa Silica Co.) through which leachate was passed (Figure 1). The columns and apparatus were constructed to simulate the slow (<2 pore volumes per month), saturated, anaerobic flow of leachate at the bottom of a landfill. More details of the column apparatus and design can be obtained from Griffin and Shimp (1975, 1976). The leachate was passed through the columns, and the effluents were collected in graduated cylinders for chemical analyses and the measurement of the flow rates.

Table 1. Chemical Analyses of Landfill Leachate from Du Page County, Illinois

Analysis	Natural (ppm)	Sterilized (ppm)
	748.	744.
K	501.	491.
Ca	46.8	43.2
Mg	233.	230.
Cu	<0.1	<0.1
Zn	18.8	16.3
Pb	4.46	4.26
Cd	1.95	1.88
Ni	0.3	0.3
Hg	0.0008	0.37*
Cr	<0.10	<0.10
Fe	4.2	3.0
Mn	<0.1	<0.1
Al	<0.1	<0.1
NH_4	862.	773.
As	0.11	0.14
B	29.9	28.5
Si	14.9	15.0
Cl	3484.	3311.
SO_4	<0.01	<0.01
PO_4	<0.1	<0.1
COD	1340.	10,603.*
Organic acids	333.	290.
Carbonyls as acetophenone	57.6	90.1
Carbohydrates as dextrose	12.	11.
pH	6.9	7.2
Eh	+7 mv	+75 mv
E.C.	10.20 mmhos/cm	10.42 mmhos/cm

* Added during passage of sterilant gas.

10,200 μmhos

SO4 reduced to HS⁻, H2S

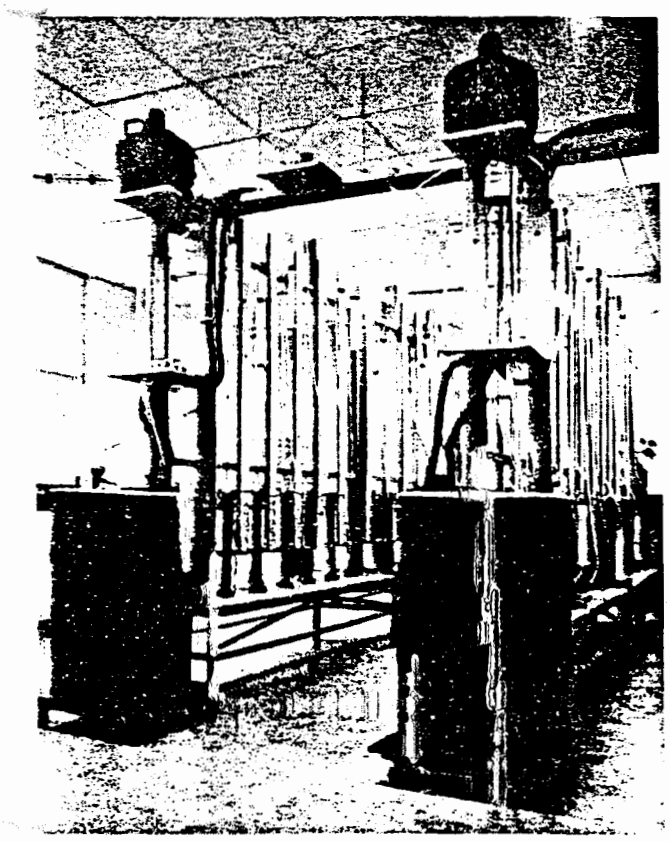

Fig. 1. The leaching apparatus. There are 44 columns controlled by two constant head inflow tanks and individually variable levels of outflow collection cylinders.

The outflow tube was maintained above the top of the columns to ensure saturated flow. The level of the outflow tube was moved either up or down to maintain relatively constant flow rates throughout the experiment. The columns were constructed of two-inch acrylic tubing to which manometer outlets were fitted at five locations on the column. To simulate field conditions the leachate containers and columns were either painted black or masked with black tape; this blackening prevents growth of organisms such as algae or photosynthetic bacteria.

Except for the 32 percent and 64 percent montmorillonite-sand mixtures, which were packed in columns 30 cm deep, the clay minerals and sand were packed into columns 40 cm deep. The columns were packed to bulk densities within the range of densities of natural glacial tills (~1.8 g/cc; Manger, 1963). The sand grains were coated with the clays, according to the methods given by Grim and Cuthbert (1945), before packing. Table 2 lists some chemical and physical properties of the various columns. Field bulk densities were achieved, and the hydraulic conductivities for each particular clay content and bulk density agree with those given by Todd (1959) for natural materials The hydraulic conductivity (k) was computed using the relationship:

$$k = \frac{Q\,dL}{A\,dH}$$

where

Q = flow rate in cm^3/sec;

A = cross-sectional area of column in cm^2;

dL = length of the column in cm;

dH = head of water in cm (dL/dH = hydraulic gradient).

The experimental design used in the study is also given in Table 2, which gives the percentages, by weight, of clay mineral(s) in each column (to which pure quartz sand was added to total 100%). The experimental design includes 100 percent sand and a complete geometric progression of clay percentages from 2 percent to 64 percent of kaolinite and of montmorillonite. Since kaolinite and illite have very similar cation exchange and lattice expansion properties, a complete geometric array for illite was not included.

Leachate was passed through the columns for periods of time ranging from 6 to 10 months, depending on the hydraulic conductivity of the individual column. During this time, effluents from each column were collected periodically and

Table 2. Physical and Chemical Properties of Various Sand-Clay Columns

Composition of column	Cation exchange capacity (meq/100 g)		Bulk density (g/cc)		Initial hydraulic conductivity (k) (cm/sec)	
	Set A†	Set B†	Set A	Set B	Set A	Set B
100% sand	0.0	.1	1.71	1.71	$1.27 \cdot 10^{-5}$	$1.80 \cdot 10^{-5}$
Sand and						
2% M*	1.4	2.3	1.71	1.72	$9.45 \cdot 10^{-6}$	$7.93 \cdot 10^{-6}$
4% M	3.2	4.3	1.77	1.74	$4.34 \cdot 10^{-6}$	$3.47 \cdot 10^{-6}$
8% M	7.3	7.2	1.79	1.78	$4.70 \cdot 10^{-6}$	$2.61 \cdot 10^{-6}$
16% M	11.9	12.1	1.87	1.86	$1.22 \cdot 10^{-6}$	$1.44 \cdot 10^{-6}$
32% M	26.8	24.0	1.55	1.52	$1.27 \cdot 10^{-6}$	$2.17 \cdot 10^{-6}$
64% M	56.2	55.5	1.23	1.11	$3.05 \cdot 10^{-7}$	$6.83 \cdot 10^{-7}$
Sand and						
2% K*	0.7	0.4	1.68	1.70	$7.44 \cdot 10^{-6}$	$4.53 \cdot 10^{-6}$
4% K	1.1	0.8	1.76	1.74	$4.78 \cdot 10^{-5}$	$2.76 \cdot 10^{-5}$
8% K	1.5	1.4	1.80	1.77	$9.90 \cdot 10^{-6}$	$8.25 \cdot 10^{-6}$
16% K	1.8	2.5	1.87	1.90	$2.86 \cdot 10^{-5}$	$1.92 \cdot 10^{-6}$
32% K	3.8	3.4	1.66	1.55	$2.40 \cdot 10^{-6}$	$4.81 \cdot 10^{-6}$
64% K	9.6	8.5	1.22	1.32	$5.45 \cdot 10^{-7}$	$4.57 \cdot 10^{-7}$
Sand and						
4% I*	0.8	0.9	1.80	1.81	$8.17 \cdot 10^{-6}$	$7.16 \cdot 10^{-6}$
16% I	3.5	3.2	1.83	1.91	$2.68 \cdot 10^{-5}$	$2.19 \cdot 10^{-5}$

* M = montmorillonite, K = kaolinite, I = illite.
† Set A - natural leachate; set B - sterile leachate.

measurements were made for Na, K, Ca, Mg, Al, Zn, Pb, Cd, Hg, Fe, Mn, NH_4, B, Si, Cl, chemical oxygen demand (COD), Eh, pH, and hydraulic conductivity. Finally, after leaching of approximately 15 pore volumes was completed, the clay mineral columns were sectioned and the contents analyzed to determine the vertical distribution of chemical constituents in each column.

Duplicate sets of columns were used in the experiment; one set of columns was leached with natural effluent while the second set was leached with sterilized effluent. Sterilization was used to determine if gross biological activity would affect hydraulic conductivity of leachate through clay minerals used as liners. The results of the experiment were statistically analyzed using the students (paired) "t" test to determine whether there were significant differences in the attenuation of each chemical constituent between sterile and natural leachate, and between clay minerals. Linear regression and moving average analysis were also performed on the column effluent data to determine relationships between hydraulic conductivity, attenuation, and clay mineral properties.

RESULTS AND DISCUSSION

Ca Elution/Hardness Halo

Results of some analyses of calcium, iron, and manganese are shown in Figure 2 plotted as relative concentration versus pore fraction. Relative concentration is the ratio of the concentration of the effluent of the column divided by the concentration of the influent. Thus, the "breakthrough" point for a given element is where the concentration of effluent equals the concentration of influent and has a value of one. A pore volume of effluent is defined as the volume necessary to displace the volume of interstitial liquid in the pore spaces in the column. The pore fraction is then given as the cumulative volume of column effluent divided by the pore volume of the individual column (Griffin and Shimp, 1976).

A wide range of attenuation was observed for several of the elements in leachate as they passed through the various columns containing different percentages of clay. The amount of reduction in concentration of a given element as it passes through the columns is discussed by Griffin and Shimp (1975, 1976).

Figure 2 illustrates the negative attenuation, or elution, of Ca, Fe, and Mn from the columns containing 2 percent, 8 percent, and 16 percent

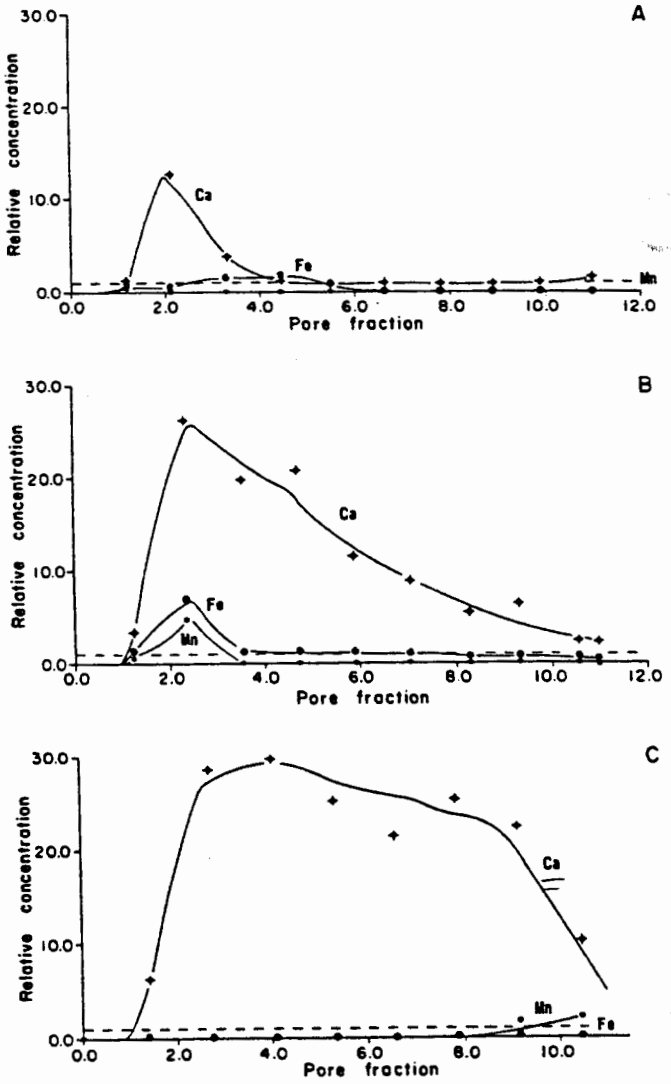

Fig. 2. Relative concentrations of Ca, Fe, and Mn as a function of pore fraction of leachate passed through columns containing (A) 2 percent montmorillonite clay, (B) 8 percent montmorillonite clay, and (C) 16 percent montmorillonite clay.

montmorillonite clay in sand. The relative concentrations greater than 1 indicate that Ca, and to a lesser extent Fe and Mn, are emerging from column at much greater concentrations than ...ose at which the influent leachate at various pore fractions. The area under the Ca curves can be seen to increase in proportion to the percentage of clay in the column. Griffin and Shimp (1976) quantified the observed attenuation represented by the area under each curve by integrating between pore fractions 1 and 11, and assigned a relative attenuation number (ATN) as shown in Table 3. Griffin and Shimp (1976) attributed the elution of Ca from the columns to an ion-exchange mechanism involving the replacing of the Ca bonded to the clays at their cation exchange positions by other ions in the leachate. The released Ca then passes onward with the ground water.

The unconsolidated surficial materials in much of Illinois are carbonate-rich, the clays generally having Ca in the cation exchange position and free carbonates in all except the leached zone. The presence of excessive hardness, reported as calcium carbonate, in the vicinity of sources of pollution has been noted in a number of articles, but its ...gin is rarely discussed. We have called this ...essive hardness the "hardness halo." Examples of it are found in Du Page County, Illinois (Figure 3), as reported by Zeizel *et al.* (1962). There are two areas of the county where the hardness, indicated by $CaCO_3$, in the shallow carbonate aquifer exceeds 1,000 parts per million

Table 3. Ranking of Chemical Constituents in Municipal Leachate According to Their Relative Mobility Through Clay Mineral Columns by Mean Attenuation Number (ATN) (from Griffin and Shimp, 1976)

Chemical constituent	ATN	Qualitative grouping
Pb	99.8	High
Zn	97.2	
Cd	97.0	
Hg	96.8	
Fe	58.4	Moderate
Si	54.7	
K	38.2	
NH_4	37.1	
Mg	29.3	
COD	21.3	Low
Na	15.4	
Cl	10.7	
B	-11.8	Negative (elution)
Mn	-95.4	
Ca	-656.7	

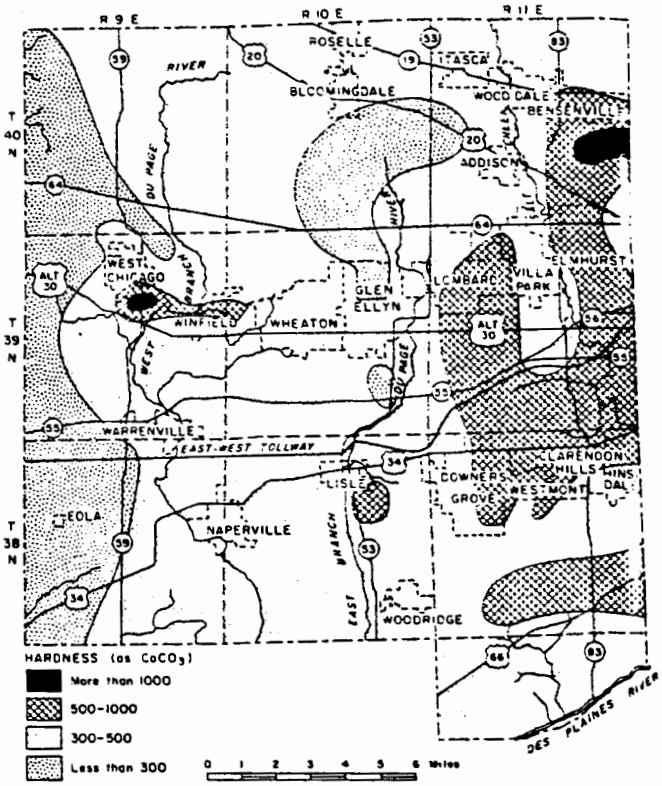

Fig. 3. Hardness of water, expressed as $CaCO_3$, in the Silurian dolomite aquifer in Du Page County, northeastern Illinois (after Zeizel *et al.*, 1962).

(ppm). The eastern area is a heavily developed residential area, where the glacial drift, which protects the aquifer from pollution, is relatively thin. No specific source can be attributed to the cause of the high hardness in this area; however, it is most likely due to a high concentration of home septic systems. The high hardness in the western area, near the town of West Chicago, is thought to have resulted from the discharge of large volumes of noncalcareous chemical waste to surface ponds (Walker, 1969). Further examples are reported in Anderson and Dornbush's (1967) study of a sanitary landfill in South Dakota and in Walker's (1969) discussion of ground-water pollution in Illinois. Most recently, Henning *et al.* (1975) found high calcium in monitoring wells very close to a landfill trench at Mentor, Ohio; the Ca concentrations decreased with distance from the fill and were lower in the refuse than in the closes wells.

Hughes *et al.* (1971) published the results of studies of five landfills in northeastern Illinois, including the Old Du Page County landfill, which was the source of the leachate used in this study. Monitoring of this landfill continued for three years after the completion of that report. Figure 4 was drawn using unpublished and published data

from the Winnetka and the Old Du Page landfills. Distances of travel from the refuse are along the inferred flow paths given by Hughes *et al.*, 1971.

There is considerable scatter in the Winnetka data (top, Figure 4). This may in part be due to a mixture of points, some being in the fine-grained alluvium and the others in the glacial till, which has somewhat different properties. However, these data suggest that the hardness approaches background values within 12 to 18 meters of the refuse, which is somewhat less than the limit to which chloride traveled (Hughes *et al.*, 1971). Note that the four datum points from piezometer nest LW5 follow this pattern.

The till under the Old Du Page landfill clearly illustrates the increase followed by the decrease in hardness. This till is separated from the refuse by 1 to 1.5 meters of sand. The hardness returns to background values within about 1.5 meters of travel, which is about half the distance of the estimated travel of the chloride ion (Hughes *et al.*, 1971). Note, in particular, the values shown for piezometer nests LW5 and LW6. These data points are all for the younger, northern part of the fill; data from the older parts of the fill do not fit the same curve (all hardness concentration values were low).

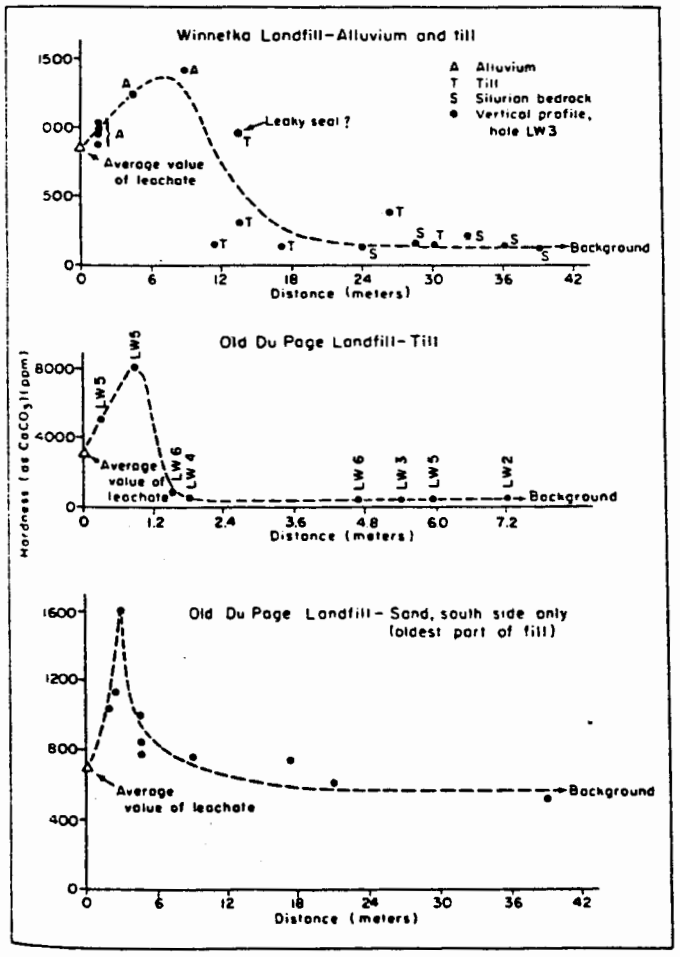

Fig. 4. "Hardness halo" effect shown as a function of distance (m) along the inferred flow path from the Winnetka and Du Page landfills in northeastern Illinois (after Hughes *et al.*, 1971).

The surficial sand (bottom, Figure 4) transmits leachate-contaminated water south from the older parts of the Du Page landfill and shows a hardness distribution similar to that of the north part of the fill. The hardness levels are much lower in the old refuse in this area, and all the values reflect that lower concentration. The hardness returns to background levels within 8 to 15 meters; however, the chloride ion has moved 240 to 300 meters in the permeable sand layer.

All these data clearly show that there is a "hardness halo" resulting from the movement of leachate into the surrounding till and sand, and that the rate of advance of the hardness front is less than that of the chloride ion. The chloride ion is probably the best tracer of this type of pollution in this environment. The distance of travel of the hardness front varied from slightly less than to approximately 10 percent of that of the chloride ion; this variation is probably controlled by the nature of the materials, cation exchange reactions, concentrations in the leachate, and ground-water flow rates. Nevertheless, all the data indicate that an increase in the chloride ion concentration followed by an increase in hardness of the water in the sediments over that in the leachate indicates that major pollution by the leachate plume will follow.

Hydraulic Conductivity

The results of initial hydraulic conductivity and bulk density measurements of the column contents are presented in Table 2. These data indicate that a wide range of hydraulic conductivities, with values in agreement with those expected under field conditions from similar materials (Todd, 1959), were observed. The relatively high bulk densities and slow flow rates used in this study closely simulate the conditions observed in the field. This procedure lends credence to the extrapolation of the results and conclusions of the laboratory studies to those obtained in the field.

During the initial stage of the experiment deionized water was circulated through the columns until hydraulic equilibrium was achieved. Hydraulic equilibrium was indicated by steady flow rates and relatively constant manometer readings from

the five manometers located over the length of the column. The columns containing low percentages of clay reached hydraulic equilibrium atively rapidly while the columns with high percentages of clay required circulation for more than a month to achieve steady manometer readings. When hydraulic equilibrium was achieved, the leachate was added to the columns.

Passage of leachate through the columns produced significant reductions of hydraulic conductivity. The natural leachate produced greater reduction than the sterile leachate. The changes in hydraulic conductivity that occurred in the kaolinite columns during the experiment have been reported previously (Griffin and Shimp, 1975). The results of hydraulic conductivity changes observed in columns containing montmorillonite, kaolinite, and illite clays are presented in Figures 5, 6, and 7. To more clearly show the trends in the data, the data presented in the Figures were statistically smoothed using the five-member moving average method. The change in hydraulic conductivity is expressed as units of the logarithmic scale. The raw data were statistically analyzed to determine whether significant differences in hydraulic conductivity sted between sterile and natural leachate. The a from the columns containing 4 percent kaolinite were rejected from the analysis when they were found to deviate by more than three standard deviations from the over-all mean change in

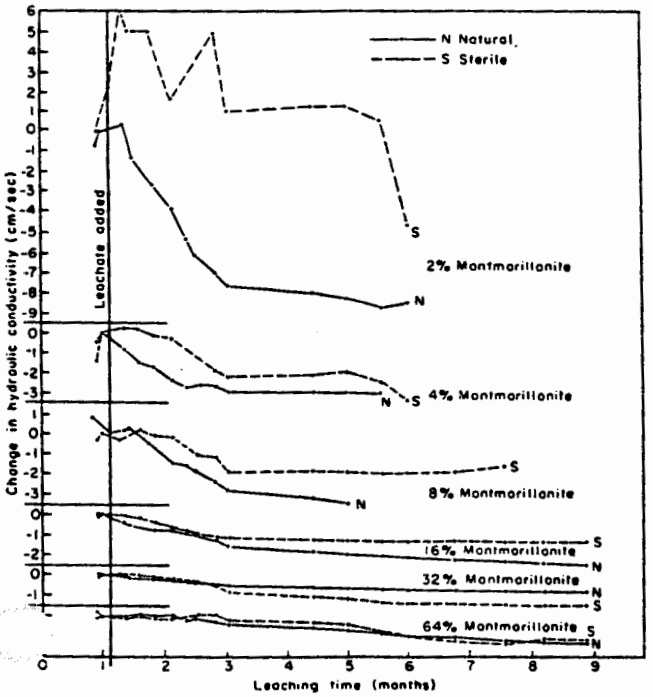

Fig. 5. Hydraulic conductivity of montmorillonite-sand columns as a function of leaching time.

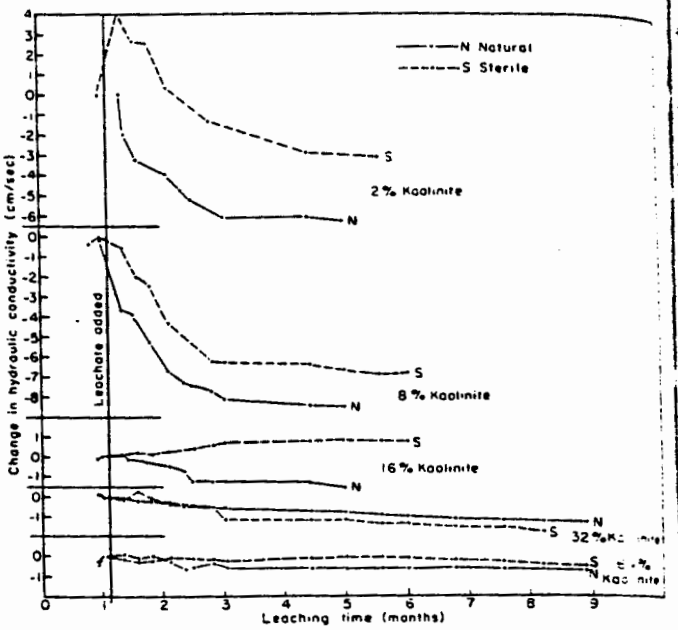

Fig. 6. Hydraulic conductivity of kaolinite-sand columns as a function of leaching time.

hydraulic conductivity observed for all other columns. The manometer readings indicated that the outflow tubes were plugged. The reason that only the columns with 4 percent kaolinite showed this problem is not clear. The results of the statistical analysis indicated that columns leached with natural leachate had significantly (.05 level) greater reductions in hydraulic conductivity than those leached with sterile leachate. This result is illustrated clearly in Figures 5, 6, and 7. Furthermore, the statistical analysis indicated that columns containing montmorillonite had significantly greater average reductions in hydrau ic conductivity than kaolinite or illite and that ther was no significant difference in the reductions in hydraulic conductivity between kaolinite and illite. Considering that montmorillonite clay swells

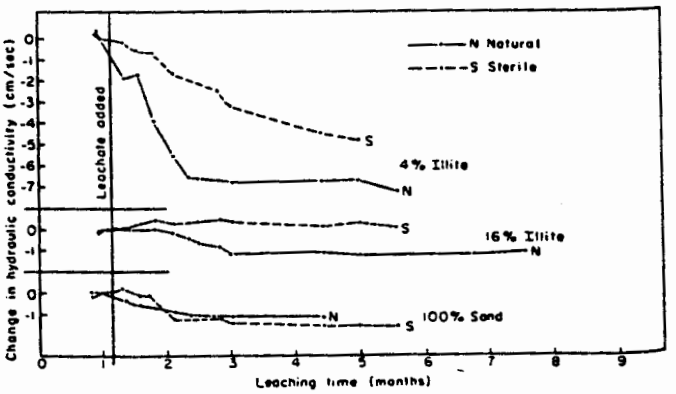

Fig. 7. Hydraulic conductivity of illite-sand columns as a function of leaching time.

when it is wetted, this result is not surprising.

Field data to support the above laboratory observation are not as clear as the data were for the "hardness halo." There have been fewer field tests for hydraulic conductivities than chemical tests for water quality. In addition, field tests may only be accurate to approximately a half-order of magnitude.

At the Winnetka landfill, 23 hydraulic conductivity tests were conducted, 7 on refuse, 4 on alluvium, and 12 on till. The data for these are too scattered to show any significant differences with respect to distance from the refuse.

At the Old Du Page County landfill, 34 field hydraulic conductivity tests were made, 14 on refuse, 14 on sand, and 6 on till. The data on the hydraulic conductivity of the sand (all south of the fill) suggest some reduction in hydraulic conductivity. The tests (10) made on monitoring wells less than 6 meters from the fill, have a mean hydraulic conductivity of 2.00×10^{-4} cm/sec (range 5.0×10^{-4} to 1.9×10^{-7}; excluding a value of 1.9×10^{-3} obtained from a gravel lens), and those monitoring wells greater than 12 meters from the refuse (4 tests) have a mean conductivity of 2.59×10^{-3} cm/sec (range 7.6×10^{-3} to 9.5×10^{-4}). The data are not statistically significant, but they do seem to indicate a possible reduction of hydraulic conductivity similar to that noted in the laboratory.

The reductions in hydraulic conductivity observed in the experiment with the Du Page leachate are particularly significant inasmuch as the Du Page leachate is an old leachate, approximately 15 years (Hughes *et al.*, 1971), and contains a relatively low percentage of organic compounds which are readily degradable by microorganisms (Table 1). In addition, this leachate has a low nutrient status, both phosphate and sulfate being absent in detectable quantities. Much higher amounts of microbial growth and plugging might be expected from a younger leachate.

These results have led to the conclusion that if clay liners, either natural or man-made of similar composition to those used in this study are used in municipal landfills, significant reductions in hydraulic conductivity can be expected because of microbial growth. Furthermore, slightly higher reductions in hydraulic conductivity can be expected from montmorillonite clays, apparently because of their tendency to swell.

Attenuation of Leachate Pollutants

During the period of time from the collection of the leachate through the period that hydraulic equilibrium was being established, the leachate was stored under refrigeration (3° to 5° C) with either argon or sterilant gas being purged slowly over the top of each drum. During the 10-month period chemical analyses were performed weekly on the leachate to monitor possible changes in composition. The value reported in Table 1 for COD, and for all the other constituents, is the average of the 37 separate analyses performed during the 10-month period of leaching.

The process of determining the attenuation of chloride and the other major components of Du Page leachate has been described by Griffin and Shimp (1976). The results of this study indicated that there was an average 6 percent greater attenuation of chloride in the columns leached with sterile leachate than those leached with the natural leachate. This greater attenuation is attributed to the reaction of chloride with the ethylene oxide to form ethylene chlorohydrin. Other than the slight increase in chloride attenuation, no other significant difference was apparently due to the increase in COD in the sterile leachate as compared to the natural leachate.

There were, however, other significant differences between the sterile and natural leachate treatments that were not attributed to the higher COD of the sterile leachate. Figure 8 illustrates the difference observed in Mn elution from the columns. The negative attenuation numbers indicate that more Mn eluted from the column than was present in the influent leachate. It can be seen that much higher levels of Mn were found in effluents from the columns leached with natural leachate. Griffin and Shimp (1976) concluded that the elution of Mn was due to reduction of surface coatings of Mn oxides on the clays by the anaerobic leachates. This conclusion is further verified by the difference in Mn elutions from the natural and sterile leachate treatment. This difference is attributed to the stronger anaerobic environment provided by the active microorganisms present in natural leachate. Inspection of the data in Table 2 shows that the average Eh (oxidation potential) reading of the natural leachate was an order of magnitude lower than the sterile leachate, even though both were in the anaerobic range (Eh readings less than 197 m.v. are considered to reflect anaerobic conditions). A similar result was obtained for Fe in that significantly (.05 level) greater mobility of Fe was found in columns leached with natural leachate than in those leached with sterile leachate. A mechanism similar to that for Mn elution is postulated as the reason for the

observed differences between the natural and sterile leachate.

Those chemical constituents for which no significant difference in attenuation between the normal and sterile leachate was found were Ca, Mg, Na, K, NH_4, Pb, Hg, Zn, and Cd. The constituents Al, Cu, Ni, Cr, As, S, and PO_4 were found in such low concentrations in the Du Page leachate that no attenuation order could be determined. Table 4 lists the constituents of the Du Page leachate and the hazard index determined by Griffin and Shimp (1976). They defined the hazard index of a leachate as the product of the Toxicity Index and the Mobility Index (see Table 4); it indicates the relative environmental hazard associated with individual ions.

Table 4. Chemical Constituents Contained in Du Page Leachate Ranked by Pollution Hazard Index [See Griffin and Shimp (1976) for the Definition and Derivation of Each Term]

Chemical constituent	Effective concentration / D. W. Standard	Toxicity Index[1]	Mobility Index[2]	Hazard Index[3]
NH_4	862/0.5	1724.	62.9	108,440.
B	(29.9 + 3.5)/1.0	33.4	111.8	3,734.
COD	1340/50	26.8	78.7	2,109.
Hg	0.87/.002	435.	3.2	1,392.
Cl	3484/250	13.9	89.3	1,241.
Ca	(46.8 + 307.3)/250*	1.42	756.7	1,072.
Cd	1.95/.01	195.	3.0	585.
Fe	4.2/0.3	14.0	41.6	582.
Na	748/270	2.77	84.6	234.
Mn	(0.02 + 0.02)/.05	0.78	195.4	153.
K	501/250*	2.00	61.8	123.
Mg	233/250*	0.93	70.7	65.7
Pb	4.46/.05	89.2	0.2	17.8
Zn	18.8/5.0	3.76	2.8	0.5
Si	14.9/250*	0.06	45.3	2.7

[1] Toxicity Index = $\frac{\text{Effective concentration}}{\text{Drinking water standard}}$

[2] Mobility Index = (100 - attenuation number)

[3] Hazard Index = Toxicity Index × Mobility Index

* Actual value not established by EPA, therefore assumed to be the same as chloride.

Design of a Landfill

The design of a landfill should take into account three factors: the hydrologic system governing direction of pollutant travel; the geochemistry of the water-sediment system; and the release rate of unattenuated pollutants to surface or ground waters. The first factor has been the subject of a number of papers and will not be discussed here.

Current landfill design and engineering practice utilizes thick liners, either natural or artificial, containing high percentages of clay. The motive is to create relatively impermeable liners that will contain the leachate and therefore protect the ground-water resources. This approach creates difficulties in humid climates where infiltration exceeds the capacity of the liner to dissipate the leachate. This causes what is referred to as the "bathtub" effect wherein the relatively impermeable clay liner fills with leachate and then overflows. The overflow manifests itself in the form of leachate springs on the surface and results in surface-water pollution instead of ground-water pollution. Neither form of pollution is necessary if proper design features are utilized in the construction of the sanitary landfill.

The results of a chemical attenuation study reported by Griffin and Shimp (1976) have indicated that most of the toxic constituents found in municipal leachates are moderately to highly attenuated by passage through laboratory columns containing relatively low percentages of clay minerals. If it is assumed that the "bathtub" effect is an undesirable feature of clay liners, then it follows that it is desirable to determine the point of "optimal" attenuation, that is, the percentage of clay in a liner which gives maximum attenuation balanced with maximum hydraulic conductivity. The data presented in Figure 9 represent examples of the percentage of montmorillonite necessary for "optimal" attenuation as determined for Cl, NH_4, and Pb. Figure 9 is a dual-scaled graph with the initial hydraulic conductivity (k) of the montmorillonite columns

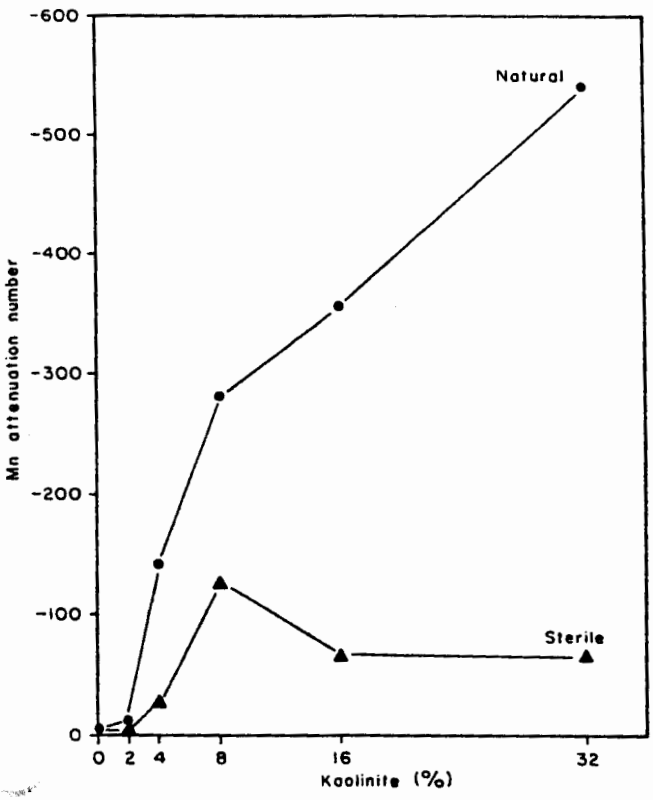

Fig. 8. Mn elution related to percentage of kaolinite leached with natural and sterile leachate.

given in Table 2 plotted as a function of percentage of montmorillonite. The other scale is the attenuation number for the chemical constituent of interest also plotted as a function of the percentage of montmorillonite. The attenuation numbers, as reported by Griffin and Shimp (1976), are the percentage of removal of the element from the leachate upon passage through 10 pore fractions of the clay-sand mixture. The attenuation scale is given as 0 at the point of minimum hydraulic conductivity and 100 at the point of maximum hydraulic conductivity. The point where the attenuation curve and the hydraulic conductivity curve cross is taken to be the "optimal" percentage of montmorillonite which gives highest attenuation and permeability.

For the heavy metals, for example Pb, even small amounts of clay gave almost total removal. The heavy metals, even though toxic, thus represent a minimal pollution hazard in municipal leachates because they are attenuated very strongly. Therefore, they can usually be ignored from consideration as far as determination of the optimal clay liner for a given leachate. At the other extreme are the relatively noninteracting constituents represented by Cl. Cl as shown in Figure 9 is relatively unattenuated by even large amounts of clay, which suggests that in order to prevent chloride migration, relatively impermeable clay liners would be necessary. Because of the nontoxic nature of the chloride ion, it also ranks low along with the heavy metals in the pollution "hazard index" (Table 4). In view of the problems associated with the "bathtub" effect, it seems unwise to design clay liners to optimize chloride attenuation. Rather, it seems prudent to design clay liners for optimum attenuation of the most hazardous constituents found in a particular

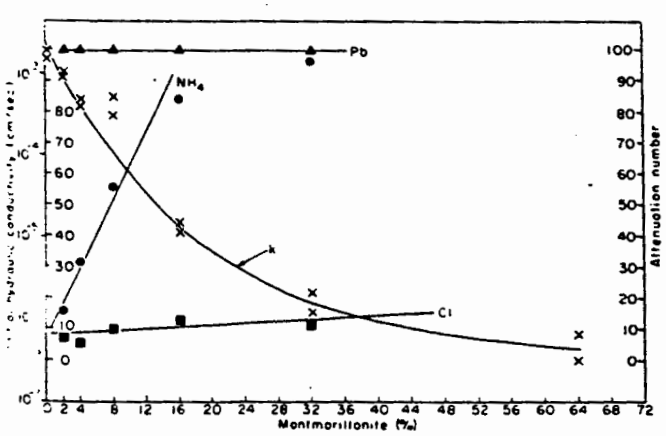

Fig. 9. "Optimal" montmorillonite percentages for attenuation of Pb, NH_4, and Cl, and the hydraulic conductivity (k) of the clay-sand mixtures.

Table 5. Estimated Thickness of Landfill Liner for Total Attenuation of Leachate for Each Meter Thickness of Overlying Refuse During a 20-Year Fill Life*

Constituent	Initial Concentration†		Cation exchange capacity (meq/100 g)					
			10		20		30	
	Ave. (ppm)	Max. (ppm)	Ave. (cm)	Max. (cm)	Ave. (cm)	Max. (cm)	Ave. (cm)	Max. (cm)
NH_4	379 (830)	1,106	32	92	16	46	11	31
Na	755 (740)	7,700	118	1,208	59	604	39	403
K	763 (530)	3,770	51	252	26	126	17	84
Mg	1,609 (240)	15,600	226	2,191	113	1,096	75	730

* Assumptions: Bulk density = 1.8 g/cc; 100 liters of leachate generated per m^3 of refuse per yr; initial concentration decreases linearly to zero at 20 yr; removal efficiencies for each constituent were estimated using the average values given by Griffin and Shimp (1976): NH_4 = 37.1%, Na = 15.4%, K = 38.2%, Mg = 29.3%.

† Concentrations taken from the 20 leachate analyses reported by EPA (1974); those in parentheses are the values of Old Du Page landfill leachate used in this study.

leachate. In the case of the Du Page leachate used in this study, the pollution hazard index ranks NH_4 as 30 times more of a pollution hazard than any other constituent found in this leachate. It therefore seems reasonable to design a clay liner for Du Page leachate that gives optimal attenuation of NH_4. With such a liner all the other constituents should also be attenuated to relatively safe levels for minimal pollution of the ground waters adjacent to the landfill site.

For the case of NH_4 (shown in Figure 9) the optimal attenuation is achieved by approximately 10 percent montmorillonite. If one extrapolates the curve, it is apparent that 18 to 20 percent montmorillonite would give nearly total removal of the NH_4 from the leachate, but would result in very low hydraulic conductivities. The ratio of the optimal percentage to the percentage necessary for nearly total removal indicates that if the liner with 10 percent of montmorillonite is doubled in thickness from the 40 cm used in this study to 80 cm, it will contain enough montmorillonite to give nearly total removal of the NH_4 in 10 pore volumes of leachate and will still retain the relatively high hydraulic conductivity of 6×10^{-5} cm/sec.

The thicknesses of mixtures of sand and clay of different cation exchange capacity (CEC) that would achieve total attenuation of selected relatively mobile ions are shown in Table 5. The removal efficiency will differ in leachates, depending upon the relative strength of ions. The efficiencies used in Table 5 are based on the Du Page leachate used in this study. There is direct relationship between the liner thickness and total attenuation; a thick liner with low CEC will have the same attenuation capacity as a thin liner with a high

CEC ∝ 1/K

CEC. Increasing cation exchange capacity generally reflects increasing clay content of the liner. Thus, a thicker liner with greater hydraulic conductivity nd lower CEC may be the optimal liner for attenuation.

Determining the release rate to aquifers or surface waters of contaminants that are not attenuated or are poorly attenuated by clay liners (natural or man-made) is necessary for proper design of landfills. In designing a landfill, a decision must be made to determine which ions should be totally attenuated and which ions eventually may be released to the environment. The chloride ion, which moves essentially unattenuated and is not noxious at low concentrations, is the most obvious example of the latter type.

The calculation of release rate of leachate from the bottom and sides of a landfill and the flow path it will take is a complex problem. At present, there are several models, existing and under development, which use the high-speed digital computer to predict the rate and path of fluids from a landfill (for example, Elzy *et al.*, 1974). These models are quite accurate and will provide very good estimates of the quantities desired. They are, of course, dependent upon the accuracy of the physical data input—infiltration rates, hydraulic conductivities, and geologic description.

Preliminary estimates of leakage can also be made by simply using Darcy's equation as suggested by Hughes *et al.* (1971):

$$Q = kA \frac{dH}{dL}$$

where:

Q = flow rate in cm^3/sec;

k = hydraulic conductivity;

A = area of the landfill in cm^2;

dL = thickness of the liner in cm;

dH = head of water across the liner in cm.

This equation is subject to some of the same limitations as the computer-solved models. Either representative or measured values of hydraulic conductivity can be used. However, the presence of a landfill generally changes the hydraulic gradient. In addition, the gradient will differ with changing hydraulic conductivities. Hughes *et al.* (1971) observed gradients as high as 1 cm/cm in glacial till with a hydraulic conductivity of about 4×10^{-7} cm/sec, and a 0.02 cm/cm gradient in dirty, surficial sand with a hydraulic conductivity of 1×10^{-3} cm/sec. Thus, one must estimate the gradient from experience.

The leakage thus calculated must then be compared in volume to the receiving waters. In the case of a surface-water body receiving the pollutant discharge, the calculation is relatively straightforward. For instance, Hughes *et al.* (1971) calculated that the Elgin landfill, located in highly permeable gravel, increased the total dissolved solids of the Fox River by 0.30 parts per million.

The leakage to aquifers is more difficult to estimate, and the computer models have considerable advantage. However, one may calculate the volume of water passing below the landfill in a simplistic way similar to the way the leakage was calculated; "A" in the equation becomes the cross-sectional area of the aquifer. However, ground-water flow is laminar, and dispersion becomes the only mechanism for dilution into the total volume of water; thus the calculation represents a total value for the receiving aquifer, and not a spot value. Nevertheless, these two simple calculations can be informative before one proceeds to more detailed hydrogeologic analyses.

Table 6 shows some representative estimates of increased ion concentration in some surface-water streams and aquifers. The Table was calculated assuming an aquifer 30 meters thick, a hydraulic conductivity of 1×10^{-2} cm/sec, and a small stream with a discharge of 1 cubic meter per second. The salinity of the discharge is assumed to be 2,000 ppm as found at the Elgin landfill by Hughes *et al.* (1971).

CONCLUSIONS

The results of this study raise some basic questions about landfill design and monitoring. The results indicate that use of hydraulic conductivity information and the pollution hazard

Table 6. Increase in Salinity of an Aquifer or Small Stream from Leakage of Landfill Leachate*

Liner (Hydraulic conductivity, cm/sec)	Increase in aquifer salinity (ppm)	Increase in stream salinity (ppm)
1×10^{-8}	0.8	0.002
1×10^{-6}	7.7	0.02
1×10^{-4}	77.	0.2
1×10^{-2}	769.	2.0

* Assumptions:

1)

	Hydraulic conductivity (cm/sec)	Gradient (cm/cm)
Aquifer	1×10^{-2}	1/1000
Liner	1×10^{-8}	1/1
Liner	1×10^{-6}	1/10
Liner	1×10^{-4}	1/100
Liner	1×10^{-2}	1/1000

2) 10^4 square meters of refuse
3) Aquifer 30 m thick
4) Leachate salinity 2000 ppm (Hughes et al., 1971)
5) Stream discharge 1 m^3/sec

ating for a given leachate or waste stream can allow a different approach to landfill design from the prevalent "containment" policy. The data suggest that over-all pollution would be decreased if landfill liners were designed to achieve higher permeability and to selectively attenuate the most toxic pollutants from the leachate. The ground waters could then dilute the nontoxic components, which can be tolerated at much higher concentrations without deleterious effects. Thus, stabilization and use of the landfill for other productive purposes can be achieved at much faster rates than can at present be achieved by containment liners.

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