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A design model for the treatment of landfill leachate with a microbially enriched soil and reed canarygrass.

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A DESIGN MODEL FOR THE TREATMENT OF LANDFILL LEACHATE WITH A MICROBIALLY ENRICHED SOIL AND REED CANARYGRASS

A Dissertation Presented

By

RONALD L. LAVIGNE

Submitted to the Graduate School of the
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of the requirements for the degree of

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Department of Plant and Soil Sciences

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DEDICATION

to

MY FAMILY

Christine

Michael, Michele, Eileen and Ron Jr.

MY PARENTS

Jeannette and Donald

MY GRANDPARENTS

When I was too young to walk you taught me how

When I stumbled from running to fast
you helped me up and soothed my pain

When I felt too tired to go on you carried me

The happiness, fullness and joy you have added
to my life will never be forgotten

Your "footprints" along the beach of my life
are etched forever in the sands of time

For all that you have been, are now and continue to be

This milestone is dedicated to you

With Love

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ABSTRACT

A DESIGN MODEL FOR THE TREATMENT OF LANDFILL LEACHATE WITH MICROBIALLY ENRICHED SOILS AND REED CANARYGRASS

MAY 1989

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The disposal of municipal solid waste continues to be one of the major environmental problems facing the world today. "Sanitary landfilling" became the accepted method of refuse disposal during the early 1970's, when open burning dumps, wind blown litter, flies and rodents were perceived to be the solid waste issue of the day. Little or no attention was given to the process of refuse decomposition and the liquid waste that is subsequently produced (i.e., leachate).

Today lined landfills are replacing older unlined facilities and the practice of collecting leachate has become commonplace. An appropriate technology to treat collected leachate, however, has yet to be developed. Current landfill designs generally require the utilization of municipal wastewater treatment facilities for leachate disposal; but this practice is costly and it may be environmentally less desirable.

This research project investigated the use of a new technique for treating landfill leachate on site. It utilizes a low technology "living filter" approach that models biological, chemical and physical processes known to be occurring in natural wetlands. The system takes advantage of the root zone aeration capabilities of reed canarygrass; and it maximizes the development of the fixed film biomass on peatmoss surfaces.

Aerobic and anaerobic environments within the treatment medium facilitate a rapid reduction of Total Organic Carbon (TOC) and Chemical Oxygen Demand (COD); which both exist at high concentrations in leachate. The environment is also conducive to the precipitation of leachate metals.

Peatmoss and reed canarygrass treatment beds were operated in both the batch and continuous flow mode to evaluate the reaction order and rate constants for leachate degradation. COD and TOC were used as modeling parameters. Mean hydraulic retention times of 3-10 days resulted in a 99%+ reduction in COD and TOC concentrations. Similar reductions were realized for heavy metals and total nitrogen. Grass clippings and peat samples were analyzed for Fe, Cu, Mn, Mg, P, K, and Ca before, during, and after 3 years of landfill leachate application. Leachate influent and effluent were also analyzed for the same metals. In each case, more than 99% of the cations measured were removed by the treatment system.

Data indicates that a root zone method using peatmoss and reed canarygrass can be an effective method for treating landfill leachate if unsaturated flow conditions are maintained.

Toxicity testing using reed canarygrass seedlings indicated that leachate is initially toxic, but with time plants are able to recover and flourish in the leachate-peatmoss environment.

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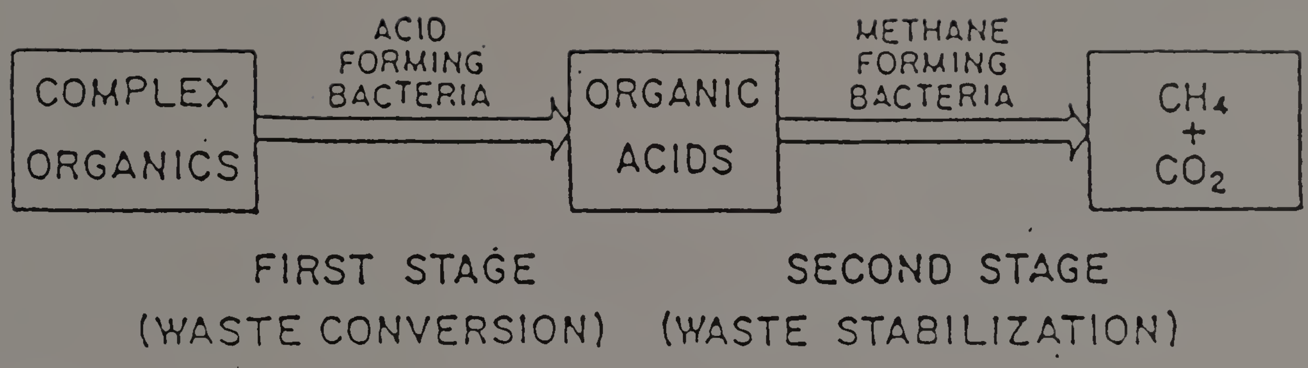
CHAPTER I

INTRODUCTION AND STATEMENT OF OBJECTIVES

When domestic, institutional, commercial, and industrial refuse are disposed of by sanitary landfilling, the encapsulated refuse quickly becomes anaerobic (1). This phenomenon is due to several factors. Firstly, the earthern cover on a landfill is poorly permeable to atmospheric O_2, especially since the primary driving force for gas exchange is diffusion from the atmosphere into the landfill interior. Secondly, refuse contains 60-75% biodegradable material, the principal constituent being paper (2). Aerobic bacterial respiration exhausts the limited O_2 supply within 30-90 days and atmospheric transfer cannot keep pace with the demand, thereby creating an optimum condition for anaerobiesis (3). Thirdly, warmth, moisture, and darkness also enhance the growth environment for anaerobes. Although many bacterial species are capable of first stage anaerobic metabolism, they have collectively been referred to as acid formers, and share the common trait of producing organic acids as their metabolic waste products (4). Acetic acid and proprionic acid represent the largest percentage of the total metabolite produced (5) (Figure 1-1). Due to the strong reducing nature of this volatile organic waste, metals and other ionic materials readily become mobile (6). This potpourri of organic and inorganic material has



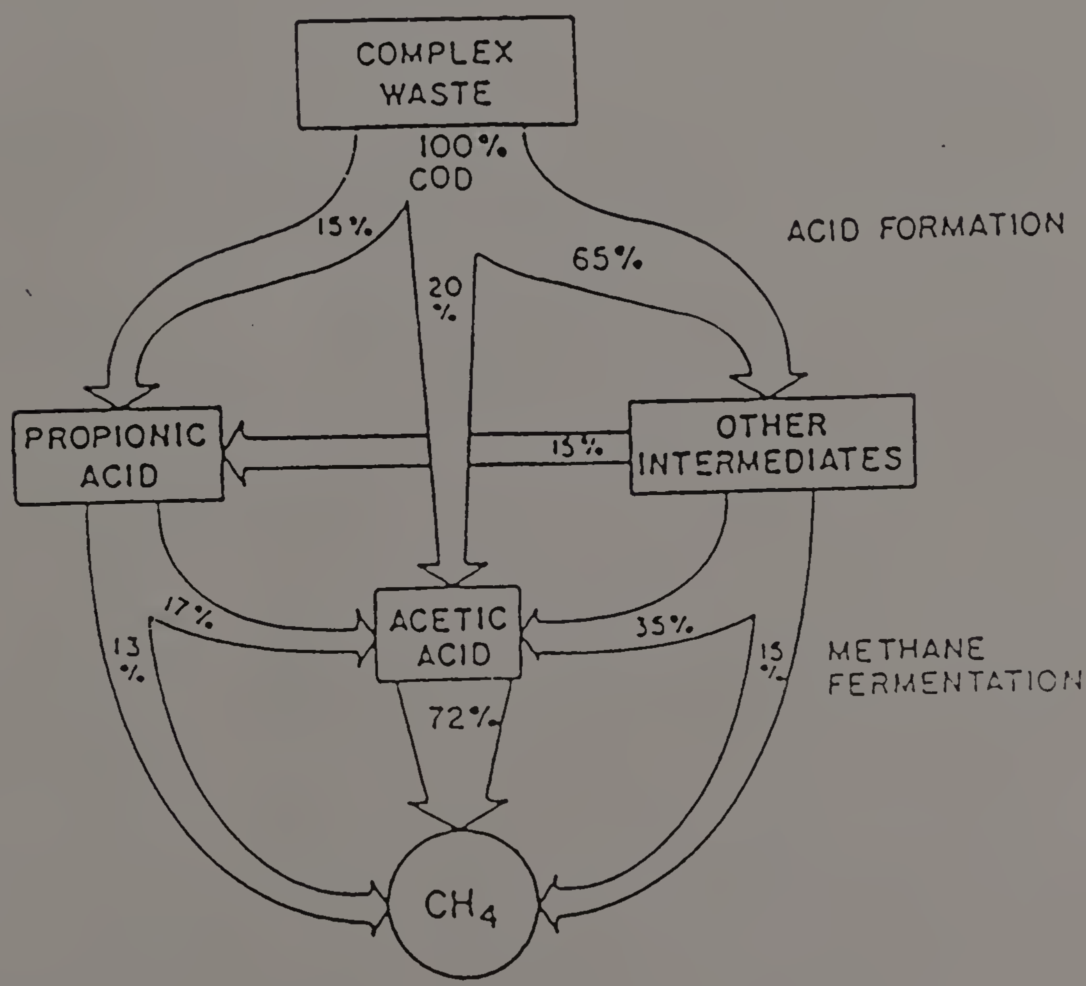


Figure 1-1. Pathways in Methane Fermentation of Complex Wastes Such as Municipal Waste Sludges. Percentages Represent Conversion of Waste COD by Various Routes. (After MaCarthy (5))

been traditionally referred to as landfill leachate. Over the years research has shown that virtually any waste material disposed of in a landfill will ultimately become part of the leachate flow stream. Other leachate pollutants include such materials as pathogenic bacteria, viruses, pesticides, hospital wastes, industrial and wastewater sludges, solvents, and hazardous wastes (7).

To what extent leachate is able to leave a disposal site by groundwater or surface water transport has been a point of interest for many years. Evidence strongly suggests that in arid or semiarid regions the transport rates are extremely slow (8). Conversely, in temperate areas such as New England and the Northeast in general, where annual precipitation exceeds 100 cm (40 inches), the movement of leachate is much more rapid. To prevent groundwater contamination design criteria for landfills usually encourage construction in soils rich in clay, located well above the groundwater table and situated some safe distance from drinking and surface water resources (9). They also include the use of impermeable materials for cover, and strongly urge that groundwater monitoring wells be placed around the site.

In spite of these environmental precautions, leachate problems pervade many landfill sites in the Northeast resulting in considerable contamination to surface and groundwater resources.

More recently, attention has been directed toward the concept of lining landfills with impermeable membranes and then treating collected leachate either off-site or on-site. Off-site treatment usually involves the piping of leachate to a nearby sewer, and combining it with the municipal sanitary sewage. This methodology requires that the community has an accessible treatment facility.

Research suggests that leachate is biologically treatable by conventional activated sludge processes if it represents less than 5% of the total wastewater flow (10), but questions have been raised relative to the treatability of nondegradable constituents in leachate that may escape the secondary treatment process (11). For large landfills on-site secondary treatment with package plants has also been tried with limited success (12). More recently increased attention has been directed toward low technology living filter type treatment systems (13, 14, 15, 16, 17). In Barre, Massachusetts, a series of oxidation ponds operated in a batch mode appears to have effectively treated collected leachate since 1975 (13). Menser et al. experienced similar treatment success by spraying pretreated leachate into deciduous forest stands in Princeton, West Virginia (17). In each case, capital and energy costs were minimal, and little or no negative environmental impact was detected.

The treatment facilities in Barre, Massachusetts, and Princeton, West Virginia were both able to achieve a 99+% removal rate for virtually all leachate pollutants. In spite of their overall successes, both projects experienced the following shortcomings.

1. The mechanisms and rates of pollutant removal were unknown, and as such the treatment processes remained somewhat of a "black box," with operating rules of thumb based on heuristic data and experiences.

2. Large amounts of land area were required for the treatment facilities. In Barre approximately 25% of the landfill area had to be set aside for treatment lagoons. During the first year of operation the Princeton project utilized 2.2 hectares (5.0 acres) of forest for leachate irrigation, and the area requirements for treatment increased during succeeding years.
3. Leachate standing in open lagoons and storage basins created odor and insect problems at each site. Their isolated locations minimized objections from the public but most landfills would not be as ideally located.

The research described herein attempted to utilize the benefits of "low technology" treatment, and it also sought to eliminate some of the problems associated with the earlier methodologies described.

In this project a design model for the treatment of sanitary landfill leachate was developed utilizing reed canarygrass growing in an organically rich soil as the treatment medium. It was assumed that microbial growth and metabolism within the soil would follow the Michaelis-Menten and Monod models for enzyme mediated biological reactions. It was also proposed that a leachate treatment design model could be developed by modifying batch and plug flow reactor models in a manner that would account for unsaturated flow in a porous biologically enriched treatment media.

The project evaluated modeling parameters using bench top greenhouse growth trays. Influent and effluent Total Organic Carbon (TOC) and Chemical Oxygen Demand (COD) changes were used to model biological activity. Sources and sinks of nitrogen and heavy metals were also monitored at regular intervals in the raw leachate, soil, reed canarygrass, and effluent. Of particular importance to the peatmoss-reed canarygrass method is the utilization of "fixed film" technology. Traditional treatment systems are generally limited by their inability to produce and sustain a large biomass. By providing an increased surface area for microbial attachment, biomass to substrate ratios can be marketly enhanced, resulting in significant reductions in detention times and related area needs.

From a practical point of view this research suggests that treatment times and areas can be reduced from months and hectares to days and square meters.

The specific objectives of this project were to:

1. Eliminate the "black box" nature of previous natural treatment systems by assuming and testing whether or not first-order microbial kinetics could be used to model leachate treatment.
2. Eliminate the need of standing bodies of leachate in ponds or lagoons; and to maintain a continuous and secured unsaturated hydraulic regime throughout the treatment process.
3. Reduce the amount of area and time required for acceptable removal rates through the utilization of "fixed film" techniques.
4. Evaluate the long term capabilities of peatmoss and reed canarygrass to effectively treat landfill leachate so that on

site disposal of treated effluent could be practiced without a significant impact on the environment or public health.

5. Evaluate toxicity problems known to exist with most landfill leachates.

CHAPTER II

LITERATURE REVIEW

An Overview of Solid Waste Disposal by Landfilling

Landfill Operation Regulations

On April 22, 1970, the United States celebrated the first in a series of "Earth Days," directed towards the improved management of the earth's resources. It was evident from the start that this movement constituted a mandate from a concerned public to governmental agencies at all levels (18). The mandate seemed rather clear and straightforward. There was a need for a comprehensive set of laws and regulations that would prohibit the pollution of land, air, and water. More importantly, federal monies and enforcement efforts were to provide the muscle needed to insure compliance.

Political survival during the early 1970's required lawmakers to sponsor and support environmental legislation at all levels of government. The ability of public pressure to impact the law-making process resulted in the scores of environmental laws that were enacted between 1970 and 1976. Unfortunately, many of these early laws had major shortcomings. First and foremost, the laws were seldom based on a sound scientific understanding of the ecological problems involved; and secondly, compliance processes and procedures usually resulted in the land, (i.e., the soil) serving as the final repository for "waste" materials.

It wasn't until 1976 that the federal government took action to correct the gross oversight of our nation's land disposal practices (19). The Resource Conservation and Recovery Act (RCRA PL 94-580) now prohibits uncontrolled "open dumping," but for millions of acres of land, the law has done too little too late. Even through the early 1980's, hazardous wastes continued to be disposed of in unlined landfills. To a somewhat lesser degree, co-disposal of hazardous waste continues today.

In 1971, the Massachusetts Department of Public Health promulgated Regulations for the Disposal of Solid Wastes by Sanitary Landfilling (20). When solid waste jurisdiction was transferred to the Department of Environmental Quality Engineering (DEQE) in the mid 1970's these minimal regulations were also transferred and remain in effect today. For the most part, they are still in their original form. It should be noted that Massachusetts was one of the first states to promulgate landfilling regulations, and therefore served as a reference for many other states (i.e., there is a close similarity among most landfilling regulations).

Generally, landfills are operated in one of two ways: The "Area Method" (Figure 2-1) utilizes large gravel pits, natural depressions, or embankments as repositories for refuse. The objective is to fill the area to original or near level topography so that the surface area can be used for parking, recreation fields, parks, etc. In some cases,

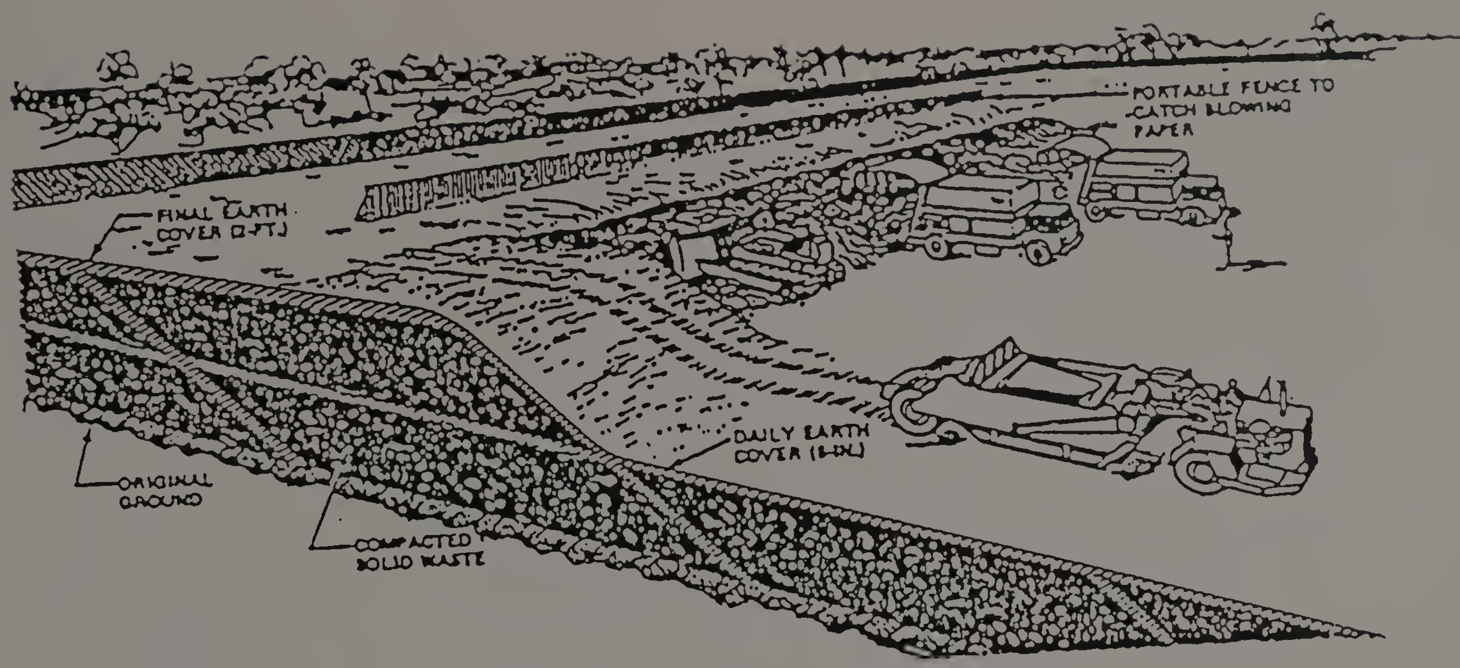


Figure 2-1. Area Method of Sanitary Landfilling. (21)

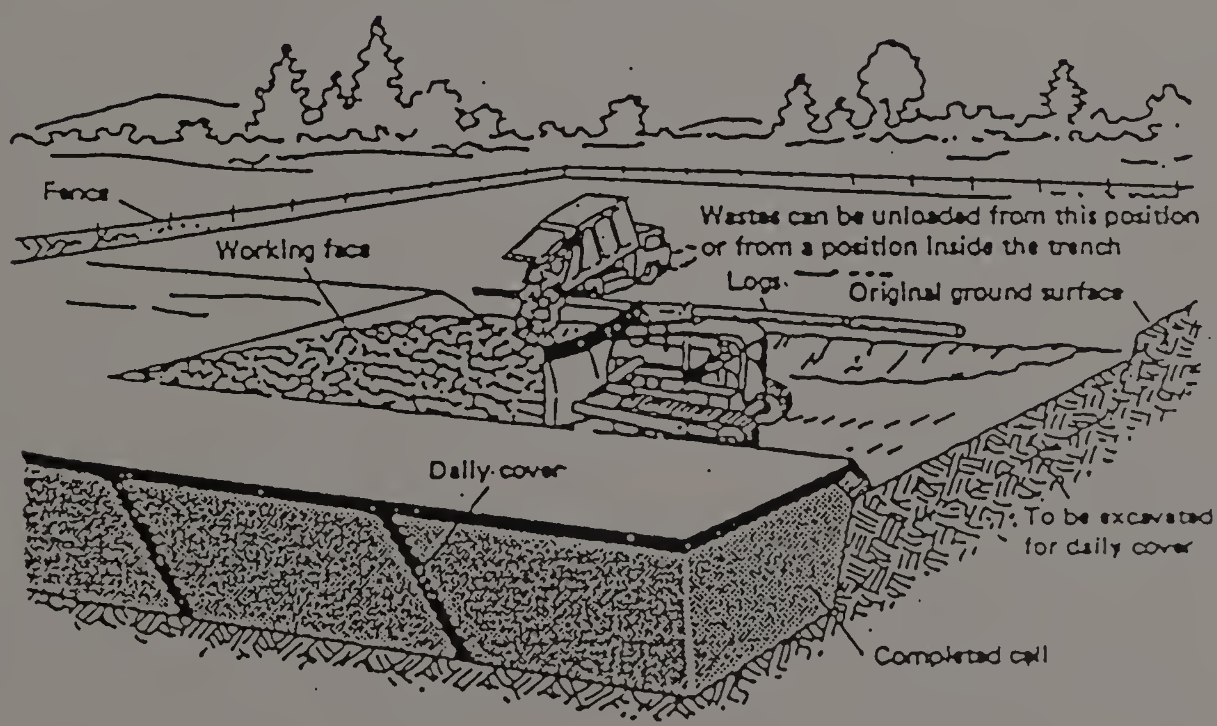


Figure 2-2. Trench Method of Sanitary Landfilling. (21)

buildings have been constructed over old fills with a fair degree of success and minimal settling problems. The "Trench Method" (Figure 2-2) is usually employed on more level land. Long trenches typically 30.5 meters (100 feet) wide and 6.1 meters (20 feet) or more deep are excavated into the ground. Filling starts at one end of the trench and moves toward the other.

In both methods it is required that refuse be compacted into 1.8-2.5 meters (6-8 feet) "lifts," and that it be covered with a minimum of 15 cm (6 inches) of soil material at the end of each day's operation. The daily sections of refuse (cells) are theoretically "fire breaks" and therefore minimize the chance of fire.

In both methods completed "sections" or "areas" are covered with less permeable soils, graded to induce runoff, loamed, and seeded. Refuse, at its lowest depth, must be at least 1.2 meters (4 feet) above the water table, and wetlands are generally excluded as disposal sites (20).

Other operational provisions often prohibit automobile tires, and require that appliances and brush be disposed of in separate sections. As previously mentioned the disposal of "special wastes" such as sludges, septage, hospital wastes, dead animals, etc., is often permitted in the regular refuse area.

Design shortcomings have often included the following:

1. Little or no evaluation of soil properties other than drainage or permeability.

2. Little or no modeling of groundwater flow and potential contaminant migration.

3. Few, if any, monitoring wells.

4. Little or no provision for dealing with groundwater pollution if it does occur.

It should be noted that virtually all landfill designs are based on the assumption that proper operation will minimize leachate production by minimizing water infiltration. Experience has shown both of these assumptions to be inaccurate from a practical point of view. One must bear in mind that landfills are operated by refuse handlers, not engineers. There are no qualifications for being a "landfill operator" other than having an ability to operate heavy equipment. Landfills seldom have impermeable soils on site, and past earth-moving experiences generally encourage the use of more permeable soils that are easily excavated with a minimum amount of wear and tear on equipment. Soils with high percentages of fine textured materials also create operational problems during wet periods, and this represents another reason for avoiding their use. The net effect here is that runoff designs are seldom implemented fully, allowing rain to percolate into the refuse, thereby producing leachate flows that often reach or exceed 27.4 m^3/day/hectare (3000 gallons/day/acre).

During the mid 1970's, the Massachusetts Division of Water Pollution Control (MDWPC) participated in developing Massachusetts' first lined landfill (22). The 1.2 hectare (3 acre) research facility was constructed at a private landfill site in Barre, Massachusetts.

In addition to having leachate collection, capabilities, the project also incorporated the States first "living filter" treatment system for landfill leachate. Since that time DEQE has used its regulatory and site review authority to require liners at virtually all new landfill sites. Most states now have similar requirements, but billions of tons of refuse still occupy older sites where open dumping and wetlands filling perpetuated themselves for many decades. Leachate production and groundwater pollution are likely to be associated with these sites for many decades to come.

Refuse Characterization and Quantity

It has been estimated that the United States produces approximately 4.1 billion metric tons of solid waste per year and that of this municipal solid waste (MSW) represents approximately 227 million metric tons (23).

MSW is generally assumed to include residential, commercial, and institutional sources, and as such, is the major waste type received at most landfills. MSW can also include sewage sludge, special wastes, yard wastes, and some demolition wastes. Many landfills have also accepted industrial and hazardous wastes, so that virtually any waste material must be considered as a possible contributor to the composition of landfill leachate. Tables 2-1 and 2-2 summarize typical sources and composition of MSW, in the U.S. and Tables 2-3 and 2-4 outline urban sources, and MSW composition. Tables 2-5 and 2-6 give typical "as discarded" densities and moisture contents for these wastes. Table 2-7 provides approximate percent elemental compositions in the degradable

Table 2-1. Typical Solid Waste Generating Facilities, Activities, and Locations Associated With Various Source Classifications.

Source	Typical facilities, activities, or locations where wastes are generated	Types of solid wastes
Residential	Single-family and multifamily dwellings, low-, medium-, and high rise apartments, etc.	Food wastes, rubbish, ashes, special wastes
Commercial	Stores, restaurants, markets, office buildings, hotels, motels, print shops, auto repair shops, medical facilities and institutions, etc.	Food wastes, rubbish, ashes, demolition and construction wastes, special wastes, occasionally hazardous wastes
Municipal*	As above*	As above*
Industrial	Construction, fabrication, light and heavy manufacturing, refineries, chemical plants, lumbering, mining, power plants, demolition, etc.	Food wastes, rubbish, ashes, demolition and construction wastes, special wastes, hazardous wastes
Open areas	Streets, alleys, parks, vacant lots, playgrounds, beaches, highways, recreational areas, etc.	Special wastes, rubbish
Treatment plant sites	Water, waste water, and industrial treatment processes, etc.	Treatment plant wastes, principally composed of residual sludges
Agricultural	Field and row crops, orchards, vineyards, dairies, feedlots, farms, etc.	Spoiled food wastes, agricultural wastes, rubbish, hazardous wastes

*The term municipal normally is assumed to include both the residential and commercial solid wastes generated in the community.
(After Tchobanoglous (23)

Table 2-2. Typical Physical Composition of Municipal Solid Wastes.

	Percent by weight			
Component	Range	Typical	Packaging materials	Davis, California
Food wastes	6-26	15	-	9.5
Paper	25-45	40	55.8	43.1
Cardboard	3-15	4		6.5
Plastics	2-8	3	3.6	1.8
Textiles	0-4	2	0.4	0.2
Rubber	0-2	0.5	-	0.8
Leather	0-2	0.5	-	0.7
Garden trimmings	0-20	12	-	14.3
Wood	1-4	2	7.8	3.5
Glass	4-16	8	18.1	7.5
Tin cans	2-8	6	14.3	5.2
Nonferrous metals	0-1	1	-	1.5
Ferrous metals	1-4	2	-	4.3
Dirt, ashes, brick, etc.	0-10	4	-	1.1

Based on measurements made over a 5-yr period (1971 to 1975).
(After Tchobanoglous (23))

Table 2-3. Average Per Capita Quantities of Solid Wastes Collected From Urban Sources in the United States, 1968*.

Source	lb/capita/day
Combined residential and commercial	4.29
Industrial	1.90
Institutional	0.16
Demolition and construction	0.72
Street and alley cleanings	0.25
Tree and landscaping	0.18
Park and beach	0.15
Catch basin	0.04
Sewage treatment plant solids	0.50
Total	8.19

*The corresponding total per capita quantities for all areas (7.92 lb/capita/day) are somewhat lower than those from urban areas.
Note: lb/capita/day x 0.4536 - kg/capita/day
(After Tchobanoglous (23))

Table 2-4. Components of Municipal Solid Wastes Generated in the United States, 1971.

Component	Total generated: Tons, millions	Total generated: Percent	Total disposed: Tons, millions	Total disposed: Percent
Paper	39.1	31.3	47.3	37.8
Glass	12.1	9.7	12.5	10.0
Metal	11.9	9.5	12.6	10.1
Ferrous	10.6	8.5	-	-
Aluminum	0.8	0.6	-	-
Other nonferrous	0.5	0.4	-	-
Plastic	4.2	3.4	4.7	3.8
Rubber and leather	3.3	2.6	3.4	2.7
Textiles	1.8	1.4	2.0	1.6
Wood	4.6	3.7	4.6	3.7
Food	22.0	17.6	17.7	14.2
Subtotal	99.0	79.2	104.8	83.9
Yard wastes	24.1	19.3	18.2	14.6
Miscellaneous inorganics	1.9	1.5	2.0	1.5
Total	125.0	100.0	125.0	100.0

Note: tons x 907.2 = kg
(After Tchobanoglous (23))

Table 2-5. Typical Densities of Municipal Solid Waste Components as Discarded*.

Components	Density, lb/ft^3 Range	Density, lb/ft^3 Typical
Food wastes	8-30	18.0
Paper	2-8	5.1
Cardboard	2-5	3.1
Plastics	2-8	4
Textiles	2-6	4
Rubber	6-12	8
Leather	6-16	10
Garden trimmings	4-14	6.5
Wood	8-20	15.0
Glass	10-30	12.1
Tin cans	3-10	5.5
Nonferrous metals	4-15	10.0
Ferrous metals	8-70	20
Dirt, ashes, brick, etc.	20-60	30

*Uncompacted.
Based on measurements made over a 5-yr period (1971 to 1975) at Davis, California.
Note: $lb/ft^3 \times 16.019 = kg/m^3$
(After Tchobanoglous (23))

Table 2-6. Typical Data on Moisture Content of Municipal Solid Waste Components.

Components	Moisture, percent	
	Range	Typical
Food wastes	50-80	70
Paper	4-10	6
Cardboard	4-8	5
Plastics	1-4	2
Textiles	6-15	10
Rubber	1-4	2
Leather	8-12	10
Garden trimmings	30-80	60
Wood	15-40	20
Glass	1-4	2
Tin cans	2-4	3
Nonferrous metals	2-4	2
Ferrous metals	2-6	3
Dirt, ashes, brick, etc.	6-12	8
Municipal solid wastes	15-40	20

(After Tchbanoglous (23))

Table 2-7. Typical Data on Total Analysis of the Combustible Components in Municipal Solid Wastes.

	Percent by weight (dry basis)					
Component	Carbon	Hydrogen	Oxygen	Nitrogen	Sulfur	Ash
Food wastes	48.0	6.4	37.6	2.6	0.4	5.0
Paper	43.5	6.0	44.0	0.3	0.2	6.0
Cardboard	44.0	5.9	44.6	0.3	0.2	5.0
Plastic	60.0	7.2	22.8	-	-	10.0
Textiles	55.0	6.6	31.2	4.6	0.15	2.5
Rubber	78.0	10.0	-	2.0	-	10.0
Leather	60.0	8.0	11.6	10.0	0.4	10.0
Garden trimmings	47.8	6.0	38.0	3.4	0.3	4.5
Wood	49.5	6.0	42.7	0.2	0.1	1.5
Dirt, ashes, brick, etc.	26.3	3.0	2.0	0.5	0.2	68.0

(After Tchbanoglous (23))

waste fraction, and it is from these materials that anaerobic landfill microbes ultimately derive their energy. It is the degradable fraction that also accounts for the array of metabolic intermediate materials (e.g., organic acids and alcohols) that are incorporated into a leachate's composition.

For many years it has also been a common practice to co-dispose of sludge and various liquid wastes with MSW. In landfills of this type the leachate produced generally takes on different characteristics. Wastewater sludge generally, accelerates refuse decomposition, and it increases nitrogen concentration within the leachate (24).

Liquid wastes, if toxic, can inhibit microbial growth (25) and they often become part of the sites general leachate composition (26). Most landfills and landfill operating regulations also identify certain waste types as special wastes that require some degree of special handling. Typical examples would include asbestos, hospital wastes, dead animals, or ash. The effect of a special waste on leachate quality could be very little, as with asbestos, or very significant, as with a "fly ash" containing substantial amounts of heavy metals. If a special waste imparts toxic properties to the leachate, there could be negative impacts to treatment systems, the environment, and the public health. The leachate selected for use in this study originated from a landfill known to be relatively free of materials such as biocides, heavy metals and toxic organics that could inhibit microbial degredation.

<u>Leachate Quantity and Quality</u>

A design for any wastewater treatment system must consider the nature and amount of material to be treated. In the case of landfill leachate, the quality and quantity produced can be highly variable (27, 28, 29, 30, 31). Some of the variability is clearly due to the lack of hydraulic controls at most landfills. Leachate sampling sites have traditionally been associated with potential pollution sites. Contaminated wetlands, brooks, small surface empoundments, and leachate "springs" are typical examples. Groundwater monitoring wells have also been used, but they generally provide data indicative of considerable groundwater dilution. When landfill sites are underlain by highly permeable unsaturated soils, the vertical downward movement of leachate has often been interpreted as not producing leachate at all.

Annual precipitation patterns also affect the quality and quantity of leachate produced (32, 33). Since the promulgation of the Resource Conservation and Recovery Act (RCRA) (34) in 1976, many more landfills have been constructed with impermeable liners, and leachate collection systems. The hydraulic controls associated with lined landfills clearly provide more reliable estimates of leachate quantity and chemical composition. The use of collection liners and discharge pipes also facilitate the application of mass balance theory to precipitation data and the various chemical parameters associated with leachate composition.

In conjunction with Figure 2-3, it can be seen that the major source of water entering a landfill site is precipitation. This can

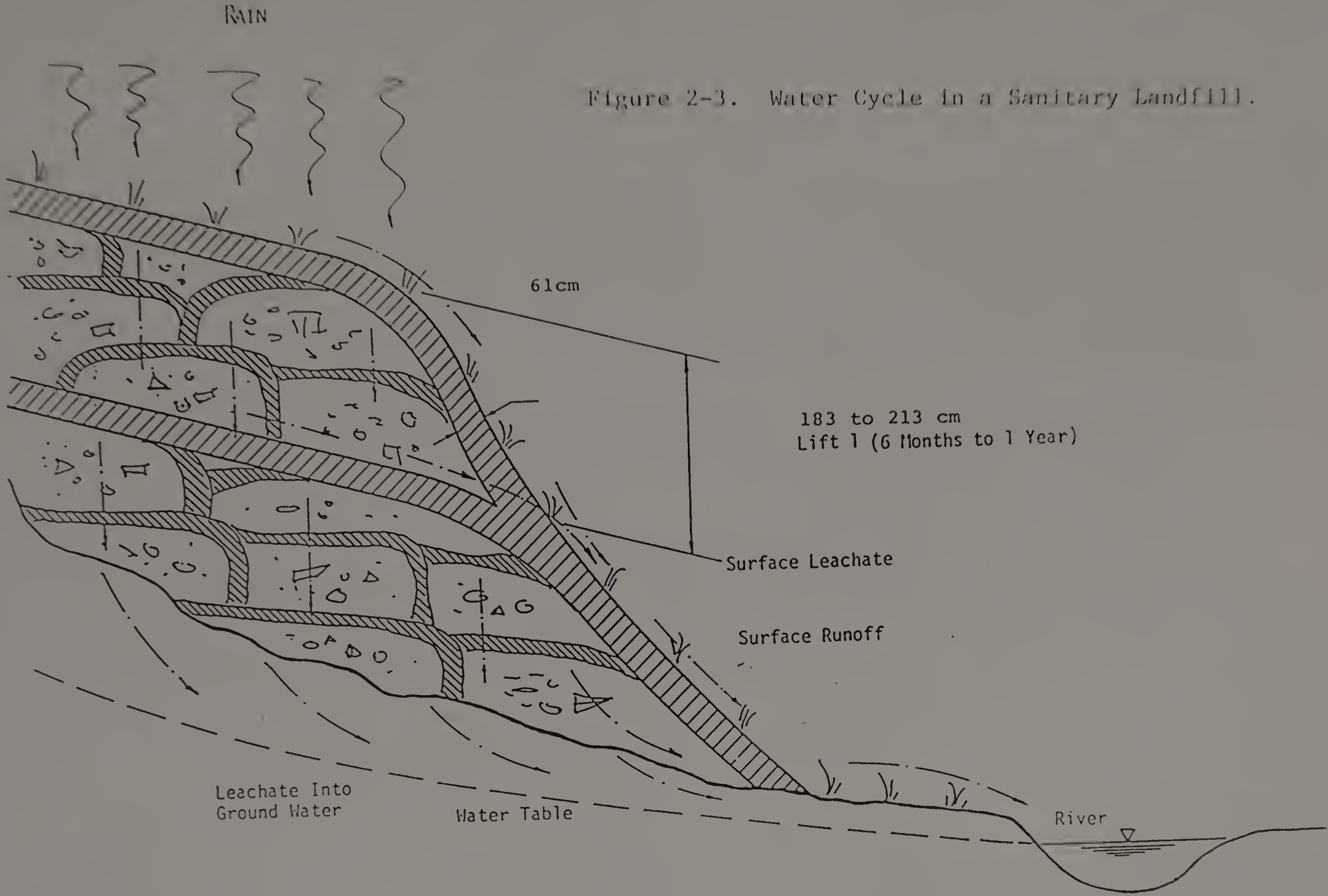


Figure 2-3. Water Cycle in a Sanitary Landfill.

come as rain, sleet, or snow, depending on the season of the year, and the geographic area being considered. Once on the land, the water can either run off, or infiltrate. Chow et al. (35, 36, 37) showed that in temperate regions, such as New England, evaporation constitutes a fairly small loss. Considering that most landfill areas are without vegetation, losses due to transpiration also may be neglected for the fill area.

Southern New England typically receives about 100 cm (40 inches) of precipitation annually. When this rain or snow falls on a landfill, the fractional part that percolates through the entire fill can vary considerably. For the lift being used at the time, it is reasonable to assume 100 percent infiltration. If the landfill site is lacking in impervious material for the intermediate cover, infiltration for the entire site will approach the 100 percent value (38). Oftentimes the use of impervious materials is avoided by the landfill operator because of the muddy working conditions associated with them during spring thaws and heavy rains. It seems worth emphasizing here, that though most codes call for impervious cover at some point in the landfilling operational scheme, more often than not it is unavailable at the site, or the operator avoids using it. Even if it is available, and used, the active fill area in every landfill is still subject to 100 percent infiltration until that lift or trench is sealed.

Under certain conditions, it is possible to reduce infiltration. If old lifts are sloped and covered with impervious fill, the percent of water run off increases. The planting of grass or trees on these areas will induce transpiration. During winter months, unused sections of the landfill freeze and this provides an impervious barrier to winter rains and snow melt. Plowing snow from the fill area will also reduce infiltration during the spring.

If one assumes 100 percent infiltration as a maximum limit for the amount of water added to refuse from precipitation, a one hectare fill area would receive 10,000 m^3/year (1.14 MGY) or 27.4 m^3/day/ha (3000 gal/day/acre). Once field capacity is reached, this figure might be interpreted as a maximum possible daily leachate flow from a hectare of landfill. If this figure is compared to measured flows from lined landfills with hydraulic smoothing (e.g., Amherst, Greenfield, Barre and Lowell, MA) the values are quite similar, and precipitation inflow is approximately equal to leachate outflow. Following the assumption that some 100 cm (40 inches) of water can infiltrate a landfill annually, some consideration must be given to water-refuse chemistry. For ease of discussion, landfill chemistry might be divided into three rather distinct stages.

<u>Chemistry of young leachates (approximately 0-2 months)</u>

During the month or two immediately following placement of new refuse, biological degradation is primarily aerobic. This being the case, metabolic by-products of the bacteria involved are mostly carbon dioxide and water. Soluble wastes will dissolve in the infiltrating water, and colloidal bacteria from animal and human feces will also be suspended. With adequate rain, leachate will flow and its composition will be quite representative of the parameters described. The following equation generally describes this early landfill chemistry.

$$\begin{matrix}\text{Paper} & & & & & & & & & \\ \text{Garbage} & & \text{Aerobic} & & & & & & & \\ \text{Feces} & + & \text{Bacteria} & + & O_2 & = & CO_2 & + H_2O & + & \text{Minerals} \\ \text{Vegetable Debris} & & & & & & & & & \text{New Cells} \\ & & & & & & & & & \text{"Wash Out"} \\ & & & & & & & & & \text{Pathogenic} \\ & & & & & & & & & \text{Organisms}\end{matrix} \qquad \text{(eq 2-1)}$$

<u>Chemistry of mature leachate (approximately 2 months-5 years)</u>

After a brief period of aerobic degradation, oxygen supplies go to zero and decomposition becomes anaerobic (39). It is during this stage that organic metabolites reach their peak. Without oxygen, a potpouri of organic acids and alcohols are produced. Under proper environmental conditions, these organic intermediates may be further oxidized to methane gas. This is especially true if wastewater treatment plant sludge is disposed of in the landfill (40). The following equation illustrates these anaerobic processes:

$$\begin{matrix}\text{Fats} & & \\ \text{Carbohydrates} + H_2O & = & \text{organic acids} + \text{alcohols} + CO_2 \\ \text{Protein} & & \end{matrix}$$

$$\begin{matrix}\text{Secondary Anaerobic Products} \\ \text{from Organic Intermediates}\end{matrix} = CH4 + CO2 \qquad \text{(eq 2-2)}$$

It is the organic putriscribe mentioned above that gives leachate its characteristic odor. During this period some reduction will also occur, resulting in the addition of metal cations to the leachate. When thin-walled containers corrode completely through, soluble wastes similar to those described for the "young landfill" will also be added to the leachate. Generally, there will be few enteric organisms leaching during this period. Most will have washed out earlier, and data seems to indicate that leachate generated during this period is toxic or inhibitory to many bacterial species (41). This problem is discussed more thoroughly in Chapter VI.

Chemistry of old leachates (approximately 5 years to 100+ years)

Sooner or later, the supply of biodegradables is exhausted within a landfill cell, and fermentation processes come to an end. This older leachate appears clearer and the repulsive odor abates. To the casual eye it would appear that the major pollution potential is over. Unfortunately this may not be the case. With adequate supplies of metals, reduced cations can flow for decades after a landfill area has closed. Hazardous wastes buried in more resistant containers (e.g., 55 gallon drums) may eventually be released as corrosive processes continue with time. If large slugs of special wastes have been landfilled, such as chromium, cyanide, PCBs or pesticides, serious damage to surface and groundwater resources can occur many years after their placement into unlined facilities (42).

In summary, it seems reasonable to conclude that a typical landfill could produce as much as 27 m^3/hectare/day (3000 gal/acre/day) of leachate and that leachate composition will vary considerably as the disposal site ages. Table 2-8 illustrates the wide range of variability in concentration that can occur when aging and dilution are involved.

Table 2-8. Characterization of Leachate from Different Sources.

Source / Parameter mg/l	Range of values from Garland and Mosher (4)	Blackwell Forest Leachate Data from Huges (2)	DuPage Leachate Used	Barre "Batch C" Super Funnel (no dilution)	Barre Batch A 1/6/76 thru ice leachate pool	Amherst Leachate exit manhole
pH	3.7-8.5	7.10	6.79	6.22	5.50	6.6
Alkalinity as $CaCO_3$	0 - 20,850	3,255	4,220	4,150	2,100	6,500
COD	40 - 89,520	39,680	1,362	13,534	11,100	10,800
TOC	256 - 28,000	-	-	4,675	ND	6,150
BOD	9 - 54,610	54,610	-	-	-	11,700
SO_4	1 - 1,826	680	<0.01	ND	128	19.2
Cl	34 - 2,800	1,697	1,070	ND	ND	700
Fe	0 - 5,500	5,500	4.40	1,095	1,020	550
Mn	0 - 1,400	1.66	<0.1	22.2	32.5	85
Ca	5 - 4,080	-	49.	778	680	600
Mg	16 - 15,600	-	204.	117	173	150
Cu	0 - 10	0.05	<0.1	-	2.65	15.9
Zn	0 - 1,000	-	0.03	-	0.71	1.35
NH_3	0 - 1,106	-	809	378	225	600
TS	0 - 59,200	-	-	-	-	4,300
Total P	0 - 154	6	<0.1	-	-	-

Considering that landfills continue to produce a variable leachate stream for years after closing, even those with liners require treatment technologies that are capable of dealing with a wide variety of pollutants and concentrations.

When discharge permits, fix maximum permissible concentrations at particular values, and when biological treatment methodologies are being considered as the treatment choice, it is generally difficult to select appropriate detention times based on microbial kinetics.

A Historical Overview of Leachate Treatment Technology

Natural Attenuation Potential of Soils

Prior to the advent of lined landfill technology, it was generally assumed that natural soils below a refuse fill would attenuate or "filter" any pollutants that might leach out. This philosophy is clearly evidenced by the four foot separation requirement in Massachusetts' regulations (20).

From a purely qualitative point of view soils do have, to varying degrees, significant treatment potentials that can be summarized as follows:

(a) Convective Dispersive Transport:

As a liquid waste moves through an unsaturated soil matrix, there is a natural tendency for it to be dispersed over larger, and larger areas. A broadening plume logically exhibits a decrease in concentration along its center line, and in time or distance, concentrations normal to the center line approach ambient conditions.

If dispersion is due principally to molecular activity or movement, the attenuative processes can be described by Fick's first and second law (43). If convective dispersion also contributes to the attenuation process then a transport model including both convection and diffusion can be used to model leachate movement and dilution as it moves from a point of origin (44).

(b) Physiochemical Processes:

Many of the contaminants transported in leachate are non-conservative in a soil media. Physicochemical processes such as ion exchange, absorption, adsorption, sieving, and reaction, all contribute to time and space factors that alter leachate composition and concentration. These changes have been described and modeled extensively by groundwater scientists (45, 46, 47, 48).

Tirsch and Jennings (49) concluded from column studies that natural soils underlying a private landfill in Barre, MA provided virtually no alteration of gross ionic strength or pollution hazard when leachate was allowed to percolate through 1.2 m (4 ft) of the material. Work by Griffen et al. (50, 51, 52, 53, 54) also showed little attenuation of organics. For this reason conservative transport models are generally used to establish the bounds of contamination potential for organic constituents in leachate. Breakthrough curves for COD as reported by Tirsch and Jennings (49) (Figures 2-4 and 2-5) illustrate the relatively conservative nature of organically rich leachate under both unsaturated and saturated flow regimes. Effluent concentrations for naturally occurring exchange species such as Ca^{2+} and Mn^{2+} often emerge as a front

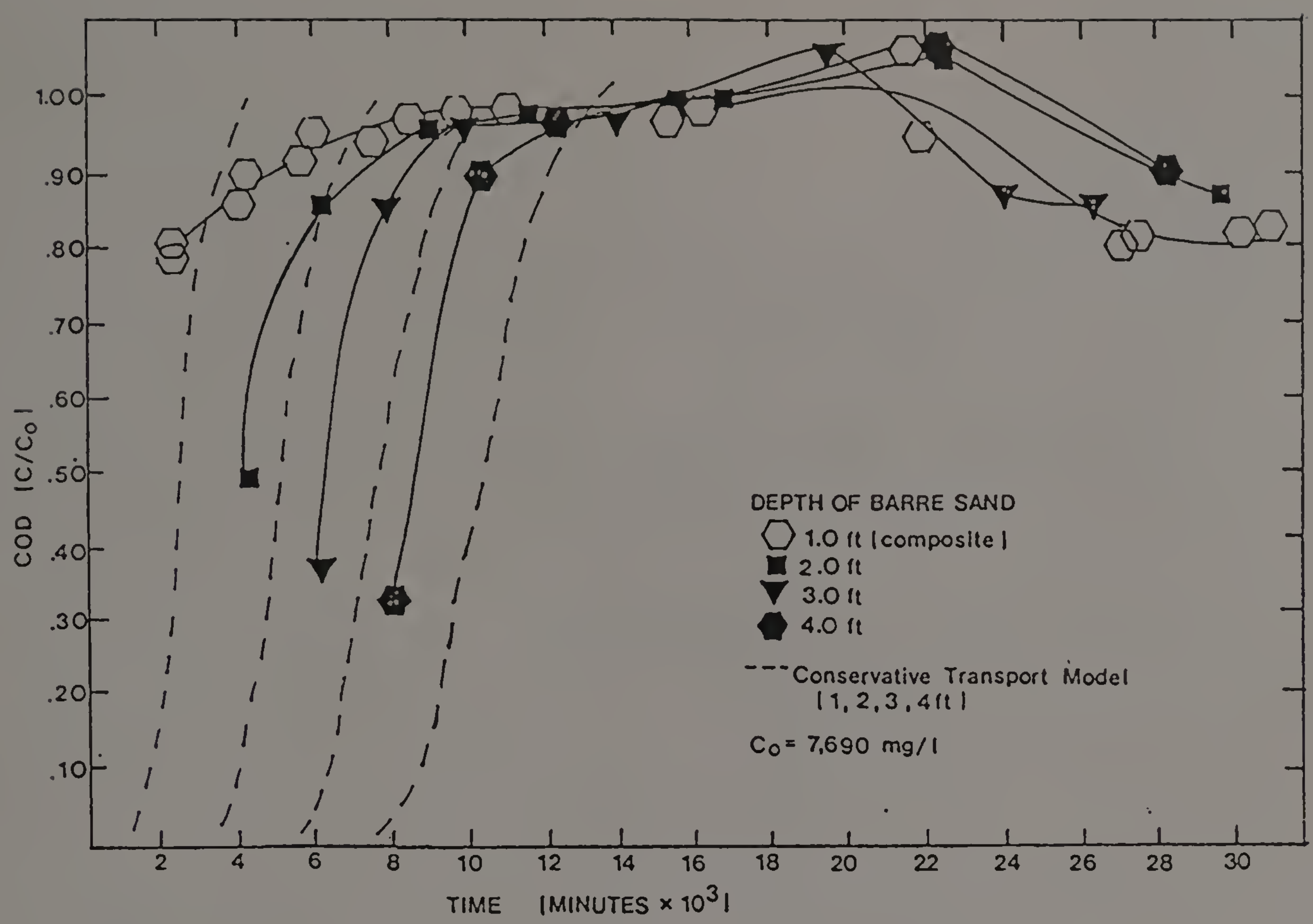


Figure 2-4. Chemical Oxygen Demand Breakthrough Curves (Unsaturated Flow). (Modified from Tirsch and Jennings (49))

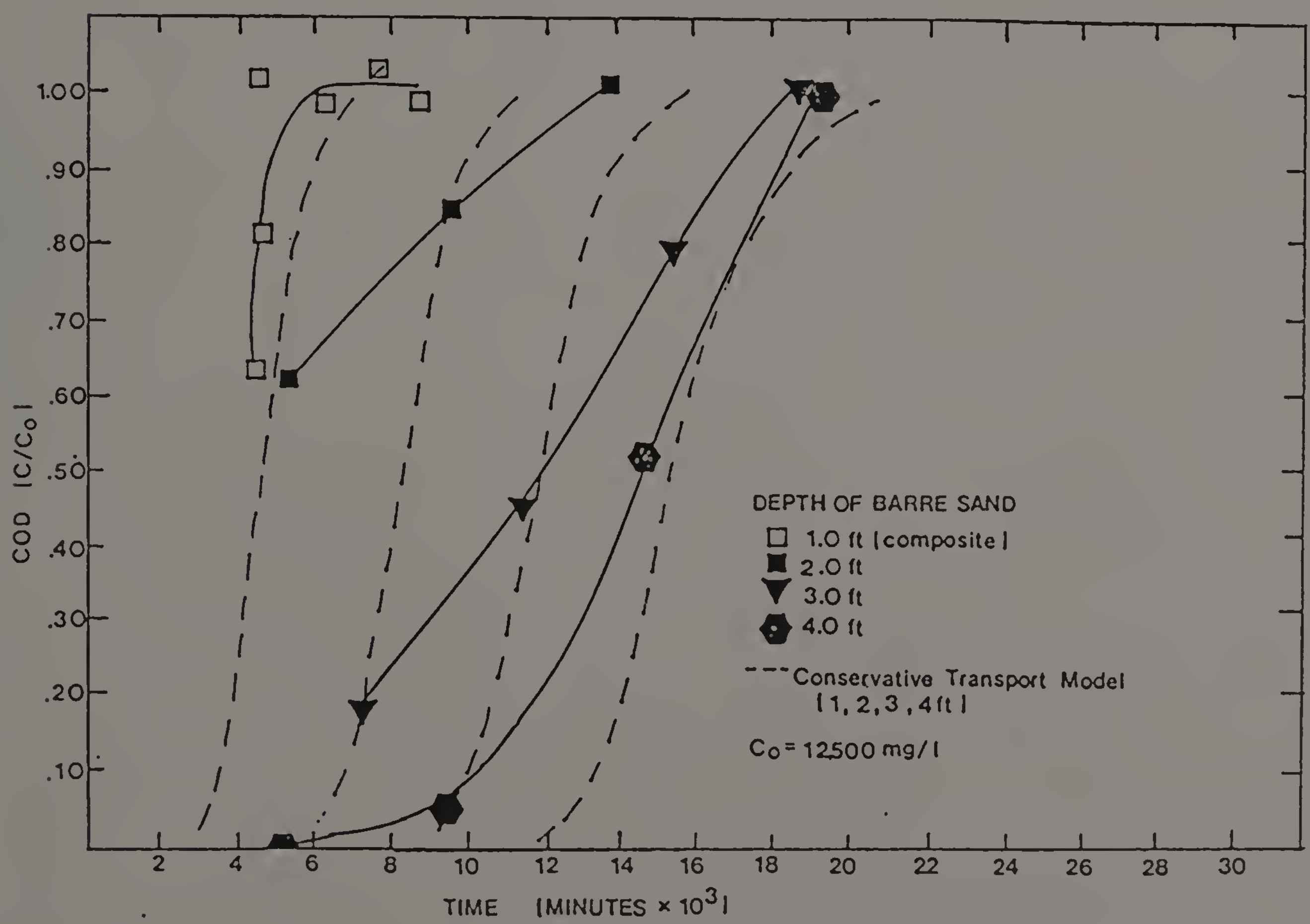


Figure 2-5. Chemical Oxygen Demand Breakthrough Curves (Saturated Flow). (Modified from Tirsch and Jennings (49))

prior to the theoretical exit times for a conservative parameter (Figure 2-6), and some cationic species appear to be removed under unsaturated flow conditions, only to emerge in time at concentrations many times that of their influent concentration. This is especially true for iron (Fe^{2+}) when the redox environment changes from an oxidizing condition to a reducing one. The iron breakthrough concentration for leachate running through 1.2 m (4 ft) of Barre sand was nearly four times that of the influent concentration (Figure 2-7).

(c) Biological Processes:

It has been demonstrated that landfill leachate contains large and diverse populations of microbial organisms (55, 56, 57). When leachate percolates into underlying soil, some of the microbial organisms are carried with it. Tirsch and Jennings (49) reported that microbial population fronts appeared to penetrate the full depth of 1.2 m (4 ft) soil columns that received approximately 3-4 cm of leachate per day over a three week period. The role of biological activity during leachate transport is of importance, because it represents a significant mechanism for attenuation or washout. When organic substrates penetrate to greater depths, microbes have the capability of reducing the organic strength. When oxygen supplies are depleted, however, and the environment becomes anaerobic and reducing, many inorganic exchange ions can be mobilized. The mobilized cations might have been indigenous to the natural soils, or they may have been previously removed from leachate that passed through the profile prior to the establishment of a microbial community. Tirsch and Jennings (49) measured the cumulative

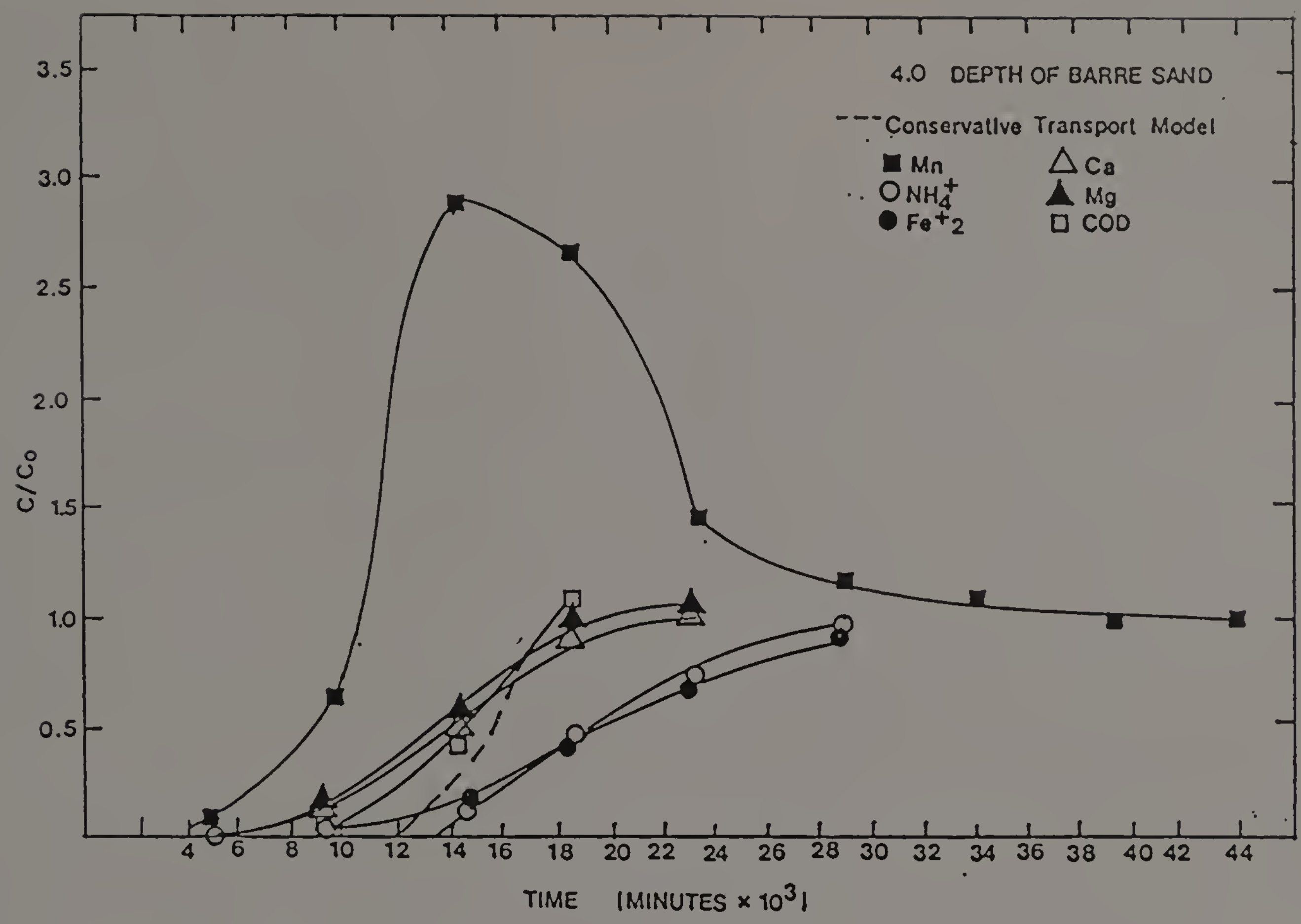


Figure 2-6. Relative Attenuations by Barre Sand (Saturated Flow). (Modified from Tirsch and Jennings (49))

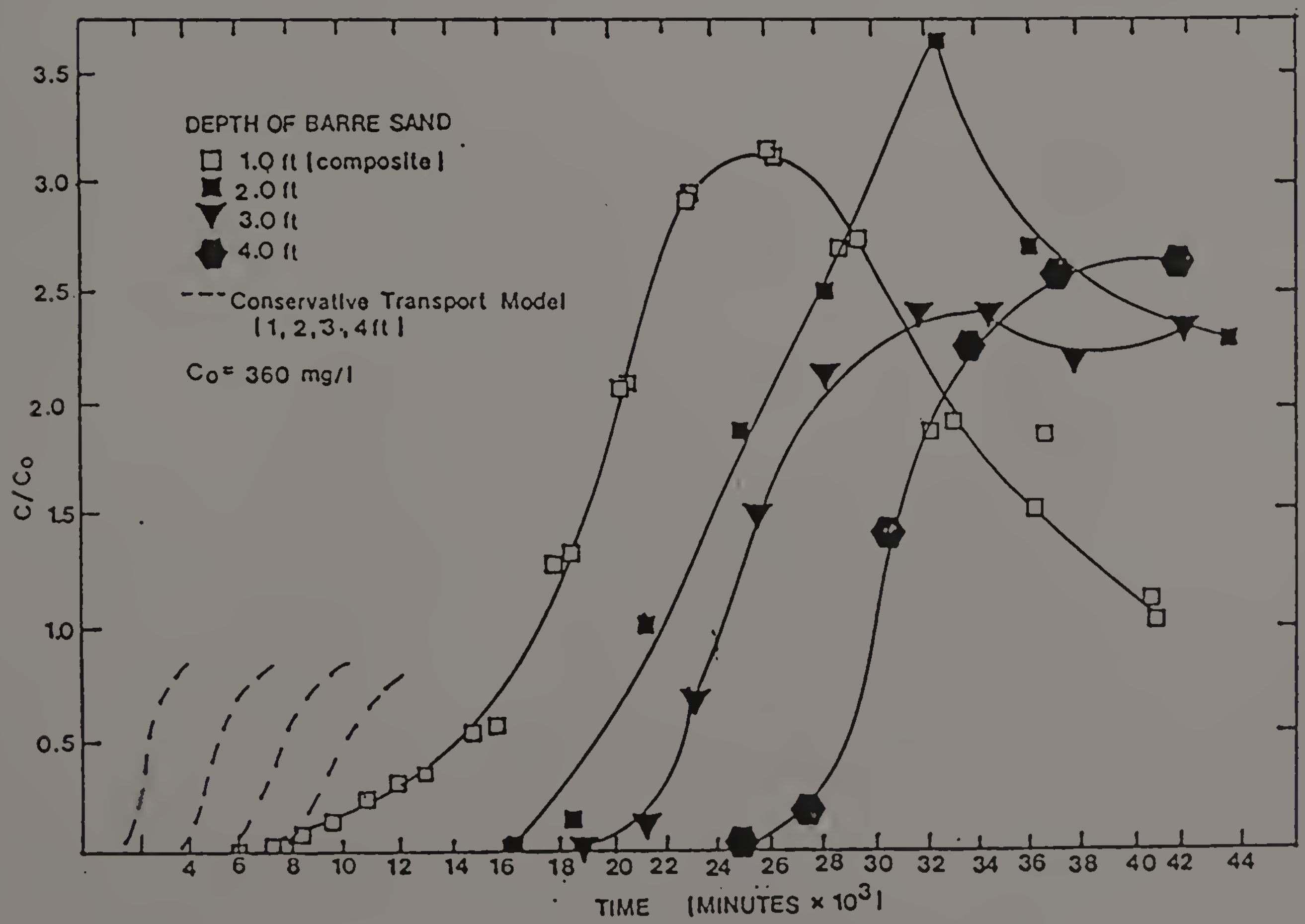


Figure 2-7. Iron Breakthrough Curves (Unsaturated Flow). (Modified from Tirsch and Jennings (49))

amount of iron that was released from their columns and concluded that the total release exceeded the amount applied. They attributed this phenomenon to a "black layer" that progressed downwards reducing natural ferric oxide coatings on the soil particles. The advance of the "black layer" in the column was closely correlated to the movement of the microbial front. Microbially mediated changes in the production of sulfate also produced extremely high effluent concentrations that were 40 to 50 times greater than that of the raw leachate. C/Co ratios greater than unity started to appear within two to three days, and C/Co ratios reached fifty to one in less than three weeks. Needless to say, the time and space variant nature of microbial activity must be considered a major component of any soil/leachate interaction model.

Existing Leachate Control and Treatment Technology

Dry Landfill Control

An awareness of the pollution potential from leachate has probably existed as long as the concept of landfilling has. Most early assumptions, however, presumed that potential pollutants would wash out of refuse in a manner and form similar to that of septage. Design regulations often used the same separation distance to groundwater for both landfills and septic system leach fields (58, 59). By incorporating separation requirements with regulations for impervious cover, it was assumed that refuse "cells" could be isolated from precipitation and groundwater. This approach or school of thought has generally been referred to as the "dry landfill theory," and it pervades most state regulations in effect today. Researchers during the early

1970's generally asserted that landfills were sanitary, and that leachate problems didn't exist, or that they were very much exaggerated by environmentalists (60, 61, 62). Unfortunately most of the early assumptions about leachate production and movement were incorrect. It wasn't until the mid to late 1970's that the nature of leachate production and migration started to become more fully understood. In 1976, Pindar et al. (63) modeled the movement of a Long Island landfill plume that had extended itself several miles from its source. Although the dry landfill approach continues to provide a basis for most facility designs, a "wet landfill" approach that utilizes impermeable liners is becoming increasingly popular (64). Leachate management under this school of thought is based on the assumption that solid waste has a finite pollution potential, and that near water tight encapsulation efforts merely postpone the ultimate release of pollutants. Advocates of the "wet landfill" approach prefer designs that maximize infiltration from rainwater, incorporate leachate recycling, or utilize wastewater effluent to degrade and flush solid waste constituents as rapidly as possible. By leaching the pollution potential out of refuse more quickly one can theoretically manage it while the landfill is still operating and while it is still cost effective to do so (65). Experience has shown that once a landfill has closed, it becomes extremely difficult to implement a pollution abatement program for leachate (66). According to Brunner (67) of the USEPA, there are thousands of landfills in the United States that were built, operated and closed following "dry landfill" design procedures. Brunner suggests that these sites may be "ticking time bombs" of pollution that will someday rupture due to differential settling, thereby releasing their pollution potential when operators are no longer around. Regardless of the long term risks and limitations, however, "dry landfilling" continues to be an accepted state of the art leachate control and management technique.

Site Selection Control

In October of 1976, section 4004 of Pl-94-580 (25), required that the Administrator of E.P.A. promulgate within one year, regulations and criteria for determining which solid waste disposal sites in the United States could be considered as "sanitary landfills." Section 4005 required that those found not to be "sanitary" had to be made sanitary or closed within a five year period.

The draft criteria and regulations proposed by E.P.A. resulted in confrontations with several states including Connecticut. Following the doctrine of riparian water law, E.P.A. proposed to require zero degradation of groundwater at the political boundary of any landfill site. In theory this approach would insure the right of "reasonable use" for downstream or down gradiant users. Unfortunately, Connecticut and several other states had implemented regulations that employed site-selection methods as a basis for leachate control (68). The site-selection method generally assumes zero or near zero reduction in leachate pollution strength due to physiochemical or biological attenuation processes. Advection and diffusion (i.e., dilution alone) are the major factors considered in the site-selection process. If it can be shown by conservative transport modeling that concentration levels will reduce to acceptable values by the time the plume reaches an aquifer or surface water body of importance; than the site is generally considered to be acceptable. This design approach generally assumes that groundwater contaminants will leave the site; and it provides little or no protection for small down gradiant users that might be located between the leachate source, and the theoretical point where concentrations approach ambient. In conjunction with these criteria, Connecticut also prohibited the use of liners for new facilities built during the late 1970's and early 1980's. By imposing rigorous constraints on a landfill's siting, several outcomes become immediately obvious. Firstly, fewer landfills are permitted. Secondly, the worse case ecological scenario is anticipated at the start when the design is being prepared. The design criteria are based on the assumption that the natural ecological systems of the area will have the capability of assimilating the total leachate pollution load for the life of the site without creating significant environmental impacts. From a practical point of view, this approach to leachate control makes the site selection task nearly impossible. When an occasional ideal site is found other political geographical and economic constraints often require that it be ruled out as a viable location.

Lined Landfills With Leachate Collection and Treatment

Most landfills currently being built incorporate the use of single or double liners with drainage networks to collect leachate and conduct it by gravity to some common manhole or storage reservoir outside the fill area. Liners are typically clay or some type of resistant man made material. Oftentimes a composite or double liner is prescribed for added groundwater protection. Tables 2-9 summarizes typical costs for common liner materials, but petroleum market prices, proximity to landfill sites and the landfill area being lined can affect these costs considerably. When associated expenses for collection drains, subgrade preparation, earthcover, and leachate storage are included, the cost of a state of the art secured landfill can exceed a half million dollars per hectare ($200,000 per acre).

Once collected, there are basically two options for dealing with leachate. One is to deliver it to some off-site wastewater treatment facility. For small landfills this can be accomplished with septic tank effluent trucks (i.e., "honeywagons"). For larger landfills with sewer lines at close proximity to the site, leachate can be piped to the wastewater treatment facility. This technique has the added advantage of diluting the potent leachate with conventional sewage and it hydraulically smoothes the BOD loading at the plant. Several environmental and economic liabilities are associated with either of these techniques, due to the special chemical nature of leachate. BOD_5 values for landfill leachate are typically 100 times that of raw sewage, and this, of course, has a direct economic impact on the cost of aeration and treatment at a typical activated sludge treatment plant (69). Secondly, there are constituents of leachate that are not amenable to conventional biological treatment (70). These constituents, in fact, may even be detrimental to the sewage treatment process due to

Table 2-9. Costs for Various Sanitary Landfill Liner Materials.

Materials	Installed Cost+ ($/sq yd)
Polyethylene (10-20 mils*)	0.90 - 1.44
Polyvinyl chloride (10-30 mils)	1.17 - 2.16
Butyl rubber (31.3-62.5 mils)	3.25 - 4.00
Hypalon (20-45 mils)	2.88 - 3.06
Ethylene propylene diene monomer (31.3-62.5 mils)	2.43 - 3.42
Chlorinated polyethylene (20-30 mils)	2.43 - 3.24
Paving asphalt with sealer coat (2 inches)	1.20 - 1.70
Paving asphalt with sealer coat (4 inches)	2.35 - 3.25
Hot sprayed asphalt (1 gallon/yd^2)	2.50 - 2.00 (includes earth cover)
Asphalt sprayed on polypropylene fabric (100 mils)	1.26 - 1.87
Soil-bentonite (9.1 lbs/yd^2)	0.72
Soil-bentonite (18.1 lbs/yd^2)	1.17
Soil-cement with sealer coat (6 inches)	1.25

+Cost does not include construction of subgrade nor the cost of earth cover. These can range from $0.10 to $0.50/$yd^2$/ft of depth.
Material costs are the same for this range of thickness.
*One mil = 0.001 inch.

Source: Haxo, H.E. Jr. Evaluation of liner materials. U.S. EPA Research Contract 68-03-0230. October 1973.

toxic effects that are described more thoroughly in Chapter VI. This difficulty becomes increasingly probable if leachate flow rates exceed 5 percent of the total wastewater flow. Boyle and Ham (10) operated six bench scale activated sludge treatment units for four months using six mixes of domestic wastewater and landfill leachate. COD, BOD, MLSS and SVI were monitored daily along with pH and alkalinity. Raw leachate BOD and COD average values were 8790 mg/l and 10,820 mg/l, respectively. Influent COD values after mixing ranged from 240 mg/l to 2,355 mg/l. As leachate concentrations approached 5% of the total wastewater stream, effluent quality deteriorated rapidly signaling severe plant upset. Tables 2-10 and 2-11 summarize influent and effluent quality for the six reactors. It is evident from Table 2-12 that reactor unit A-4 which represented the 5% leachate study experienced rapid increases in effluent concentrations (i.e., treatment efficiency deteriorated) for all parameters being evaluated. Increased oxygen requirements were evidenced at the 1% leachate concentration and BOD effluent quality deteriorated at the 2% leachate concentration. Data from this study indicates that conventional activated sludge processes are extremely sensitive to the addition of even small amounts of landfill leachate. Even if toxicity and microbial upset are not encountered, there is still reason for concern regarding leachate components that escape conventional treatment, and pass through a sewage treatment plant to a point of discharge into some lake or river. Heavy metals, pesticides, hospital wastes, and hazardous wastes are just a few of the substances that routinely find their way into landfill leachate (72, 73, 74).

Table 2-10. Extended Aeration Leachate (Domestic)--Unit Loading.

Parameter	A-1	A-2	A-3	A-4	A-5	A-6
Leachate,* %V/V	0	1	2	5	10	20
COD influent, mg/l	240	350	450	770	1,300	2,355
BOD influent, mg/l	140	225	310	570	1,000	1,870
Organic Load, lb BOD/day/100 lb, MLSS	3.7	6.0	8.3	15.2	26.9	50.0
Volumetric Load, lb BOD/day/1,000, cu ft	5.7	9.4	13.0	23.8	41.8	77.6

*Leachate Strength: COD = 10,820 mg/l, BOD = 8,790 mg/l, TVS = 4,400 mg/l.
Note: lb/day/1,000 cu ft x 16 = g/day/cu m.
After Boyle and Ham (71).

Table 2-11. Extended Aeration Leachate--Domestic*.

Unit	Leachate (% V/V)	COD effluent (mg/l)		Oxygen Uptake Mean (mg/day)	Sludge production (mg/day)		SVI	
		Mean	95% CI		Mean	95% CI	Mean	95% CI
A-1	0	30	5	24.5	82	23	49	2
A-2	1	24	5	25.6	110	35	62	2
A-3	2	31	6	43.3	148	44	69	4
A-4	5	38	7	83.5	178	53	100	5
A-5	10	59	6	132.0	332	118	166	16
A-6	20	113	12	230.0	722		526	72

*Performance Data for 9/28/71 to 11/25/71.
CI--confidence interval
After Boyle and Ham (71).

Table 2-12. Extended Aeration Leachate (Domestic)--Effects.

Unit	Leachate (% V/V)	Increase in Parameter (%)				
		BOD effluent	COD effluent	Oxygen Uptake	Solids Produced	SVI
A-1	0	--	--	--	--	--
A-2	1	0	0	4.5	34.7	26.5
A-3	2	8.2	0	76.7	80.5	41.9
A-4	5	53.5	26.4	241	117	104
A-5	10	160	96.8	440	305	239
A-6	20	1,040	276	840	780	975

After Boyle and Ham (71).

The alternative to "off-site" treatment is "on-site" treatment. This has been practiced with limited success using conventional activated sludge package plants (71, 75, 76). Logically, if toxicity and microbial upset are encountered when leachate is diluted, as just described, attempting to treat leachate by the same process in a concentrated form is even more difficult. At the GROWS landfill site described by Steiner et al. (76), treated leachate effluent must be recycled back into the landfill by spray irrigation because it fails to meet permit requirements for point source discharges. The inherent shortcomings of conventional treatment technology become evident if one considers the fundamental principles involved:

1. Activated sludge processes are designed to remove 90%$\pm$ of BOD_5 and suspended solids (55). For conventional wastewater with a typical BOD_5 of 250-300 mg/l, a 90% reduction meets acceptable discharge requirements (i.e., 30 mg/l BOD_5 and 30 mg/l SS).
2. Landfill leachate generally has suspended solids and BOD_5 concentrations in excess of 25,000 mg/l (13). If 90% removals are achieved by conventional treatment, the effluent quality will approximiate that of raw sewage (i.e., 250-300 mg/l). This is clearly unacceptable for discharge.
3. Secondary treatment technology is defined as "biological treatment." As such it would be unreasonable to expect advanced (tertiary) treatment effluent quality from a process that was never intended nor designed to remove or treat more than conventional biodegradable wastes (i.e., sanitary sewage).

Clearly what is needed by the solid waste industry is a treatment technology that is able to realize at least a 99% reduction in conventional pollutants, while at the same time being able to remove some of the more exotic wastes, such as heavy metals and toxic substances. If a final effluent is intended for surface water discharge, or groundwater infiltration, appropriate water quality standards and objectives must also be considered. To achieve these treatment objectives new technologies may have to incorporate multiple methodologies, and/or develop techniques still untried. Economic considerations are equally important, and generally impose real world constraints.

Emerging Technology

In 1976 the first lined landfill in Massachusetts was constructed in Barre. More than two years of previous characterization work had indicated that landfills operating in natural wetlands seemed to benefit by the "natural" treatment potential of the wetlands ecosystem (13). Based on this observation, it was decided that a "constructed wetlands" approach would be used to treat Barre's leachate. Figures 2-8 and 2-9 illustrate the general configuration of the two hectare (5 acre) Barre facility, and Table 2-14 summarizes the general treatment performance of the "constructed wetlands" (i.e., lagoons) for the second full year of operation (1977). The landfill and treatment system continue to function in an effective manner today even though total landfill and lagoon areas have been expanded considerably. This small prototype provided basic design criteria for several larger applications elsewhere

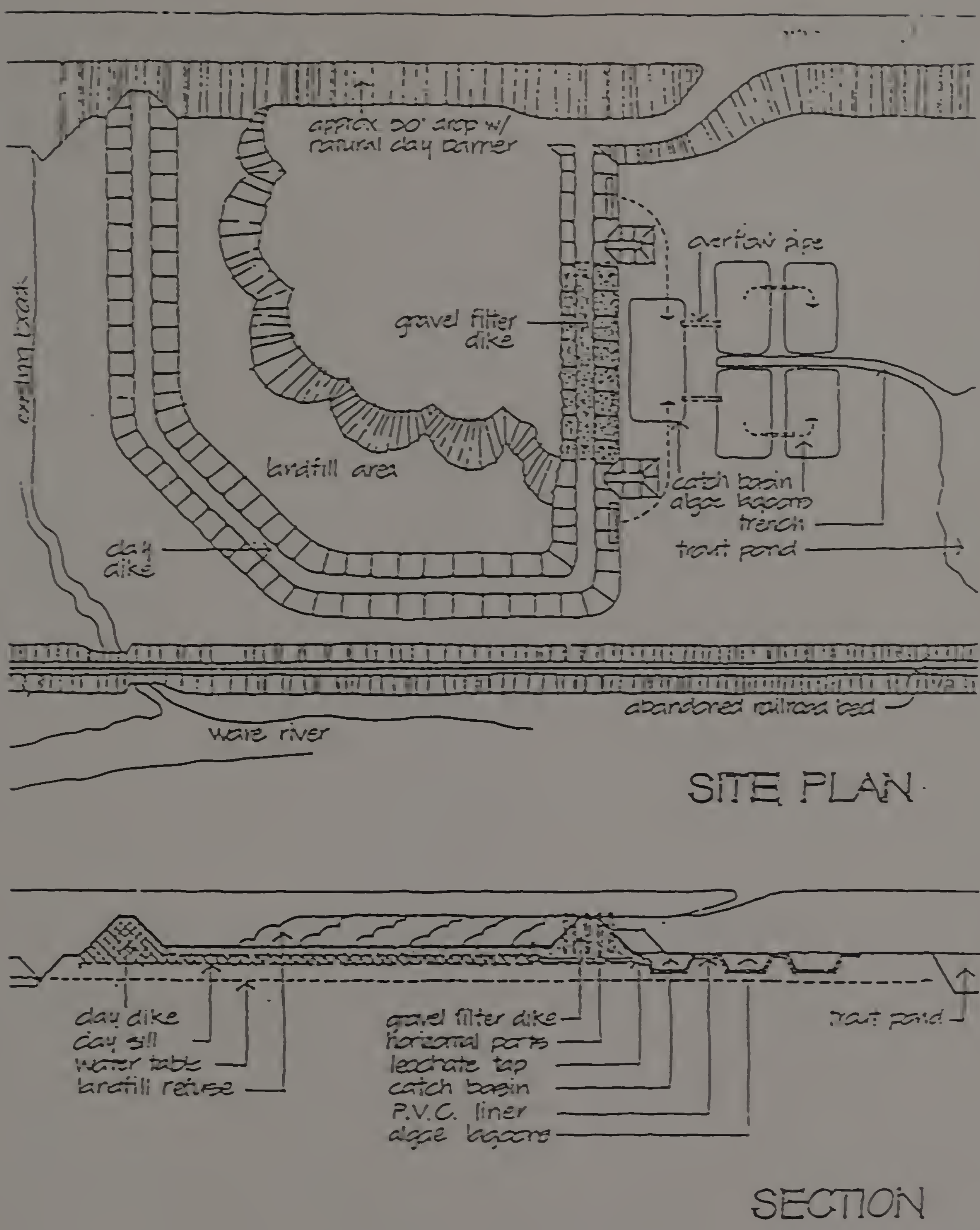


Figure 2-8. Barre Landfill Plant View and Section.
(After Lavigne (13))

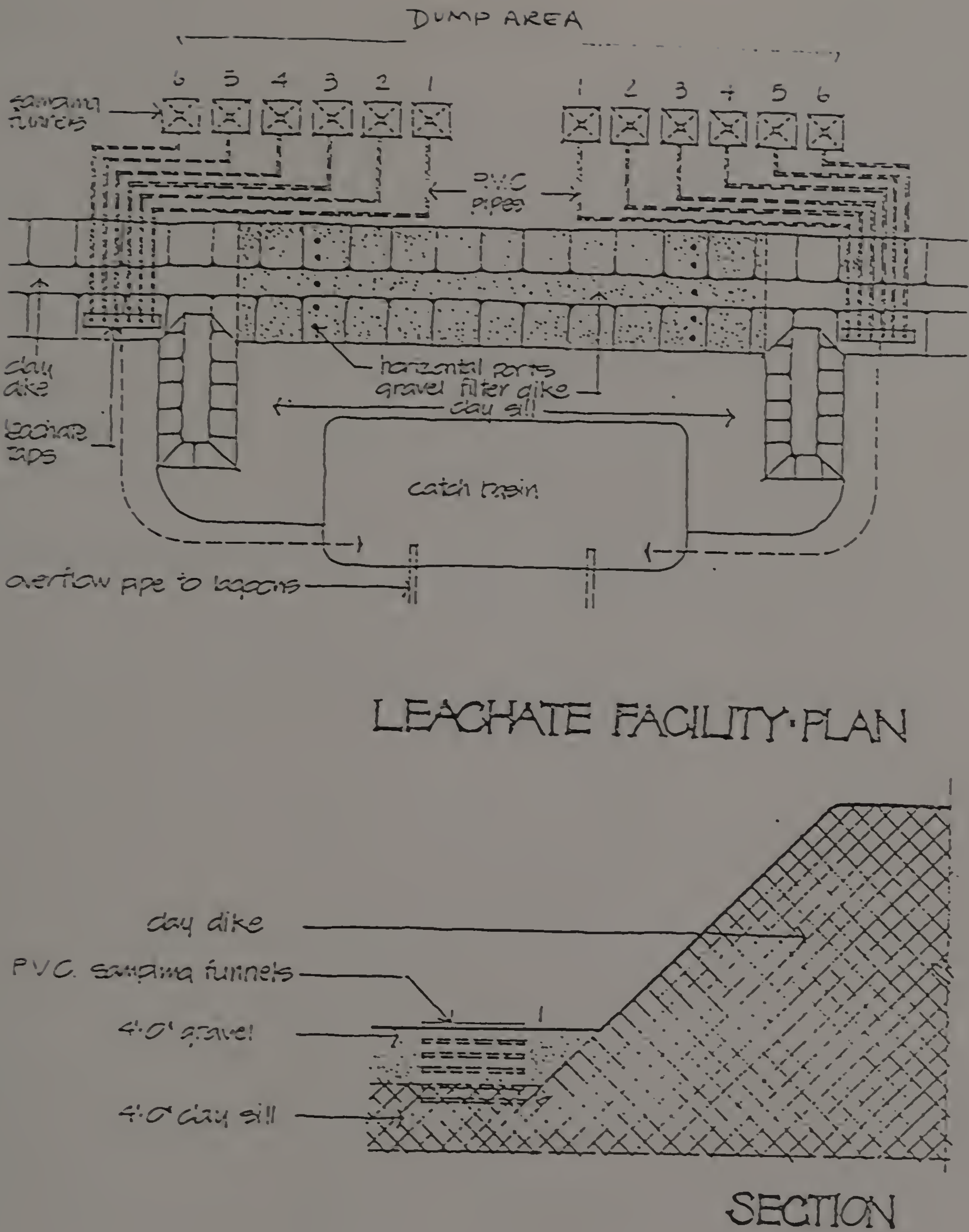


Figure 2-9. Barre Leachate Sampling Funnels and Ports.
(After Lavigne (13))

Table 2-13. Leachate treatment - Martone Landfill, Barre, MA.

Parameter mg/l	Quality of Leachate Collected from Interior of Barre Landfill	Quality of Leachate After 30 Days of Treatment	Quality of Leachate After 60 Days of Treatment	Quality of Leachate at Time of Discharge (90 days)
BOD5	21,060	4,650	220	10.3
COD	35,680	9,500	400	117
pH	5.15	6.50	6.80	7.3
Alkalinity as $CaCO_3$	4,600	980	315	32
Sulfate	2,330	450	79	29
Ammonia	437	130	70	3.5
Chloride	372	350	317	200
Total Solids	11,600	4,300	2,580	1,430
Volatile Solids	3,900	1,900	1,050	320
Iron	1,400	318	120	1.0
Zinc	24	14	12.0	1.0
Manganese	28	11.0	3.3	1.6

Values represent averages for the 1977 sampling year.

After Lavigne (13).

applications elsewhere in Massachusetts. Several different operational alternatives are still being evaluated. Patents for this system were awarded in June of 1981 and January of 1983 (77, 78), and the process is marketed by the Barre based firm Resource Control Inc.

Other uses of wetlands for the treatment of wastewater have been practiced for more than a century. Stanbridge (79) reported that in 1877, 6 m^3 of sewage per day were being applied to a swamp in Great Britain producing an offensive odor and highly polluted effluent. By providing suitable underdrainage it was possible to treat effectively about .05 $m^3/day/m^2$ without the soil becoming clogged.

The use of aquatic plant species to assist with treatment by maintaining aerobic conditions in the soil was not understood until studies were started in Europe, the U.S.A. and Canada about 15-20 years ago (80). Dr. Reinhold Kickuth at the University of Hessen in Germany developed a reed (Phragmites) (root zone method) and sand bed system to treat about .1 $m^3/m^2/day$ of sewage at Othfreseu, and the system has been operational since 1974 (81). The United States, Canada, Denmark and Holland have used man-made and natural wetlands to treat wastewater effectively for more than a decade now and the effluent quality generally is excellent, including removal of phosphorus (82). Cooper et al. (80) summarized the key features of a Phragmites and sandbed system as follows:

- Rhizomes of reeds (normally Phragmites) provide a 'hydraulic pathway' through the rhizosphere (the annular space between rhizomes and roots and surrounding soil) along which wastewater can flow;

- Wastewater is treated by bacterial action (aerobic in actively growing rhizospheres and anoxic/anaerobic in dead and decaying rhizospheres and in the surrounding soil);
- Atmospheric oxygen is provided to the rhizosphere via the leaves and stems through the hollow rhizomes and roots;
- Aerobic composting of sludges in wastewaters occurs in the above-ground layer of 'straw' derived from dead leaves and stems.

He also indicated that the addition of hydrated lime to bring calcium contents to 2-2.5% is generally practiced. Hydraulic conductivities in sand beds that range between 10^{-3} and 10^{-5} meters/sec are desirable, and bed depths are generally less than a meter (83). Kiekuth (81) reports that hydraulic conductivity increases with bed age as roots and rhizomes develop, and that within 2-4 years steady state rates of 10^{-3} meters/sec are generally achieved. It shall be reported later that similar root and rhizome hydraulic benefits were observed in the peatmoss-reed canarygrass system used for this study.

The Max Planck Institute Process (MPIP) developed by Seidel (84) uses Phragmites in a "constructed" marsh. The marsh is generally made of reinforced concrete, with dimensions typically ranging from 2-4 m wide, up to 100 m long and .5 to 1 m deep. Scirpus and Typha plants have also been used. The vegetal support media is generally sand and gravel. The performance of a MPIP system installed in Laguna Niguel, California was monitored for nearly a year under EPA contact, and the system did not perform as well as those used elsewhere (79).

The Lelystad Process, developed in the Netherlands, evolved from the MPIP process. In 1967 the IJsselmeerpolders Development Authority started to use wetlands to treat sewage (85). deJong et al. (86) reported that <u>Phragmites</u> and <u>Scirpus</u> performed equally well in these older systems. Detention times of about ten days seemed most effective, and this value agrees well with times used for other Root Zone Methods (RZM). It was also reported by Greiner et al. (87), that marshes containing clay soils with high Al and Fe components contributed to improved removals of phosphorus through adsorption and precipitation. Nitrification and denitrification were attributed to bacterial activity, and little effect of nitrogen removal was attributed directly to uptake by the plants. The Lelystad Process does not attempt to utilize root zone ecology, and a significant proportion of the wastewater being treated flows above ground over the beds, often producing problems with flies, mosquitos, and odors. Surface freezing during winter months and short-circuiting are also common problems. The more "natural" construction and operational practices associated with the Lelystad Process are still capable of producing a high quality effluent, but treatment area requirements are approximately double that of the RZM for comparable wastewater flow rates and concentrations. It should be noted that at the present time none of these technologies have been used for landfill leachate treatment, though some researchers have suggested that there may be future applications in this regard. None of the systems reviewed have attempted to use peatmoss or reed canarygrass in any combination themselves nor with any other materials.

Column studies by Rock et al. (88) were used to evaluate the effectiveness of sphagnum peat for the removal of metals from landfill leachate, but the study made no attempt to utilize living plants, and there was no effort to evaluate any biological transformation of leachate constituents. As such peatmoss was evaluated purely as a physicochemical treatment medium. Metal removal rates of 50-98% were reported with average values generally being about 75%. Rock concluded that peatmoss could be used as a prefilter to conventional treatment or spray irrigation.

In summary, it seems that although regulatory agencies at all levels of government continue to require the use of liners, there are very few technologies that can cost effectively treat landfill leachate to a desired effluent quality. The practice of using existing wastewater treatment facilities has severe limitations, and raises many questions regarding untreated parameters and toxicity. Low technology "Living Filter" systems seem to hold the most promise, but natural wetland systems currently subject to protection may, for all intents and purposes, be excluded from the treatment options.

If cost effective and space efficient, engineered wetland systems can be constructed and operated at or near a landfill site, and they may provide the technology needed to solve the problem of leachate treatment.

Successful systems will probably satisfy most or all of the following requirements:

1. Low capital costs for construction.
2. Low operating costs.
3. Low maintenance costs.
4. An ability to process a waste that is variable with flow rate and constituent concentrations.
5. Minimum land requirements.
6. Minimum nuisances (i.e., flies, mosquitos, odors).
7. An effluent quality that permits on-site discharge or infiltration into existing surface or groundwater resources.

CHAPTER III

GREENHOUSE BENCH SCALE BATCH REACTOR ANALYSIS

Introduction

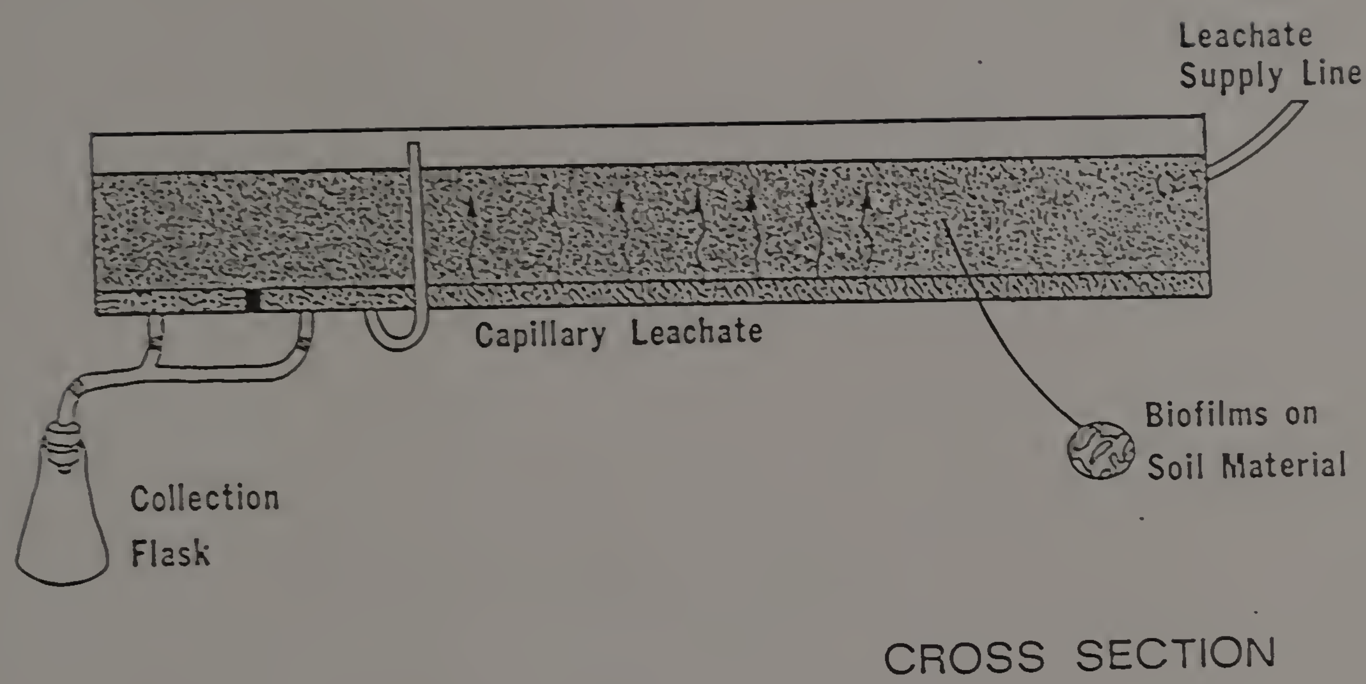
The design of any wastewater treatment facility is generally based upon knowledge of the wastewater to be treated, limitations of the systems being considered, and space or land area available at the treatment site. When an atypical wastewater, such as landfill leachate, is to be treated it is generally prudent to conduct bench top and pilot plant studies prior to contemplating the design of a full scale facility.

Previous researchers have concluded that landfill leachate is biologically treatable (5, 10, 71, 75), but there have been considerable limitations with the application of conventional technologies in the field (89, 90, 76).

As discussed earlier, the chemical composition of leachate varies considerably with the age of a landfill. This characteristic makes the treatment design process difficult. For traditional treatment technologies to operate effectively, the wastewater composition and flowrate must remain relatively constant with time. When steady state conditions cannot be maintained, treatment effectiveness is reduced, along with effluent quality. For domestic wastewaters, flowrates are generally smoothed by extensive sewerage networks, and the concentrations of treatment parameters remain fairly consistent with time. Conventional secondary treatment processes (i.e., biological treatment) have, for the most part, been developed to principally remove or reduce BOD and suspended solids concentrations. It is not surprising, therefore, that regulatory agencies have questioned the advisability of adding landfill leachate to domestic wastewater for conventional biological treatment.

The purpose of this investigation was to evaluate the feasibility of utilizing a low technology root-zone method as a pretreatment or total treatment alternative to a conventional activated sludge process.

Although batch treatment methodologies continue to have application in some areas of process design, they are generally considered to be impractical for full scale wastewater treatment facilities. The continuous flow alternative that theoretically has comparable kinetic benefits for first order reactions is the plug flow (PF) system. Ideal plug flow systems are, however, more difficult to control hydraulically, and for this reason have limited research applications. The treatment reactors (beds) designed for this study incorporated both batch and plug flow operational capabilities. It was assumed that if batch treatment data supported the theory that a peatmoss and reed canarygrass system could be used to effectively treat landfill leachate, then the reactors could be switched to the continuous flow mode without major system interruptions. Figure 3-1 illustrates the general configuration of the reactors used in this study.



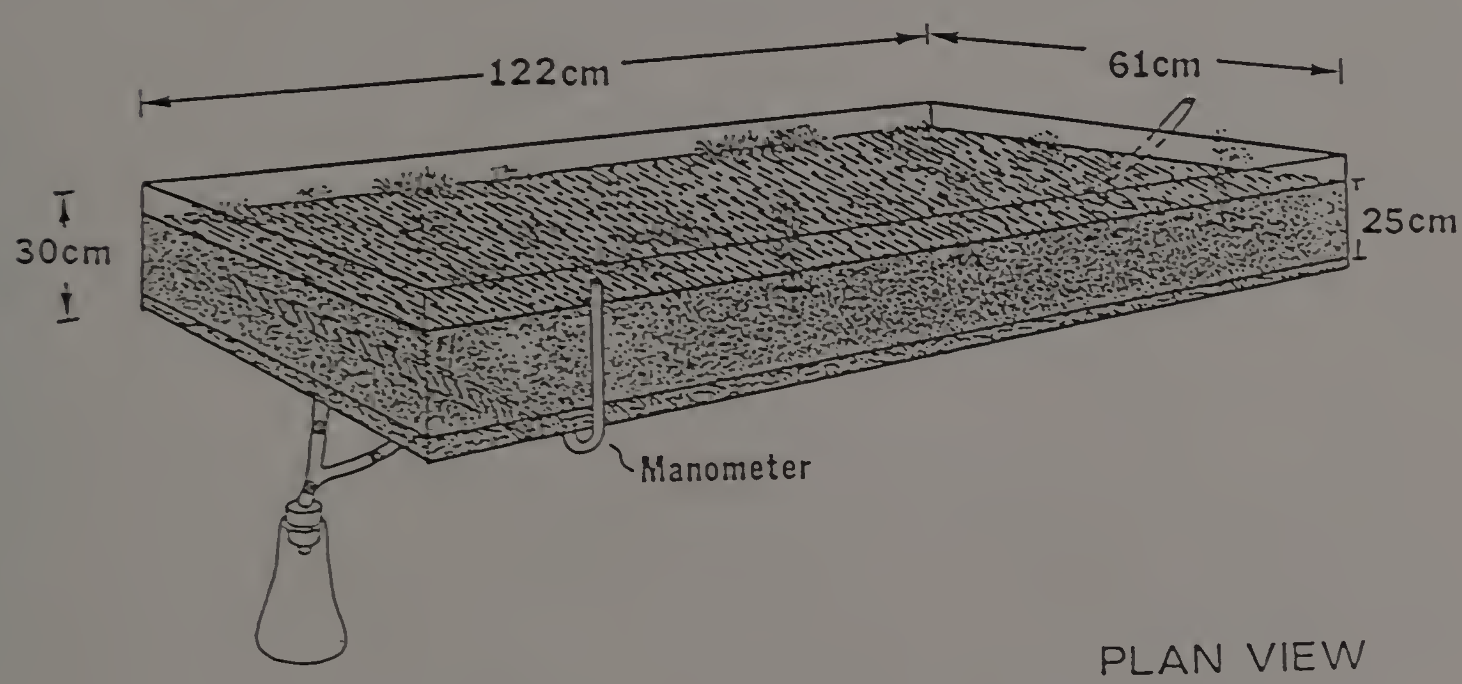


Figure 3-1. Bench Scale Treatment Tray for Batch and Plug Flow Modeling.

For the purposes of ascertaining design parameters it was proposed that overall changes in leachate Total Organic Carbon (TOC) and/or Chemical Oxygen Demand (COD) could be described by first order kinetics (eq 3-1).

$$-\frac{dC}{dt} = kC \qquad \text{(eq 3-1)}$$

where:

$-\frac{dC}{dt}$ = rate of change in the concentration of TOC or COD with time (mass-volume^{-1} time^{-1})

C = concentration of TOC or COD at any time t (mass-vol^{-1})

k = reaction rate constant time^{-1}

It should be noted that for ideal batch reactor analyses there are no additions or removals of liquid volumes. This condition is nearly impossible to maintain in a greenhouse environment, however, where evapotranspiration can contribute significantly toward increasing final treatment concentrations.

Table 3-1 summarizes solved mass balance equations for various reaction orders and reactor types. Each equation has been written in terms of t (nominal hydraulic retention time). If variables are separated in equation 3-1 and then integrated with appropriate initial conditions the resulting calculations will give the linear form of a first order batch reaction (eq 3-2).

Table 3-1. Nominal Hydraulic Retention-Time Equations for Reactions of Different Order in Continuous Flow Stirred Tank Reactors (CFSTRs) and Plug Flow (PF) Reactors.

	Nominal hydraulic retention time	
Reaction order	t^{CFSTR}	t^{PF}
0	$\frac{1}{K}(C^o - C^t)$	$\frac{1}{K}(C^o - C^t)$
1	$\frac{1}{K}(\frac{C_o}{C_t} - 1)$	$\frac{1}{K}[\ln(\frac{C_o}{C_t})]$
2	$\frac{1}{KC_t}(\frac{C_o}{C_t} - 1)$	$\frac{1}{KC_o}(\frac{C_o}{C_t} - 1)$

(After Weber (91))

$$\ln C_t = -kt + \ln C_o \qquad \text{(eq 3-2)}$$

where C_o = initial concentrations at t=0 mass-volume^{-1}

C_t = final concentrations for any time t mass-volume^{-1}

t = time

k = reaction rate constant time^{-1}

Solving eq 3-2 for t will result in a retention time formula for a first order reaction (eq 3-3).

$$t = \frac{1}{k} \left[\ln \left(\frac{C_o}{C_t}\right)\right] \qquad \text{(eq 3-3)}$$

It can be seen that eq 3-3 describing first order kinetics applies to both batch and plug flow reactor detention times (See Table 3-1, reaction order 1).

If equation 3-2 is tested as a model, it is evident that first order kinetic data should plot as a straight line with ln C_t as the ordinate and t as the abscissa. The slope of this straight line would represent the rate constant k. Initial testing would, logically, be conducted using the batch method. By then selecting a desired C_o/C_t value, and knowing k, it would be possible to control the flow rate in a continuous plug flow system to achieve the desired retention time t.

Objectives

The objectives of this batch study were as follows:

1. To evaluate pre and post treatment leachate concentrations of twelve batch reactors of similar design operated at twelve

different retention or treatment times ranging from one to twelve days.

2. To maintain an unsaturated but near field capacity moisture profile in each of the reactors.
3. To observe possible toxic effects of landfill leachate on reed canarygrass.
4. To test the hypothesis that first order kinetics could be used to describe or model the overall attenuation of Total Organic Carbon (TOC) and/or Chemical Oxygen Demand (COD) in the leachate.
5. To determine an appropriate kinetic rate constant for the purpose of plug flow modeling and an eventual full scale leachate treatment system design.
6. To evaluate physicochemical processes associated with heavy metal removal.
7. To evaluate the uptake of heavy metals by plant tissues.
8. To observe any ecological interactions that might be unique to a system comprised, presumably, of noncompatable substrates support media and living organisms.

Methods and Materials

Seed Bed Preparation

The treatment reactors illustrated in Figure 3-1 were constructed of pine boards with plywood bottoms. The insides were sealed and coated with several layers of fiberglass resin, and the manometer and drainage system was made of conventional maple syrup tubing and connectors. Drainage ports for the reactors were drilled through the plywood bottoms. Maple syrup spigots were cemented and driven by hammer so as to provide water tight fittings. Spigot ends inside the reactor were flared with a countersink drill bit, and covered with a tuft of glasswood held in place by Elmer's waterproof glue. Each reactor had 4 pairs of drainage spigots set in 30 cm (12 in) intervals along the 120 cm (48 in) base. Pairs of spigots were connected to each other by maple syrup tubing (6 mm I.D.), and the entire drainage system was connected to a single discharge tube at the terminal end of a manifold system. When raised and secured at the top edge of the reactors, each drainage pan formed a manometer so that saturation depth within the reactor could be monitored from the outside. When the manifolds were lowered the reactor's contents were able to drain from the eight spigots through the common discharge tube. It was intended that the continuous flow studies manifolds could be sectioned by cutting them into four separate drainage lines, each capable of collecting treated leachate from an individual 30 cm section (Figure 3-2).

Each treatment bed was filled with 27 kg of air dried sphagnum peatmoss (source Fafard of Canada), resulting in the dimensions depicted in Figure 3-1. It was assumed that the weight of wetted peatmoss and eventual rhizome growth would collectively determine the long term operating densities, so there was no attempt to establish or maintain them. Gentle shaking of the air dried peat as it was added to each reactor did result in a fairly uniform depth of 25 cm (10 inches) per

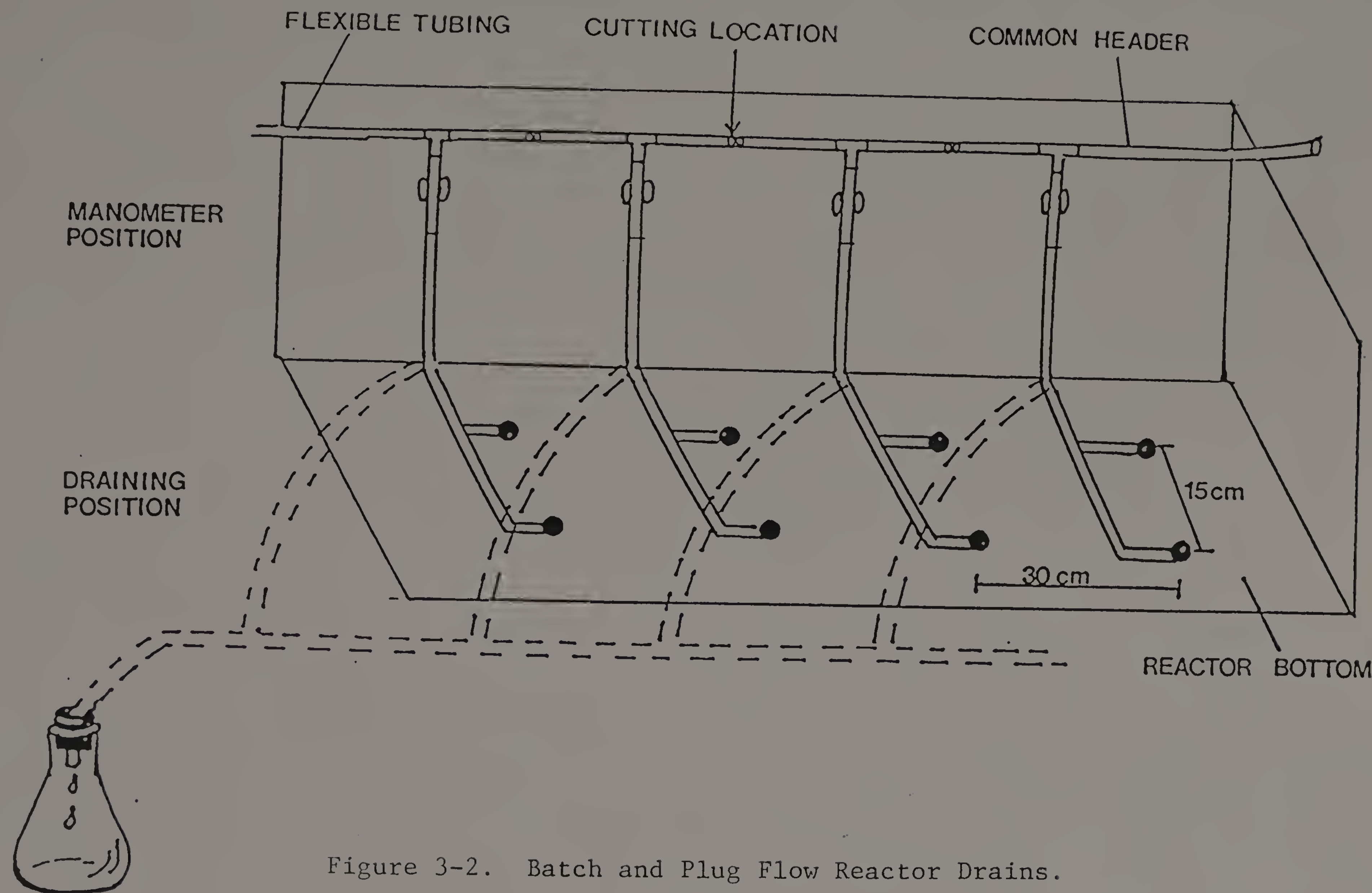


Figure 3-2. Batch and Plug Flow Reactor Drains.

bed. As such an approximate initial density of .14 g/cm^3 can be assumed, and it seems reasonable to assume that this density remained about the same during the entire period of experimentation. McLellan et al. (92) used air dried peat densities of .12, .15 and .18 g/cm^3 in column studies of leachate treatment using sphagnum moss. Clogging was reported at the two higher densities, but their work did not involve rooted plants. Reed canarygrass was selected, in part, for this study because of its expected ability to maintain the high permeability rate associated with an unclogged peat matrix. Two samples of peat were taken at random from each reactor during the peatbed filling process so that oven dried weight and cation exchange capacities could be determined. Values for the 32 peat samples were averaged and are reported in Table 3-2.

Near field capacity saturation of the peat beds was accomplished by daily watering so as to maintain a 2.5 cm (1 inch) head in the reactor manometers. After two weeks of wetting, each reactor was seeded with 5 g of reed canarygrass (Phalaris arundinacea L.). The seeds were gently tapped into the saturated peatmoss and allowed to germinate. By the sixth week seedling height had reached approximately 5 cm (2 inches). During that time watering was continued on an as needed basis to maintain saturated conditions in the bottom 2.5 cm (1 inch) of each bed. After reaching a height of 5 cm batch leachate applications were initiated. It should be noted that previous pot studies had attempted to germinate reed canarygrass seeds in leachate saturated peat. For all dilutions used, the seeds were unable to germinate, and it was assumed

Table 3-2. Summary of Treatment Bed Specifications.

Item	Description
Length	122 cm
Width	61 cm
Depth bed	30 cm
peat	25 cm
Peat Weight	27 Kg
Peat Volume	.186 m^3
Peat Density	.14 g/cm^3
Peat Type	Canadian Sphagnum
Cation Exchange Capacity*	290 meq/100g
Seeding Density Reed Canarygrass _Phalaris_ _arundinacea_ L.	5 g/reactor (.7440 m^2) (60 lbs/Acre)
Reactors #1-#12	Treatment Times 1 day-12 days

*Ammonium Acetate method using 5 g peat sample and 500 ml NaCl (10%) exchange volume.

Modified procedures from John H. Baker (personal communication).

that some type of acute toxicity was rendering the seeds non-viable. It was therefore decided that seedlings should be established before leachate applications started. At the end of the batch study, all leaf material above the reactor lip was harvested, dried at 95^{o}C and milled in a Whiley mill. Representative half gram samples from each reactor were digested using perchloric acid and stored for analyses by atomic absorption spectrophotometry.

Leachate Collection and Application

Leachate was collected on an as needed basis from an Amherst landfill which is equiped with a liner. The particular source was selected because the landfill was relatively new, and generally free of industrial or hazardous wastes. Table 2-8 includes a general characterization of the leachate used and compares it to sources from other studies. For the purposes of modeling, TOC and/or COD were analyzed for the raw leachate each time it was collected. During the months of March-June of 1985, forty-one collections of raw leachate were made from the landfill leachate source. Early attempts to pump leachate from a landfill manhole were thwarted by extreme foaming of the leachate and there were repeated difficulties in maintaining the portable pump's prime. Although more difficult and primitive, the collection process had to be modified to utilize a bucket and rope. With practice it was possible to swing the bucket under the 25 cm (10 in) landfill drainage collection pipe in a manner that would allow it to lodge and fill. A slight tightening of the rope would dislodge full pails of leachate that could be hoisted out hand over hand, and funneled into 22 liter plastic gasoline jugs. The air tight gasoline cans were transported approximately 5 miles back to the greenhouse where they were either used immediately or temporarily refrigerated @ 2^{o}C.

Prior to each days application of leachate to the various treatment beds, a 150 ml sample was taken from the raw leachate container and saved for analyses. Raw leachate and treated samples were analyzed for pH, alkalinity, Mn, K, Fe, Cu, Zn, Pb, Ca, Mg, Cl, TOC or COD.

Two weeks prior to treatment start-up, the 16 reactors were drained of their remaining irrigation water, and then watered daily with 4 liter alequots of tap water. Baseline samples of irrigation leachate alone were taken from all 16 reactors before leachate application began. These baseline samples actually represented approximately 8 months of bed ripening (i.e., peatmoss, reed canarygrass, and tap water interaction). It was presumed that these regular watering and drainage samples would provide the best basis for comparison with landfill leachate effluent after treatment in the same reactors. Twelve of the sixteen reactors were assigned batch detention times of one to twelve days, respectively. The remaining four beds were all operated with a one day detention time. All leachate applications were diluted by 50 percent with Amherst tap water prior to application. The assigned applications were all 4 liter treatments. Each application volume was prepared by mixing 2 liters of leachate with 2 liters of tap water. The mixture was sprinkled with a plastic watering can evenly over the appropriate reactor. Prior to the first application (March 20, 1985) the 16 reactors were allowed to gravity drain completely so as to eliminate the 2.5 cm sump of regular irrigation water. This was accomplished by lowering the flexible manometers below the reactor bottoms. After several hours of draining the manometers were returned to their upright positions, and leachate dilutions were added. It had been previously determined by preliminary evapotranspiration studies that a minimum of 4 liters would be needed to insure enough sample volume from the 8-12 day treatment beds. It had also been determined that 4 liters of liquid added to near field capacity beds would saturate the bottom 5 cm (2 inches) leaving approximately 20 cm (8 inches) of unsaturated peat.

Figure 3-3 illustrates the general configuration of the 16 reactors in the greenhouse. As previously mentioned, reactors #13-16 were operated with the same retention time as reactor #1 (i.e., 24 hrs). Reactors #2-12 were assigned retention times in days that corresponded to their numbers. Leachate addition and removal continued for a period of 38 days so as to insure a minimum of four samples from reactor number 12. Prior to each new addition of leachate, the treated leachate was removed by lowering the manometer which allowed the sump to drain freely into plastic collection jugs. After draining, the manometers were returned to the upright position and the new batch of leachate was added. Volume losses for each reactor were recorded for comparison with pan evaporation results that were being determined simultaneously in the greenhouse. Each composite sample was split into two 150 ml aliquots. After determining effluent pH, 1 set of samples was acidified with concentrated HNO_3 to a pH of between 2.0 and 2.5 for TOC and heavy metal

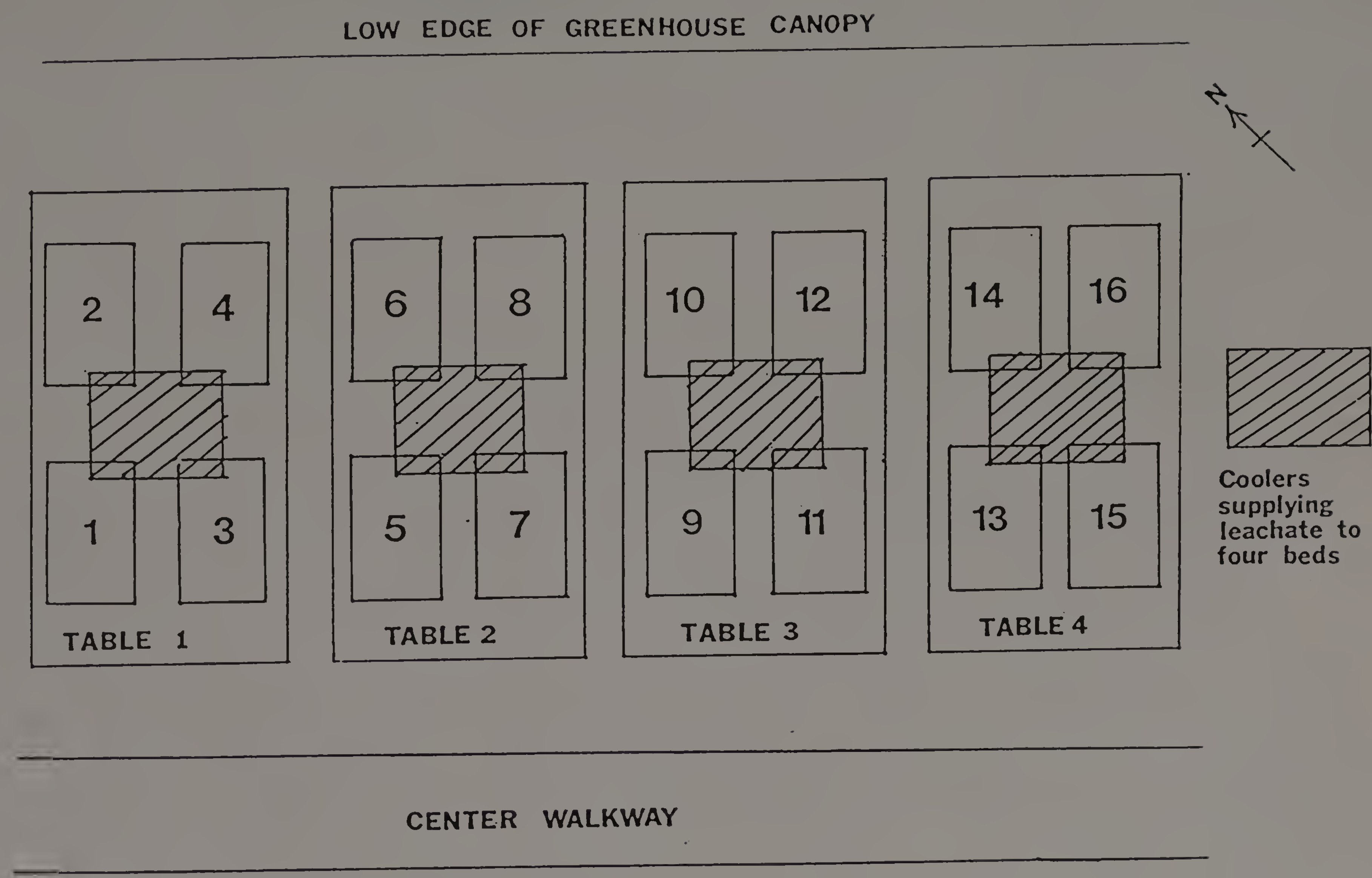


Figure 3-3 Configuration of 16 Greenhouse Reactors.

analyses. Both sample sets were refrigerated at 2°C for future analyses.

The following outline summarizes techniques and equipment used for chemical analyses:

1. Total Organic Carbon (TOC) - A Dohrmann Model DC80 Total Organic Carbon Analyzer was used for all TOC analyses. Acidified samples (pH 2.0-2.5) were either diluted or run full strength through the 40 ul or 200 ul channels. Each sample was injected 3 times, and the results were averaged.
2. Chemical Oxygen Demand (COD) - COD analyses were performed using the HACH (HACH Company, Loveland, CO) wide range (0-1500 mg/l) 2 ml microsample technique. Digestion and reflexing were accomplished using a HACH aluminum heat block (150°C, 302°F) and all digested samples were titrated with .125N Ferrous Ammonium Sulfate Standard Solution. Spectrophotomatic techniques were not used because of anticipated interferences with other leachate constituents.
3. Mn, K, Fe, Cu, Zn, Pb, Ca and Mg were analyzed with an Instrumentation Laboratory IL 551 A.A./A.E. Spectrophotometer (Instrumentation Laboratory Inc., Lexington, MA).
4. pH values were determined using a Fisher Accumet Model 805MP pH meter with standard pH electrodes (Fisher Scientific, Pittsburg, PA).
5. Chlorides were measured using a Buchler-Cotlove chloridometer with automatic titrater.

6. Other wet chemistry procedures followed EPA's Compilation of Methodology Used for Measuring Pollution Parameters of Sanitary Landfill Leachate, EPA 600/3-75-011 (Oct. 1975).

Results and Discussion

TOC Modeling

Table 3-3 summarizes the mean influent and effluent concentrations from twelve batch reactors with treatment times of one to twelve days, respectively. Although each reactor reduced TOC concentrations by more than 99%, the data is not representative of first order removal rates. Figure 3-4 illustrates ideal first order decay models for the 12 treatment periods assuming an initial TOC of 3075 mg/l (i.e., 50-50 dilution). Based on the data collected it must be concluded that if first order reductions of TOC did occur in the batch reactors, the reductions occurred in less than 24 hours. Additional runs with shorter detention times would clearly have been appropriate, but it was decided that the project proceed to continous flow applications so that the system could be stressed with higher application rates and shorter treatment times. Table 3-4 summarizes theoretical rate constants for each reactor based on influent and effluent concentrations. These values will be compared to plug flow treatment results later.

Leachate Toxicity

Within 24 hours of 50% strength leachate application, all of the reed canarygrass leaf blades browned and appeared dead. It should be noted that the grass was generally in a poor condition prior to any

Table 3-3. Summary of Mean TOC Concentrations for 12 Batch Reactors During 40 Days of Continous Operation.

Treatment time	Raw Leachate TOC	Mean Effluent TOC	Baseline TOC	Net Leachate TOC Remaining	Fraction of Net Leachate Remaining	TOC Removal
(days)	mg/l					(%)
1	6150	85.7	50.7	35.0	.0057	99.43
2	6150	83.5	50.7	32.8	.0054	99.46
3	6150	86.5	50.7	35.8	.0059	99.41
4	6150	76.2	50.7	25.5	.0041	99.59
5	6150	73.2	50.7	22.5	.0037	99.63
6	6150	65.2	50.7	14.5	.0024	99.76
7	6150	73.6	50.7	22.9	.0037	99.63
8	6150	69.0	50.7	18.3	.0030	99.70
9	6150	65.3	50.7	14.6	.0024	99.76
10	6150	67.1	50.7	16.4	.0027	99.73
11	6150	76.4	50.7	25.7	.0042	99.58
12	6150	45.5	50.7	0	0	100.00

TOC units expressed as mg/l
Baseline TOC = irrigation effluent before leachate treatment

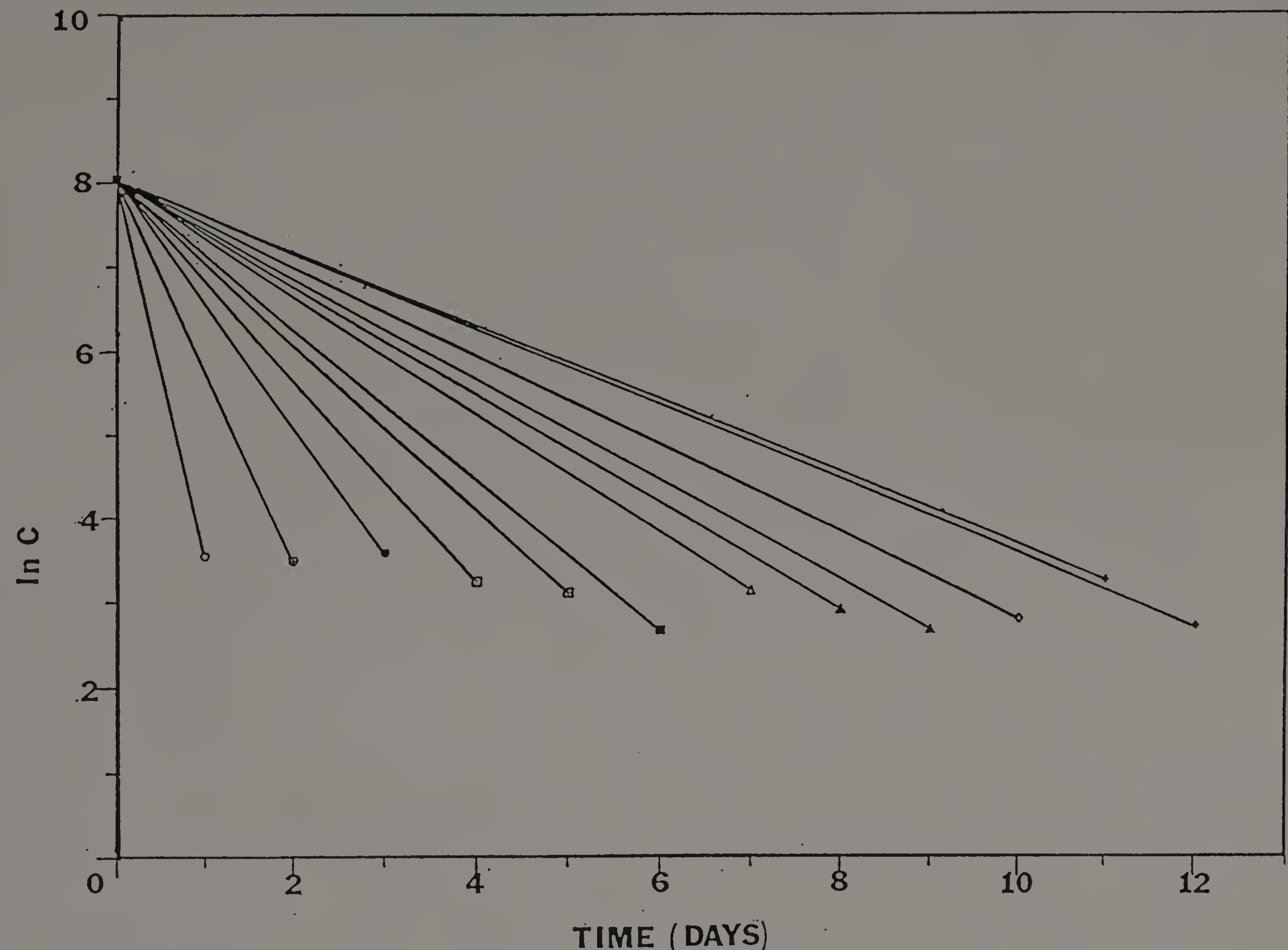


Figure 3-4. Ideal First Order Decay Models (0-12 Days Treatment Times).

Table 3-4. Theoretical Rate Constants for First Order TOC Decay.

Reactor $\#\bar{t}$ days	Effluent TOC mg/l	Baseline TOC mg/l	Net TOC mg/l C_t	Net $\ln C_t$	$-k$ $days^{-1}$
1	85.7	50.7	35.0	3.55	4.5
2	83.5	50.7	32.8	3.49	2.3
3	86.5	50.7	35.8	3.58	1.5
4	76.2	50.7	25.5	3.24	1.2
5	73.2	50.7	22.5	3.11	1.0
6	65.2	50.7	14.5	2.67	0.9
7	73.6	50.7	22.9	3.13	0.7
8	69.0	50.7	18.3	2.91	.64
9	65.3	50.7	14.6	2.68	.60
10	67.1	50.7	16.4	2.80	.52
11	76.4	50.7	25.7	3.25	.43
12	45.5	50.7	15.0	2.71	.44

First Order Rate Equation (Batch) $-k = \dfrac{\ln \left[\frac{C_t}{C_o}\right]}{\bar{t}}$

C_o = 3075 mg/l : $\ln C_o$ = 8.03

leachate application. During the 8 months of pretreatment preparation, the grass was generally chlorotic, and grew in small bunches covering approximately 50% of each growth bed. Its height never exceeded 15 cm (6 in). Pitcher plants and venus fly traps were as numerous naturally as the reed canarygrass. Clearly the environment provided by the peatmoss was not conducive to good grass growth. The addition of leachate seemed to make a poor condition worse. In spite of the sudden grass loss, leachate application continued based on the assumption that microbial activity within the peatmoss would be a principal factor in reducing leachate TOC.

After several weeks of repeated treatments, new reed canarygrass leaf blades began to emerge around the perimeter of most of the reactors. After several more weeks of leachate application, new grass shoots covered the entire seedbed. By the end of the study, a deep green succulent growth pattern characterized all of the reactors and stem height was nearly a meter in length. Clearly the leachate had changed the growing environment in a very positive way. The grass continued to flourish for the duration of the study and leachate toxicity responses never occurred again. Why the canarygrass behaved as it did remained unclear, but a special pot study was designed to evaluate the effects of other leachate dilutions and methods of application. That study and its subsequent findings are described in Chapter VI.

Table 3-5. Batch Treatment Analyses of Metal Concentrations in Reed Canarygrass Leaf Blades.

Reactor Number	Mn	K	Fe	Cu	Zn	Pb	Ca	Mg	pH
	mg/kg x 10^3								
Raw Leachate[d]:	85	-	875	19.5	85	80	600	150	5.7
Reactor:									
1	2.51	155.50	2.60	.445	.410	.34	24.35	1.12	
2	2.46	152.80	4.02	.353	.294	.34	25.25	.95	
3	1.63	122.20	2.70	.327	.282	.32	17.17	.90	
4	1.38	131.40	5.22	.249	.253	.34	16.01	1.14	
5	2.49	114.70	3.35	.289	.265	.32	16.90	1.35	
6	1.94	96.10	3.16	.346	.176	.33	11.00	.74	
7	1.84	108.00	3.07	.244	.196	.31	12.05	.64	
8	1.96	109.00	3.82	.336	.228	.32	16.82	1.06	
9	1.92	116.50	2.36	.305	.238	.31	13.30	.67	
10	1.42	115.90	2.67	.261	.202	.30	9.92	.65	
11	2.19	138.70	2.76	.306	.220	.34	13.24	1.14	
12	1.51	113.00	1.74	.234	.224	.32	9.49	1.70	
Intercept	**[a]	**		**	**		**		
Slope	*[b]	**	NS[c]	**	**	NS	**	NS	

Linear Regression Analyses
[a]Significant at P = 0.05
[b]Highly Significant at P = 0.01
[c]No Significance
[d]Raw leachate values as mg/l

Metals Concentrations

Table 3-5 compares metal concentrations found in raw leachate to those of reed canarygrass harvested at the end of the batch study. The data suggests that Mg and Pb concentrations were relatively unaffected by the various applications of leachate among reactors #1 through #12. For Mn, K, Fe, Cu, Zn and Ca there was a continuous decrease in metals concentrations as leachate application rates decreased from reactors #1 through #12. Comparing metal concentrations from this first cutting to those of the final cutting (Table 4-5) after 3 years of leachate application it seems evident that reed canarygrass is an effective sink for metals in the leachate (Table 3-6).

Summary

Although the batch study failed to provide data to support the first order decay model hypothesized, metals and TOC were effectively removed for all of the treatment times evaluated. The data suggested that if first order removals were occurring, it would be for treatment times less than 24 hrs. As expected, there seemed to be a severe toxicity response of the reed canarygrass to leachate. First indications were that the initial leachate sprinkling had killed all of the grass. After several weeks of continued treatments, however, young leachate tolerant leaf blades emerged first around the perimeter of each reactor, and eventually throughout the entire seedbed. Lush green color and succulent growth was observed for all of the treatments and grass grew to a meter in length by the end of the 40 day treatment period.

Table 3-6. Batch Treatment Analyses of Metal Concentrations in Raw Leachate and Treated Effluent.

Reactor Number	Mn	Fe	Cu	Zn (mg/l)	Pb	Ca	Mg	pH
Raw Leachate:	85	875	19.5	85	80	600	150	5.7
Reactor Effluent:								
1	.21	.76	.26	.30	.19	10.10	8.03	3.5
2	.13	.77	.47	.13	.21	12.61	11.63	3.6
3	.15	.74	.70	.93	.24	12.60	5.82	3.8
4	.10	.65	.60	.06	.25	11.54	6.56	3.8
5	.14	.72	.52	.05	.26	11.14	8.58	3.7
6	.13	.55	.60	.06	.40	13.80	7.41	3.6
7	.15	.53	.54	.05	.42	15.92	10.01	3.5
8	.14	.67	.46	.08	.39	16.15	9.66	3.5
9	.15	.51	.41	.08	.29	10.86	8.70	3.5
10	.15	.46	.49	.08	.23	19.11	9.40	3.5
11	.18	.57	.42	.10	.19	18.52	10.02	3.5
12	.43	.54	.66	.16	.18	30.99	11.20	3.3
								$\bar{x}$ = 3.6 SD = .14

Note: Reactor pH prior to leachate application: $\bar{x}$ = 3.7
SD = .13

The following conclusions were drawn from the batch study experiment.

1. The batch TOC treatment data did not model the first order model proposed, and as such, a single rate constant (k) could not be determined.
2. 99% removals of TOC for all of the treatment times suggested that removals were occurring very rapidly (i.e., less than 24 hrs).
3. There was a 99%$\pm$ removal of the leachate metals Mn, Fe, Cu, Zn, Pb, Ca and Mg for all of the treatment times (i.e., 1-12 days).
4. Reed canarygrass leaf tissues accumulated increased amounts of the metals Mn, K, Fe, Cu, Zn and Ca as the total volume of leachate application increased.
5. The metals Mg and Pb did not show an accumulation response to increased volumes of leachate.
6. Although the reed canarygrass showed immediate signs of "burning" after the first leachate application, it recovered extremely well and reached final harvest heights of nearly 1 meter. It was therefore concluded that peatmoss and reed canarygrass provided an excellent treatment media for landfill leachate.

CHAPTER IV

GREENHOUSE BENCH SCALE CONTINUOUS FLOW REACTOR ANALYSIS

Introduction

Previous batch studies indicated that the peatmoss and reed canarygrass treatment media could effectively remove 99% of the TOC in landfill leachate; and that this could be accomplished in a relatively short period of time (i.e., 1-12 days). These preliminary findings agreed well with treatment times and application rates found in the literature (93, 94, 82, 87), but they did not fit the first order model hypothesized earlier. Based on Figure 3-4 it seemed reasonable to conclude that if first order reductions for TOC were occurring, they had to be occurring during the first 24 hour treatment period. Considering the potent nature of landfill leachate, there was difficulty in accepting the theory that treatment could occur that rapidly. Other root zone methods (RZM) have typically treated wastewater in a 5-10 day period (80, 82, 84) and the concentration of TOC in wastewater is about one order of magnitude less than the batch study leachate. There were, however, some compensating considerations that suggested less than 24 hour retention times were possible:

1. The degradable constituents of leachate are basically volatile organics in solution form. As such they provide a substrate

that is very amenable to microbial degradation. Domestic wastewater, on the other hand, has considerably lower organic concentrations and much of the organic material is incorporated into wastewater solids, making it more difficult for microbes to degrade.

2. During the first few hours of leachate exposure to the atmosphere, it has been observed that rapid changes in leachate chemistry occur (38). Even the leachate pumping experiences described in Chapter III provide testimony to the fact that leachate is very unstable, and in part extremely volatile. Considering the watering technique used in applying the batch leachate, it seems reasonable to conclude that a considerable fraction of the solution TOC could have volatilized during the application process and immediately after application. Odors in the greenhouse would also support that theory.

3. If the first order model (eq 3-2) is inspected, it becomes immediately evident that the efficiency of treatment (i.e., C_t/C_o) for any unit of time is directly related to substrate concentration. That is, in fact, a property of the first order reaction that sets it apart from other rates that are not concentration dependent (i.e., zero order). If leachate TOC did decay according to first order kinetics in the batch study, the rate of reduction $\frac{-dC}{dt}$ (eq 3-1) would be most rapid during the first few time increments. Considering that the TOC of the applied leachate mixture was more than 3,000 mg/l, the majority

(i.e., 99%) of this concentration could be treatable in hours instead of days as evaluated.

4. Most RZM systems operate the treatment bed under saturated or flooded conditions. The only oxygen supply available to facilitate aerobic degradation comes from plant roots that translocate excessive amounts of O_2 from the leaf area. The peatmoss and reed canarygrass beds were always operated in an unsaturated mode. The 2-5 cm sump at the bottom of each reactor transported leachate by capillarity into the 20 cm of peat above. As such the transport of O_2 and the ease of organic volatilization, as previously discussed, may have been greatly enhanced.

For the above reasons it was decided to move forward with continuous flow studies in two simultaneously different ways.

Earlier personal communications with soil microbiologist, Dr. Haim Gunner, suggested that a perfusion study might provide more reliability and control in the treatment process. Little was known, at the time, of the role reed canarygrass would play in treatment, so it was decided to use 3 bench scale perfusion units without the canarygrass to evaluate treatment times that could be easily monitored on an hourly basis. It was hoped that after establishing a microbial population, the perfusion units could be used to provide more reliable kinetic data. The results of this project are discussed in Chapter V.

While continuing with the short treatment time-small volume perfusion study, the 16 larger greenhouse reactors were modified for plug flow applications of landfill leachate that would stress the system to failure, and that would provide data for less than 24 hour treatment times. To accomplish these objectives it was decided to conduct two separate plug flow studies that would evaluate high rate applications, and more moderate rates. It was also decided that application rates would be replicated so that reactors with the same rates could be compared. The limited number of reactors, and difficulty in collecting leachate necessitated that the studies be conducted in sequence rather than at the same time. The high application rate study was conducted over a 60 day period during the fall of 1987, and the moderate or low rate study followed over a similar time period in the spring of 1988. For each study, application rates were replicated four times in four different greenhouse locations. As such, each study was only able to evaluate 4 different rates. For the high rate treatment, rates of 200, 400, 600 and 800 cm^3/hr were selected. These rates extrapolate to 4.8, 9.6, 14.4 and 19.2 liters/day, respectively. If the rate of the shortest treatment time batch reactor is compared (2 liters/day), it is evident that the high rate plug flow application rates were approximately 2.5, 5, 7, and 9.5 times greater.

The moderate or low rate study used application rates of 100, 200, 300, 400 cm^3/hr, and these values extrapolate into rates of 2.4, 4.8, 7.2 and 9.6 liter/day. It is from the combination of rates selected that treatment applications ranged from approximately that of the 24 hr batch reactor to one that was nearly an order of magnitude greater.

Objectives

This study attempted to accomplish two general objectives simultaneously. Firstly, there was a need to operate the treatment units in a continuous flow manner. Earlier difficulties with establishing a healthy stand of reed canarygrass resulted in the decision not to dismantle the treatment reactors for peat analyses; but instead, to use the acclimated batch systems for continuous flow applications.

Secondly, there was a need to evaluate retention times of less than 24 hours. Batch experiments had not provided kinetic data to support the first order model originally hypothesized, because 99% removals had occurred even with the shortest treatment times. By using variable flow rates, it was anticipated that effluent TOC could be controlled and selected over a full range of concentrations between that of raw leachate and a highly treated effluent. The specific objectives of this study were as follows:

1. To develop a reliable method for applying leachate in a continuous manner to 16 greenhouse reactors.
2. To develop techniques that would provide reliable control of leachate flow rates.
3. To select application or flow rates that would be representative of a wide range of effluent concentrations and treatment times.
4. To test the first order model, and to determine an appropriate rate constant (k).

5. To measure any additional accumulation of the metals Mn, K, Fe, Cu, Zn, Pb, Ca and Mg in leaf tissue of reed canarygrass.

Methods and Materials

High Rate Continuous Flow Study

Following the completion of the batch treatment study, the sixteen reactors were modified slightly to facilitate a continuous application of leachate. This was accomplished by assigning each reactor within clusters of four a different application rate, and by repeating the pattern for the remaining clusters (Fig. 3-2). The figure also illustrates where leachate supply reservoirs were located for each of the greenhouse benches being used. Table 4-1 summarizes flow rates assigned to each reactor.

Corresponding daily rates of 4.8, 9.6, 14.4 and 19.2 liters per reactor meant that a 24 hour leachate supply of 48 liters (12 gal) per reservoir was needed, or a total of nearly 200 liters (50 gal) per day for all 16 reactors. This required that daily trips to the same landfill used for the batch study be made. The same 22 liter (5.5 gal) plastic gasoline cans were used to transport leachate in an air tight manner. Each storage reservoir (48 liter coolers) needed to be filled twice daily in the greenhouse. Leachate was supplied to the four reactors in each cluster by a manifold that split a main reservoir discharge line into 4 smaller feed lines. Each reactor was equiped with a Clayton-Mark Model 1700A Chemical feed pump calibrated to deliver the prescribed flow.

Table 4-1. Application Rates for High Rate Continuous Flow Study.

Reactor Number	Flow Rate (cm^3/hr)	Reactor Number	Flow Rate (cm^3/hr)
1	200	9	200
2	400	10	400
3	600	11	600
4	800	12	800
5	200	13	200
6	400	14	400
7	600	15	600
8	800	16	800

NOTE: 200 cm^3/hr = .65 cm/day = 235 cm/yr.

Inlet ends of the reactors were fitted with 7 mm "T" shaped distribution headers that extended approximately 60 cm (24 in) across the reactor's width. The headers were drilled on two sides with 1 mm holes at 2.5 cm (1 in) intervals along their entire length. Ends were plugged to insure uniform distribution. The headers were set approximately 2.5 cm (1 in) below the surface of the peat. Tubing and connectors used for this study were also of the standard maple syrup collection type, and they performed extremely well over the entire period of the project. The same cannot be said for the Clayton-Mark pumps, which repeatedly malfunctioned due to poor foot value designs.

High rate leachate application continued for more than 60 days, but sample collection and analysis was restricted to the last two weeks. During the first 45 days reactors processed leachate continually and treated leachate was discharged onto the sand greenhouse floor. It was assumed that during these 45 days each reactor would reach a steady state condition with respect to microbial growth rates, leachate influent and effluent concentrations and leachate flow rates. Sampling was accomplished using 120 ml plastic containers with plastic screw caps. These wide mouth beaker type containers and lids worked extremely well and made sampling very easy. By drilling a 7 mm hole through the center of 16 screw-caps and then force fitting them to the ends of the maple syrup discharge lines it was possible to screw on the 120 ml base which could then be left dangling as it filled. It should be noted that each discharge line was held 2.5 cm (1 in) above the reactor base by an electrical staple driven into the reactor side. This provided a 2.5 cm

(1 in) sump to insure uniform capillary wetting for all of the peat. It often took more than one hour to collect a sample from the slower reactors, but a screw lid and container could be left to overflow, for later pick-up. By carefully removing the filled sample vials and securing them with an undrilled air tight screw cap they could be labeled and stored at $2^{o}C$ for analyses.

Moderate Rate Continuous Flow Study

In the spring of 1988, when leachate flows at the landfill increased, a moderate rate continuous flow study was conducted. Nearly identical procedures were used as those in the high rate study, but the Clayton-Mark pumps were replaced with Chem Feed Flex flow model A-114-4 units. The replacement pumps were of a peristaltic design free of values and blockage points. The units were easy to calibrate, and held close flow tolerances for extended periods of time. Table 4-2 summarizes the 4 application rates used for the 16 reactors.

Corresponding daily application rates for the 100, 200, 300, 400 cm^3/hr treatments were 2.4, 4.8, 7.2 and 9.6 liters/day, respectively. Total daily leachate requirements were only 24 liters per day, which provided slightly less demanding collection and supply requirements. After 45 days of continuous operation, sampling was again begun on an alternate day basis. Samples for both continuous flow studies were analyzed for COD using the Hach microsample (2 ml) technique.

Metals Accumulation

Following the two continuous flow studies, grass samples were cut, dried, milled and digested with perchloric acid so that the metals Mn,

Table 4-2. Application Rates for Moderate Rate Continuous Flow Study.

Reactor Number	Flow Rate (cm^3/hr)	Reactor Number	Flow Rate (cm^3/hr)
1	100	9	100
2	200	10	200
3	300	11	300
4	400	12	400
5	100	13	100
6	200	14	200
7	300	15	300
8	400	16	400

NOTE: 200 cm^3/hr = .65 cm/day = 235 cm/yr.

K, Fe, Cu, Zn, Pb, Ca and Mg could be analyzed and compared with batch study values. The same IL 551 atomic absorption spectrophotometer was used for all samples.

Results and Discussion

High Flow Study

Treatment reactors for each study received leachate applications for a period of 45 days before any sampling was attempted. During that time period it was assumed that microbial populations were being established based on the available substrate. High rate applications of leachate (200, 400, 600 and 800 cm^3/hr) were applied first, and a fixed head of 2.5 cm (1 in) was maintained in each reactor to insure uniform wetting of the unsaturated peat.

Although the Clayton-Mark Model 1700A chemical feed pumps performed extremely well during the initial flow calibration period using tap water, they proved to be extremely poor units, for applying leachate to the treatment beds. The problem seemed two fold. Firstly, the volatile components of leachate, readily come out of solution as leachate warms. The vapor pressure has been known to deform leachate storage containers, and it is strong enough to force lids from plastic milk jugs. In the leachate supply lines, and in the head assemblies of the Clayton-Mark pumps, vapor bubbles regularly formed, breaking the liquid prime which would then produce either altered flow, or none at all. Secondly, reduced iron in the leachate (approximately 1000 mg/l), oxidizes quite rapidly after being removed from the landfill environment. Iron oxide precipitate on the value seats and "O" ring value stems regularly caused problems with pumping. The combined effect of precipitation of Fe and volatilization of organics made leachate flow control extremely difficult. Each pump had to be checked several times a day and cleaned when needed. To reduce the supply line vapor problem, dry ice was set in the tops of the supply coolers, and supply lines were shortened to a minimum length. It also became necessary to schedule the two studies during periods of the year when greenhouse temperatures tended to be cooler (Fall & Spring). In time, it was possible to anticipate flowrate upset and take appropriate remedial action in advance. The collection of more than 200 liters per day of leachate and the twice a day filling schedule also provided an ideal opportunity to check the pumps regularly. Table 4-3 summarizes the effluent COD data for the 4 flow rates used. Each mean value represents an average of 24 samples. Table 4-4 summarizes flow rate data used for both the high and moderate treatment studies.

Moderate Flow Study

Due to the pumping difficulties encountered with the high rate study, pumps for each treatment reactor were replaced by ones with a peristaltic design. The new pumps performed extremely well during the entire study. The more moderate application rates of 100, 200, 300 and 400 cm^3/hr reduced total daily leachate needs by 50% and vapor binding in the supply lines were minimized.

Treatment beds were again allowed to reach steady state conditions over a 45 day period. In April of 1988 sampling began on an every other

Table 4-3. COD Effluent Concentrations for 6 Continuous Flow Applications of Landfill Leachate to Peatmoss and Reed Canarygrass Treatment Beds.

Flow Rate cm^3/hr	Flow Rate l/day	Effluent COD mg/l	ln C COD	Hydraulic retention time t = days
100	2.4	225	5.42	.80
200	4.8	452	6.11	.40
300	7.2	1068	6.97	.26
400	9.6	1401	7.24	.20
600	14.4	2448	7.80	.13
800	19.2	2820	7.94	.10

NOTE: COD are based on multiple samples from replicated reactors.

$$t = \frac{\text{volume}}{\text{flow rate}}$$

Table 4-4. Summary of Six Application Rates Used in Two Continuous Flow Studies.

— Pump Rate —		——— Application Rate ———			Annual Precipitation Ratio
cm/hr	l/day	cm/day	cm/year	in/yr	
100	2.4	.32	120	47	1.2
200	4.8	.65	235	93	2.3
300	7.2	.97	353	140	3.5
400	9.6	1.29	470	185	4.7
600	14.4	1.94	706	278	7.1
800	19.2	2.58	942	370	9.4

NOTES: Annual Precipitation Ratio
(1) based on 100 cm/yr of rainfall
(2) reactor areas = 7442 cm^2

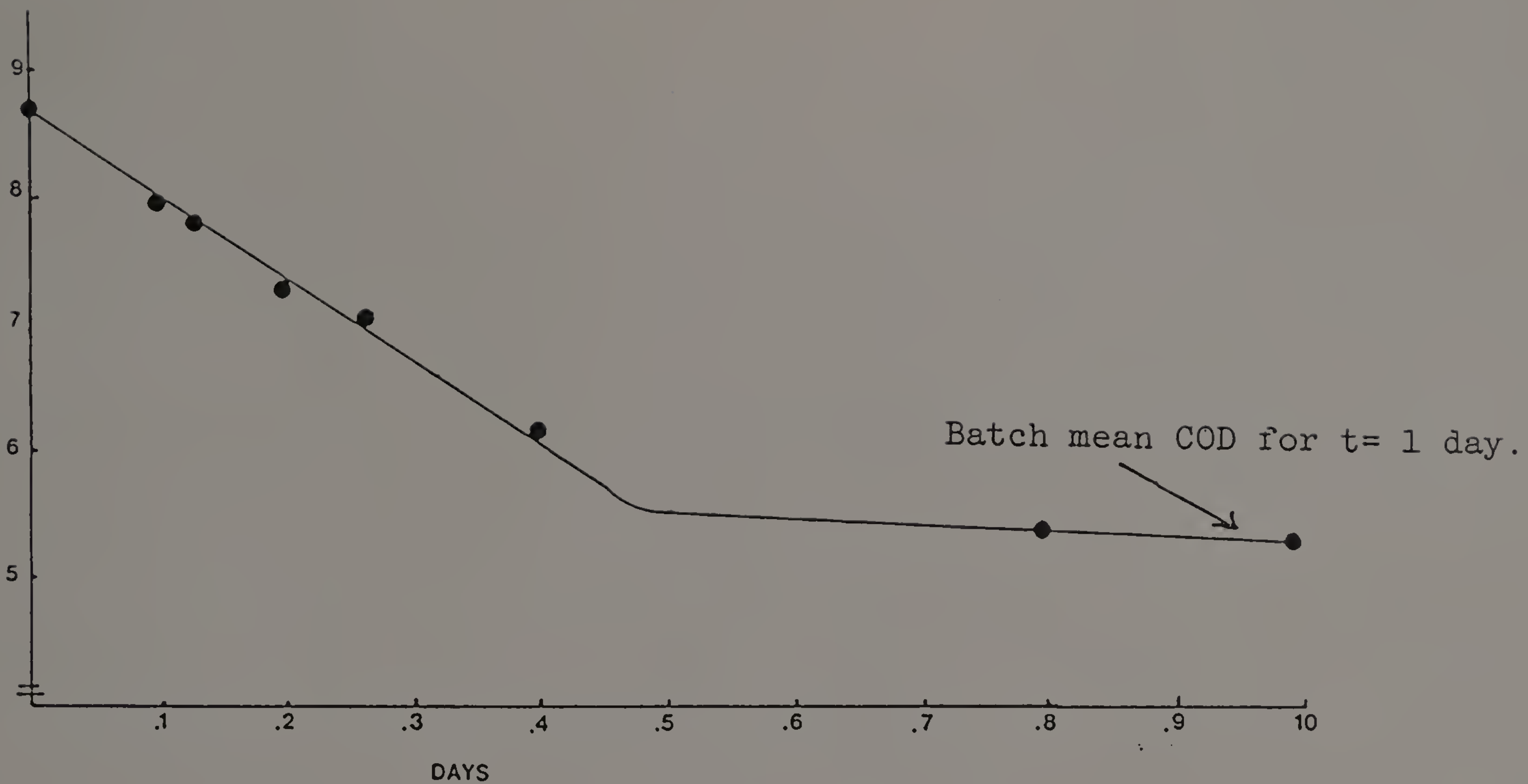
day basis for a period of approximately 3 weeks. Thirty-six samples were collected for each application rate and COD results were averaged. Sample collecting from the 100 cm^3/hr reactors was often difficult, due to the high rate of evapotranspiration. It may be reasonable to conclude that at this application rate (.32 cm/day), most of the applied liquid is converted to vapor. Pan evaporation studies in the greenhouse during the same time period indicated that an average .3 cm/day was evaporating from the water surface, so that a total evapotranspiration loss of .32 cm/day would not be unreasonable.

Another problem occurring at increasing frequencies during the final stages of plug flow treatments was clogging. The reactor manometers had to be watched closely to insure that liquid levels in the reactors did not exceed 2.5 cm (1 in). On several occasions higher saturation heights were observed and excess fluids had to be removed by hand suctioning. When the reactors were finally dismantled, reed canarygrass roots were found in the drain lines of the clogging units.

COD results for each of the application rates were averaged and are reported in Table 4-3.

Upon completion of the second continuous flow (plug flow) study, the 16 reactors were allowed to drain and dry. Reed canarygrass was harvested, oven dried, milled and digested with perchloric acid as before and analyzed for metals. The leaf concentrations of Mn, K, Fe, Cu, Zn, Pb, Cu and Mg are reported in Table 4-5.

Figure 4-1 illustrates COD data from the two continuous flow studies using the first order model (eq 3-2).



Note: First order decay in first 0.4 days i.e. 9.4 hours as proposed.

$$k = \frac{\text{Rise}}{\text{Run}} = \frac{8.72 - 6.11}{0.4} = 6.53 \text{ days}^{-1}$$

Figure 4-1. First Order Decay of Landfill Leachate COD in Two Plug Flow Reactor Studies.

Table 4-5. Analyses of Metal Concentrations in Reed Canarygrass Leaf Blades After 3 Years of Leachate Application.

	Mn	K	Fe	Cu	Zn	Pb	Ca	Mg	pH
	mg/Kg x 10^2								
Raw leachate	85	-	875	19.5	85	80	600	150	5.7
Reactor #									
1	4.03	316.00	10.68	.244	1.19	.02	69.90	23.40	
2	3.48	191.50	7.78	.176	.865	.01	42.30	16.65	
3	2.73	195.63	5.08	.242	.953	.01	46.35	22.80	
4	4.23	310.25	41.38	.272	.925	.02	107.10	32.95	
5	2.98	386.88	5.90	.199	.898	.03	57.40	28.40	
6	3.80	267.38	4.40	.168	1.06	.03	50.35	31.45	
7	3.10	246.75	14.95	.321	.988	.04	58.30	22.80	
8	5.08	204.50	9.23	.126	.950	.02	55.45	22.20	
9	3.10	321.13	5.58	.177	.803	.02	52.85	21.55	
10	4.38	165.75	7.75	.162	1.11	.02	53.60	23.90	
11	2.80	239.00	5.13	.221	.773	.03	44.50	24.20	
12	2.73	158.38	8.98	.177	.820	.01	43.30	20.40	
13	3.30	392.75	4.50	.187	.820	.03	42.40	26.15	
14	2.93	195.25	6.25	.155	.770	.02	23.40	18.80	
15	3.15	225.75	7.95	.142	.923	.03	52.55	19.50	
16	3.40	306.13	14.40	.154	.930	.02	61.65	19.20	

Note: Mean effluent pH from reactors during continuous flow operations = 7.7 SD = .912.
Raw leachate values as mg/l.
Reactor pH prior to leachate application: $\bar{x}$ = 3.7
SD = .13

$$\ln C_t = -kt + \ln C_o \qquad \text{(eq 3-2)}$$

The data indicates that for treatment times of less than .5 days the model adequately describes changes in landfill leachate TOC. A rate constant of 6.53 $days^{-1}$ was determined. After .5 days, the model became asymptotic as with the soil perfusion data reported in Chapter V, Figure 5-2. It should be noted, however, that the rate constant for this study with reed canarygrass present was more than an order of magnitude greater than that of the perfusion study (6.53 $days^{-1}$ vs. 43 $days^{-1}$). Figure 4-1 also shows the ln C data point for the batch reactor with a 24 hour treatment time. It seems evident that the longest treatment time for the continuous flow study (i.e., .8 days) had a COD effluent that approached the value reported for the shortest treatment time in the batch study (i.e., 1 day). As proposed in Chapter III it also appears that first order decay occurs very rapidly after leachate has been applied to the reed canarygrass and peatmoss treatment units. Comparing these results to those reported by Cooper et al. (80) for treatment of domestic sewage, the time required for a 90% reduction of leachate COD is generally an order of magnitude less. Realistically, however, this study was conducted in a greenhouse environment under near optimum conditions, and it may be unrealistic to compare data with in situ systems.

<u>Metals Analyses</u>

Metal concentrations found in reed canarygrass leaf tissue increased considerably over the life of the project. Unfortunately it was not possible to associate tissue concentrations with leachate application rates due to the fact that each reactor was used for 3 different studies. Figure 4-5 summarizes values for Mn, K, Fe, Cu, Zn, Pb, Ca and Mg after 3 years of intermittent leachate application. If these values are compared to those reported in Table 3-5, it is evident that concentrations of Mn and K increased nearly 2X, Fe concentrations increased more than 3X and Zn concentrations increased approximately 4X. Calcium increased 3X and Mg increased 20X. Pb and Cu remained relatively unchanged over the entire study and this may be due to low initial concentrations in the leachate (Figure 2-8).

As concluded earlier reed canarygrass appears to be an excellent sink for the removal of metals analyzed in this leachate source.

Summary

This continuous flow study demonstrated that first order kinetics can be used to model COD reductions in peatmoss and reed canarygrass greenhouse reactors. Treatment times of less than .5 days were needed, however, which was significantly shorter than originally expected. Comparing the rate constant for this study (6.53 $days^{-1}$) with that of the soil perfusion study (.43 $days^{-1}$), it is evident that reed canarygrass was a vital component to the treatment process. The only clogging encountered with the greenhouse reactors occurred in the outlet lines where roots had grown. It therefore appears that in the root zone area, reed canarygrass also helped in maintaining a desirable hydraulic conductivity for the higher application rates.

It was not possible to correlate application rates with metal concentrations in the canarygrass leaf blades, but cuttings before and after the continuous flow study clearly indicated that reed canarygrass may be an excellent sink for the storage and removal of Mn, K, Fe, Zn, Ca and Mg.

The specific conclusions reached may be summarized as follows:

1. Leachate COD reductions measured fit the first order model hypothesized.
2. The kinetic rate constant was determined to be 6.53 $days^{-1}$.
3. First order reduction in TOC occurred in reactors with less than .5 days retention time.
4. The leaf blades of reed canarygrass concentrated the metals Mn, K, Fe, Zn, Ca and Mg most effectively. Pb and Cu did not appear to concentrate in the grass shoots.
5. Reed canarygrass rhizomes proliferated and spread through all of the treatment beds.

The network was so extensive that the peatmoss came out of the reactors as a single "block" when they were dismantled. Based on the importance associated with root zone ecology in other RZM studies (80, 82, 84, 85, 87) it seems reasonable to conclude that Phalaris contributed significantly to the removal rates measured.

CHAPTER V

CONTINUOUS FLOW PERFUSION STUDY

Introduction

Perfusion Apparatus

Development of the soil perfusion technique occurred during the late 1940's and early 1950's, for the purpose of accurately studying the metabolic events that take place in a soil environment (95). In 1946 Lees and Quastel (96) described an apparatus developed by Audus (97) that applied differential suction across a soil column in such a way that intermittent volumes of bubbles and solution would be lifted and dropped onto the column. The solution was then able to percolate down through the soil column to a recycle reservoir. Figure 5-1 illustrates the general arrangement of components in the Audus soil perfusion apparatus. Continuous suction is applied at A, and the two resistance tubes R_1 and R_2 (usually thermometer tubing) distribute a differential suction across the soil column P. The greater negative suction at G is transmitted through the soil column to the delivery tube T. A column of solution is drawn up T from S until S is empty. When S empties, atmospheric air enters through S breaking the vacuum or suction in T. During the following time incriment (approximately one second), a 10$\pm$ cm column of solution travels over the top of T into the small reservoir

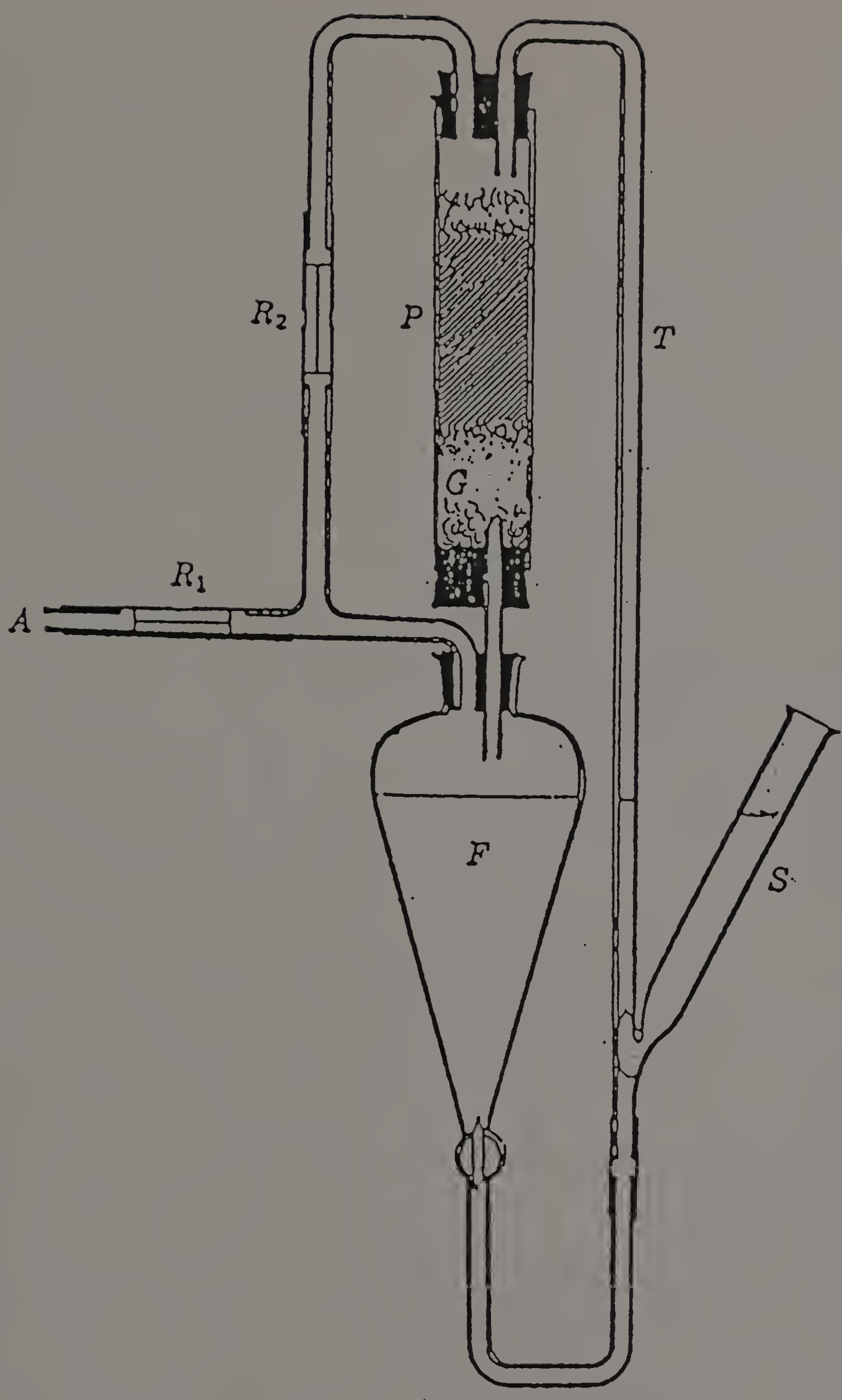


Figure 5-1. Soil Perfusion Apparatus. (After Audus (97))

A = Vacuum line, R_1 & R_2 = Resistance tubes,

P = Soil column, F = Leachate supply reservoir,

T = Delivery tube, S = Air intake and sampling port,

G = Porous packing

above the soil column. Simultaneously fluid flows from F to S re-establishing the vacuum and the cycle is repeated. Under ideal conditions, each column of solution is followed by a 10± cm column of ambient air, and another 10± cm column of solution. The frequency of 10 cm solution doses to the soil column is typically 25-30 per minute. By knowing the diameter of T, the length of the solution and air bubbles and the frequency of each, it is theoretically possible to calculate the flow rate of solution and air applied to the perfusion column. The rate of flow can be controlled, somewhat, by adjusting the suction force at A. Soil column P is typically 5-7 cm in diameter and some 30-50± cm in length. A 500 ml separatory funnel F, can be used as a reservoir. By inspection it can be seen that the liquid levels in F and S are the same and the apparatus must be watched regularly to insure that S neither overflows nor runs dry because of low reservoir depths. Samples (small) may be removed by pipette from S for analyses, but sampling will alter the total volume present.

Objectives

As mentioned in Chapter IV, it was decided to use a perfusion type apparatus to evaluate landfill leachate treatment in a peatmoss column. It seemed reasonable to conclude that the closed nature of the system would simulate a batch reactor where changes in COD concentration could be monitored over short increments of time. The primary objective for using the apparatus was to collect a series of samples that would measure concentration changes with time. The data would be used to test the first order decay model discussed in Chapter III.

The objectives may be summarized as follows:

1. To construct a laboratory treatment system modeling the greenhouse reactors that would provide improved control of the treatment process.
2. To collect treated leachate samples over an uninterrupted treatment period of 1-10 days.
3. To analyze treatment data by testing it with a First Order Kinetics model.
4. To evaluate the rate constant (k) based on the First Order Model $\ln C_t = -kt + \ln C_o$

Methods and Materials

Three soil perfusion units similar to the one illustrated in Figure 5-1 were constructed using regular laboratory glassware. Vacuum was provided by a variable suction pump. Glass columns 4.7 cm in diameter were packed with 95 g of air dried sphagnum peatmoss to a depth of 45 cm. The peat filled column volume was 780.7 cm^3 giving a density of .12 g/cm^3 which was approximately the same as that of the greenhouse reactors (.14 g/cm^3).

Previous wetting studies had indicated peatmoss would absorb nearly 80%$\pm$ of its volume, so it was concluded that a wetting period would be needed prior to data taking. A 500 ml aliquot of landfill leachate was added to each of the three separatory funnels, and all 3 columns were allowed to saturate for a 2 week wetting period. The wetting was accomplished by lowering the columns so that their tops were level with the leachate surface in the separatory funnels. After wetting, an additional 500 ml aliquot was added to each reservoir so that a total of 1000 ml of landfill leachate was contained within each unit, with approximately 500 ml in the soil column itself and 500 ml in the separatory funnel reservoir. In July of 1986 sample taking began.

All 3 units were run as replicates of each other and 5 ml samples were always taken at the same time. The Hach microsample technique was used to analyze for COD. Replicate sample data was averaged for plotting purposes. The units were allowed to run for 10 days and sample volumes totaling 50 ml for each unit were removed. Unless otherwise specified all materials and methods used were the same as those described more thoroughly in Chapters III and IV.

Results and Discussion

The perfusion apparatus illustrated in Figure 5-1 performed well during early operation. Attempts were made to quantify an application rate for the recycled leachate into the peat. Counting leachate bubbles ascending the column indicated an average frequency of 25 bubbles per minute. Typical bubbles were 10 cm long and they traveled through a glass tube 3 mm in diameter. It was calculated that an average bubble contained .7 cm^3 of leachate and that 25 of them a minute would deliver approximately 1000+ cm^3/hr to the peatmoss column. Given that the column cross sectional area was 17.35 cm^2, a mean velocity through the peat was calculated based on the Continuity Equation 5-1.

$$Q = A_{xs} \times \bar{v} \qquad \text{(eq 5-1)}$$

where: Q is a flow rate cm^3/hr

A_{xs} is the cross-sectional area cm^2

$\bar{v}$ is the average pore velocity cm/hr

therefore: If $Q = 1000$ cm^3/hr

$A_{xs} = 17.35$ cm^2

$\bar{v} = 57.64$ cm/hr

The column length was 50 cm long which would indicate that approximately one unit volume would pass through the peat each hour. It was therefore concluded that in the course of a day 24 liters of leachate would pass through the column. The entire system originally contained 1 liter of leachate, which indicates that a recyle ratio of 24:1 was occurring. Alternatively it could be stated that the average drop of leachate spent a half hour in the peat column, a half hour in the reservoir and that it repeated this cycle 24 times a day.

Unfortunately, after a week of continuous operation, the units became extremely difficult to operate. Bubble movement was slowed considerably and eventually stopped. All indications were that clogging had occurred and that the column was impermeable to leachate or air or both. At approximately the same time it was observed that sections of the peatmoss column began to separate leaving horizontal cracks in the previously uniform peatmoss. It was concluded that the differential suction was actually moving sections of peat instead of air and liquid.

Fortunately, 10 samples had been collected from each of the columns before they failed and that data is reported in Table 5-1. Figure 5-2 illustrates a ln C vs. t plot of the averaged COD values. The plot clearly suggests two periods of COD change within the columns. During the time period of 0-3 days, changes in COD concentrations fit the first order decay equation proposed in the objectives, with a k value of .43 $days^{-1}$. Following day 3, the data became asymptotic with little or no apparant change. There are two possible explanations for this. Day 1-3 treatment exposed the entire leachate volume (1 liter) to the peatmoss column once each hour. At the same time alternating bubbles of air were drawn into the column, facilitating some oxidation activity and possibly some aerobic decay of the leachate by microorganisms. During that time period COD values decreased from 6300 mg/l to 1800 mg/l. By day 2 iron oxide precipitate was clearly forming in the glass wool tuft at the top of the column and by day three leachate bubbles had slowed considerably. The columns appeared clogged and the addition of air had nearly stopped. Although sampling continued for the next several days, it appeared that treatment had ceased and that the system had gone anaerobic. The lack of regular aeration through the S tube intake (Figure 5-1) and the absense of reed canayrgrass which aerates and maintains hydraulic conductivity in the peat clearly impacted the treatment process negatively. After two additional weeks of trying to restore normal bubble movement by adjusting suction amounts, the apparatus was

Table 5-1. Seven Day Changes in Landfill Leachate COD Treated in Perfusion Columns.

Date	Lapsed Time $\bar{t}$ days	Initial COD mg/l	COD at Time $\bar{t}$	Percent Reduction
7/14/86	0.0	6310	6310	0.0
7/14/86	0.2	6310	4896	22.4
7/15/86	1.0	6310	4020	63.7
7/15/86	1.2	6310	3750	40.6
7/16/86	2.0	6310	2693	57.3
7/17/86	3.0	6310	1766	72.0
7/18/86	4.0	6310	1800	71.5
7/19/86	5.0	6310	1539	75.6
7/20/86	6.0	6310	1690	73.2
7/21/86	7.0	6310	1333	78.9

*Values not adjusted for peatmoss contribution to COD.

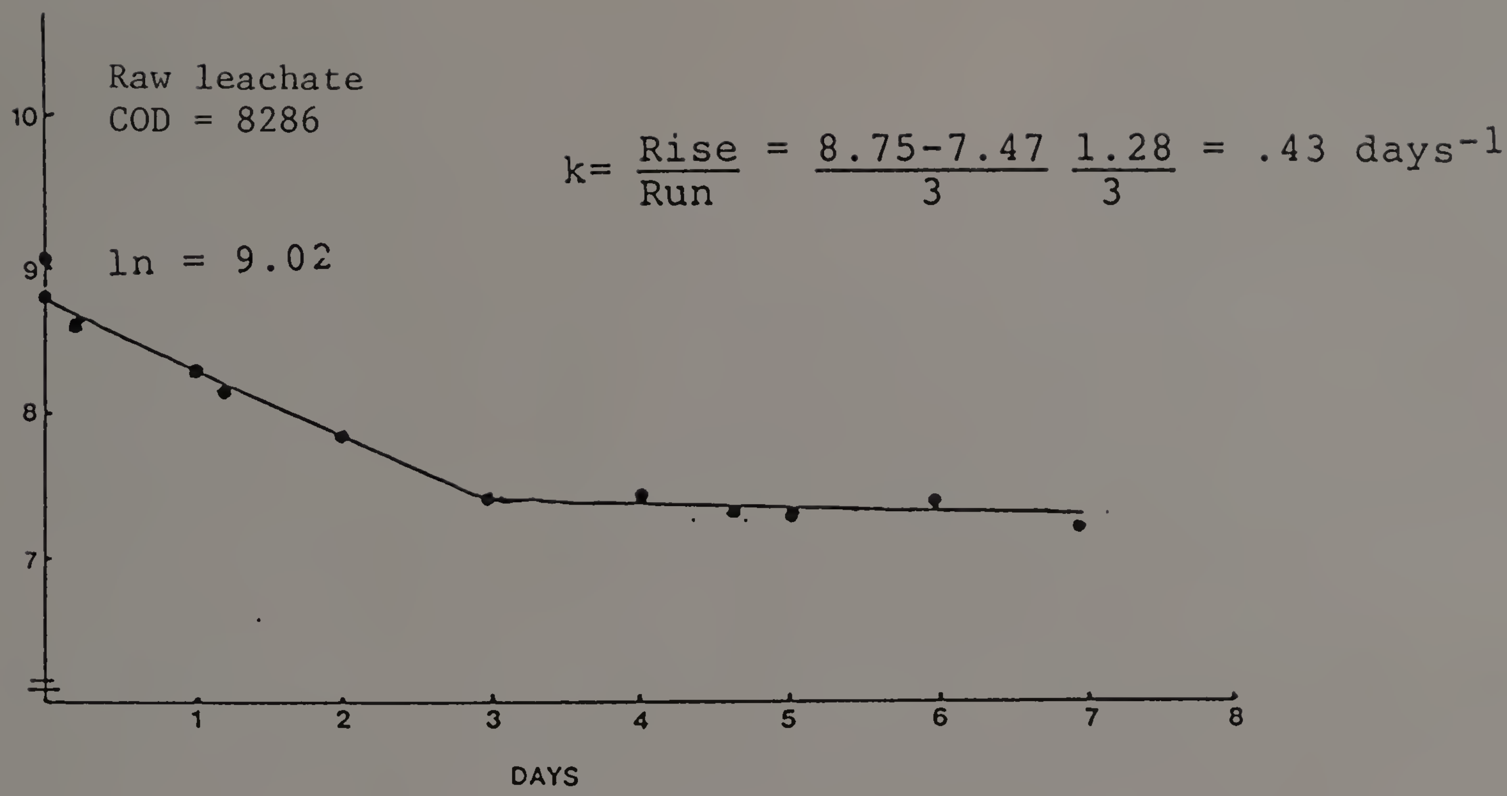


Figure 5-2. First Order Decay of Landfill Leachate COD in Perfusion Columns.

disassembled. A final sample was taken and analyzed from each column. COD values had increased to 550$\pm$ mg/l suggesting that the columns were making leachate instead of treating it. If one considers that sphagnum moss is itself organic, it is not surprising that under anaerobic conditions it might degrade producing leachate of its own.

Summary

Although the soil perfusion apparatus has the potential of providing excellent kinetic data for microbically mediated soil processes, limitations do seem to exist when clogging occurs. Pre-filtering of landfill leachate might extend treatment duration times, but iron precipitation may always be a problem. Future applications of the technique might incorporate a solids filtering device at the top of the reactor that could be changed daily with only minor interruptions to the process.

The use of a common vacuum manifold for multiple columns may also be unwise and should be avoided in future studies with leachate. As clogging begins there is some flexibility with system operation, but it requires that vacuum and line resistances be controllable. Individual vacuum pumps for each column would facilitate total suction control, but variable resistance devices at R_1 and R_2 in Figure 5-1 would greatly improve operational flexibility.

General conclusions that can be drawn from the limited experimentation that was conducted are as follows:

1. Early COD reductions (between t_o and day 3) did follow first order reduction kinetics.
2. The rate constant for the first 3 day period was .43 $days^{-1}$.
3. Asymptotic reductions between day 3 and day 7 may have been due to reduced oxygen intake through the sampling port S.
4. If peat columns are allowed to become anaerobic, their COD removing capability may be reversed, as measured by increased COD concentrations in the perfusate.
5. The absence of reed canarygrass from this system appears to impact hydraulic conductivity and overall treatment efficiency in a negative way.

CHAPTER VI

LEACHATE TOXICITY

Introduction

For each of the biological treatment technologies reviewed, there was a need to dilute or pretreat the leachate before living organisms could be exposed to it. Toxicity studies by Plotkin and Ram (98) using four freshwater aquatic species, fathead minnows (Pimephales promelas), zooplankton (Daphnia magma), green algae (Selenastrum capricornutum), and aerobic luminescent bacteria (Photobacterium phosphorium), clearly demonstrated that leachate from a Fitchburg, Massachusetts sanitary landfill was toxic to all four organisms and "highly toxic" to the test bacteria. The Fitchburg leachate is allowed to flow into Flagg Brook where it dilutes to approximately 7% of the total summer flow. Even with this 10 fold plus dilution, acute toxicity levels remained high enough to impact the diversity of aquatic life within Flagg Brook.

McBride et al. (99) evaluated the acute and sublethal toxicity effect of landfill leachate on rainbow trout (Salmo gairdneri). Their study involved a 65 hectare sanitary landfill in Vancouver, British Columbia that discharges its leachate into the Fraser River Estuary, an important component of the migration route utilized by 5 species of Pacific salmon, steelhead trout and cutthroat trout. The estuary is also utilized as a nursery for several species of salmonids including rainbow trout. McBride et al. (99) reported that 96 h LC_{50} values for 2 leachate samples on the day of sampling were 6.5% and 7.5% and after 14 days of storage were 5.9% and 5.8%, respectively. In each case the lethal toxicity properties of the leachate increased during the 14 day storage period. Table 6-1 illustrates the change in leachate parameters over the 14 day period. Although some minor variability in concentrations was observed, a clear cut indication of why older leachate was more toxic was not indicated by the data. It is worth noting that the landfill studied is underlain by 2.7 m of peat through which the leachate must percolate before being intercepted by a 1.2 m natural clay liner. If the McBride leachate is compared to Barre, Massachusetts leachate (Table 2-11), it can be seen that the concentration of COD is considerably less for the leachate that percolated through the 2.7 m of peatmoss. Should toxicity be a function of COD, an unfiltered leachate sample could be considerably more toxic than that indicated by the McBride study. Other studies by LeBlanc (100), Polprasert et al. (101) and Walker et al. (15), all demonstrated landfill leachate to be a potent toxic mixture to various forms of aquatic life. Considering that these acute toxicity studies implicate leachate at concentrations as low as .1% of its original strength and that a 4 hectare (10 acre) landfill site will usually generate some 110 m^3 (30,000 gallons) of leachate per day (102), there may be reason for concern regarding the impact of such a waste on surface and groundwater resources, especially if they are used as drinking water sources. A

Table 6-1. Effect of Storage on the Stability of Landfill Leachate.

Parameter (mg/L)	Storage Period (days) 0	3	7	10	14	Mean	S.D.
COD	710	730	690	700	640	694	± 33.6
pH	8.0	8.1	8.1	8.2	8.2	8.1	± .1
Chloride	735	740	750	755	768	749	± 12.9
Suspended solids (NFR)	32	31	23	23	20	25.8	± 5.3
Dissolved solids (FR)	3200	3200	3400	3300	3300	3280	± 83.7
Total alkalinity	2490	2670	2600	2640	2640	2608	± 70.5
Total hardness	480	--	421	--	--	450	± 42
Nitrite-N	.124	.067	.066	.074	.078	.082	± .024
Nitrate-N	.311	.433	.464	.404	.403	.403	± .057
Ammonia-N	370	395	363	425	389	388	± 24.3
Non-ionized NH_3							
at 15°C	9.9	13.2	12.1	17.7	16.2	13.8	± 3.1
at 5°C	4.6	6.1	5.6	8.2	7.5	7.8	± 3.9

(After McBridge et al. (99))

potentially more relevant issue relates to the chronic toxicity (i.e., mutagenicity) of landfill leachate that might exist at even more dilute concentrations. If more dilute concentrations of landfill leachate possessed mutagenic properties, the practices of permitting the operation of unlined landfills, or of collecting, treating and discharging landfill leachate through conventional wastewater treatment facilities may be questionable.

Menser et al. (17) applied approximately 155 cm (60 inches) of leachate per season to six forage grasses at a Princeton, West Virginia landfill over a 6 month period during 1975 and 1976 (October - April). The species used were orchardgrass (*D. glomerata* L.), reed canarygrass (*P. arundinacea* L.), bromegrass (*B. inermis* L.), tall fescue (*F. arundinacea* Schreb.) cv 'Ky 31', and bermudagrass (*C. dactylon* (L.) Pers.) cvs. 'Midland' and 'Tufcote'. The bermudagrasses are considered warm-season grasses while the others are considered cool-season grasses. Soil amendments and plantings were accomplished in May, 1974.

Table 6-2 provides analyses data of the 1974 leachate used in the forage application study. The individual species exhibited differential survival tendencies. Soil amendments beneficially influenced tolerance to leachate, especially with lime applications. Stands of all grasses were moderately-to-severely depleted by the 60-inch leachate application, however, rootstocks persisted and excellent recoveries were made. Reed canarygrass and tall fescue showed better tolerance than orchardgrass and bromegrass. Bermudagrasses effectively survived leachate treatments but were damaged by early and late summer frosts.

Table 6-2. Leachate Quality of Samples Taken From Drains Beneath the Mercer County Sanitary Landfill, Princeton, WV.

Parameter	Year 1971	Year 1974	Year 1976
	------- $mg.kg^{-1}$ (ppm) ------		
Kjeldahl N	63	62	101
SO_4	107	55	--
Cl	274	230	--
Ca	458	605	602
Mg	188	174	156
K	67	45	117
Fe	303	424	562
Mn	182	55	61
PO_4	--	4.5	1.1
Zn	--	2.5	1.4
Al	--	2.1	1.5
Sr	--	2.6	2.5
Na	257	265	283
	------- $mg.g^{-1}$ (ppb) --------		
Ni	52	400	225
Cr	34	100	336
Pb	2.5	133	386
Co	--	370	337
Cu	30	19	38
Cd	--	17	58
COD, mg/liter	5757	8973	3371
EC, umhos	2958	4092	4485
pH	5.6	5.3	5.5

(After Menser et al. (17))

Species differentially accumulated many of the mineral contaminants of leachate, especially manganese and iron. Soil amendments influenced the uptake of these elements. Menser concluded that forage grasses appeared to be an acceptable concentrating mechanism for leachate pollutants. Significant tolerance improvements were for ammended plots, especially for those treated with lime and in all cases tolerance improved during the second year of leachate application. Menser also reported pronounced increases in soil pH for various depths that resulted during the two year study period. Even the non-ammended spray plots (i.e., leachate only) experienced significant increases to depths of 30 cm or more.

Toxicity was not considered to be a limiting factor for leachate application to forages, but it clearly had an inhibiting effect that seemed to decrease with plant age and time. The mechanisms effecting inhibitions or survival tendencies were not considered in this study, but it was concluded that soil amendments caused significant differences in the elemental contents of grasses, especially for manganese and iron (17). It was also concluded that leachate alone influenced the accumulation of all macroelements, but that it did not cause significant accumulations of the toxic heavy metals cobalt, nickel, chromium, lead, or cadmium (17).

Steiner et al. (76) attempted several combinations of conventional wastewater treatment technologies to treat leachate from a 12 hectare (50 acre) commercial landfill in Falls Township, Pennsylvania. Although the leachate character was typical of others reported, researchers were unable to develop an "activated sludge" culture even after 6 months operation. It was generally concluded that phosphorus limitations and ammonia toxicity inhibited microbial growth. Subsequent pretreatment involved lime addition followed by air stripping of ammonia. Phosphoric acid was then added as a neutralizing agent for the lime treatment.

Once these pretreatment processes were implemented, an activated sludge could be developed in approximately four weeks and BOD and COD removal efficiencies reached 98.8% and 94.1%, respectively.

Tirsch and Jennings (49) and Lavigne (22) reported that efforts to enumerate coliform bacteria by the membrane technique were frustrated by inhibited growth whenever concentrated leachate was tested. Serial dilutions of the same sample often resulted in countable plates occuring at the 10^{-4} to 10^{-6} dilution range.

Lombardo (103) also reported considerable difficulty in performing standard 5 day BOD tests on leachate known to be high in degradable constituents. Standard wastewater "seed" was unable to survive in the more concentrated dilutions and techniques were modified to use an acclimated seed prepared by aerating raw leachate over a period of weeks. Lombardo attributed the seed problem to leachate toxicity, as did Tirsch, Jennings and Lavigne (49, 22).

Walker (22) reported that leachate concentrations as dilute as 10^{-3} proved inhibitory to batch treatment studies using the green algae <u>Scenedesmus</u> <u>dimorphous</u>. Surprisingly, when leachate dilutions were extended to 1/2000, the environment became stimulatory to the algae.

Immediately following the first application of Amherst leachate to the treatment beds used in this peatmoss reed canarygrass study, there appeared to be a total destruction of the foliar part of the grass. Assuming that the primary mechanism of leachate treatment would be due to a fixed film of microbes on the peatmoss, the experiment was continued. It should be noted that all leachate was applied with a watering can to the top surface of the treatment beds during this batch study, thereby exposing the young grass seedlings to direct foliar contact with the leachate.

After several weeks of continued leachate application, resistant foliar grass growth emerged and by the end of the first batch study, (approximately 6 weeks) grass blades had reached a height of nearly one meter in all treatment beds. The largest and most succulent plants appeared to grow in the beds having batch treatment times of three to six days.

While treatment research continued in the beds using a plug flow mode of operation, a separate pot study was initiated to further investigate the possible concentrations and methods of leachate application that might effect toxicity damage and recovery for reed canarygrass seedlings and their shoots. Discussions with turf and forage specialists (R. Cooper and W. Torello, personal communications) suggested that short term damage to; and subsequent recovery of the reed canarygrass might have been due to osmotic imbalances at the leaf-leachate interface. This imbalance could be due to the high ionic strength of the leachate that was applied foliarly during the batch study. After the initial "burning," it was theorized that undamaged roots and rhizomes became acclimated to the ionically strong leachate and in time modified their internal chemistry to a point that was compatable with the leachate environment. Subsequent growth of new shoots would reflect this adaptation, and they would be more tolerant to the potent leachate.

Objectives

Based on the above hypothesis it was decided to treat reed canarygrass seedlings grown in replicate pots both foliarly and through the root zone with several dilutions of Amherst leachate. It was anticipated that for seedlings treated foliarly with the more concentrated dilutions (e.g., 100% and 50%) "burning" would occur as was previously observed and that at some point in time the plants would recover and thrive. It was also theorized that for more dilute foliar applications (e.g., 25%, 12.5% and 6.25%) initial burning might be reduced or eliminated completely. Applications with Amherst tap water alone would serve as controls.

For the root zone treated plants it was expected that the "shoots" would not burn and that in time most plants would show some degree of positive response to leachate nutrients.

The experimental design attempted to accomplish the following objectives:

1. To select an optimum leachate concentration for maximum reed canarygrass growth.

2. To identify leachate concentrations that would produce acute toxicity effects.
3. To identify advantages and disadvantages associated with foliar and root zone application methods.
4. To identify a range of leachate strengths that could be applied to reed canarygrass without inducing an acute toxicity response.
5. To see if the temporary "burning" and recovery response observed in the batch treatment study could be reproduced.

Methods and Materials

Two replicate sets of 30 pots, 25 cm in diameter, were filled with 1000 grams of air dried sphagnum peatmoss. The pots were watered daily to near field capacity, so that their moisture content would be similar to that of the batch treatment beds. Each pot was seeded with one gram of reed canarygrass seeds. Subsequent watering was accomplished using capillarity from pie plate reservoirs located under each pot. When seedling growth reached approximately 5 cm (2 inches), leachate application was initiated.

Applications were continued for a period of sixty days during the summer of 1985. At the end of the treatment period maximum stem height was measured for each pot, and shoots were harvested and oven dried at 95°C for weighing.

<u>Set 1 treatment</u>:

The 30 pots in set #1 were subdivided into six subsets of five each. Each of five subsets received 100 cm^3/day of Amherst leachate diluted to prescribed concentrations of 100%, 50%, 25%, 12.5% and 6.25% leachate. The sixth subset received Amherst tap only serving as a control. All 30 pots in this treatment set received their daily applications foliarly. A small 120 ml wide mouth vial with small holes drilled in the screw on cap was used as the watering device. Care was taken to apply each 100 cm^3 treatment so that all seedlings were wetted equally. The choice of 100 cm^3/day was selected following green house evaporation studies that indicated this amount to be approximately equal to the daily evaporation rate for a 25 cm (10 inch) pot. This rate was also similar to the application rate used in the batch study experiment where toxic effects were first observed.

<u>Set 2 treatment</u>:

Set two contained the same number of pots and subsets as treatment 1. The same dilutions of Amherst leachate were applied at the same rate of 100 cm^3/day. One subset also received 100 cm^3/day of Amherst tap as a control. The only difference between set one and set two was the method of application. For all set 2 pots, the daily leachate dilutions were applied by pouring the 100 cm^3 into an aluminum pie pan at the bottom of each pot. Continuous wetting was accomplished by capillary action in an upward direction through the root zone of the pots.

Stem height and weight were used to evaluate the response of each treatment.

Results and Discussion

Foliar Treatment

For the foliarly treated pots, reed canarygrass seedlings watered with any dilution of leachate "burned" within 24 hours. 100%, 50% and 25% treatments appeared to have their entire stems and leaf blades destroyed. 12.5% and 6.25% treatments generally had damaged or burned leaves; but their stem sections appeared more resistant. Within 3 weeks all foliar treatment pots showed some sign of recovery, except for those treated with pure leachate (100% treatment). It seemed evident that the foliar application of pure leachate stressed the seedlings beyond any point of recovery, and as such, must be considered fatally toxic to reed canarygrass in an undiluted form.

After 60 days of treatment, it was clearly evident that the best recoveries were associated with the 25% and 12.5% foliar treatments. Seedlings foliarly treated with 50% leachate and with Amherst tapwater did more poorly, but for different reasons.

Recovered shoots generally grew to a height of 25-50 cm before harvesting, not unlike the growth pattern previously observed in the batch reactor study. Table 6-3 summarizes individual grass weights for each pot. Treatment averages for the 12.5% and 25% treatments were significantly different from other treatments, at the .01 probability level, but there was no statistical difference in mean grass weights between these two treatments.

Table 6-3. Weights of Reed Canarygrass Harvested From Foliarly Treated Pots.

Percent Leachate in 100 ml Applications	Average (g)	Standard Deviation
Amherst Tap	.9	± .35
6.25%	3.74	± .64
12.5%	4.66	± .96
25%	4.64	±1.58
50%	1.34	±1.20
100%	0.0	± .04

Notes: Application rate = 100 cm^3/day
Total application = 6 liters over 60 days

Capillary Treatment

Early growth patterns were as predicted, with a lush green growth characterizing the more concentrated leachate treatments. Plants grown in Amherst tap water and the 6.25% leachate were less productive and in the case of plants treated with Amherst tap, chlorotic conditions were clearly evident. After six weeks of treatment, a totally unexplained reversal pattern developed, resulting in a loss of nearly all plants subjected to leachate treatment. A sudden loss of color and vigor was followed by browning and senescence. Growth changes were most rapid for the 100%, 50% and 25% treatments. The 12.5% and 6.25% treatments senesced more slowly and the Amherst tap control continued to do poorly.

The only obvious change in the greenhouse was a reduction in ventilation due to a broken blower fan belt. Within a week almost all of the treatment pots were a deep brown and by the end of the 60 days all vegetation appeared to be dead. A preliminary conclusion was that heat stress due to high greenhouse temperatures ($\pm 40^{\circ}C$) and poor ventiliation was the cause of die-off.

It was decided to repeat the same study during the summer of 1986. In preparing new pots and discarding the old aluminum pie plates used as support during the previous study, it was observed that the pie plate bottoms were almost completely deteriorated for the root fed plants. Weight comparisons with new plates indicated that more than 50% of the aluminum had been lost from most of the plates used during the first study. Previous assumptions of heat stress were modified to include the possibility of aluminum toxicity. Clearly the strong reducing nature of the leachate in contact with the aluminum had mobilized it, so that it was able to move up and into the peat and root areas inside the pots. New pie plates for the second study were placed inside ziplock bags. The plastic membrane acted as a barrier between the leachate and aluminum. Plants responded favorably again to increased leachate strength when added through the pot bases, but sudden die-offs did not occur the second time. Heat did seem to retard the overall growth of all plants and final harvest weights for root treatment plants were not significantly different after 60 days at the .01 probability level.

Summary

The seedling pot study was able to effectively duplicate the toxic response and recovery phenomenon observed in the batch study. The mechanisms involved, however, are still unknown. Most importantly, the more than normal recovery of the grass after repeated applications of leachate to the leaf blades strongly supports the hypothesis that a peatmoss and reed canarygrass system may be able to effectively treat landfill leachate. The effectiveness of any root zone method is generally dependent upon the ability of the plant root and rhizome system to deliver oxygen to aerobic bacteria living within the rhizosphere (80, 82, 84, 85). Final dismantling of the treatment reactors at the end of the plug flow study revealed a dense network of rhizome growth throughout the bed (Chapter VII).

Additional research is needed to determine the particular mechanisms involved, but for this study the combination of peatmoss reed canarygrass and leachate worked effectively to attenuate leachate TOC.

The specific conclusions reached may be summarized as follows:

Foliar Study

1. Foliar "burning" of leaf blades occurred within 24 hours for all leachate dilutions applied.
2. After a period of 2-3 weeks, leachate tolerant shoots appeared in all of the leachate treated pots, repeating what had been observed in earlier batch studies.
3. The 25% and 12.5% treatments recovered more rapidly than did the 50% or 6.25% foliar treatments.
4. Poor growth in all control pots supported the conclusion that peatmoss alone is not a good media for growing reed canarygrass.
5. Seedlings treated with pure leachate (i.e., 100%) never recovered, and this could be a problem with system start up if foliar applications are ever used on a full scale treatment system.
6. The less severe impact of diluted leachate suggests that a recycle system for dilution may be appropriate in the field.

Capillary Treatment

1. The addition of leachate through the root zone instead of foliarly appears to eliminate "burning," regardless of the concentration applied. Subsurface application of leachate in

future studies may, therefore, be more appropriate than foliar application.

2. The aggressive nature of leachate appeared to mobilize aluminum in the pie pan bottoms, so that it was able to move up and into the root zone area. The subsequent loss of vigor and eventual death was presumed to be due to aluminum toxicity. When the capillary study was repeated, using plastic pie plate liners, toxicity responses did not occur.

CHAPTER VII

DISCUSSION

General Summary

This project has provided encouraging preliminary data relative to the application of peatmoss and reed canarygrass for the treatment of landfill leachate. Batch treatment studies failed to verify the proposed kinetic model, but data indicated that a 99% reduction in TOC and selected metals was possible using the modified RZM treatment system. Reed canarygrass initially showed toxic responses to landfill leachate, but after four weeks of repeated batch applications, leachate acclimated shoots emerged and proliferated. The toxicity problem did not repeat itself for the duration of the study. Analyses of leaf blade concentrations of Mn, K, Fe, Cu, Zn and Ca at 3 different times, ranging over the entire study period suggested that reed canarygrass is an excellent sink for the removal of these materials from landfill leachate. Subsequent continuous flow reactor studies indicated that first order kinetic modeling was possible and that a rate constant of 6.53 $days^{-1}$ described reasonably well the application rates and treatment times evaluated. It is not clear, however, to what extent poor pump performance during the first plug flow study contributed to the somewhat erratic behavior of the various reactors.

To insure steady state conditions for each reactor, flow rate calibration and microbial equilibration were extended over a six week pre-sampling period. It was intended that during this period microbial populations would reach a maximum for the flow rate and substrate concentrations being provided. Each flow rate (i.e., 100, 200, 300, 400, 600 and 800 cm^3/hr) was also replicated four times in four different reactors that were located in different areas of the greenhouse.

Once sampling began, it was continued on an every other day basis until 10 samples had been collected from each reactor. Results from replicate flow rates were pooled to minimize "scatter," and are reported as composite values in Table 4-3.

During final weeks of treatment in the moderate rate plug flow study, the clogging of effluent lines became increasingly a problem. Final dismantling of the reactors revealed that reed canarygrass rhizomes had proliferated so extensively that they had actually grown into the discharge lines. Future use of the same reactors would require modifications to the drainage system so that root blockage is not repeated. Final air dried weighing of the peatmoss indicated that there was no significant change in weight for any of the reactors (Table 7-1), but accurate measurements were not possible for the following reasons:

1. The rhizomes and root stock had proliferated so extensively throughout the peat, it seemed evident that their weight was significant. Many tedious hours were spent sorting peatmoss from roots and rhizomes, but only a fraction could ultimately

be separated. Based on oven dried corrections the vegetative weight of reed canarygrass parts represented an average of 2 1/2% of the total remaining reactor weight.

2. Visual inspection of the surface peatmoss and clod like units deeper in the reactors indicated that a considerable amount of iron had precipitated out and therefore would contribute to final peat bed weights. From visual observations, it seemed obvious that some peat had decomposed. The near full 25 cm (10 in) beds at the beginning of the study subsided to approximately 20 cm (8 in) at the end and that included the peat additions made in November of 1987 (see Column 2, Table 7-1).

It is possible that the loss of depth was due to consolidation and increased density; but the assumption that some peat decomposition did occur with off-setting weight increases from iron precipitation and reed canarygrass root and rhizome material seems more reasonable.

On a more positive note, it was encouraging to observe the extensive rhizome and root network that had developed in each of the reactors. Gersberg (104) and Armstrong (105) have shown that vascular aquatic plants, such as <u>Scirpus validus</u> (bulrush), <u>Phragmites communis</u> (common reed), <u>Typha latifola</u> (cattail), and <u>Phalaris arundinacea</u> (reed canarygrass), are able to translocate oxygen from shoots to roots. Bacteria attached to the roots or rhizomes are then able to utilize the surplus oxygen for aerobic decomposition of substrates. Considering the dense root and rhizome networks observed in this study, it is not

Table 7-1. Peat Weight Losses Over 4 Years of Intermittent Leachate Application.

Reactor #	4/12/84 Initial AD weight kg	Initial OD* corrected weight kg	11/1/87 Added Peat OD corrected kg	Total OD Peat	9/13/88 Final OD Peat	Net 4 yr. gains & losses %
1	27	11.34	3.71	15.05	16.565	+1.51
2	27	11.34	2.85	14.19	16.907	+2.72
3	27	11.34	2.85	14.19	15.859	+1.67
4	27	11.34	2.85	14.19	15.128	+0.94
5	27	11.34	2.85	14.19	14.529	+0.34
6	27	11.34	2.85	14.19	13.646	-0.54
7	27	11.34	4.06	15.40	17.726	+2.33
8	27	11.34	3.93	15.27	16.722	+1.45
9	27	11.34	3.57	14.91	17.545	+2.64
10	27	11.34	2.85	14.91	16.016	+1.83
11	27	11.34	3.86	15.20	16.725	+1.53
12	27	11.34	4.284	15.62	16.813	+1.19
13	27	11.34	4.284	15.62	16.681	+1.06
14	27	11.34	3.93	15.27	16.825	+1.55
15	27	11.34	3.57	14.91	15.164	+ .25
16	27	11.34	3.28	14.62	14.327	- .29

*Oven drying temperature 95°C
AD - Air dried
OD - Oven dried

surprising that TOC and COD removals were generally better than originally expected. The absence of root material in the soil perfusion study was most likely a major contributing factor to the generally poor and slower reduction of leachate COD.

Future Research Needs

There should be little doubt as the world moves into the 21st century, that changes in technology, life style, and general resource utilization will continue to produce new and perhaps larger types and amounts of solid waste that will need to be managed. Recycling and resource recovery continues to be the current options of choice, but salvage market prices and air quality issues impede their full scale implementation. Sanitary landfilling has also fallen subject to the watchful eyes of environmentalists, enforcement agencies and of course the "NIMBY" people. Somewhere along the way environmental education has failed to teach the lessons that the entire planet is "in my back yard," and that ecologically speaking there is no such thing as "away." If the United States continues to produce 250 million metric tons of municipal solid waste per year, world tours on refuse barges will not avoid the fact that the ecosystems of planet Earth must ultimately be the repository. If our industry and agriculture continue to produce 4.3 billion metric tons of non municipal solid waste each year, the same planet must ultimately be "our dump." When "the good life" social practices of western civilization spread to less developed corners of the Earth, new solid waste disposal problems will certainly follow.

There exists a challenge, then, to the engineering and scientific communities of the world to provide solutions to a problem that grows and changes almost daily. The educational lesson to be taught is equally difficult. People must learn that if humankind chooses to extract raw materials from the earth and to fashion them into daily "necessities," then "their back yard" is the only place for unwanted items (i.e., waste) to go. With the exclusion of radioactive decay losses, every gram of resource used must ultimately be managed or disposed of.

This research effort assumed from the start, that "in my back yard" "IMBY" technology held the most promise for a cost effective environmentally sound solution to the problem of landfill leachate pollution. "Back yard" wetlands have served as dump sites for decades. Previous land reclamation projects paid little or no attention to the renovating potential of the ecosystems that they sought to eliminate. It may well be said, that the anti-mosquito landfilling programs of years past have contributed significantly to minimizing the true pollution potential of our solid waste stream. It should not be concluded, however, that landfills belong in natural wetlands. More realistically, "constructed wetland" environments that are temporarily isolated from natural ecosystems provide the most promise. Small constructed wetland units provide hydraulic controls that maximize treatment efficiency. Secured wastefills and treatment units can utilize the benefits associated with natural decay and assimilation without threatening surface or groundwater resources.

Most importantly, the large economy of scale associated with more elaborate technologies can be significantly reduced, permitting the construction of smaller and hopefully less environmentally threatening disposal sites.

The peatmoss and reed canarygrass system studied in this project suggests that root zone technology may provide solutions to current leachate treatment problems. The system seems relatively easy to construct and operate, and costs are minimal compared to more advanced treatment methods. Over the years similar RZM's have been successful in treating domestic wastewater and special industrial wastes. As such, basic design criteria are emerging for the technology. Before full scale application of this type system can be considered however, some remaining questions need to be answered:

1. What is the transport pathway and fate of solutes in peatmoss?

Mass transport by convection clearly moves solutes through the macropores in a way similar to that of any porous media, but a considerable amount of leachate is also absorbed by the hollow sphagnum cells. Dissolved organics are able to diffuse in and out of the dead cell walls, but no one has evaluated this "reservoir" as a source or sink for substrate. When microbes attach themselves to the solid matrix, they may be able to utilize diffusing substrates in the peatmoss cells as well as the convective material moving through the macropores. As such the substrate reservoir capacity of sphagnum cells could "smooth" changes in leachate organic concentration that occur regularly at most landfill sites.

2. How does the micro environment in a root zone system vary in space and time and how does it effect effluent quality?

The literature clearly suggests that zones of aerobic and anaerobic activity exist in any RZM system. This project did not directly address the ecology of root zone fauna and flora, but the root zone environment would clearly impact factors such as precipitation and solution, nitrification and denitrification, and the nature and fate of organic materials found in leachate or produced by the treatment media.

3. Are other operational modes more effective?

Greenhouse space and size limited study of alternative operational modes, but they should be investigated with in situ pilot plants. Greater aspect ratios (i.e., length to width), reactors in series, chemical pretreatment of leachate (e.g., liming) open as opposed to covered systems and effluent recycle lines to dilute pure leachate are all alternatives that could ultimately improve effluent quality and operational flexibility of the system.

4. Does RZM effluent retain the toxic properties of landfill leachate?

Many studies have implicated landfill leachate as a toxic material (see Chapter VI) to plants and aquatic animals. Most toxic substances known to science also possess mutagenic and carcinogenic properties at lower concentrations. If landfill leachate retains any of these properties after treatment, the practice of surface water or groundwater discharge may be unadvisable. It would therefore be prudent to conduct toxicity and/or mutagenicity screening tests to evaluate peatmoss and reed canarygrass effluent quality.

5. Is treated effluent acceptable for discharge?

The current discharge permit program (NPDES) does not address many of the pollution parameters associated with leachate. Although this study demonstrated an ability to remove 99%+ of TOC, COD and metals, it is questionable whether the effluent is safe to release into the environment. Future research should attempt to develop a final "polishing" process that could bring constituent concentrations of concern down to those found in ambient surface waters and groundwaters.

6. What will be the effect of applying leachate to a peatmoss and reed canarygrass system over a long period of time?

After three years of operation there appeared to be little or no net loss of peatmoss from the system, but clearly there is the potential for microbial attack if substrate in the leachate becomes limiting. There is also a maximum limit for the adsorptive capacity of peatmoss. Considering that landfills produce leachate for many decades and that organic and inorganic concentrations continue to change even after closing, the question of longevity must be addressed. If peatmoss and reed canarygrass systems have a finite life, then provisions for replacement, renovation and monitoring will have to be included as part of any permitting process. It will most likely take many years of in situ operation to adequately evaluate the functional life and routine maintenance requirements of a full scale system.

CHAPTER VIII

CONCLUSIONS

As hypothesized, peatmoss and reed canarygrass served as a suitable media for the treatment of landfill leachate in a greenhouse environment. Batch studies demonstrated that 99% removals of TOC, COD, Mn, Fe, Cu, Zn, Pb, Ca and Mg were possible with 1-12 days treatment time. The batch data failed however to fit a first order kinetic model. It was concluded that if first order reductions did occur the time period involved was less than 24 hours.

Eight months of pre-leachate growth indicated peatmoss to be a poor growth media for reed canarygrass. Acute foliar toxicity responses resulting from the first leachate application also suggested that reed canarygrass might not be able to survive in a leachate environment. After several weeks of continued leachate application, however, resistant shoots emerged and flourished. By the end of the 60 day study, dense succulent leaf growth had reached a meter or more in height, and the plants had a lush green color. The acclimated reed canarygrass continued to do well for the duration of the project.

Leaf blade analyses of the grass after the batch study showed that metal concentrations for Mn, K, Fe, Cu, Zn and Ca were all higher in the reactors receiving more frequent applications of leachate. There were no concentrating effects observed for Pb and Mg. The data suggested that reed canarygrass could be an effective sink for the removal of metals in landfill leachate.

Subsequent plug flow studies indicated that a peatmoss and reed canarygrass system could effectively treat leachate with a continuous flow operation. COD data for six application rates each replicated 4 times generally followed first order kinetics, with a rate constant (k) equal to 6.53 $days^{-1}$. Grass cuttings taken and analyzed between the high flow and moderate flow experiments further substantiated the hypothesis that reed canarygrass could be an effective sink for the metals Mn, K, Fe, Cu, Zn and Ca.

Three soil perfusion units were used to evaluate leachate treatment with peatmoss alone, but all three systems failed after seven days of continuous operation, apparently due to clogging. Leachate recycle and aeration rates started to decrease after 3 days and COD concentrations remained unchanged at 1300-1500 mg/l. Although this represented only an 80% reduction in concentration, the system was unable to reduce COD further (Figure 5-2). After seven more days of operation COD concentrations started to increase suggesting that under anaerobic conditions a peatmoss system might create leachate instead of treating it. The first three days of normal operation did, however, provide COD data that fit the first order model proposed, but a rate constant of .43 $days^{-1}$ clearly indicated that without the reed canarygrass, peatmoss alone did not perform as well.

Toxicity studies on potted reed canarygrass seedlings were able to duplicate the "burning" and regrowth pattern observed at the onset of the batch study. It was also observed that plants treated foliarly with 12.5% and 25% leachate dilutions recovered more rapidly than those treated with other concentrations. Seedlings receiving similar leachate dilutions by capillary upflow from pie pan reservoirs at the base of each pot did not experience foliar "burning," but instead seemed to have a positive growth response to increased leachate concentrations. After 6 weeks of successful growth, virtually all of the plants started to loose color and to brown. Later inspection of the aluminum pie plate reservoirs suggested that leachate had reduced much of the aluminum allowing it to move upward into the root zone region. Aluminum toxicity was therefore suspected as causing the rapid deterioration of the affected plants.

After exposing reed canarygrass and peatmoss to more than three years of intermittent leachate applications, the 16 reactors were dismantled and inspected. Extensive rhizome growth had spread to every corner of each treatment unit and a knife was needed to section the peat. The entire mass was interwoven so extensively with rhizomes, that it retained its shape after removal from the reactor units that contained it. Rhizomes were even found growing well into the drainage tubes.

In conclusion, there is no immediate explanation of why the initially poor growth patterns of reed canarygrass in peatmoss were eventually reversed so dramatically with the addition of leachate. Once established, however, the root zone system seemed to perform remarkably well. TOC and COD concentrations were reduced to less than 1% of initial raw leachate values. The data suggests that a first order kinetic model may be appropriate for modeling a peatmoss reed canarygrass RZM. Based on the application rates evaluated, it may be possible to construct on-site leachate treatment systems using only a fraction of the space required by other methodologies.

Many of the problems associated with standing bodies of leachate ponds and lagoons are eliminated by the unsaturated treatment environment used in this method. Although leachate tends to be initially toxic, repeated application produces a hearty stand of reed canarygrass with a root and rhizome system that pervades every part of the subsurface environment. The long term capability of this system, especially under in situ conditions is still unknown, but following more than three years use of a greenhouse, the system continued to perform well.

A larger scale in situ application is clearly indicated by these preliminary findings, so that "real world" effectiveness can be evaluated.

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