Gas and Leachate from Landfills

Formation, Collection and Treatment

U. S. ENVIRONMENTAL PROTECTION AGENCY
CINCINNATI, OHIO   45268

GAS AND LEACHATE FROM LANDFILLS

FORMATION, COLLECTION, AND TREATMENT

Proceedings of a Research Symposium held at
Rutgers University, New Brunswick, New Jersey
March 25 and 26, 1975, and cosponsored by the
U.S. Environmental Protection Agency
Solid and Hazardous Waste Research Division and Region II
and by the Department of Environmental Science
Cook College, Rutgers University

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FOREWORD

Man and his environment must be protected from the adverse effects of pesticides, radiation, noise, and other forms of pollution, and the unwise management of solid waste. Efforts to protect the environment require a focus that recognizes the interplay between the components of our physical environment--air, water, and land. The Municipal Environmental Research Laboratory contributes to this multidisciplinary focus through programs engaged in

• studies on the effects of environmental contaminants on the biosphere, and

• a search for ways to prevent contamination and to recycle valuable resources.

Sanitary landfills are an environmentally acceptable method to dispose of an increasing volume of municipal waste. These Proceedings identify research aimed at minimizing the impact of landfills and providing solutions to unique problems.

Louis W. Lefke
Acting Director
Municipal Environmental
Research Laboratory

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ACKNOWLEDGMENT

INTRODUCTION TO SYMPOSIUM ON GAS AND LEACHATE FROM LANDFILLS: FORMATION, COLLECTION, AND TREATMENT

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A well-known truism is that wastewater treatment is only as effective as its solids handling capability. Whether the source of solids is wastewater, air pollution, or undiluted waste solids (sanitary or industrial landfills), the basic principles of stabilization, that is the natural principle processes, apply similarly in each case. Organic solids degradation or stabilization proceeds through a series of less known biological and chemical processes. Environmental factors such as temperature, moisture content, mixing, reaction, pH, buffer capacity, kinds and number of microorganisms, toxicity and intermediate compound formation, and removal all have a bearing on the rate of stabilization. The real problem or challenge arises when we must project byproduct accumulation and concentration at a given time and place resulting from the decomposition of complex municipal waste solids under a series of environmental conditions not always under full control.

The number of variables involved, coupled with the myriad of potential future intermediate compounds, provides almost a limitless list of possibilities. Not too long ago we would have been satisfied to achieve a reasonable degree of reproducibility of results in our investigations when optimizing a number of the environmental factors. Now, however, the name of the game is ultimate disposal, which must be acceptable within a total environmental assessment and impact framework. The problem is compounded because land availability, zoning, public acceptance, economic feasibility, and establishment of water quality standards with a margin of safety when scientific information is lacking are just a few of the additional stumbling blocks preventing quick and/or realistic solutions. To these we must add compliance schedules corresponding to public demand for action long overdue, available funding notwithstanding. A massive effort is underway through strong legislation for a vast monitoring effort to establish environmental quality standards for different waste control effluents now being discharged into the environment. The tendency to use such information for standardization purposes without sufficient consideration for the apparent variables can conceivably result in realistic control guidelines. We must not forget our prime objective must be a workable system which permits a controlled release without unreasonably impairing the use of the environment.

All processes of ultimate solids degradation usually result in a byproduct or byproducts consisting of gases and innocuous residues. We must recognize and include the important sources of contamination derived from natural processes such as leaching of soils, erosion of rocks, washing the atmosphere of impurities, and decomposition of organic matter such as all forms of vegetation and food. We must also ask the

question: what portion of animal and man's activity should we include as background or baseline? It would appear extremely important to establish a realistic baseline of constituent concentration level at different locations before imposing an environmental standard.

A brief search of the literature on studies relating to sanitary landfills leads one to comment on complexities over and above having a feasible baseline. As researchers we must always be cognizant of the possible use of our findings directly as basic ingredients for the drafting of standards and eventual enforcement. Constituent concentrations found during investigations should be carefully qualified by reporting fully the environmental conditions under which decomposition took place. Concentrations observed immediately beneath a solids loading area are of academic interest only when reported as a single number. A reported constituent level at distance from the source without including the type of soil or medium through which the leachate traveled, the time of transit, or vegetative cover could be misconstrued if used without the inclusion of important "qualifications." If we are to make a significant contribution toward solutions of solids disposal problems, we must recognize that our findings must fit into the total solution of ultimate disposal with all its environmental interactions. At the current pace of enforcement and new legislation, we must be doubly cautious that findings are appropriately identified and limitations on conclusions delineated.

CURRENT EPA RESEARCH ACTIVITIES IN SOLID WASTE MANAGEMENT

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ROLE OF THE U.S. ENVIRONMENTAL PROTECTION AGENCY

The overall role of solid waste management programs for the U.S. Environmental Protection Agency (EPA) involves:

- Controlling the quantity and characteristics of wastes

- Efficiently collecting those that must be removed

- Recycling, through materials and energy recovery, all that can be used

- Properly disposing of the residuals that have no further use in such a manner as to protect and enhance the land

To fulfill this role, the following efforts are being pursued:

- To develop the data base and assemble the elements necessary for a national program to properly control and manage those wastes that are inherently toxic or hazardous because of their particular use, application, or environmental impact

- To support research and provide technical assistance to local and state governments to improve productivity in collection operations and ensure the use of disposal practices that will protect the land and prevent degradation of the air and water, while expanding the recovery of energy and materials from mixed wastes

- To conduct studies and analyses to help reveal the total environmental consequences of our traditional mining, manufacturing, marketing, and distribution practices as these relate to the waste of resources and the pollution of air, land, and water

- To conduct studies and analyses and monitor ongoing resource recovery demonstrations to ensure the development of appropriate technology and workable institutional arrangements to bring about maximum recovery and reuse of resources from solid waste

- To assist state solid waste management agencies to move toward a stronger role in protecting the land and a broader role in managing all residual materials

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SOLID WASTE RESEARCH ROLE

The solid waste research program initially concentrated on problems associated with municipal solid wastes, but recently, emphasis has shifted in anticipation of hazardous wastes regulation. The primary objectives of the solid waste research and development program are:

- To perform research to establish the basis for a solid waste management regulatory program for hazardous and nonhazardous wastes

- To perform comprehensive research leading to drafting performance standards for sanitary landfill operation

- To develop and evaluate resource recovery technologies to provide options for energy and materials recovery

- To perform research and development leading to improved municipal waste management practices

These objectives are presently being met by research concerning:

- Known health effects of hazardous wastes

- Pollutant transport processes

- Known and new technologies for hazardous waste disposal, including salt mine, deep-well injection

- Environmental effects of municipal sanitary landfills; leachate, gas, sludges, and hazardous wastes are included

- Economic analysis of alternative hazardous waste control policies

The Solid and Hazardous Waste Research Laboratory (SHWRL) has the prime responsibility for this research. This Laboratory is one of six laboratories in the EPA, National Environmental Research Center (NERC) at Cincinnati, Ohio. For ease of identification, SHWRL has classified its research program into five categories, which will be discussed briefly:

- Health/environmental effects research

- Pollutant migration research

- Assessment of control technology for hazardous waste disposal

- Resource recovery

- Energy

HEALTH/ENVIRONMENTAL EFFECTS RESEARCH

This research is being performed to determine human health and environmental effects as they relate to the management and land disposal of selected hazardous substances/ wastes to develop criteria documents that summarize, assess, and interpret health and ecological effects of specific hazardous wastes. The hazardous waste criteria documents will contain:

- Comprehensive effects data for:
  - all forms of life, both human and other living organisms, for the air, water, and land

- Environmental aspects of hazardous materials for:
  - environmental distribution
  - transport through soil; through soil to water or air; and through water or air to humans or other organisms

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— transformation

— fate

— accumulation and magnification

Criteria documents are currently being developed for the following hazardous materials and related compounds:

arsenic   endrin
asbestos   fluorides
benzidine   lead
beryllium   mercury
cadmium   methyl/parathion
chromium   PCB's
copper   selenium
cyanides   toxaphene
DDD/DDE/DDT   zinc

These criteria documents will serve as background information for establishing "Hazardous Waste Disposal Standards."

POLLUTANT MIGRATION RESEARCH

This research is being performed to:

• Study migration of hazardous materials through soils and in water transport

• Document movement of such materials to establish the link to health/environmental effects

• Establish role of soil in controlling or reducing harmful substances reaching water or air

These pollutant migration studies are being performed simultaneously in the areas of (a) industrial hazardous wastes, (b) municipal refuse, (c) specialized wastes, and (d) polyvinylchlorides.

Industrial Hazardous Wastes

The Industrial Hazardous Waste test program consists of:

• Literature review

— State-of-the-art document and bibliography

• Controlled laboratory program

— Waste characterization

— Soil column leaching studies

— Pollutant identification, migration, retention, and transformation

• Field verification program

— Well monitoring and soil sampling of full-scale industrial and municipal waste disposal sites to verify lab results

The industrial wastes currently being researched in the laboratory are obtained from the following waste streams:

electroplating

chlorine production

nickel-cadmium battery production

inorganic pigment manufacturing

water-base paint production

FGD sludge (flue gas desulfurization--coal burning)

titanium pigments

hydrofluoric acid

aluminum fluoride

phosphorous

lead-acid batteries

carbon-zinc primary batteries

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Two more wastes from the mining industry are currently being identified for inclusion into the FY '75 research program.

The compounds of concern and those currently being monitored in our soil columns and field investigative efforts are:

arsenic   lead
asbestos   mercury
beryllium   nickel
cadmium   selenium
chromium   vanadium
copper   zinc
cyanides

Municipal Refuse

The municipal refuse test program primarily relates to the two decomposition products, gas and leachate. This program is similar to that being performed for hazardous waste:

* Literature review

    - Water quality bibliography

    - Gas bibliography

    - Summary report, gas and leachate from sanitary landfills

    - Compilation of methodology for leachate identification

* Laboratory program

    - Leachate and gas characterization

    - Soil column leaching studies: pollutant identification including viruses, migration, retention, and transformation

    - Gas migration: modelling prediction of production/migration

    - Leachate and gas generation: leachate and gas production from laboratory lysimeters

* Field verification program

    - Monitoring simulated test cells for leachate and gas generation from raw, shredded, baled, and mixed refuse environments

    - Monitoring full-scale raw, shredded, and baled municipal refuse disposal sites to verify lab results

Specialized Wastes

The specialized waste test program relates to hexachlorobenzene and pesticides:

* Hexachlorobenzene wastes are being investigated to determine the volatilization aspects of the material and to evaluate the effectiveness of various materials for covering these wastes to reduce volatilization

* Pesticide wastes of high concentrations are being investigated to determine pollutant migration potential when these wastes are disposed onto the land

Polyvinylchloride (PVC)

The PVC test program is assessing the environmental effects of current disposal practices.

* Incineration of PVC is being investigated to determine detrimental effects

* A study is being conducted to develop projections of future PVC production

ASSESSMENT OF CONTROL TECHNOLOGY FOR HAZARDOUS WASTE DISPOSAL

Research performed in this area relates either to treatment or isolation technology to minimize pollutants from entering into man's environment. Treatment technologies being investigated are:

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- Treatment by natural soil processes

- Treatment by physical/ chemical/biological processes

- Treatment by thermal decomposition

Isolation technologies being investigated are:

- Underground cavities

- Liners/membranes

- Encapsulation

- Chemical fixation

Treatment Technology

- Natural Soil Processes: The treatment of pollutants from hazardous waste and municipal refuse disposal sites by natural soil processes is basically being performed under the "Pollutant Migration" studies whereby various raw soils are being evaluated in column studies for their pollutant attenuation capabilities. The U.S. Department of Agriculture (USDA) soils series currently being investigated are: Anthony, Ava, Chalmers, Davidson, Fanno, Kalkaska, Mohave, Molokai, Nicholson, and Wagram. These soils encompass the range of soil types--from sand to clays to silts. Other soils are also being investigated whereby various percentages of the clay mineral, kaolinite, montmorillonite, and illite are mixed with pure sand to form various mixtures of sand and clay soils.

- Physical/Chemical/Biological Treatment Processes: The treatment of hazardous waste materials primarily relates to the chemical treatment/ degradation methods for pesticides. Basically, laboratory studies are being performed to confirm safe

procedures recommended for disposal. Release of toxic gases is being evaluated from various treatments, i.e., combustion, peroxides, caustic soda, acids, and hypochlorite. Also, a field manual is being developed for recommended disposal for small quantities of pesticides (1 to 5 lb or 1 to 5 gal) for use by home owners and farmers. Various new technologies are being reviewed for recommendation as to future studies. Promising processes recommended are:

chlorinalysis

wet air oxidation

decomposition by acids and bases

chemical oxidation and other chemical treatments

catalysis

batch and continuous ion exchange

photochemical processes

low-temperature microwave discharge

biological degradation, i.e., enzymes, trickling filters, and activated sludge

osmosis/ultrafiltration

activated carbon adsorption

The treatment of municipal refuse leachate is being pursued in the areas relating to:

— Physical-chemical treatment: various physical-chemical treatment schemes were investigated in the lab, and the most promising on-site pilot treatment plant was being developed. This pilot plant consists of an anaerobic lagoon

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followed by aerobic polishing. Additional studies are being performed to determine the effects of leachate disposal into domestic wastewater treatment plants.

— Biological treatment: various unit processes including biological treatment schemes are being investigated to determine process kinetics, the nature of the organic fraction of leachate, and the degree of treatment that may be obtainable using conventional wastewater treatment methods.

— Recirculation: recycling of leachate is being investigated to determine the beneficial aspects of recirculation as a means of leachate control and accelerated landfill stabilization. Recommended design, operation, and control methods applicable to conventional sanitary landfill practice will be developed.

— Spray irrigation: spray irrigation of leachate is being investigated as a low-cost, on-site treatment scheme. Optimum leachate loading rates and removal efficiencies for organic and inorganic constituents are being determined for two soil types.

• Thermal Decomposition: Treatment by thermal decomposition relates to the establishment of time-temperature relationships for incinerating pesticides. Specifically, through the test program, existing information will be summarized into a state-of-the-art document, and experimental incineration/decomposition studies will

be conducted on upwards of 40 pesticides. A lab-scale evaluation/confirmation study and a pilot scale incinerator study are being performed. The candidate pesticides for thermal decomposition are:

Completed (FY 1974)	Planned (FY 1975)
DDT	Toxaphene
Aldrin	Captan
Picloram	Zineb
Malathion	Atrazine

Isolation Technology

• Underground Cavities: The isolation technology for underground cavities is being performed to evaluate the adequacy of:

— deep-well injection (for liquid waste disposal), including drilled wells and permeable formations

— salt mines

— hard-rock mines for storage of solid, fixed, or encapsulated wastes

• Liner/Membranes: The liner/membrane technology is being studied to evaluate suitability for eliminating or reducing leachate from landfill sites of municipal or industrial hazardous wastes. Under investigation are:

— six synthetic membranes (including polyethylene [PE], plasticized polyvinyl chloride [PVC], butyl rubber sheeting, chlorosulfonated polyethylene sheeting Hypalon , ethylene propylene rubber [EPDM], and chlorinated polyethylene [CPE]

— six admixtures (including asphalt concrete, hydraulic asphalt concrete, soil cement, soil asphalt, bituminous seal catalytically

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blown asphalt, and
bituminous seal-fabric
plus asphalt emulsion)

– eight soil types (including
sands, silts, and clays)

The test program will evaluate the
chemical resistance and durability
of the liner materials over a 12-
and 24-month exposure period to
industrial waste leachates and to
actual landfill leachates in a land-
fill environment. Acidic, basic,
and neutral solutions will be
utilized to generate industrial
waste leachates.

• Encapsulation: The
encapsulation technology program
is evaluating promising organic
and inorganic processes for
both fixing and coating
hazardous materials of
pesticides, soluble organics,
and heavy oil residues. The
process relates to fixing the
material in a 55-gal drum or
up to 500-lb block and then
encapsulating the drum or block
with a nonporous plastic
coating.

• Chemical Fixation: The chemical
fixation technology relates
to transforming the waste into
an insoluble or very low

solubility form to minimize
leaching. The test program
consists of investigating five
industrial waste streams, both
in the raw and fixed state.
Each waste stream will be
treated with five separate
fixation processes and be
subjected to leaching and
physical testing. These lab
studies will identify which
processes should be evaluated
in the field. The five
industrial wastes being
investigated are the same as
those being researched under
the pollutant migration study:

– electroplating

– chlorine production

– nickel-cadmium battery
production

– inorganic pigment manu-
facturing

– calcium fluoride
(electronics)

The following fixation processes
will be utilized with either
industrial waste or flue gas
desulfurization waste. The
assignment of processors to
sludge categories is shown
below:

	Sludge category	
	---	---
Processor	Industrial waste	Flue gas desulfurization
1. International Utilities Conversion System, Inc. (IUCS)	X	X
2. Chem-Fix, Division of Environmental Sciences	X	X
3. Nuclear Engineering Co.-- Tiger-Lok Process	X	
4. Wehran Engineering-- Krete-Rok Process		X
5. TRW Systems Group, Inc.-- Organic Binder	X	
6. Lancy Lab		X
7. Dravo	Calcium fluoride only	X

RESOURCE RECOVERY

Research performed in this area relates to source reduction, recycling, and energy conversion of waste materials:

- Source Reduction: The source reduction activity has primarily related to municipal refuse incineration. Currently, there are no active research studies being performed in this area, and the program is basically finalizing reports of previous studies.

- Recycling: The recycling activity primarily relates to the reuse of carbon black from tires. Specific activities being pursued under this test program are:

  - evaluation of economics and technical feasibility of using ground tires for road surfaces

  - evaluation of economics of recovering carbon black from tires by destructive distillation, hydrogenation, and carbon black feed stock.

Also, the production of methanol from solid waste is being investigated. Specifically, a biological system incorporating enzymatic conversion of waste cellulose to glucose and its subsequent fermentation to ethyl alcohol is being investigated. Some research is being conducted on the utilization of char, a byproduct resulting from the pyrolysis of solid wastes. Thermochemical cycles are being developed to gasify char into synthesis gases, which ultimately are expected to be converted economically to methanol for use as fuel.

- Energy Conversion: The energy conversion activity primarily relates to: the use of waste combustibles as partial fuel in coal-fired boilers; the use of waste combustibles as total fuel for producing electricity from burning shredded waste; and the development of waste as a supplementary fuel. The use of waste as a partial fuel in coal-fired biolers is being performed on a full-scale operation to evaluate/optimize the particle size of shredded refuse required and the firing point location for inputting shredded refuse. Burning characteristics and particulate emission information is being obtained. The use of waste as a total fuel relates to the CPU-400 project, which incorporates the use of shredding, classifying, metal/glass separation/recovery, fluidized bed combustion, granular bed filter, and turbine utilization to produce electricity. The use of waste as a supplementary fuel is being investigated by combusting waste with PVC and lump coal with high and low sulfur contents. The corrosion of fireside metal and the effects of particle size on burning efficiency are being evaluated in this study.

ENERGY

This research program is being expanded considerably throughout EPA since the current energy crisis developed. The two specific energy-related categories that have been assigned to SHWRL relate to waste as fuels and sulfur oxide control.

- Waste as Fuels: The program, presently being funded, will utilize energy conversion processes previously discussed. These processes will be expanded and new activities initiated to identify fuel sources from our various wastes. Specifically, tentative plans allow for program efforts to be pursued in the following areas:

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- waste preprocessing
  assessment

- utilization of municipal
  solid wastes as a
  supplementary fuel with
  coal-fired boilers

- utilization of municipal
  solid wastes as a
  supplementary fuel with
  oil-fired boilers

- co-incineration of municipal
  solid wastes and sewage
  sludge

- pilot-scale pyrolytic
  conversion of mixed waste
  to fuel

- assessment of portable
  pyrolysis for waste
  utilization as a fuel

- assessment of bioconversion
  processes for waste
  utilization as a fuel

• Sulfur Oxide Control: This
program, which is presently
being funded, will allow us
to expand our current efforts
in chemical fixation, soil
attenuation, and liner
evaluation for flue gas
desulfurization (FGD) sludges.
Specifically, tentative plans
allow for program efforts to
be pursued in the following
areas:

- FGD waste leachate/soil
  attenuation studies

- FGD waste leachate/liner
  compatibility studies

- evaluation of alternate
  FGD waste disposal sites

- lab and field evaluation
  of FGD waste treatment
  processes

- development of guidelines
  for FGD waste disposal
  standards

- lab and field evaluation
  of FGD waste disposal
  options.

CONCLUSION

The laboratory-sponsored project
areas and studies discussed here
reflect the SHWRL overall effort
in solid waste management research.
Some of the programs will be
discussed in much more detail by
the following speakers. More
information about a specific study
or project is available from Robert
L. Stenburg, Director, Solid and
Hazardous Waste Research Laboratory,
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CURRENT OFFICE OF SOLID WASTE MANAGEMENT PROGRAMS LANDFILL ACTIVITIES

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Since its inception, the Federal solid waste program has recognized the need to use the land as a waste sink. We have focused a large part of our efforts on improving land disposal practices. This is evidenced by our grant and contract activities and publication of the state-of-the-art document "Sanitary Landfill Design and Operation." This document served as the basis of the "Guidelines for Land Disposal of Solid Wastes" which we promulgated last year.

Most of our earlier efforts were directed at improvement of operational practices of land disposal. This made sense in that the greatest impact could be achieved at the least cost by concentrating on operations.

It has become clear that although operational improvements are desirable, they alone do not provide for environmentally acceptable use of the land as a waste sink. This has been supported by continuing and recently intensified research in the areas of leachate and gas formation and migration. Even where operational improvements have been made, leachate and gas problems can exist, especially where site selection and design are inadequate.

With regard to leachate and gas, I wish to describe several of our efforts which we hope will provide data and technology to alleviate potential problems associated with the land disposal of wastes.

LEACHATE

Our leachate program is structured along three lines: Technology Demonstration and Evaluation, Damage Assessment, and Institutional Mechanisms for Preventative and Remedial Action.

We anticipate a future need for expanded land disposal standards or guidelines at the Federal and/or state level. These standards or guidelines would address leachate in a more definitive manner than our existing guidelines. Logically, before drafting such standards, we would develop an issue paper justifying the standards and setting forth their rationale. Several projects have been defined to provide assessments of leachate damage and leachate control technology. These assessments would form the basis of the issue paper.

Under contracts, we are instrumenting and will be monitoring 16 existing disposal sites. From this effort, we hope to learn about leachate generation and movement under various field conditions. This will provide us with an estimate of the extent of actual or potential damage.

Other field monitoring projects which will provide similar information include:

1. Continued monitoring of the Orange County, Florida, demonstration sanitary landfill. This is a special case in which the fill was constructed in a high water-table area.

2. Continued monitoring of field lysimeters constructed in conjunction with our milling demonstration project in Madison, Wisconsin. These lysimeters will provide information on leachate from millfill, both covered and uncovered, and sanitary landfill cells.

3. Monitoring of a balefill test cell constructed in St. Paul, Minnesota.

The next part of the damage assessment is to assess the economic impact of poor waste disposal practices on groundwater contamination. This activity is an in-house examination of damage cases and will continue through 1975.

The final link in the damage assessment is the determination of groundwater movement and the extent to which soil attenuation acts as a mechanism in reducing harmful effects of leachate. Input to this area is being provided by studies conducted by EPA's Solid and Hazardous Waste Research Laboratory in Cincinnati, Ohio.

Our thrust in the area of institutional mechanisms is to develop a model enforcement procedure for use by state and local governments. This effort will address administrative procedures for use in enforcement, as well as provide guidance in field monitoring for problem detection and evidentiary purposes. Development of this guidance document will be by contract with guidance

from a team of state solid waste management agency directors.

In many cases, the ability to control leachate requires that it be collected and treated prior to discharge into the environment.

To make available information on the collection of leachate, we have compiled a state-of-the-art paper on various artificial liner materials. Studies on the durability and longevity of several such materials are being conducted under the auspices of the Cincinnati Laboratory. Through funds provided by the Appalachian Regional Commission, we will be able to support the construction of a lined landfill in Lycoming County, Pennsylvania. This project will provide documentation of the construction techniques and costs related to the development of a well-designed, environmentally controlled sanitary landfill. In the future, we hope to be able to instrument and monitor existing lined landfills for leak detection.

In the area of leachate treatment, we hope to demonstrate the following basic approaches:

1. Anaerobic filter

2. Spray irrigation

3. Activated sludge

4. Municipal sewage treatment facility.

Of these four basic approaches, we are farthest along with the anaerobic filter demonstration. This project will be conducted in Enfield, Connecticut, in cooperation with the State of Connecticut, the Town of Enfield, the University of Illinois, and A. W. Martin Associates, Inc. The project has been conceived as consisting of five phases. Preliminary analyses have been initiated to define the characteristics of the leachate to be treated. Bench-scale testing will be used to determine the design

parameters for the filter system. The site's hydrology will be analyzed and described, providing an assessment of leachate flow rates on which to base system design. The system will then be constructed and its performance evaluated.

We are presently negotiating a project which would provide us with an evaluation of combined physical/chemical and biological treatment systems. The facility is presently under construction in the State of Pennsylvania. This project would provide information on the field-scale efficacy and costs of physical/chemical and biological unit processes operated independently and in combination. We are also continuing to monitor the leachate recirculation system constructed in Sonoma County, California. A report on the initial 3 years of this project is now in press. In the future, we hope to be able to identify and document various leachate treatment systems in use across the country.

GAS

We view landfill-generated gas as both a potential hazard and a potential energy source.

At Winston-Salem, North Carolina, the gas proved to be a hazard. Gases allegedly migrated from an adjacent closed dump into a National Guard Armory. The gases accumulated to an explosive concentration, and, on ignition, resulted in a flash fire. We have recently completed a project which resulted in construction of a barrier system, with back-up alarms, to protect the building from migrating gases. The barrier consists of a series of wells installed between the dump and the building. A pump runs continuously to maintain a negative pressure in the wells, thereby intercepting and exhausting migrating gases. Monitoring wells were used to evaluate the barrier at various

pumping rates. A report describing system design, construction, costs, and evaluation is in press.

We have considered that withdrawal of landfill gas for use as an energy source could have a dual benefit: providing energy from an otherwise wasted source, as well as venting the gas which otherwise might pose a hazard through lateral migration. Thus, we have funded a pilot effort at the Mountain View, California, sanitary landfill to achieve the following goals:

1. Determine optimum gas withdrawal rate and area of influence of a production well at a given flow rate

2. Determine marketability of gas

3. Determine effects of moisture addition on gas production/recovery.

The original gas extraction system consisted of a three-stage well from which gas was withdrawn using a truck-mounted pump. Once withdrawal was initiated, the truck engine which powered the pump was converted from gasoline to landfill gas simply by directing a portion of the gas flow through the carburetor. Gas pressures are monitored at various depths and distances from the two production wells now in use. Gases are being withdrawn from the three-stage well systems at various pumping rates in order to achieve the desired objectives.

A more permanent self-contained system has been installed for continued pumping. The pump is driven by a Volkswagen engine which runs on the landfill gas. During this testing, the gas is being flared. Gas composition at the various pumping rates is being closely monitored by the Pacific Gas and Electric Company. PG&E is also conducting a marketability study as part of the project. This will provide information on the potential economics of marketing

this energy source in various forms.

We are presently negotiating a project which would expand on the Mountain View withdrawal studies and provide information on the cleansing of landfill gas to pipeline quality.

Thank you for the opportunity to summarize our efforts in the areas of landfill leachate and gas.

CURRENT SOLID WASTE RESEARCH ACTIVITIES IN NEW YORK STATE

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Before getting into the research activities, I would like to give you some idea of the magnitude of the solid waste problem in New York State. In the past 12 years, the number of disposal sites has decreased from about 1600 to the current 779. Regionalization of local solid waste activities has been the main reason for this reduction. For example, there are 92 multi-municipal systems serving 617 municipalities. Included are 10 systems operated on a county-wide basis.

Currently, there are 22 proposals for developing resource recovery systems, which will enhance the trend of regionalization and at the same time, recover large quantities of wastes for reuse or energy production. One point should be made here: regardless of the development of major resource recovery systems, there will still be a need to provide for land disposal of wastes in areas not economically served by resource recovery and for residues from resource and other processing techniques.

Since the landfill cannot be phased out entirely, we must minimize its impact upon the environment. For this reason, we are now developing new comprehensive rules and regulations which will provide for construction and operational permits and will require certified operators at all landfills. In addition, there will be requirements for groundwater monitoring wells for all new sites.

In reviewing designs for new and revised landfills, we want to be assured that impacts upon groundwaters are minimized. In some areas, such as Long Island, the only way to be assured of protecting the groundwaters is by requiring liners; however, we do not want to require liners in all cases as some states are doing now. We would much rather minimize the amount of water reaching the refuse using proper materials and construction techniques and then rely upon the soils to attenuate what leachate may be produced. Unfortunately, we do not know enough about the natural attenuation capabilities of New York State soils. I am very pleased to hear of the meaningful research underway in this area; however, additional work is necessary before we are at the point where we can include soil attenuation as a part of landfill design. The next phase of research we would like to sponsor is in this area.

Currently, the only solid waste research which we are sponsoring is being performed at the Civil and Environmental Engineering School at Cornell. In this project, leachate from two landfills in the central part of the state is being extensively sampled at a series of surface sample points in springs

and streams below the disposal sites. By evaluating these data, we hope to learn more about the conservative nature of various pollutants. Flow measurements in the streams allow an evaluation of the influence of dilution. This information will then be used to select a set of leachate indices. In making this selection, ease of analysis and interpretation of results will also be considered.

This project is being run concurrently with a remote sensing project funded by EPA. Various types of films and a thermal scanner are being used at different elevations, during daylight and at night, in an attempt to detect the presence of leachate from landfills. The ground sampling program acts as the ground control. The results to date are encouraging in that leachate springs and polluted streams can be readily located on the remote sensing imagery.

In summary, leachate is the last unknown aspect of landfilling. Some of the work now underway should prove to be very helpful; however, much more research is needed before we will truly understand leachate and how to cope with it. If we can find the funding, New York State hopes to continue sponsoring this needed research.

CURRENT SOLID WASTE MANAGEMENT ACTIVITIES IN PUERTO RICO

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San Juan, Puerto Rico

Puerto Rico is the smallest, most easterly island of the Greater Antilles. It is roughly rectangular in shape, with an area of 3,339 sq. mi. The island is bounded by the Atlantic Ocean to the north, the Caribbean Sea to the south, Vieques Sound to the east, and the Mona Passage to the west.

The island is characterized by rugged, mountainous terrain, with the principal topographic feature being the Cordillera Central, a mountain range that runs east-west across its center. Portions of this range rise to elevations of more than 4000 ft, and it is the principal drainage divider of Puerto Rico. Alluvial soils characterize the coastal areas.

The climate is affected by steady trade winds and is subtropical. Rainfall averages 69 in./yr.

Much of Puerto Rico's land is being intensively utilized, and urban and industrial development has been increasing during the last 20 yr at a tremendous rate.

With nearly 2.8 million people, Puerto Rico's population density of about 800 persons/sq mi is one of the highest of the world. The island's rapid population growth is expected to continue at a high rate. These trends toward accelerating population growth and a shift to the cities strongly indicate that solid waste generation will increase. Production of household and commercial solid waste by individual municipalities is presently overburdening in its scope, at a current production of over 5700 tons/day, and an anticipated production of over 14,000 tons/day in the next 20 yr.

Today, land disposal is the method widely used in Puerto Rico for disposing of municipal and industrial solid waste.

We have been advocating and using for the last 5 yr the sanitary landfill method of solid waste disposal. Twenty-seven of the seventy-eight municipalities had developed sanitary landfills, and by January 1, 1976, all municipalities will have to comply with the requirements established by Environmental Quality Board Regulation for the Control of Solid Waste.

Recognizing that land disposal of solid waste poses many problems to local government agencies, the Environmental Quality Board has made special efforts to examine the environmental effects of leachate from dumps and other improperly designed and located landfills. It has become clear that leachate from municipal solid waste land disposal sites can cause serious and expensive problems. Also we recognize the need for factual information about the actual or potential threat that sanitary landfills may present to the water quality of the local aquifers and surface waters. We have initiated

research activities in coordination with the U.S. Geological Survey on leachate formation at different sanitary landfill sites, either closed or in operation.

A proposed long-range plan for a continuing monitoring program of solid waste disposal sites has been discussed with personnel from the U.S. Geological Survey. Groundwater quality monitoring will be conducted at points relative to groundwater and in areas of high utilization of groundwater. Also, studies will be conducted on sanitary landfill sites located on specific geological formations throughout the island. It will include, too, sites used for the disposal of industrial toxic and hazardous solid wastes.

Presently, we are performing an island-wide survey on industrial hazardous and toxic wastes. This study is oriented toward the formulation of a state plan for the management of hazardous and toxic waste with respective regulations. More than 300 industries are being surveyed.

Also, during the period year 1975, efforts have been concentrated on the development of advanced resource recovery with the following activities:

1. Economic feasibility study for using the metropolitan San Juan solid waste as an auxiliary fuel. A contract has been signed with the Water Resources Authority to conduct this study.

2. Investigation of existing markets for segregated solid wastes, paper, glass, and other metals.

3. Plans for the collection, transportation, processing, and final disposal of junked autos.

Other activities have been developed for the continuing

implementation of the solid waste program: enforcement activities, and the Public Image Education Program and Community Information (The ENCESTE Program).

BAYAMON LANDFILL STUDY--
A CASE STUDY

In 1970, the Municipality of Bayamon started the operation of a sanitary landfill for the disposal of municipal solid waste. The landfill, located at Barrio Buena Vista, about 8.0 km east of Highway 178, was closed in 1974. Operations in the landfill were discontinued by order of the U.S. District Court in San Juan, Puerto Rico, as a result of a lawsuit by residents of the area.

Prior to the closure of the landfill, and at the request of the U.S. District Court, the U.S. Geological Survey (WRD) conducted a field test and collected and analyzed samples of the leachate flowing from the landfill (June and July, 1972).

The leachate problem was corrected after the municipal administration was ordered to correct deficiencies noted (July 1972). Engineering works were performed providing drainage facilities and operating the landfill satisfactorily.

The leachate problem appeared again after closure of the landfill in 1974 and was aggravated during the months of October through December 1974 after a heavy rainy season. Excessive leachate continues to emanate from the landfill, contaminating adjacent water bodies. This situation was due primarily to the following conditions: improper operation of the site, elimination of the drainage system, and poor maintenance of the site after closure of the sanitary landfill.

In September 1974, the Federal District Court named a committee to study the conditions of the

landfill and to recommend alterna-
tives for the rehabilitation of
the site and surface waters being
contaminated by leachate produced
by the excessive infiltration of
runoff waters into the landfill
site.

Field tests of the leachate
flowing from the landfill were
conducted by the U.S. Geological
Survey at the request of the
Environmental Quality Board and
the Municipality of Bayamon.

The Bayamon landfill site study
sought to evaluate the physical
and chemical characteristics of
the leachate. In addition, testing
was conducted to evaluate the ability
of various soil filtration media
to filter and retain some of the
contaminants in the leachate, if any.

Reconnaissance

A reconnaissance of the Bayamon
landfill at Barrio Buena Vista was
made January 27, 1975. The
topographic characteristics of the
area indicated that the landfill
is located on a ravine over the
headwaters of a small creek, which
receives base flow and surface
runoff from the adjacent areas.
A state of supersaturation has
occurred within the fill due to
groundwater movement from percolation
of precipitation and from surface
runoff. Leached fluids produced
drain through three main sites
at the tail of the landfill.

The aerobic decomposition of
organic matter produces offensive
odor as a result of the formation
of gases, principally methane,
carbon dioxide, ammonia, and hydrogen
sulfide.

A survey was made on June 8
and 9, 1972, in which U.S. Geological
Survey personnel collected samples
of the surface and groundwater in
the vicinity of the landfill.

Sampling and Field Test

Samples of leachate were
collected from the three main
draining sites and a combined flow
sample was collected at about 100
ft downstream from the tail of the
dump site (Figure 1). A 40-gal
leachate composite sample was
collected to be used for a filtration
laboratory test employing different
filtering media.

Field data for the collected
samples are shown in Table 1.
Results of laboratory analyses
for the different pollutional
parameters were tabulated.

Laboratory Filtration Test

The purpose of this test was
to determine the ability of different
materials to remove or retain
contaminants from the leachate.
The experimental filtration units
used consisted of four 2.5-ft-long,
6-in.-diameter plastic pipe filters
containing 1.5 ft of porous media--
-grain-size particles ranging from
0.019 in. to less than 2.0 in. in
diameter. The following materials
(suggested by geologist Mario
Soriano) were used as filtering
media: limestone from Bayamon,
crystalline limestone from Mayaguez,
siltstone from Guaynabo, and sand
from San Juan.

Five gallons of the composite
leachate sample were filtered through
each column. The filtrate was
refiltered until a constant pH and
conductance value were obtained
(Table 2).

Significance of Parameters

The following section briefly
presents the significance and certain
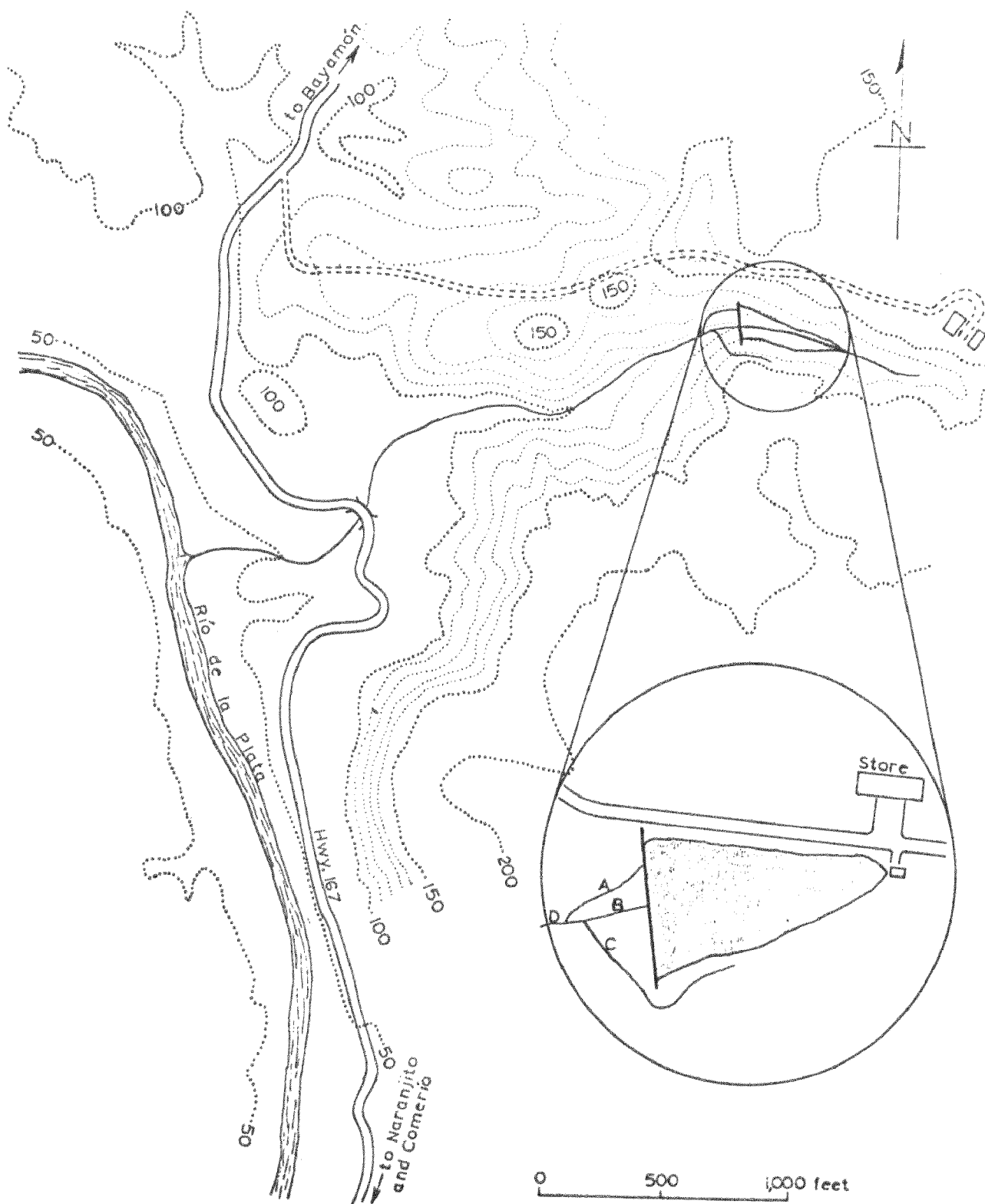permissible limits of the various
pollution parameters determined.

Figure 1. Location of landfill and sampling stations.
Contour in meters.

Table 1.   BAYAMON LANDFILL SAMPLING STATION FIELD DATA,
JANUARY 27, 1975.

Station	Nature	Time	Flow, gal/min	Temp, C	pH	Alkalinity, HCO_3	Specific conductance, mhos at 25 C
A	Leachate	1110	7.5	25.0	7.8	1880	6600
B	Leachate	1130	8.6	25.5	7.9	1200	3000
C	Leachate	1145	4.8	25.0	7.7	840	2880
D	Combined flows	1210	20.2	25.5	8.1	1440	3490
40-gal composite sample	Leachate	--	--	25.0	7.8	1540	3700

Table 2.   BAYAMON LANDFILL LABORATORY DATA: FILTRATION
EXPERIMENTS, JANUARY 28, 1975.

Filtering media	Trial	Temp., C	Specific conductance, mhos/25 C	pH	Filtration rate, gal/min.
None	Original	25	3700	7.80	--
Limestone, Bayamon	1	26	3520	7.65	0.67
" "	2	26	3500	7.70	0.86
" "	3	26	4320	7.75	0.57
" "	4	26	3450	7.70	0.82
None	Original	25.5	3650	7.80	--
Crystalline limestone, Mayaguez	1	26	3500	7.65	1.33
" "	2	26	3400	7.65	1.25
" "	3	25.5	3300	7.65	1.15
" "	4	25.5	3290	7.65	1.35
None	Original	25	3700	7.75	--
Limestone, Guaynabo	1	25.5	3475	7.65	1.14
" "	2	25	3400	7.60	1.12
" "	3	25	3400	7.65	1.04
None	Original	26	3720	7.80	--
Sand, San Juan	1	26	3700	7.60	0.25
" "	2	25.5	3875	7.60	0.20
" "	3	26	3700	7.60	0.22
" "	4	25.5	3720	7.60	0.23

BOD (5-day). Biochemical oxygen demand (BOD) for most practical purposes represents a measure of the carbonaceous organic material usable as a source of food by aerobic organisms. High BOD values usually result in depressed dissolved oxygen values. Low BOD values are a characteristic of clean water.

pH. Most natural waters on the island range from 6.8 to 8.6 pH. Departure from this range in either direction is indicative of some form of acid or basic pollution.

Coliform (E. coli). Membrane filter (MF) procedure at 35 C. The sanitary significance of this test is that some of the bacteria detected, but not necessarily all, are from the gut of warmblooded animals. Recommended raw water maximum count is 10,000/100 ml. (National Technical Advisory Committee, 1968, Water Quality Criteria: Federal Water Pollution Control Administration, p. 20).

Coliform (fecal). MF procedure at 44.5 C. At this controlled temperature, only bacteria from the gut of warmblooded animals survive. Recommend raw water maximum count is 2,000/100 ml. (National Technical Advisory Committee, 1968, Water Quality Criteria: Federal Water Pollution Control Administration, p. 20).

Streptococci. MF procedure at 35 C. This group indicates pollution since streptococci are generally from the intestines of man and other animals. Some authorities state that ratios between fecal coliform and streptococci can be used to differentiate between man and the various types of animals from which the bacteria are derived.

TOC. Total organic carbon (TOC) is a direct measure of carbon in organic substances such as domestic and industrial wastes. Low values are desirable. High values of TOC would be expected with high values for BOD or COD and vice versa.

Nitrate. Represents the final stage of the oxidation of nitrogenous material. It is seldom found in Puerto Rico surface water in a very high concentration, even though the water analyzed may be highly polluted. Short stream travel time does not permit sufficient time for oxidation to proceed to completion. It is far more significant as a pollution indicator in groundwater supplies. High nitrates, 45 mg/ℓ, preclude the use of the water in infant's food intake. (U.S. Public Health Service, 1962, Drinking Water Standards. PHS Publ. No. 956, p.7-8).

Ammonia nitrogen. Bacterial decomposition of nitrogenous organic substances produces ammonia nitrogen, so its presence is taken as evidence of pollution.

Organic nitrogen. This is a Kjeldahl procedure that measures nitrogen derived by chemical decomposition of nitrogenous organic substances such as amino acids and proteins. Its presence is evidence of pollution.

Kjeldahl nitrogen. If the Kjeldahl procedure is made without prior removal of ammonia nitrogen, it is equivalent to the sum of ammonia nitrogen and organic nitrogen and is sometimes referred to as total Kjeldahl nitrogen.

Phosphate. An oxidation product of sewage, a component of detergents and textiles. Promotes undesirable algal growth. Because of the complexity of phosphate relationships, no generally acceptable limits have been established.

Specific conductance. Sixty-five percent of the specific conductance roughly equals the dissolved solids present. Maximum desirable limit for public supplies is 500 mg/ℓ. (U.S. Public Health Service, 1962, Drinking Water Standards. PHS Publ. No. 956, p. 7-8).

Toxic metals. Because of recent interest and concern regarding certain toxic metals, determination of mercury and arsenic have been made. A maximum of 50 micrograms/ℓ arsenic is permitted in drinking water. (U.S. Public Health Service, 1962, Drinking Water Standards. PHS Publ. No. 956, p. 7-8). No maximum standard has been designated for mercury. Mercury and arsenic would not be in significant quantities in ordinary sewage but might be expected in certain industrial and agricultural effluents.

Conclusions

Results of laboratory analyses of samples collected in the 1975 study indicate a progressive improvement of the mixed leached fluids due to dilution and aeration of the leachate on its way downstream.

Data collected at site D in 1972 and 1975 demonstrated that the mineral composition of the leachate has not changed. However, several mineral and biological parameters exhibited lower concentration values in 1975. These variations suggest that periodic surveys of selected parameters should be made to monitor possible quality of water changes in the downstream flow and in landfill fluids.

Flow measurements are needed to determine the effects of variable climatological conditions in the landfill and streamflow.

Preliminary results of the filtration experiment denoted that none of the materials used has any ion-exchange capacity. The reduction of iron in the limestone from Mayaguez and sand from San Juan is attributed to occlusion or adsorption of ferric hydroxide precipitates formed when groundwater containing iron in the ferrous state comes in contact with air and when bicarbonate ions are present.

The following engineering works and actions were recommended as possible rehabilitation measures in correcting to a certain degree the degradation conditions created by the landfill.

1. Alteration of leachate chemistry using several geologic materials to regenerate the quality of leachates percolating through it.

2. Stabilizing landfill fill and filtering media, based on the results of the study performed by the U.S. Geological Survey.

3. Runoff water control through engineering works.

4. Control of water infiltrating into the landfill.

5. Site grading through application of 24 in. of clay materials to reduce the infiltration of runoff water. Maintaining the grade at a minimum slope of 1 to 3%.

Land disposal of solid waste is needed today and will be needed in the future. Even the most complete and effective system of resource recovery that can be envisioned still will leave residues which must be disposed of on land, in ways that do not pollute. It is further recognized that a sanitary landfill, unless properly engineered and on a suitable site, can pollute surface and groundwater resources.

BIBLIOGRAPHY

1. Blannon, J.C., and Peterson, M.L. Survival of Fecal Coliforms and Fecal Streptococci in a Sanitary Landfill. News of Environmental Research in Cincinnati, U.S. Environmental Protection Agency, Cincinnati, Ohio. April 12, 1974.

2. Brunner, D.R. and D.J. Keller. Sanitary Landfill Design and Operation. Washington, U.S. Government Printing Office, 1972.

3. Garland, G.A., and D.C. Mosher. Leachate Effects from Improper Land Disposal. February 1975.

4. PR Environmental Quality Board. Comprehensive Solid Waste Management Plan for Puerto Rico, 1971.

5. U.S. Environmental Protection Agency. Recommended Methods of Reduction, Neutralization, Recovery or Disposal of Solid Waste. Vol 1.

6. U.S. Environmental Protection Agency. Thermal Processing and Land Disposal of Solid Waste; Guidelines Federal Register, 39 (158)-29327-29338. August 14, 1974.

7. U.S. Geological Survey, San Juan Office. Bayamon Landfill Site Study. January 1975.

8. U.S. Public Health Service Drinking Water Standards, revised 1962.

LANDFILL RESEARCH WORK IN PROGRESS AT HARWELLS HAZARDOUS MATERIALS SERVICE

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INTRODUCTION

I would like to take this opportunity of thanking the conference organizers for the invitation to attend this meeting. Within the United Kingdom we have a considerable amount of landfill research in progress and I hope to explain in this lecture Harwell Laboratories' involvement in the work so that a more effective interchange of information can take place in the future.

The Atomic Energy Research Establishment of the United Kingdom Atomic Authority, like many nuclear research establishments throughout the world, has diversified into nonnuclear projects and now prefers to be known as the Harwell Laboratory. About half of the establishment's income currently comes from nonnuclear work, such as the Hazardous Materials Group, which was initiated by Dr. Frank Feates. The group consists of 30 staff, but it uses the services of some 60 other specialists in other divisions of Harwell including software programmers, information officers, chemical analysts, environmental health physicists, and civil engineers. We also have a team of hydrogeologists attached from the Institute of Geological Sciences to help us with a large research programme.

Before describing the landfill programme in some detail, I have been asked to describe some of the other services and activities of the group. Although not all directly concerned with landfill, they provide us with a wide range of knowledge and expertise in the general area of toxic waste disposal, which has been invaluable during the landfill research programme.

LANDFILL SITE RECLAMATION

The group has provided all the chemical advice required by the Cheshire County Council during the conversion of the Malkins Bank landfill into a golf course. This large landfill, dating back to the very early days of the Imperial Chemical Industry, was in a very hazardous condition. During the reclamation, it was necessary to provide continuous surveillance of the civil engineering operations, which involved moving many thousands of drums of hazardous chemicals, and advice on whether they could be safely buried or treated in other ways. A mobile laboratory on the site was required to enable on-the-spot decisions to be made on the method adopted and to help identify the wastes. A culvert running through the whole site was seriously

contaminated, and its banks were covered in places with an almost attractive multi-colored slime of chemical wastes from the years gone by!

The same team of experts has also been involved in supervising the demolition of a large zinc smelter to ensure that the dust, some of which was arsenical, did not contaminate surrounding property and that safe working conditions were provided for demolition personnel working at the site. Health physicists, using air samples, carried out much of this work.

Following on from these tasks, we have been involved in mercury decontamination, beryllium decontamination, and looking for spent cyanide drums in an area required for motorway developments. Though this was not directly landfill research, the people involved in these jobs have obtained practical experience working with hazards in the field over a wide range of toxic wastes, from the different types of hazards they present and the different physical forms in which they arise.

THE CHEMICAL EMERGENCY CENTRE

Many people have sought our advice over the years in the safe disposal of dangerous chemicals. The Chemical Industries Association has also been concerned about the safe transport of chemicals around the United Kingdom. As a result, we have set up a Chemical Emergency Centre at Harwell approved by both the Chemical Industries Association and the Department of the Environment. There is a chain of similar Emergency Centres throughout the United Kingdom operated by various chemical manufacturing firms as well. Each centre advises the police and fire brigades on actions to be taken in the event of an emergency. We act for the areas near Harwell, but also act as a central source of information on the hazards associated with trade-

named chemicals from all over the world. Much of these data are already on a computer file, and it can be searched for by a software programme that provides a visual display (e.g., all chemicals in 2-gal blue plastic drums). This data bank is also useful to us in identifying chemicals in landfill sites.

It is presently being extended to give much more detail on certain pesticides and herbicides under a contract with the European Economic Community.

Practical experience has been obtained in cleaning spills in motorways, blowing up peroxides that were unsafe, removing dangerous chemicals using remote handling equipment, safe igniting of hazardous and toxic chemicals, explosive destruction of unsafe gas cylinders, emptying severely corroded oleum containers of many tons weight, sea dumping of four train loads of toxic chemicals (allowed under the international regulations), repacking explosives that cause dermatetic effects, removing drums from beaches, etc. Some of these have been on landfill sites. The practical experience of the real live hazards is again a useful adjunct to advising the central government on the real hazards involved with some rather unusual wastes that, in the past, were probably discreetly dumped somewhere!

We have a specially equipped vehicle and mobile laboratory, manned 24 hours of the day, to go out to chemical emergencies near Harwell, if asked to do so by the Police or Fire Services.

THE INDUSTRIAL WASTES INFORMATION BULLETIN

To ensure that there is a source of information available to industry on the disposal of industrial wastes, we produce, under Department of the Environment sponsorship, a monthly bulletin

of 200 abstracts of articles on industrial wastes. These articles are grouped together and one of the groups is "landfill." In addition, those of us involved in landfill research do have our own printout each month of a somewhat wide range of articles on landfill research topics, which would not be of interest to the average reader of our bulletin.

All the documents are key worded, and hard copies are held in our library. Subscribers to the bulletin (current subscription: United Kingdom subscribers--30; overseas--40) can have keyworded searches carried out. A photocopy service is also available. There are currently some 400 subscribers, many from overseas, and we answer two or more telephone enquiries on industrial waste topics each day. The impending implementation of the Control of Pollution Bill that, amongst other things, controls landfill operations, has resulted in a general increase in enquiries.

DATA BANK ON MATERIALS DEPOSITED IN LANDFILLS

Under a previous Act of Parliament (The Deposit of Poisonous Wastes Act), a person placing a potentially hazardous material in a landfill had to notify the appropriate authorities. Data from over a million such notifications made in 1972 have been filed at Harwell. We can search the data to produce printouts of a wide range of information for the central government to aid them in developing future landfill legislation. The place at which a particular chemical has been deposited can be determined, or the ratio of liquid-to-solid toxic wastes, etc.

WASTE SURVEYS

As a follow up of this exercise, we have done surveys of the waste arising in the Mersey Valley for the Runcorn Development Corporation and also have developed jointly with Pencol consultants a long-range plan for waste disposal sites in Cheshire. A digest of our experience in long-range planning has been written for central government and now a course is run regularly at Harwell to train waste disposal officers of the County Council Authorities who will, in the future, have to prepare plans for their own areas when the Control of Pollution Bill is fully implemented.

We have also analyzed large numbers of soil, waste, dust, and leachate samples from various landfill sites. In the light of these analyses, we have advised on potential reclamation of sites and reuse for reduction of hazards from leachate by recirculation.

THE LANDFILL RESEARCH PROGRAMME

The Control of Pollution Bill, which has been passed by Parliament but not yet fully implemented, says in one of its sections that landfill sites will have to be licensed for particular types of waste and that, eventually, long terms must be developed by each county to dispose of its own waste, either in sites operated by the County authorities or in commerically operated sites.

The Landfill Research Programme was initiated to provide the Department of the Environment with the technical information required for such a site licensing system. The programme, costing £1,200,000 at 1972 price, is carried out by two teams--one based at the Water Resources Centre and the other at Harwell. Our Harwell team has been strengthened by the addition of a team of hydrogeologists from the Institute of Geological Sciences, under Dr. John Mather.

Some 20 landfill sites are being examined by the combined teams to provide actual field information on the complex interactions that

occur in a range of different types of landfill. There is also a supporting lysimeter programme. The studies in the behavior of selected toxic wastes within the refuse and the generation of leachate are being carried out by the Waste Resources Centre. Studies of the subsequent movement of the leachates through the rock structure in the unsaturated zone to the aquifer are being carried out at Harwell.

LANDFILL SITE INVESTIGATIONS

Before the Landfill Research Programme had commenced, the Institute of Geological Sciences had carried out a survey of some 3000 landfill sites in the United Kingdom and identified, mainly from a desk study, some 50 or so sites where a possible pollution hazard might exist. Many of these sites have been visited, and sites have been chosen for detailed study. The aim has been to select sites that cover the different kinds of toxic waste and that are also situated on a wide range of geological strata likely to be used for landfill sites.

Once a site is chosen, the records of what has been deposited are studied in more detail, and the detailed hydrogeology of the area is established. We then take samples of refuse, surface waters, and any lagoons of visible leachates. Then a drilling rig is used to obtain samples of the rock structure and the refuse at a matrix of points chosen to enable us to follow any pollution plume from the landfill. The samples are sealed in the field, removed to the laboratory, and centrifuged in a cooled centrifuge to give leachate or water samples for analysis.

Work has not been completed at Eastfield Quarry in Midlothian in Scotland, and a report, being prepared for the Department of the Environment, will be published as soon as possible. Work on a chalk site in Oxfordshire, a lagoon site

in the midlands, and a site containing food industry wastes has been completed. A further site in Scotland is still under investigation. The results are tending to show that many toxic materials present in the landfilled material have migrated hardly at all and that fissures, mine shafts, faults in rock structures, stratification of permeable zones, and perched water tables make the simple hydrogeological concept of leachate flowing through a simple unsaturated zone to an aquifer an oversimplification in many cases.

A report covering the early steps of this work was issued by the Department of the Environment in September 1974 entitled "Programme of Research into the Behavior of Hazardous Wastes in Landfill Sites."

In addition to the rather extensive study of the above-mentioned sites, some small-scale field studies have been made on the movement of mercury wastes from a range of different industries. The specification of the migrating mercury species and the mechanism pointing its movement are quite complex within a landfill situation. The absorption of trace metals in particulate materials needs to be investigated and great care taken over filter-paper pore sizes if tone ionic concentrations are required. Adsorption of vessel walls and the time and place of acidification of samples all need careful attention in this kind of work.

LYSIMETER EXPERIMENTS

It is not for me to describe the Water Resources Active experiments, except for the sake of completeness. They operate bays of pulverized refuse both aerobically and anaerobically. To the bays they have added either layers of cyanide waste, or a metal hydroxide wastes, or a solution of cutting oils. They hope to obtain mass balances to account for each of the materials

when they eventually take the bays to pieces. They use natural rainfall conditions and also simulate the same experiments in the laboratory.

At Harwell, we have a complimentary programme studying the movement of toxic leachate through sandstone, chalk, and gravel. We also have one large-scale experiment in each rock structure backed up by appropriate smaller scale laboratory column studies.

The sandstone lysimeter experiment is situated some 17 miles from Harwell. Meter-wide trenches were dug through the 2 m of greens and 1 m into the underlying clay and filled with cement to create four, 4-m by 4-m blocks of in situ sand, 2 m deep, within a concrete walled box. Synthetic leachate solutions sprayed on the top surface move through the sandstone to the impermeable clay layers, where they flow out through an exit pipe system installed from an adjacent underground laboratory. In addition, an extensive array of piezometers and suction probes measure suction potentials and collect water samples from the unsaturated zone. The suction probes are continuously peristaltically pumped at a flow rate equal to one-twentieth of the flow that would pass the cross sectional area of the porous zone of the probe. This ensures the minimum of disturbance to the flow conditions. Such probes have now operated continuously for over 6 months. The four bays and underground lab are housed in a large, farmers barn!

The synthetic leachate solution is applied from 100 outlets over each bay. The first solution contains metal ions up to 100 ppm concentration, together with fatty acids at concentrations similar to those found in landfill leachates. Steady-state unsaturated flow will be established with top water at constant inlet rate equivalent to about four times normal rainfall. Then a sudden switch is made to a continuous supply of leachate. We hope to follow the movement of

the pollution front through the bay, both by examination of liquid from the suction probes and from small cored samples of sand removed from one corner of the experimental plots. Small-scale column experiments in the laboratory have shown that the likelihood of any movement of the metals to any great depth is small. The presence of the fatty acids tends to enhance movement, and in practice, particulate matter blocks the pores at the refuse/rock interface. Ferric hydroxide gels and microbial growths that form also block the interface and adsorb many metal species. However, we are attempting to show how cleanup of leachate occurs once the leachate has entered the unsaturated zone so these other processes are a further bonus to reducing pollution.

It is not possible to study percolation through fissured chalk in exactly the same way. We have, therefore, adopted the following approach. A pumped well has been constructed in the chalk at Harwell. Initially, we are going to follow the rate at which water, applied at different rates to the surface, reaches the water table. Very slow water addition may move, by intergranular flow, through the bulk of the chalk, but if the same volume of water is added suddenly, much of it will flow rapidly through the chalk fissures to the water table. In addition to these studies in water flow, we have been pouring synthetic leachate into plots of chalk and taking chalk samples after varying periods of time to see if the pollution front has moved at all under natural rainfall conditions. It may be possible to a limited extent to combine the two approaches and apply polluted leachates at different rates to the ground near our pumping wells and recover the pollutant by pumping.

Gravel is even more complex. It contains sandy, clayey horizons, and leachates may move by complex flow paths through gravel below a landfill site. To study this, we have a ring of wells in a gravel-

over-a-clay base, and we intend
to apply a leachate to the centre
point and monitor its movement past
the ring of wells. Laboratory
backup experiments in the adsorption
properties of the sandy, clayey,
and gravely horizons are in progress.

Experiments have also been
carried out using undisturbed columns
of soil, which are taken by a
technique developed at Wageningen.
A 2-m-high fibre-glass cylinder,
about 1 m in diameter, has a sharp
metal cutting edge in its lower
end. It is hydromatically drawn
into the ground and then a plate
is slid underneath the cylinder.
The rock core is lifted out in the
fibre-glass tube and removed to
the laboratory. We are hoping that
this technique may be successfully
used to obtain areas of the bases
of many potential landfill sites
for laboratory assessment of their
potential sorptive capacities without
being subject to the edge effects
of smaller laboratory column experi-
ments.

Currently, cores of this type
of Uffington greens and chalk and
disturbed gravel are being exposed
to our first synthetic leachate
mixture.

One incidental observation
that has been made is that when
the clear liquid centrifuges from
the rock cores taken from under
Eastfield Quarry landfill is left
standing exposed to the atmosphere,
a crystelline deposit of jansite
forms. Jansite is formed under
certain conditions from solutions
containing $K^+SO_4^{--}$ and Fe^{+++} ions
and is well known for its ability
to contain other trace metals in
its structure. In the absence of
the K^+ and SO_4^{--} ions, we would
have expected ferric hydroxide gels
to be formed. They, of course,
are also capable of adsorbing other
metal ions. We do not know to what
extent the precipitation of jansite
or ferric hydroxide occurs when
the anaerobic leachates from the
base of a landfill site meet the
more oxygenated aquifer. It is
possible that such precipitation

reactions may still further enhance
the removal of trace metals from
leachates within the rock structure.

THE WASTES RESEARCH UNIT

Finally, I would like to mention
the Department of the Environment
Wastes Research Unit, which is also
part of our Hazardous Materials
Group. Its function is to answer
the many technical enquiries from
the Department of the Environment.
Sometimes these enquiries involve
a paper-work study of the extent
to which liquids are deposited with
solid refuse at different sites.
On the other hand, the volatility
of certain solvents when deposited
in a landfill or their adsorption
on the waste itself may require
detailed esperimental investigation.
This type of information is used
by the various study groups that
have been set up to draw legislation
on different aspects of the handling
and disposal of toxic chemicals.

Studies have also been made
on the relative rates at which
various toxic materials are leached
from actual wastes and on the effect
of encapsulation procedures on the
amounts that will be released to
a landfill under different
conditions.

With such a wide range of
experience available on hazardous
materials and in the landfill, we
have now turned full circle. We
have found that the radioactive
waste disposal programme has now
started to ask us to help them solve
some of their problems. The original
concept of the Hazardous Materials
Service was to do the reverse and
use nuclear experience in non-
radioactive waste problems. Is
that what you call recycling?

I hope you have found this
sketch of our work interesting and
that when we next meet I will be
able to make a more detailed
contribution to your program with
emphasis on the actual results we
have obtained. Thank you.

THEORETICAL APPROACH TO GAS MOVEMENT THROUGH SOILS

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INTRODUCTION

To predict gas concentrations around sanitary landfills, appropriate mathematical analogues must be developed and solved. To be of value to potential users, these analogues should:

• be based upon parameters which can be obtained by relatively routine field investigation and laboratory testing;

• allow decoupling of parameters used to describe the gas, the surrounding soil, and the regional geology;

• use analytical techniques that are economical and that are understood by the general scientific community;

• result in predictive techniques taht are amenable to graphical presentation for use by planners and designers.

This paper describes such an analogue and presents some typical results. These results are excerpted from a broader study currently being conducted at The Ohio State University under USEPA Contract No. 68-03-0326: Development of Predictive Models for Sanitary Landfill Gas Movement. Other Ohio State Civil Engineering faculty who are associated with the project and whose contributions to the present work are hereby acknowledged are Professors T. H. Wu, Robert M. Sykes, and Ranbir S. Sandhu.

THEORY OF GAS MOVEMENT IN POROUS MEDIA

Diffusional Flow in a Capillary Tube

As a gas flows in a capillary tube, its progress will be impeded by collisions with other gas molecules and with the capillary walls. Figure 1 delineates mechanisms involving only collisions with the walls (Knudsen diffusion), only collisions with other gas molecules (molecular diffusion), both wall and intermolecular collisions (transition region diffusion), hopping or sliding along the capillary wall (surface flow), and finally flow involving all mechanisms (general flow). The type of flow which is likely to predominate depends upon the relative magnitudes of the capillary radius, r, and the mean free path of the molecule.

Pressure Flow in a Capillary Tube

If a total pressure gradient exists in a capillary tube, transport will occur in the direction of decreasing pressure by Poiseuille flow and by slip flow along the capillary walls.
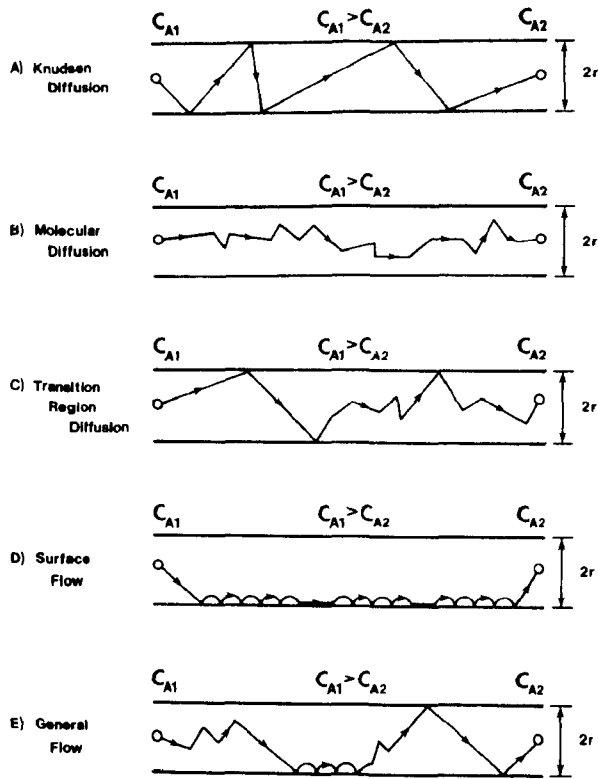
Figure 1. Flow mechanisms for a single gas in a capillary tube.

Figure 2. Flow of multicomponent gas through a capillary tube.

Combined Pressure and Diffusional Flow in a Tube

If both partial pressure and total pressure gradients are present, a combination of diffusional and pressure flows will occur. Figure 2 depicts this situation. In Figure 2A, no total pressure gradient exists but the partial pressure of gas A decreases to the right and gas B will flow to the left by diffusional flow. In Figure 2B, both a total pressure gradient and partial pressure gradients of gases A and B occur to the right. Gases A and B will flow to the right by pressure flow. In Figure 2C, a total pressure gradient and a partial pressure gradient of gas A exist to the right while a partial pressure gradient of gas B exists to the left. Gas A will clearly flow to the right by both diffusional and pressure flow. Gas B will tend to flow to the left by diffusional flow and to the right by pressure flow. The actual flow direction cannot be determined a priori.

Generalization to Porous Media

In porous media (see Figure 3), the flow is reduced due to the inability of gas to flow through the portion occupied by the solids. This reduction is reflected by the effective porosity

$$n' = \frac{V_g}{V_t} \qquad (1)$$

where V_g = gas volume and V_t = total volume.

In a dry medium, the effective porosity is equal to the conventional porosity, while in a partially saturated medium, the effective porosity is related to the conventional porosity by

$$n' = \frac{n}{100} \left(1 - \frac{S}{100}\right) \qquad (2)$$
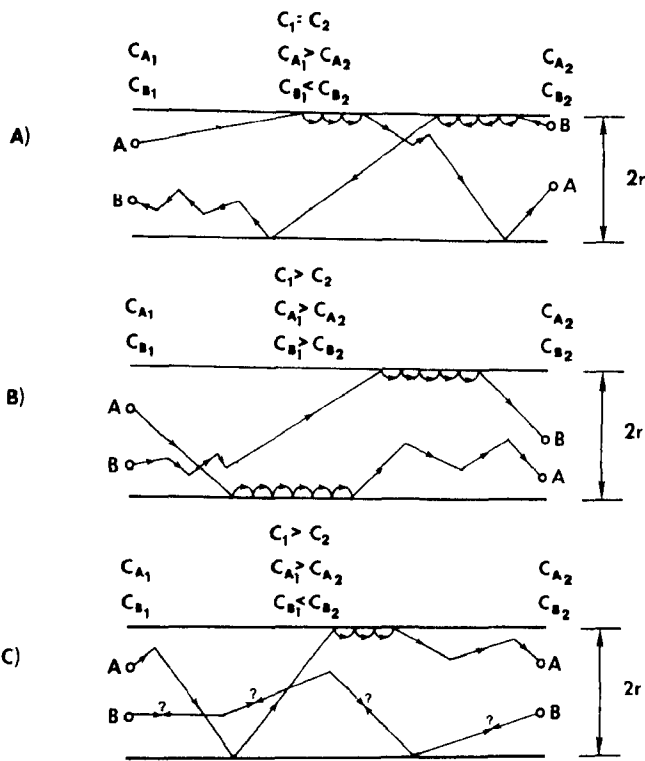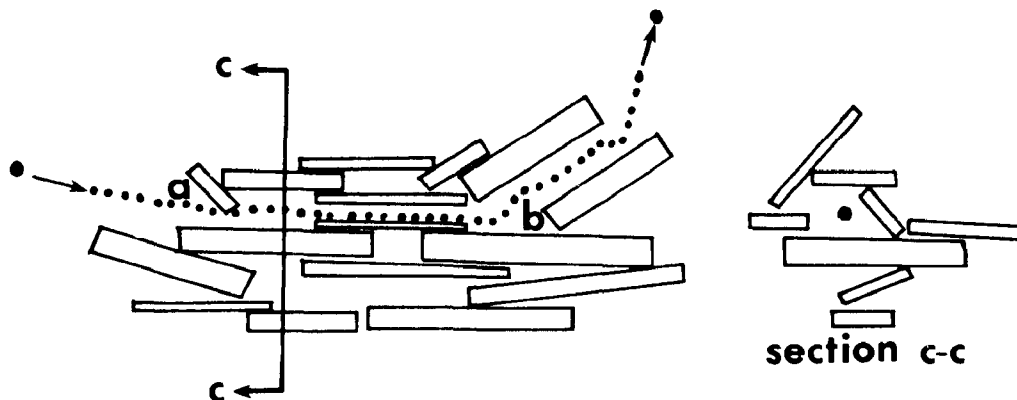
where S = degree of saturation.

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section c-c

Figure 3. Gas flow path through a porous medium.

A second characteristic of porous media which reduces flow rates is the necessity for gas molecules to circumvent the solid particles. This is reflected in the tortuosity, τ, of the porous medium.

Because the predominating flow mechanism is related to the radius of a capillary tube, special consideration must be taken of the wide range of pore sizes encountered when approximating the pores of a soil as a bundle of parallel capillary tubes. This pore size distribution must be incorporated in the differential equations describing the flow process.

Flow with Chemical Reaction

Gases produced by sanitary landfill decomposition processes consist principally of methane and carbon dioxide. While methane is relatively unreactive, carbon dioxide dissolves in groundwater through the following reactions:
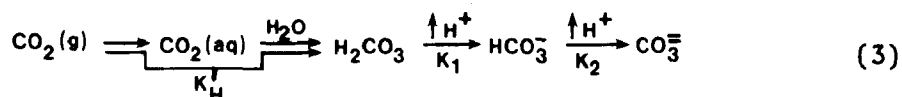
Normally, CO_2 (aq) cannot be distinguished analytically from H_2CO_3 so that a single equilibrium constant is used for this step of the reaction. Simultaneous algebraic equations must be solved to calculate the redistribution of the several constituents at each time step.

Combined Flow with Reaction in a Porous Medium

Moore and Alzaydi (1975) have developed equations to described flow of gases around sanitary landfills. A flux equation must be written for each component:

where N^o_A = total (pressure plus diffusional) flux of gas A relative to fixed coordinates

τ = tortuosity factor

r = pore radius

$$CO_2(g) \rightleftharpoons CO_2(aq) \underset{K^T_H}{\overset{H_2O}{\rightleftharpoons}} H_2CO_3 \overset{\uparrow H^+}{\underset{K_1}{\longrightarrow}} HCO_3^- \overset{\uparrow H^+}{\underset{K_2}{\longrightarrow}} CO_3^= \qquad (3)$$

$$-\overset{o}{N}_A = \frac{1}{\tau} \sum_{r_{min}}^{r_{max}} f_v(r) \left| \frac{\dfrac{\bar{D}_{AB}}{RT} \cdot \dfrac{\partial C_A}{\partial Z}}{C - \left(1 - \sqrt{\dfrac{M_A}{M_B}}\right)C_A + \dfrac{\bar{D}_{AB}}{RTK_A^T r}} + \frac{C_A}{4}\left[\frac{RTr^2}{2\mu} + \frac{\eta K_A \cdot r}{C_A + C_B \sqrt{\dfrac{M_B}{M_A}}}\right] \frac{\partial C}{\partial Z} \right| \qquad (4)$$

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$r_{min} \; r_{max}$ = limits on pore size distribution approximation

$f_v(r)$ = volume fraction having pore radius, r

$\overline{D}_{AB}$ = (constant) diffusion coefficient for gases A and B

R = gas constant

T = temperature

C_i = concentration

Z = spatial distance

C = total gas concentration

M_i = molecular weight of gas i

K_Λ = (constant) Knudsen diffusion coefficient for gas A

μ = gas viscosity

In addition, conservation of matter must be reflected in continuity equations:

$$\frac{\partial c_A}{\partial t} = -\frac{\partial N_A}{\partial z} + S_A \qquad (5)$$

where S_A = amount of component A created (or destroyed).

Note that in these equations the properties of the gases and of the porous medium are decoupled:

gas properties: $\overline{D}_{AB}$, M_A, M_B, K_Λ, μ.

porous medium properties: r, r_{min}, r_{max}, $f_v(r)$,   .

APPLICATIONS TO SANITARY LANDFILLS

Geometrics of Landfill and Surrounding Soil

Simplified geometric conditions (see Figure 4) are assumed in applying the theory to sanitary landfills. The landfill itself is assumed cylindrical in shape and of radius, r_f, and depth, d_f. The soil surrounding the landfill is assumed to have a horizontal surface varying in perviousness from completely pervious to completely impervious. At some depth, d_s, an impervious stratum (bedrock) is assumed to be encountered. At some radial distance, r_s, another soil stratum of varying perviousness is assumed to be encountered. (Note: the mathematical model does not extend beyond r_s.)

At a radial distance, r_p, vents in which the methane concentration is maintained at zero are spaced equally around a circumference and penetrate to depth, d_p. The circumferential spacing of the vents
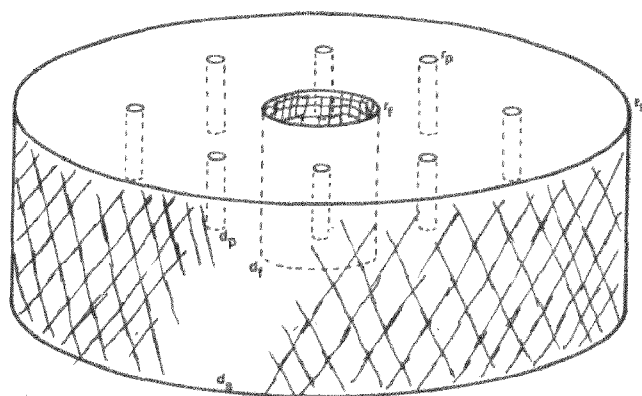

Figure 4. Simplified landfill geometry.

may be any integral fraction of 2π radians. Vents spaced infinitely closely constitute the limiting case of trenches.

A solution has been provided (Moore and Rai, 1975) for these cases and for zero total pressure gradient. This situation was chosen since no field evidence points to the existence of pressure gradients around currently instrumented landfills.

Design Charts without Vents

A set of design charts has been prepared for the geometry shown in Figure 5. Because equations 4 and 5 are nonlinear, complete normalization could not be achieved. Instead solutions were obtained for average landfill dimensions (d_f = 16 m, r_f = 160 m); for average gas compositions and decomposition times (70% methane, 30% carbon dioxide; decomposition time = 5 years); and for typical soil properties (n = .4, r = 4×10^5 Å for granular soil, γ = 2.25). A pressure of 1 atmosphere and a temperature of 25°C were used.

The philosophy used in constructing the design charts was to assume typical properties and develop correction charts for extending the results to atypical situations.

Example Design Charts

Figures 6 through 8 show typical charts for granular soil with different combinations of pervious and impervious surface and radial boundaries. On each chart a spectrum of ratios d_s/d_f and r_s/r_f is shown. Values of r_t/r_f can be obtained where r_t is the farthest radial extent of the 5% methane level observed above the base of the landfill.

Figure 6. Design chart for methane in granular soil (pervious ground surface and radial boundary).

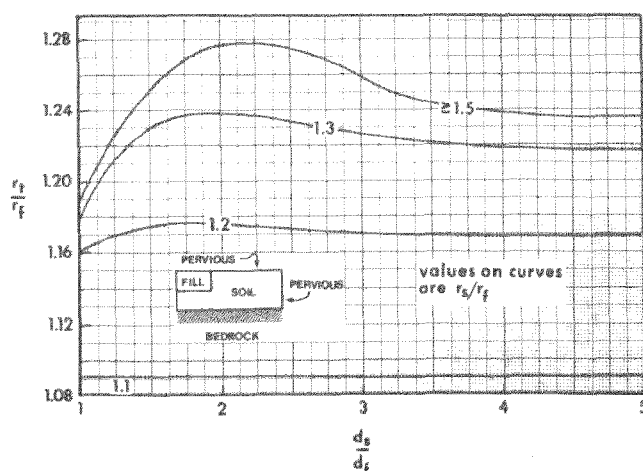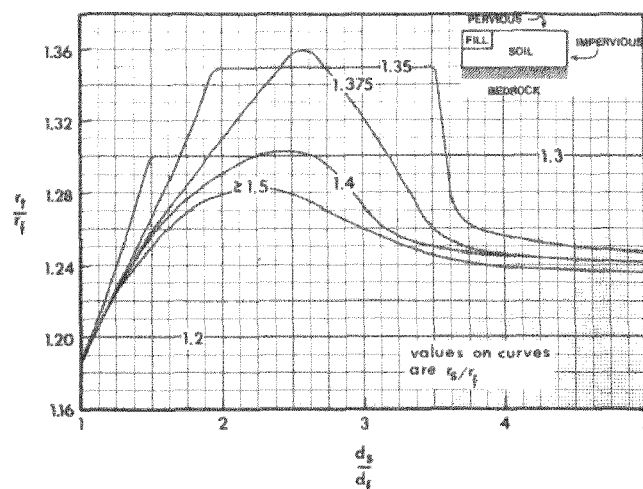Figure 7. Design chart for methane in granular soil (pervious) ground surface and impervious radial boundary).

Figure 5. Landfill configuration for design charts.

Figure 8. Design chart for methane in granular soil (impervious ground surface and pervious radial boundary).

Note that for a pervious ground surface and a pervious radial boundary, the maximum extent of a 5% methane level is only 1.28 times the landfill radius. For a pervious top and an impervious radial boundary, the maximum r_t/r_f is 1.36. Finally, for an impervious top and pervious radial boundary, the maximum extent r_t/r_f is 2.23.

For design considerations, the time required for permeation of gas is important. Figures 9 through 11 show time overlays for Figures 6 through 8, respectively. The time overlays give the time at which the maximum excursion of the 5% level of methane was observed.

Note that for the pervious ground surface and radial boundaries, the maximum 5% excursion occured on the order of 1 year after decomposition terminated. Conversely, for the impervious ground surface, the times for maximum excursion are in excess of 150 years.

To provide information for shorter time periods, a set of curves is presented in Figure 12 for $r_s/r_f \geq 5.0$ and for values of r_t/r_f for the maximum excursion of the 5% methane level after 50 years, 100 years, and full duration.
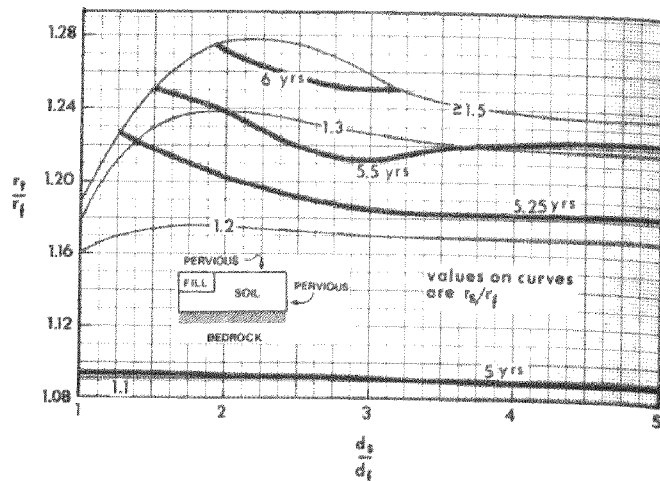
Figure 9. Time overlay for design chart for methane in granular soil (pervious ground surface and radial boundary).
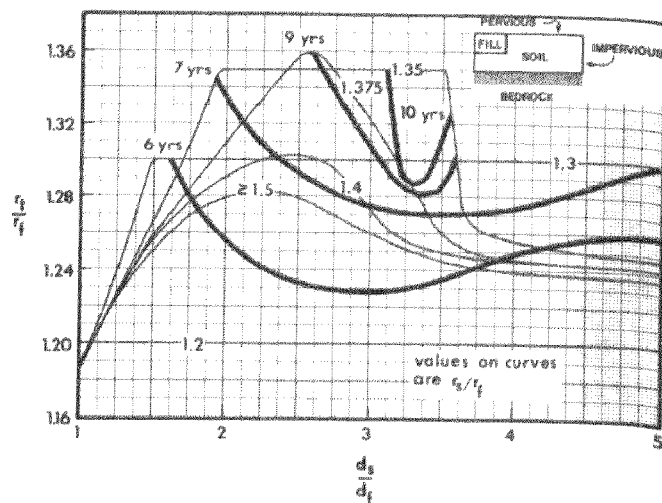
Figure 10. Time overlay for design chart for methane in granular soil (pervious ground surface and impervious radial boundary).
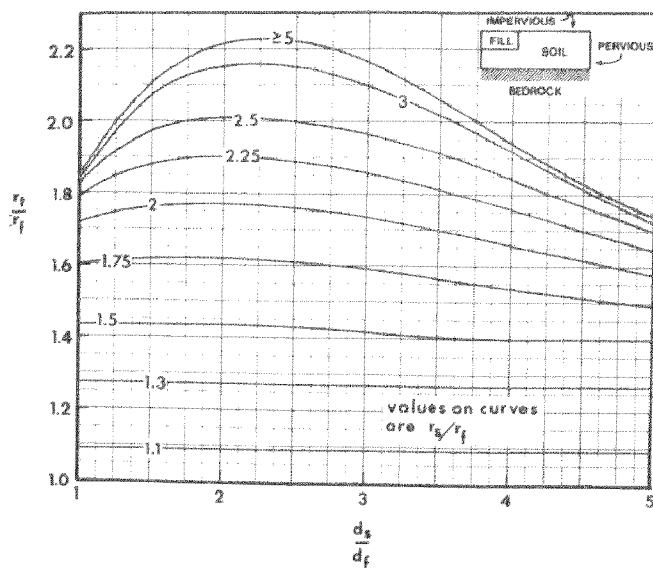
38
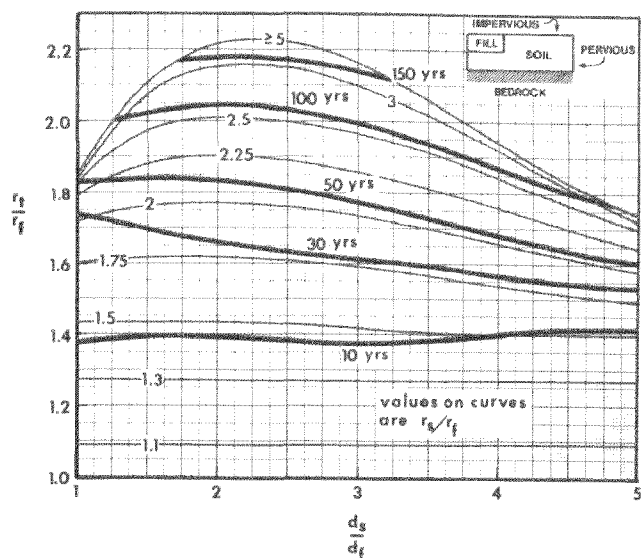
Figure 11. Time overlay for design chart for methane in granular soil (impervious ground surface and pervious radial boundary).
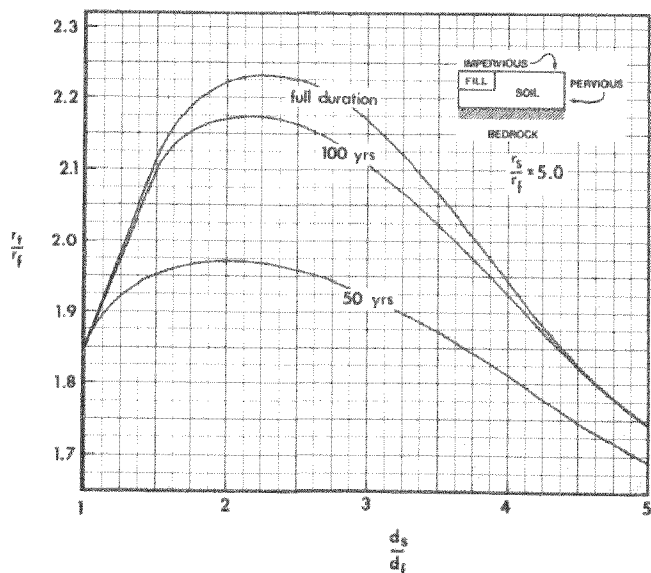
Figure 12. Excursion of 5% methane level in granular soil after different lengths of time.

Effects of Varying Assumed Parameters

A study was made of the effects of varying the typical values assumed for decomposition time, soil porosity, and temperature.

The effects of varying decomposition time are shown in Figure 13. For short decomposition times, corrections of as much as -12% were required. However, decomposition times of up to 15 years required corrections of only +7%.

Figure 14 shows the effects of varying porosity. Here correction factors are quite small, with factors of -10% being required for porosities of 0.1.
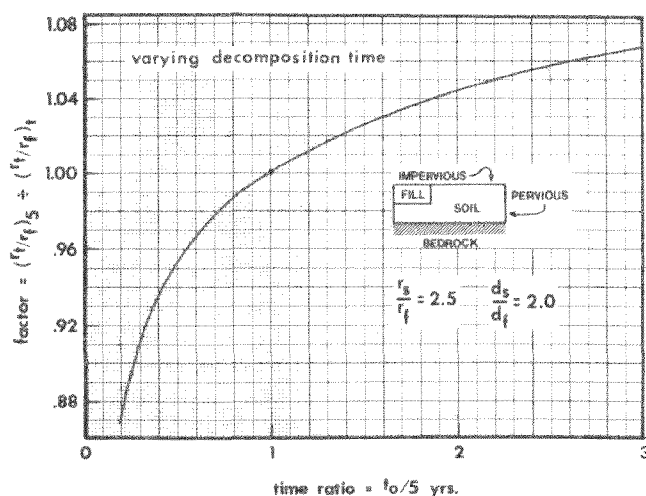
Figure 13. Effect of varying decomposition time on maximum excursion of 5% methane level in granular soil.
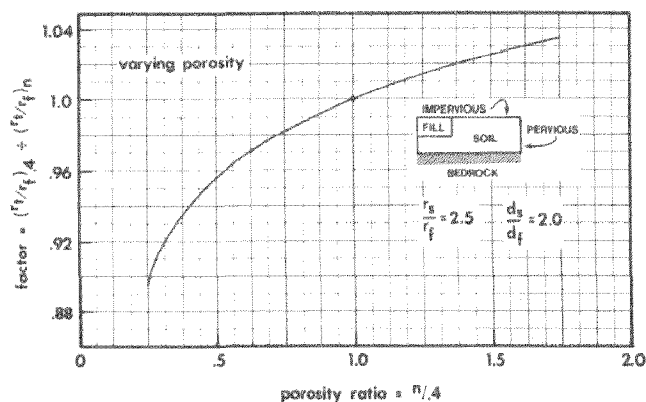
Figure 14. Effect of varying soil porosity on maximum excursion of 5% methane level in granular soil.

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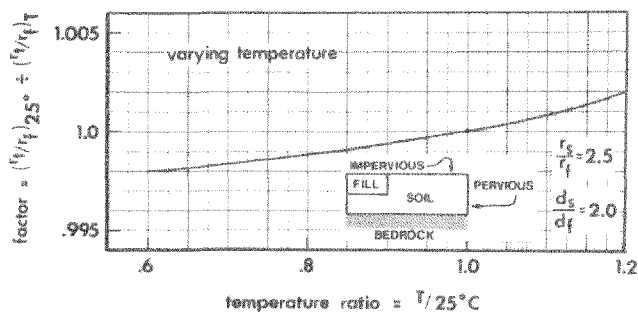
Figure 15. Effect of varying tempera-
ture on maximum diffusional
excursion of 5% methane
level in granular soil.

Finally, the effect of tem-
perature variation is shown in
Figure 15. The effect is negligibly
small.

Effects of Varying Perviousness of Ground Surface

Factors such as rainfall
infiltration, frost, vegetation,
paving, etc., can result in the
ground surface having perviousness
varying between completely pervious
and completely impervious. The
effect of relative perviousness
of the ground surface boundary is
shown in Figure 16. The variation
can be seen to be nonlinear with
a high degree of imperviousness
being required to approach the
totally impervious permeation limits.

Effectiveness of Vents

To study the effectiveness
of venting systems, several
configurations were studied for
an impervious ground surface and
a pervious radial boundary with
$r_s/r_f = 2.5$, $d_s/d_f = 2.0$, and t
= 5 years. In these studies, the
effectiveness of the vent was
determined by taking the ratio of
the methane concentrations at a
given time and place with and without
the vent.

Figure 17 shows the variation
of effectiveness of the vent as
a function of the radial distance
away from the vent. High reductions
are observed in the radial vertical
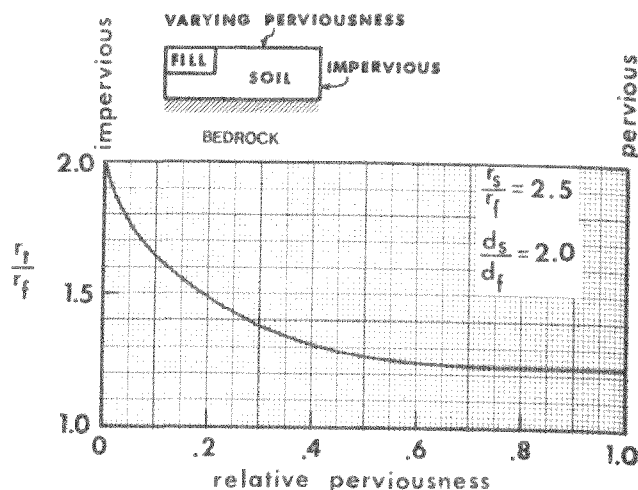
Figure 16. Effect of varying ground
surface perviousness on
maximum excursion of 5%
methane level in granu-
lar soil.
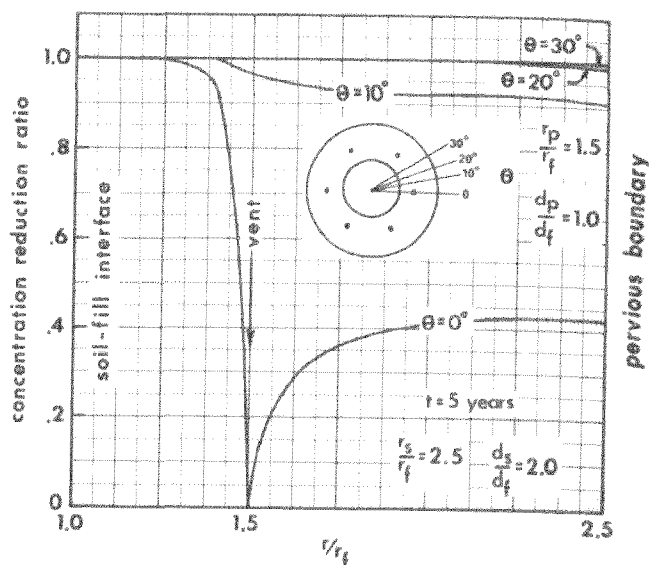
Figure 17. Circumferential variation
of effectiveness of vents.

plane of the vent; however, effec-
tiveness reduces rapidly along
radial vertical planes away from
the vent. Figure 18 shows the
effect of varying depth of
penetration of the vent. It may
be seen that beyond the vent, added
penetration is effective along the
radial plane extending through the
vent.

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Finally, Figure 19 shows the effect of varying the radial distance to the vent.
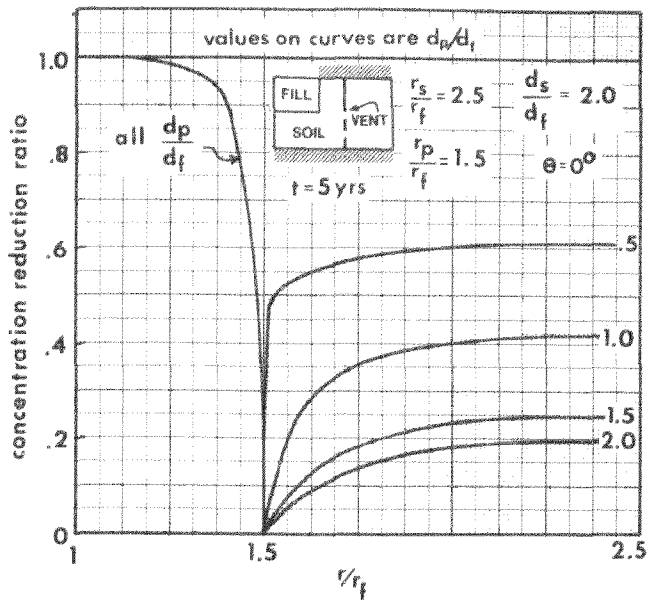
Figure 18. Effect of depth of penetration of vent on effectiveness in reducing methane concentrations.
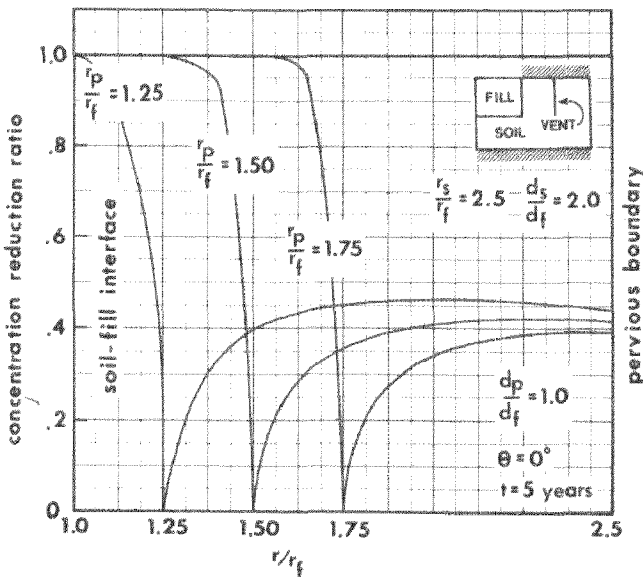
Figure 19. Effect of radial placement on effectiveness of vents in reducing methane concentrations.

It may be seen that the effectiveness of the vent is increased by placing the vent closer to the landfill-soil interface.

Effectiveness of Trenching

To study the effectiveness of trenching around landfills on reduction of methane concentration, the configuration chosen was the same as that previously used to study the effectiveness of vents (r_s/r_f = 2.5, d_s/d_f = 2.0, r_p/r_f = 1.5, impervious ground surface, pervious radial boundary). Figure 20 shows the effect of depth of penetration on reduction of methane concentration at the elevation of the bottom of the landfill. It may be seen that reductions of from 50% to over 90% are obtainable for penetration depths varying from 0.5 d to 1.5 d.

Effects of Efficiency of Vents or Trenches

For the studies reported in the previous two sections, it was assumed that the concentration of methane in the vent or trench was maintained at zero. In practice, gases collected at the vent or trench may not be completely released to the atmosphere.
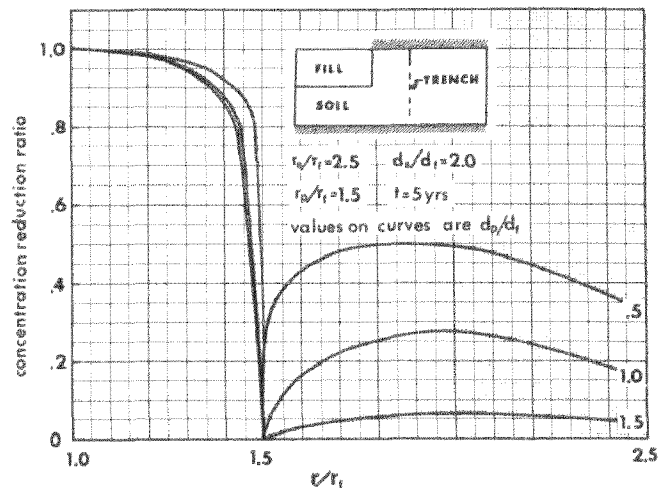
Figure 20. Effect of depth of penetration of trenches on effectiveness in reducing methane concentrations.

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Figure 21 shows the concentration reduction ratio at $r/r_f = 2.0$ as a function of trench efficiency. A nearly linear effect of trench efficiency is noted. This same effect is noted in the relationship between r_t/r_f and trench efficiency as shown in Figure 22. This figure shows that as long as the trench remains 20% efficient, the 5% methane level at the elevation of the bottom of the landfill remains on the landfill side of the trench. At lower efficiency levels, the 5% methane level extends beyond the trench.
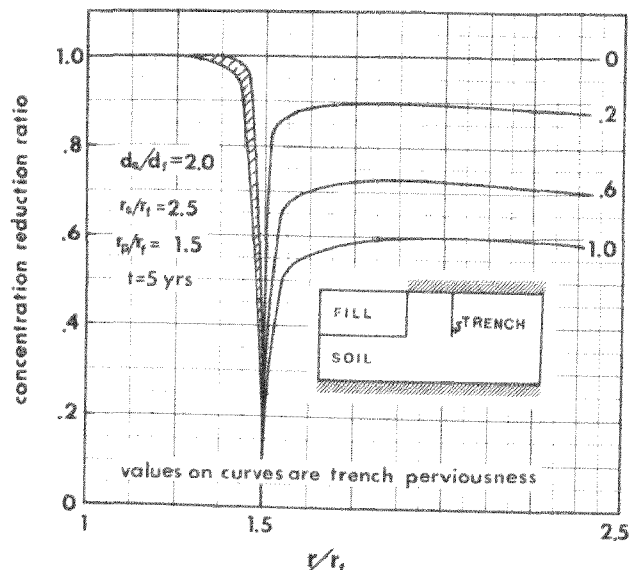
Figure 21. Effect of efficiency of trenches on effectiveness in reducing methane concentrations.
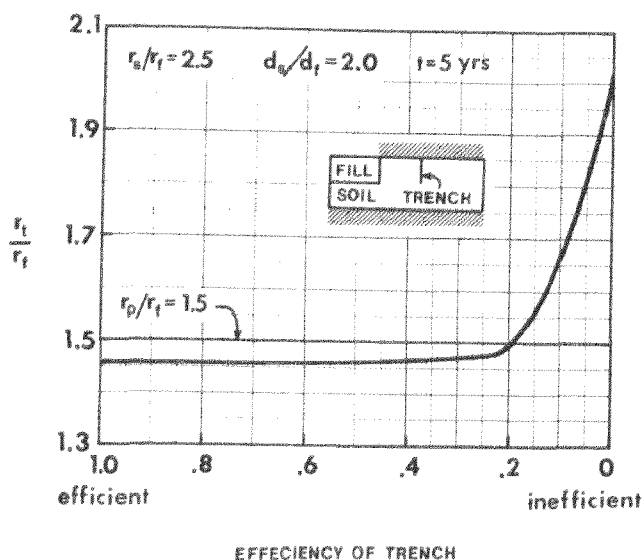
Figure 22. Effect of efficiency of trenches on maximum excursion of 5% methane level.

SUMMARY

The work reported in this paper provides a physical basis and a mathematical tool for predicting the flow of gases around sanitary landfills. In addition, quantitative relationships have been presented for a number of typical configurations. The effects of variations in assumed parameters for landfill, soil and gas properties have been examined. Finally, limited data have been presented to aid in design of venting and trenching systems and to allow the evaluation of the effects of the inevitable imperfections in constructed vents and trenches.

CONCLUSIONS

The reader is cautioned against extrapolating the results presented in this paper to other field situations. However, based on this work and the assumptions employed, certain broad conclusions may be drawn:

1. It is possible to develop a mathematical analogue to describe the flow of gases around sanitary landfills which decouples the physical characteristics of the gases, the porous medium, and the areal geology.

2. Interaction between the gas and the liquid or solid phase can be incorporated in the analogue through equilibrium equations.

3. A solution to the analogue, assuming a 70% methane level in the landfill for 5 years and no excess total pressure, yields the following results:

   a. Depending upon the perviousness of the ground surface, the maximum radial extent of the 5% methane level above the elevation of the base of the landfill extends for 1.28 to 2.25 times the radius of the landfill, and decay to below 5% methane requires from 1 to in excess of 150 years.

   b. Decomposition times in excess of 5 years result in further excursion of the 5% methane level.

   c. As the porosity of the soil increases, the 5% methane excursion distance increases.

   d. Temperature does not appreciably affect diffusional transport.

   e. A small degree of perviousness of the ground surface is very effective in reducing the 5% methane excursion.

   f. Vents are effective in reducing methane concentrations along a radial line passing through the vent but are essentially useless in reducing concentrations along radial lines a few degrees of arc away.

   g. Continuous circumferential trenching can reduce methane concentrations significantly. Deeper penetration increases effectiveness.

   h. Significant methane reduction can be obtained even if trenches are not perfect (i.e., the methane concentration is not kept at zero).

REFERENCES

1. Moore, C. A., and A. Alzaydi (1975). Theoretical Considerations of Movements of Gases Around Sanitary Landfills, Report to U.S. EPA, in press.
2. Moore, C. A., and Iqbal Rai (1975). Three Dimensional Solutions to Gas Flow Around Sanitary Landfills, Report to U.S. EPA, in press.

ANALYTICAL METHODOLOGIES FOR LEACHATE AND GAS ANALYSIS

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INTRODUCTION

The oldest method of solid waste disposal, that of landfilling, is still the most widely used. This method can have an adverse effect on the environment unless sound engineering principles are used during design, operation, and long term maintenance of the solid waste fill. When infiltration of rainwater is not restricted, the water will dissolve organic and inorganic substances from the solid waste. The leachate thus generated may move out of the fill into the surrounding soil and subsequently pollute groundwaters or nearby aquifers. In several instances, regulatory agencies (State of Illinois, 1973) have, therefore, required monitoring of the environment to determine the impact of landfilling. To minimize such impact, clay layers, asphalt, plastic, or rubber liners are placed at the bottom of the fill before the disposal of the solid waste (Anon, 1972) to prevent leachate migration. Leachate treatment facilities are then a necessity and have, therefore, been installed at several landfills (Cressman, 1973; Schoenberger et al., 1971).

The environmental impact of leachate (dependent on leachate strength, attenuation in surrounding soils, biodegradation and efficiency of leachate treatment) requires the accurate and consistent determination of several water contaminants. It was the purpose of the present study (Chian and DeWalle, 1975a and 1975b) to review the analytical methods to determine contaminants as reported in the literature. The methods compiled and evaluated in this study were generally reported in the literature; additional information was obtained by contacting the principal investigators. Interferences in the chemical analysis due to the complex nature of the leachate as enumerated in the reported studies are listed in this report.

The compilation showed that different methods subject to different interferences are used to determine a certain parameter. For each parameter, only that method that was found to have the smallest interference was evaluated in this laboratory. The laboratory evaluation tested the method for its susceptibility to certain interferences commonly found in leachate. In addition, the accuracy of the method was tested. All laboratory analyses were performed using a high-strength leachate sample obtained from a recently installed lysimeter filled with milled refuse. Recommendations made in this report, therefore, only apply to leachate of similar strength. No evaluation was made

of the precision and sensitivity
of each method since this was beyond
the scope of the work. Realizing
the above restrictions, recom-
mendations were made in the present
study for the selection of those
methods least subject to
interference. Further recommen-
dations were made concerning
modifications of the selected methods
(Chian and DeWalle, 1975b).

SAMPLING PROCEDURES

Sample Collection

Principle. Leachate can be
collected from subsurface soil
strata by using wells or piezometers
placed in drilled holes. A
piezometer is a screen or permeable
plastic tip fastened to the end
of a pipe or tube, installed in
a boring. The annulus above it
is sealed so that the water-level
measurements or water samples
obtained from this installation
apply only to a restricted area
in the bottom part of the boring
below the seal in the annulus.
A well point is similar to a
piezometer except that there is
generally no seal in the annulus,
and therefore, measurements or water
samples obtained from a well point
may reflect conditions over a large
vertical interval (Hughes et al.,
1971). In some instances, the
annulus of a well point is also
sealed but the well point still
reflects conditions over a larger
vertical interval than those obtained
with piezometers. Pore water samples
above the groundwater table are
collected with suction lysimeters
(Apgar and Langmuir, 1971). As
leachate permeates through the soil
in relatively thin strata of higher
permeability, the collection device
should be placed at such a depth
that it includes such permeable
strata.

Leachate collected above ground
may appear in springs or collection
trenches at the toe of a solid waste
disposal site. Such samples may
contain eroded soil and will have
reacted with the soil to
significantly affect its quality.
On such samples, a suspended solids
determination should be performed.
The soil should be removed by
sedimentation and not by filtration
as the latter method may remove
significant quantities of heavy
metals and phosphates. Filtration
(0.45 µ) should only be used when
the suspended solids interfere
significantly with the chemical
analysis.

Leachate may also reach the
surface and enter the surface waters
directly through groundwater
discharge zones. In such instances,
an estimate will have to be made
of the approximate extent of the
dilution.

Interference. Characteristics
of leachate can be affected by the
methods and materials used. Apgar
and Langmuir (1971), for example,
obtained their sample from suction
lysimeters with an effective pore
diameter of 1 µ. Hughes et al.
(1971) used predominantly No. 10
brass well screens with an opening
larger than 5 mm. Fungaroli (1971)
did not use drain pipes but collected
leachate through sand and glass
beads of increasing size. Most
studies, however, do not mention
the type of sampling device through
which the leachate was collected.
One study, for example, only
mentioned the soil material used
to back fill the leachate collection
trenches in which perforated pipe
was embedded, but did not specify
the openings in the leachate
collection pipes.

The effective pore diameter
of the material through which the
leachate is collected may have an
appreciable effect on the
concentration of several parameters
such as suspended solids, phosphates,
and heavy metals since precipitates
or coatings may be formed near the
collection device which may filter
out these materials.

The construction of the
collection device should be such

that the device maintains anaerobic conditions and entry of air is minimized, as this will enhance aerobic degradation of the sample. The material used for the construction of the collection device may also have some effect on the leachate characteristics and should, therefore, be specified.

Recommendation. It is, therefore, recommended that leachate sampling conditions be specified when results of chemical analysis are presented. Such specifications should include a description of the soils, the construction, depth and characteristics of the sampling device, the effectiveness of the device to maintain anaerobic conditions, and the approximate detention time of the leachate in the collection device. When the leachate is collected from the surface, the soil conditions should be specified as well as the time that the leachate is exposed to aerobic conditions. Other characteristics that will help to explain the results of the chemical analyses should also be reported.

Sample Preservation

Principle. Leachate collected from a recently installed solid waste fill will have a translucent light brown color, but it will turn dark green or black and become turbid immediately after collection due to aerobic exposure and the subsequent oxidation of heavy metals and organics. The odor is very disagreeable and nauseating and is generally due to the presence of free volatile fatty acids such as butyric and valeric acid. Leachate samples collected from an older solid waste site are generally light brown to light yellow and do not change color directly after sampling since they are more stabilized and have lower metal concentrations. The odor is not offensive since free volatile fatty acids are generally absent due to active methane fermentation. Collection of leachate samples below

the surface at older sites will result in the liberation of carbon dioxide gas bubbles when the sample is exposed to atmospheric pressures, which will cause a reduction in bicarbonate alkalinity.

Cook (1966) showed that storage of leachate, with a COD of 1200 mg/liter and a pH of approximately 7.5, in a quart jar capped with aluminum foil and maintained at room temperature caused a 55 percent COD reduction after 3 weeks, with most of the decrease occurring after 1 week. A leachate sample in extensive contact with the atmosphere before sampling showed a 61% reduction due to the presence of more adapted aerobic microbial population.

None of the other studies reported in the literature quantitatively measured the changes of specific contaminants with time of storage, although some studies reported visual changes in the sample. None of the studies evaluated the effectiveness of different preservation techniques.

Evaluation of sample changes. Several characteristics listed in Standard Methods (APHA, 1971), which are used to characterize the nature of leachate, are subject to changes immediately after sampling. To illustrate the necessity of establishing strict sampling procedures, these parameters have been studied simultaneously as a function of time immediately after the sampling of leachate from a lysimeter located at the University of Illinois. The leachate was collected in a 4-liter bottle filled to the top. Small samples were withdrawn from the bottle at regular intervals for analysis, while the capped sample bottle was stored in the coldroom at 4 C. The parameters monitored during this period included COD, turbidity, color (absorbance at 400 nm of the 1:10 diluted sample), pH, suspended solids (SS), oxidation reduction potential (ORP), and conductivity (Figures 1, 2). The most pronounced

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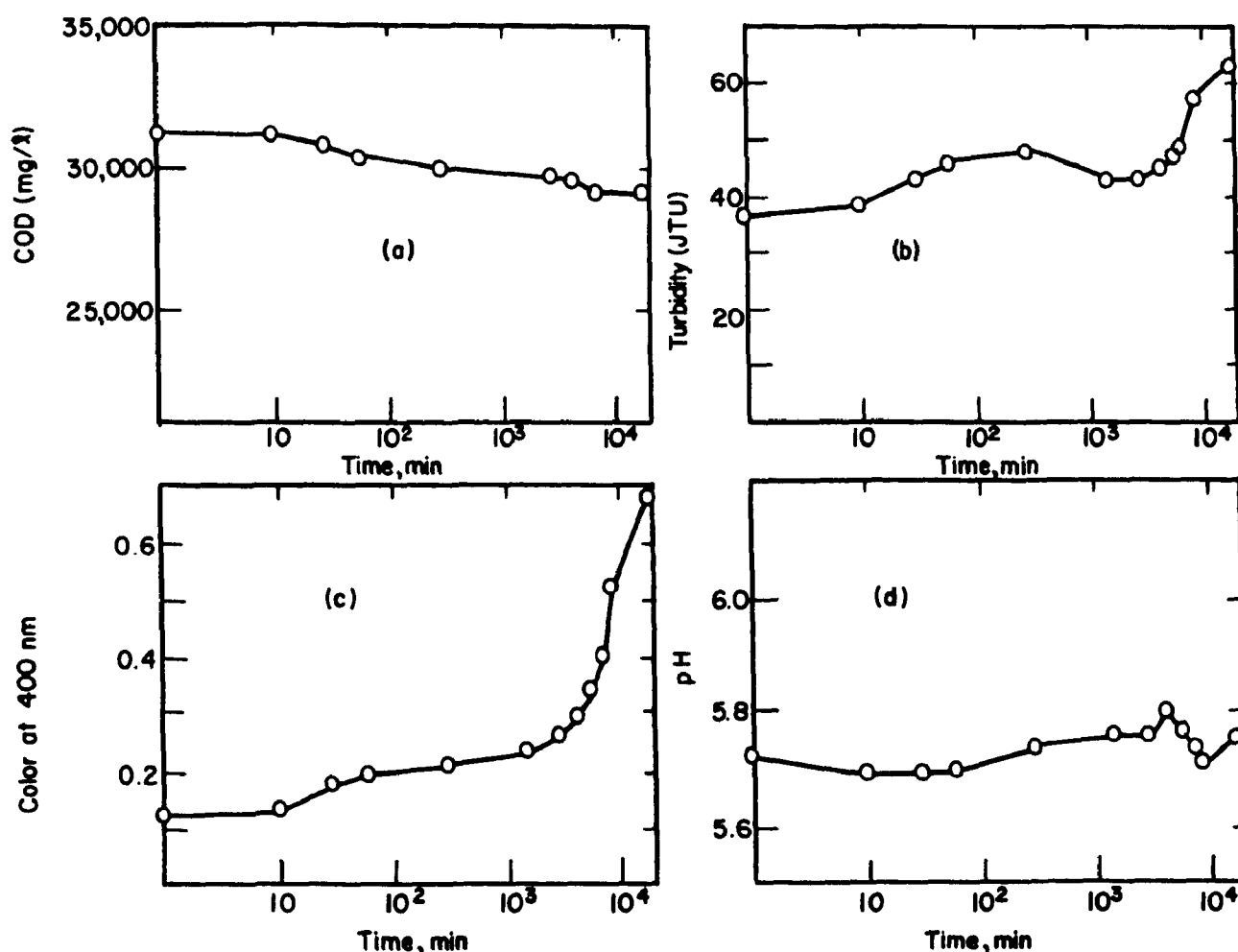
Figure 1. Change of COD, turbidity, color, and pH with time of storage at 4° C.

changes took place with the SS, ORP, turbidity, and color of the sample; the latter corresponded to a visual change of the sample color from light yellow to dark brown. This is caused by the oxidation of the ferrous iron to ferric hydroxide. The formation of ferric hydroxide contributes to the increase of color, turbidity, and SS. The increasing ferric hydroxide content of the SS was reflected by the percent of fixed SS which increased from an initial 18.2% to 50.3% at the end of the monitoring period. Since some of the apparent volatile SS is contributed by the loss of bound water in ferric hydroxide at 550 C, the percent of inorganics in SS at the end of the monitoring period may be higher than 50.3%.

The oxidation of the organic matter and the iron caused a decrease of the COD by 6.8% and the formation of a precipitate. It also caused the conductivity to decrease. Figures 1 and 2 depict results of this study within a 12-day period. Eighty minutes after the first sample was withdrawn from the lysimeter, a second sample was collected. Comparison of the data showed that the second sample had a higher initial COD of 31,600 versus 31,000 mg/liter, whereas the turbidity, pH, and color were slightly higher in the first sample. The ORP was more negative with the first sample than in the second. The initial SS were approximately equal in both samples. The final COD in the two samples after 13 days of storage was approximately

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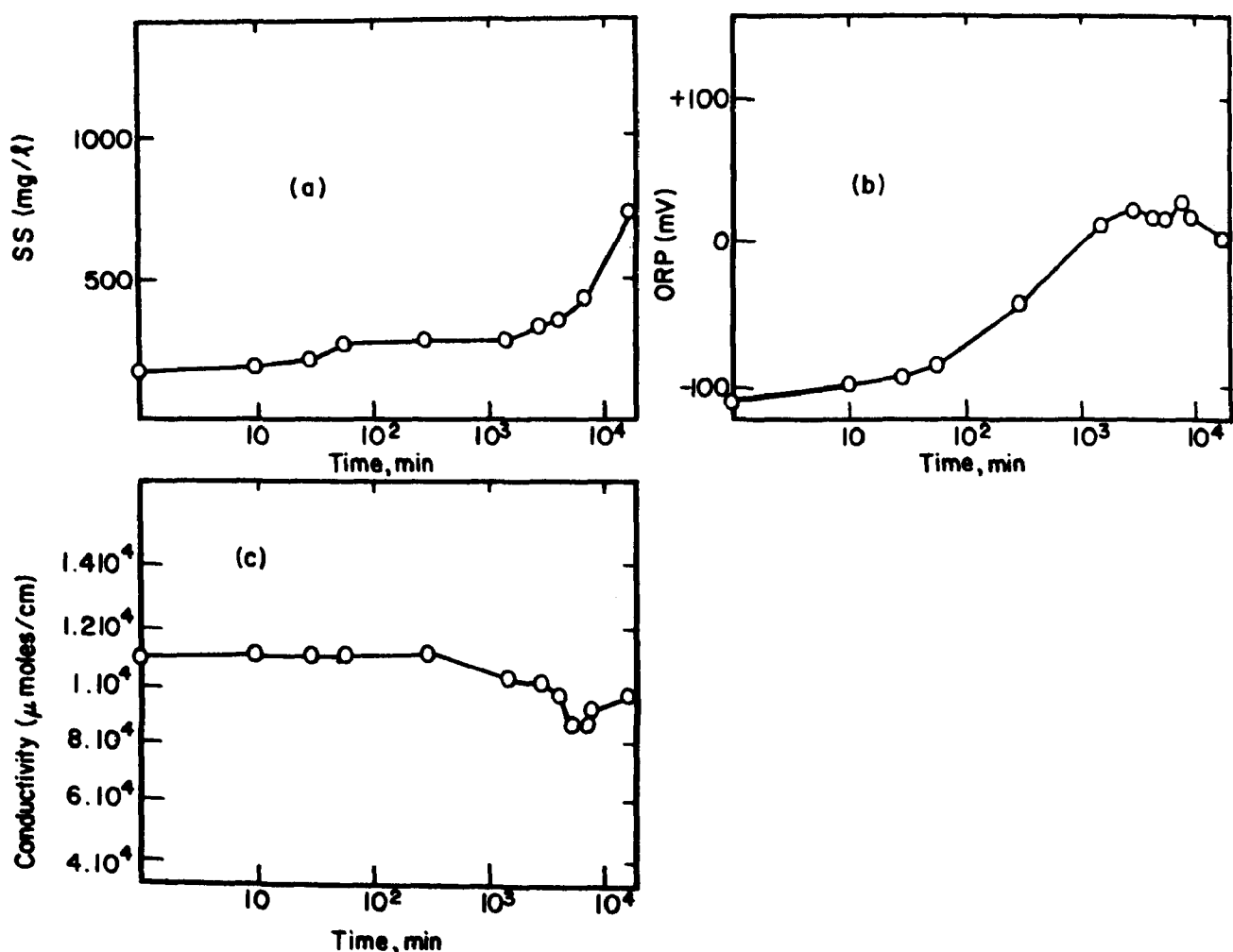
Figure 2. Change of SS, ORP, and conductivity with time of storage at 4°C.

equal, i.e., 29,000 mg/liter for the first sample and 29,071 mg/liter for the second sample. The final SS were higher for the second sample. Since a fraction of the COD decrease is contributed by the oxidation of the iron, it is expected that an initial higher COD would result in a higher SS and turbidity after prolonged storage. This was indeed observed. In conclusion, the same results of rapid increase of turbidity, SS, and color and stabilization of ORP were confirmed with the second sample.

Recommendations. Based on these observations, it is recommended that several parameters be determined directly after collection of the sample or, it this cannot be

accomplished, the leachate should be collected under anaerobic conditions in a tightly stoppered bottle. The sample should be stored in a glass bottle for organic analysis, as this keeps the sample more anaerobic. For heavy metal analysis, the sample is preferably stored in a polyethylene bottle, as it prevents adsorption of heavy metals onto the wall of the container. The sequence of parameter analyses should be ORP, color, turbidity, SS, pH, and conductivity. Other parameters such as COD and organic N may also change directly after sampling, but these changes may be reduced when the sample is acidified. Acidification and storage at 4 C will stop the methane fermentation, which process is

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responsible for free volatile fatty acid removal, while they also slow the bacterial acid fermentation of complex organic substrates. Acidification, however, enhances volatilization of undissociated fatty acids, precipitates humic-like organics, and facilitates hydrolysis of complex organics. Preservation with 40 mg/liter mercuric chloride is not recommended as it is probably not effective. The mercury will be precipitated under anaerobic conditions as mercury sulfide and lose its bactericidal properties. When results of organic analysis are presented, the time lag between sample collection and analysis should be stated.

SELECTION OF PARAMETERS TO BE MEASURED

The remainder of the final report (Chian and DeWalle, 1975a) contains analyses that can be performed to characterize the leachate sample. When a large number of samples have to be analyzed, it is not feasible to measure all parameters, and those parameters will be determined that are easy to measure. It is felt that most information is obtained by measuring conductivity (attributed by salts and volatile free fatty acids), color or absorbance at 400 nm (attributed by iron and organics), and pH (a low pH indicates presence of volatile free fatty acids). When more parameters are to be measured, they should include COD (reflects concentration of organics) and total solids (reflects presence of organics and inorganics). Only after the above five parameters are measured is determining other parameters (such as TOC, free volatile fatty acids, BOD, organic nitrogen, or specific anions and cations) warranted.

When an organic parameter such as TOC or organic nitrogen is measured, it is recommended that the inorganic equivalent, such as the bicarbonate concentration and the ammonia concentration, also

be included. The ratio of organic-C/(organic-C + inorganic-C) then reflects the degree of biological stabilization of the sample, since acid fermentation followed by methane fermentation converts the complex organics to free volatile fatty acids, which are then converted into methane and carbon dioxide. The latter dissolves for a significant fraction into the leachate and is reflected in the increased bicarbonate concentration. However, it should be realized that the titration method is not applicable for the bicarbonate determination in leachate since free volatile fatty acids are also included. The only accurate way is, therefore, to measure the inorganic carbon with the dual channel organic carbon analyzer using the inorganic channel.

The ratio of organic-N/(organic-N + ammonia-N) does not represent major analytical problems and both measurements can be made subsequently with the Kjeldahl apparatus. The ammonia is first distilled off, whereafter the organic nitrogen is digested, converted to ammonia, and subsequently distilled off.

METHODOLOGY OF METHOD EVALUATION

Since most of the leachate studies have been conducted by researchers in the sanitary or environmental engineering field, the methods that are used closely reflect those of Standard Methods (APHA, 1971). Studies between 1960 and 1965 used the 11th edition; between 1965 and 1971, the 12th edition; and after 1971, the 13th edition. Laboratories not employing complicated instruments sometimes use methods listed by Hach Chemical Company (Hach Chemical Company, 1973). Methods used by geologists are generally those reported in: Techniques of Water Resources Investigation of the U.S. Geological Survey (U.S. Geological Survey, 1970). Recent studies use the U.S. Environmental Protection Agency

procedures in Methods for Chemical
Analysis of Water and Wastes (U.S.
EPA, 1974) which also contain
optional procedures for automated
analysis. Most studies employing
automated chemical analysis, however,
use methods recommended by Technicon
Industrial Systems, Industrial
Methods (Technicon, 1973).

The different parameters that
have been determined in the studies
reported in the literature are
listed in the final report. It
contains a survey of the different
methods used to analyze a certain
parameter and the obtained
experiences. The method least
interfered with by the matrix of
the leachate sample was selected
and then evaluated in greater detail
in the present study. The method
was evaluated with the standard
addition method and by using
progressively increasing dilutions
(Chian and DeWalle, 1975a).

Standard Addition Method

The standard addition method
is widely used in chemical analysis
when interferences present in the
sample cannot be avoided. An
advantage of this method is that
it avoids the necessity of preparing
synthetic standards of a composition
similar to that of the sample (U.S.
Geological Survey, 1970). In this
method, equal volumes of sample
are added to a water blank and
standards containing increasing
but known amounts of the test
element. The blank and the standards
must have the same volume to result
in a similar dilution of the sample.
The diluted samples containing
increasing amounts of the test
element are then analyzed according
to the standard procedures. The
obtained values are then plotted
on the vertical axis of a graph
while the concentration of the known
standards are plotted on the
horizontal (Figure 3). When the
resulting line is extrapolated to
zero measured concentration, the
point of interception of the abscissa
is the concentration of the unknown
element. The abscissa on the left
of the ordinate is scaled the same
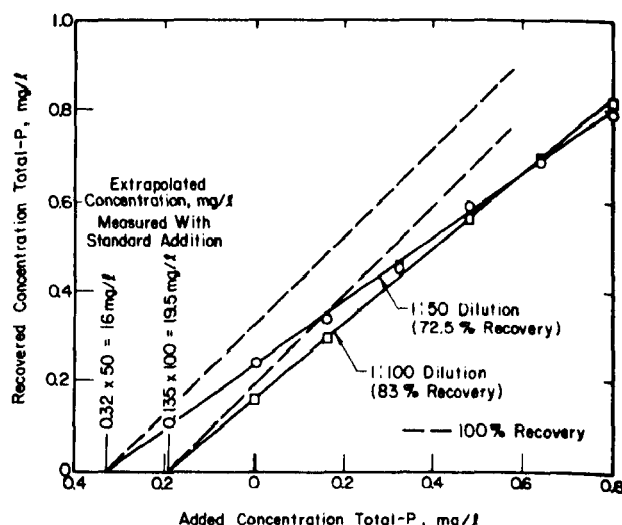
Figure 3. Standard addition method
for total-P determination
in 1:50 and 1:100 diluted
leachate sample.

as on the right side, but in the
opposite direction of the ordinate.
Since the scale of the ordinate
and abscissa are indentical, a line
drawn under 45° from the extrapolated
point on the abscissa to the ordinate
represents a 100% recovery of the
added element. Thus, 100% of the
known amount added to the diluted
sample is recovered. If the actual
line connecting the points has a
slope lower than 45°, the recovery
of the added element is less than
100% a slope higher than 45°
represents a higher than 100%
recovery. The percentage recovery
can be calculated from the tangent
of the line connecting the data
points. This is identical to taking
the intersect of the ordinate and
dividing it by the extrapolated
value on the left side of the
abscissa.

Dilution Method

In the dilution method, the
sample is diluted with increasing
amounts of distilled deionized water
and analyzed according to the
standard procedures. The obtained
concentration is then adjusted for
its dilution effect to give the
apparent initial concentration
before dilution (Figure 4).

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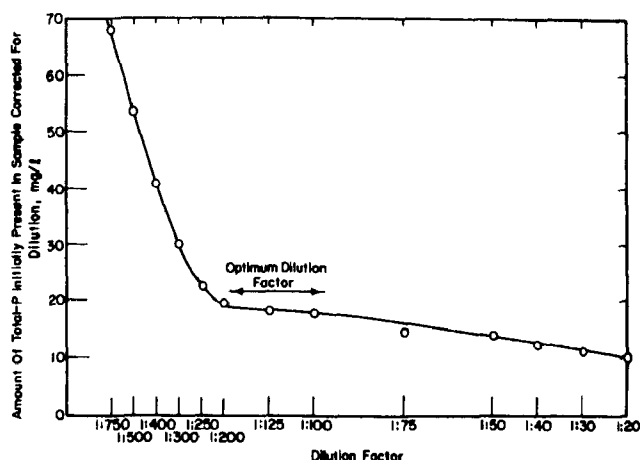
Figure 4. Effect of dilution on
calculated initial total-
P concentration.

The apparent concentration is then
plotted vertically and the dilution
factor, horizontally. The apparent
concentration generally increases
at increasing dilutions till it
reaches a plateau value. This
indicates that matrix interferences
of the sample are reduced with
increasing dilution. The dilutions
associated with this plateau value
are then used to determine the other
leachate samples in the batch.
Comparison of the dilution method
and the standard addition method
as evaluated for the total phosphate
determination showed that the latter
is generally more accurate. The
former, however, is easier to
administer and requires less time.

CONCLUSIONS

It was concluded that several
parameters of samples collected
from recently installed solid waste
fill undergo extensive changes
immediately after collection unless
strict anaerobic sampling and storage
conditions are maintained. Pre-
liminary laboratory evaluation of
physical, chemical, and biological
parameters showed that chemical
analysis using colorimetric methods
is strongly interfered by color,
suspended solids, and high salt

content present in leachate.
Interfering effects can be reduced
by using a standard addition method
in which increasing quantities of
the specific parameter are added
to the sample, after which its
recovery is determined. The obtained
percentage recovery is then used
to readjust the measured value.
A less accurate method is to dilute
the leachate sample with increasing
amounts of dilution water to
determine whether the interfering
effect can be sufficiently reduced
by progressive dilution. One of
the above approaches should be used
by the analyst prior to the analysis
of a series of leachate samples
for those parameters most subject
to interferences.

An extensive compilation of
the different analytical methods
used by researchers, consulting
firms, and regulatory agencies in
the United States showed that
numerous methods are used to
determine a specific parameter.
Based on research conducted at the
University of Illinois,
recommendations were made to use
those methods least subject to
interferences (Chian and DeWalle,
1975a).

RECOMMENDATIONS FOR RESEARCH

The present study conducted
a preliminary evaluation to determine
those methods to measure a certain
parameter which was least subject
to interference. Since only the
accuracy of each selected method
was evaluated, further research
will have to establish the precision
and sensitivity. Since all analyses
were conducted with a relatively
concentrated leachate sample,
additional leachate samples of
different strengths collected from
landfills of different ages will
have to be evaluated.

Methods that are less
complicated and time consuming,
but subject to larger interferences
than the recommended methods should
be evaluated, as they are likely

to be used under field conditions.
For example, although the Kjeldahl
distillation is recommended for
accurate ammonia determinations,
the more rapid Nessler method will
often be used under field conditions
and should, therefore, be evaluated.

All automated methods as recom-
mended by EPA (1974) for water and
wastewater and Technicon (1973)
for industrial waste should be
evaluated for possible interferences
since most tests are based on color-
imetric analysis and are generally
subject to strong interferences
by the color and SS present in
leachate. Such evaluation is
necessary since increasing numbers
of leachate samples will be analyzed
by automated methods at a future
date.

It is recommended that further
research should be conducted, in-
cluding a literature search, to
establish correlations between
specific constituents and general
parameters such as conductivity,
absorbance at 400 nm, and pH. These
three parameters are easy to
determine and are, therefore,
valuable for monitoring and
enforcement purposes, as they can
be used to screen large numbers
of samples. Only when these
parameters exceed a certain value,
to be determined by further research,
will further and more costly chemical
analysis be warranted.

It is finally recommended that
further research establish the exact
nature of the interfering substance.
When the interference is caused
by a common parameter that is
generally included in the measure-
ments, such as total solids,
chlorides, or sulfates, the knowledge
of the concentration of the
interfering parameter can be used
for the calculated concentration
of the interfered measurement.
This eliminates the need for the
time consuming standard addition
curves for the interfered
measurement.

REFERENCES

1. Anon, "Abandoned Limestone
   Quarry at Montgomery County
   Recreated Into Showplace
   Landfill Operation,"
   Constructioneer, Jan. 10 (1972).
2. Apgar, M. A., and D. Langmuir,
   "Groundwater Pollution Potential
   of a Landfill Above the Water
   Table," Groundwater 9, 6, 76-
   96 (1971).
3. APHA, "Standard Methods for
   the Examination of Water and
   Wastewater," American Public
   Health Association, Washington,
   D.C. (1971).
4. Chian, E. S. K., and F. B.
   DeWalle. "Compilation of
   Methodology Used for Measuring
   Pollution Parameters of Sanitary
   Landfill," Contract 68-03-2052,
   Solid and Hazardous Waste
   Research Laboratory, USEPA,
   Cincinnati, Ohio (1975a).
5. Chian, E. S. K. and F. B.
   DeWalle. "Treatment of Leachate
   from Landfills," First Annual
   Report, Contract 68-03-0162,
   Solid and Hazardous Waste
   Research Laboratory, USEPA,
   NERC, Cincinnati, Ohio (1975b).
6. Cook, H. A. "Biological and
   Chemical Investigation of
   Seepage from a Sanitary
   Landfill," M. Sc. Thesis,
   Department of Civil Engineering,
   West Virginia University,
   Morgantown, W. V., Ap. 71
   (1966).
7. Cressman, B. "Officials Tour
   Bucks Leachate Plant," The
   Morning Call, Allentown, Pa.,
   Friday, October 5 (1973).
8. Fungaroli, A. A. "Pollution
   of Subsurface Water by Sanitary
   Landfills," U.S. Environmental
   Protection Agency, Washington,
   D.C., Report SW-12g, pp. 186
   (1971).
9. Hach Chemical Company, "Water
   Analysis Handbook," Hach
   Chemical Co., Ames, Iowa (1973).
10. Hughes, G. M., et al.,
   "Hydrogeology of Solid Waste
   Disposal Sites in Northeastern

Illinois," U.S. Environmental Protection Agency, Washington, D.C., Report SW-12d, pp. 154 (1971).

11. Schoenberger, R. J. et al., "Treatment and Disposal of Sanitary Landfill Leachate," Proceed. 5th Mid-Atlantic Industrial Waste Conference, Drexel University, Philadelphia, Pa. (1971).

12. State of Illinois, "Solid Waste Rules and Regulations," Illinois Environmental Protection Agency, Division of Land Pollution Control, Springfield, Ill. (1973).

13. Technicon, "Technicon Autoanalyzer II," Technicon Instruments Corp., Tarrytown, N.Y. (1973).

14. U.S. Environmental Protection Agency, "Methods for Chemical Analysis of Water and Wastes," U.S. Environmental Protection Agency, Office of Technology Transfer, Washington, D.C. (1974).

15. U.S. Geological Survey, "Techniques of Water Resources Investigation of the U.S. Geological Survey; Methods for Collection and Analysis of Water Samples for Dissolved Minerals and Gases," Book 5, Chapter A-1, Washington, D.C. (1970).

LEACHATE ATTENUATION IN UNDISTURBED AND REMOULDED SOILS

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INTRODUCTION

From a technological point of view, the most difficult problem associated with the design of sanitary landfills is the prediction of contaminant migration in the soil. In a flurry of research since 1970, it has been shown that the extent of groundwater pollution resulting from the burial of solid wastes wherein the water is rendered nonpotable is generally limited to a very small region immediately adjacent to the refuse (1-4). This region is frequently contained within a distance of 75 m. It is generally held that the degree to which groundwater pollution occurs is related to the capacity of the soil to attenuate contaminants discharged from the landfill. However, capabilities to predict this phenomenon do not presently exist.

Accordingly, several research programmes (5-9) are currently in progress to examine the interactions between various soil types and typical landfill leachate and industrial waste contaminants. It is intended that these programmes provide information suitable to allow the modelling of contaminant migration in soil.

Much of the experimentation involves the use of columns of remoulded soil through which samples of leachate are passed.

Measurements of contaminant strength in the effluent from the columns are used to calculate contaminant attenuation in the soil. The intention then is to simulate as closely as possible conditions as they exist in the field in order that the data may be transported from the laboratory to the real situation.

Two conditions exist which limit the effectiveness of column studies. The first of these is related to the difficulty in simulating within the column a flow pattern representative of the field. This is particularly a problem when the field flow condition is other than intergranular. In this case, the ratio of leachate to soil contact decreases as the deviation from the condition of intergranular flow increases. To overcome this would necessitate the use of large undisturbed soil samples in the column studies. Research to this point in time has not addressed itself to the laboratory study of attenuation under conditions of fissure flow.

The second major difficulty in the use of column analysis is the time required to collect experimental data. Particularly in soils of significant clay content, the time required for leachate to flow through a laboratory scale soil column is measured in months and may extend well beyond the period of a year.

However, it is reasonable to propose that laboratory techniques could be used to investigate the suitability of potential waste disposal sites. Representative soils from the site could be collected and exposed to typically strong leachates, and the extent to which contaminants are attenuated by the soil could be measured. The resulting data then could serve to predict the extent of contamination in the field. However, to be useful in this regard, the time required to conduct the laboratory investigation should be much less than that required for column flow.

The research described in this paper was undertaken in response to these two situations. The objectives of the research were twofold. The first objective was to compare the flow and contaminant removal patterns in columns containing both undisturbed and remoulded samples of the same soil. The purpose was to determine whether or not, for the soils under investigation, similar patterns could be exhibited. It has been reported (10, 5, 6) that these do exist, but Webber (11) indicates that the availability of suitable supporting data are limited. The significance of this work, if successful, would be a justification for using the more easily managed remoulded condition for laboratory experiments for soils exhibiting intergranular flow.

The second objective of the research was to investigate the use of dispersed soil experiments for examining soil contaminant interactions. Such experiments require only a matter of hours for completion and, within this time frame, would serve effectively as a technique for site evaluation. It was proposed that columns and dispersed soil experiments be conducted in parallel using the same soil and leachate. The ability to reproduce the column performance by using the dispersed soil data was to be investigated.

CONTAMINANT ATTENUATION IN SOIL

General

The attenuation of contaminants in the soil has been described by the following physical, chemical, and biological processes (1, 10, 12-15):

1. Mechanical filtration

2. Precipitation and co-precipitation

3. Sorption

4. Gaseous exchange

5. Dilution and dispersion

6. Microbial activity.

Mechanical filtration is the physical restriction by soil to flow of suspended contaminants. Precipitation and co-precipitation involve the formation of insoluble compounds resulting from, among others, a change in temperature, pH, and/or solution composition as the leachate moves through the soil. Sorption includes the processes of adsorption, absorption, and ion exchange where the sorbing medium may be the soil itself, organic compounds in the soil, microbial growths, or chemical precipitants. Gaseous exchange describes the exchange with air of gaseous contaminants and decomposition products. Dilution and dispersion consist of the processes by which contaminant concentrations are decreased by intermixing with the soil water. Microbial activity defines the uptake and utilization of inorganic and organic contaminants by the soil microbial community.

The complex nature of the attenuation process is demonstrated by microbial activity and its participation in contaminant attenuation. Alexander (15)

identified the following contributions to attenuation which could be made by the soil microbial community:

1. Mineralization

2. Immobilization

3. Oxidation

4. Reduction

5. Volatilization or fixation

6. Geological deposit formation

7. Production of organic chelating or complexing agents

8. Adsorption

9. Isotope fractionation

In Laboratory Experimentation

In the column experiments used in this research, it was assumed that all of the six processes listed as contributing to attenuation could potentially be operative, with the exception of lateral dispersion as it might exist in the field. The columns were sealed under a nitrogen gas pressure to ensure anoxic conditions and consequently the influence of gaseous exchange would be limited.

In the dispersed soil experiments, as with the columns, the influence of lateral dispersion would not be exerted, and because of similar sealed conditions with the presence of nitrogen gas, the full effect of gas exchange would not be observed. In addition, because of the short duration of the dispersed soil experiment, it was unlikely that any appreciable microbial activity would be initiated.

EXPERIMENTAL MATERIALS

Leachate

The leachate used in the experiments was generated in a laboratory lysimeter charged with domestic refuse and in continuous operation since 1971. The quality characteristics of the leachate are shown in Table 1. These data are averages of analyses performed over the duration of the experimental programme. Deviations from mean values were generally less than 10%.

Table 1. LEACHATE CHARACTERISTICS*

pH	5.8
Alkalinity as $CaCO_3$	1,040
Chloride as Cl	900
Sulphate as SO_4	500
Calcium as Ca	600
Magnesium as Mg	250
Sodium as Na	370
Potassium as K	625
Total suspended solids	400
Volatile suspended solids	240
Fixed suspended solids	160
Total dissolved solids	15,200
Volatile dissolved solids	8,560
Fixed dissolved solids	6,640
Free ammonia N	1,106
Organic N	196
Total Kjeldahl N	1,302
Nitrite as N	0.01
Nitrate as N	0.1
Phosphorous as P	1.7
Manganese as Mn	25
Tannins and lignins	300
Phenols, in ppb	8,000
Iron as Fe (total)	47
Iron as Fe (soluble)	---
Chromium as Cr	0.07
Nickel as Ni	0.61
Lead as Pb	1.3
Zinc as Zn	50
Copper as Cu	0.13

*All analyses except pH and phenols are reported in mg/liter. Analyses were performed according to "Standard Methods for the Examination of Water and Wastewater," American Public Health Association, 13th ed., 1971.

Soil

Three soils were used in the experimental programme. Soils 1 and 2 were used in the column experiments to compare the flow and contaminant attenuation patterns. They were chosen to provide information about undisturbed and remoulded conditions over a broad range of grain size distribution. Soil 3 was used in a parallel column and dispersed soil experiment. Only one soil has been reported upon for the column and dispersed soil experiments. However, the complete experimental programme currently in progress examines 10 soils. The properties of the three soils are shown in Table 2.

Table 2. SOIL PROPERTIES

Property	Soil 1	Soil 2	Soil 3
Grain size (% by weight)			
<0.002 mm	10.2	3.9	1.72
0.002-0.074 mm	39.0	6.0	0.58
>0.074 mm	50.8	90.1	97.70
Cation exchange capacity (meq/100 g)	4.3	1.4	3.3
Oxygen demand (mg/g)	nd	nd	0.84
pH	7.37	7.43	7.35
Resident ions (mg/ℓ)			
Na	nd	nd	61
K	nd	nd	1.7
Ca	nd	nd	153
Mg	nd	nd	4.0
Clay mineralogy	*	*	†

* Minerals identified.
† Identified were major chlorite, montmorillinite, and illite; minor vermiculite and kaolinite; and quartz (non-clay mineral).

UNDISTURBED VERSUS REMOULDED SOIL STUDIES

Study Methods

The experiments designed to compare the flow and contaminant removal patterns between undisturbed and remoulded soil were performed in sealed acrylic columns as shown in Figure 1. The columns were 7.6 cm I.D. and 0.6 m in length.
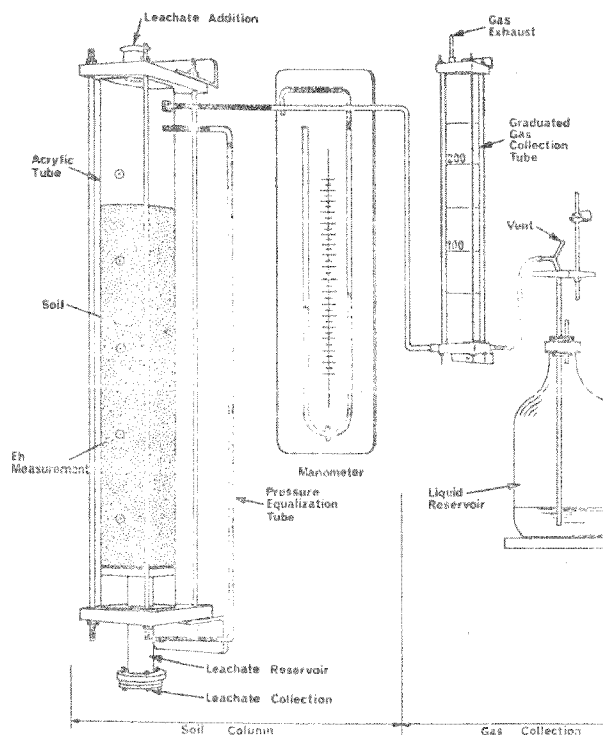
Figure 1. Soil column and gas collection apparatus.

As shown in Figure 1, the columns were fitted with gas collection and pressure equalization facilities. The latter apparatus was necessary to dissipate pressure resulting from gas produced by anaerobic microbial activity.

Undisturbed soil samples were collected in a standard 7.6 cm I.D. Shelby tube. These were extruded into the acrylic columns through the use of an extruder shown in Figure 2.
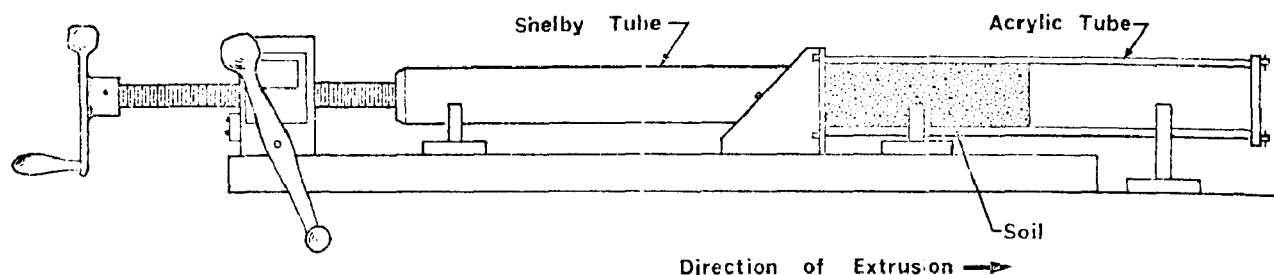
Figure 2. Extrusion of undisturbed soil sample into an acrylic tube prepared for attenuation studies.

The columns were selected to ensure the existence of a uniform inside diameter. The walls of the column were coated with silicone grease to facilitate extrusion and to impede fluid streaming along the column wall/soil interface. No visible change in the soil occurred during the extrusion process.

The same soils were used in the remoulded columns. These were packed into the columns to yield approximately the degree of compaction as in the undisturbed columns. The parallel columns contained approximately the same soil weight. Table 3 lists the properties of the soils.

A volume of 2400 ml of leachate was applied to each column at a weekly rate of 50 ml during the period June 1973 to May 1974. Following percolation through the soil, the leachate was chemically analysed.

Data Analysis and Interpretation

The chemical character of the column effluent is represented in Figures 3 to 6. These figures represent breakthrough curves by showing the concentration ratio, C/C_0, plotted against the column discharge volume in ml where C is the effluent concentration in mg/liters and C_0 is the influent concentration in mg/liters.

Table 3. SOIL COLUMN CHARACTERISTICS

Characteristic	Soil 1		Soil 2	
	Undisturbed	Remoulded	Undisturbed	Remoulded
Bulk density, g/cc	1.66	1.82	1.65	1.70
Moisture content, %	15	15	14	14
Dry weight soil, g	2941.3	2950.8	3199.3	3206.1
Column number	1U	1R	2U	2R
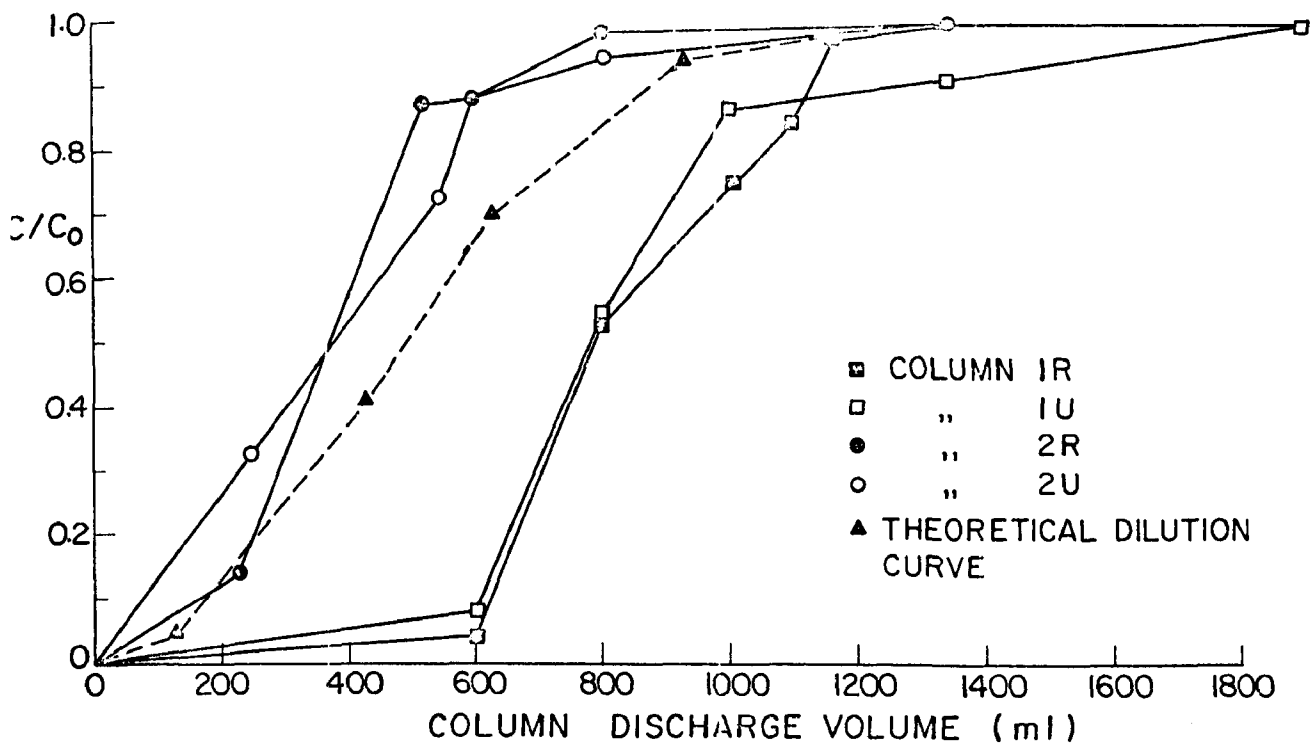
Figure 3. Comparison between undisturbed and remoulded soils. Breakthrough curves for chloride.
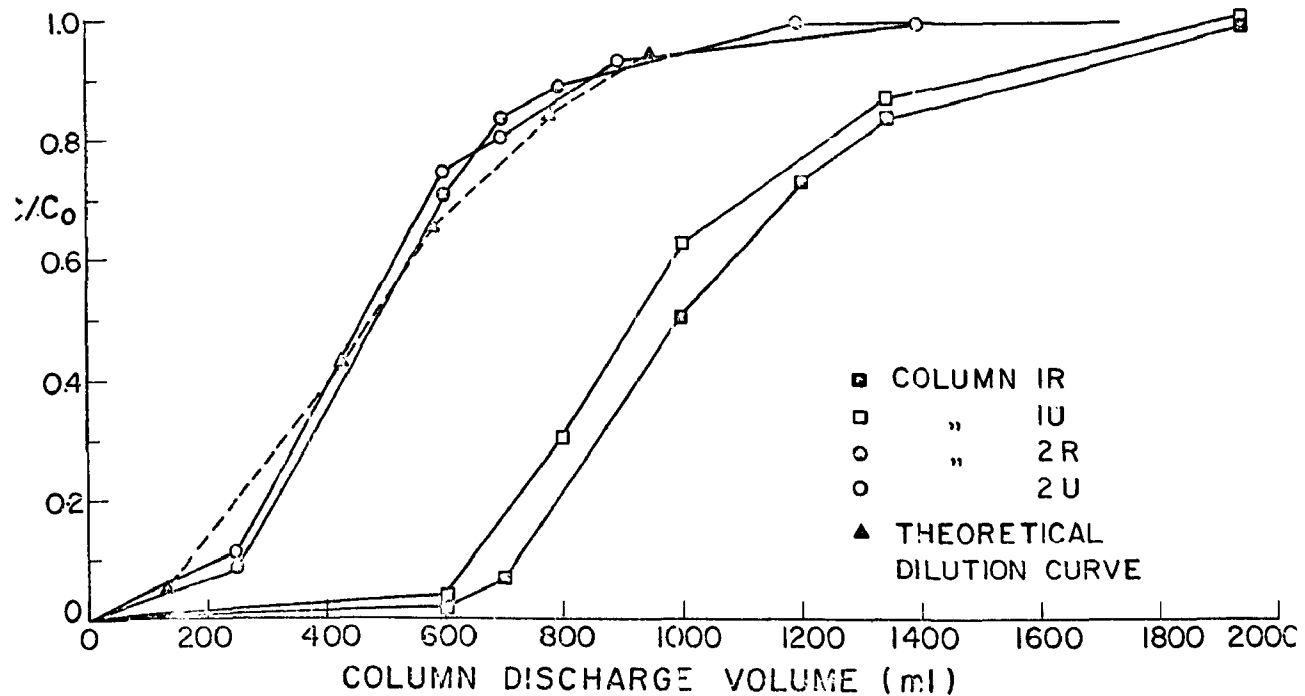
Figure 4. Comparison between undisturbed and remoulded soils. Breakthrough curves for potassium.
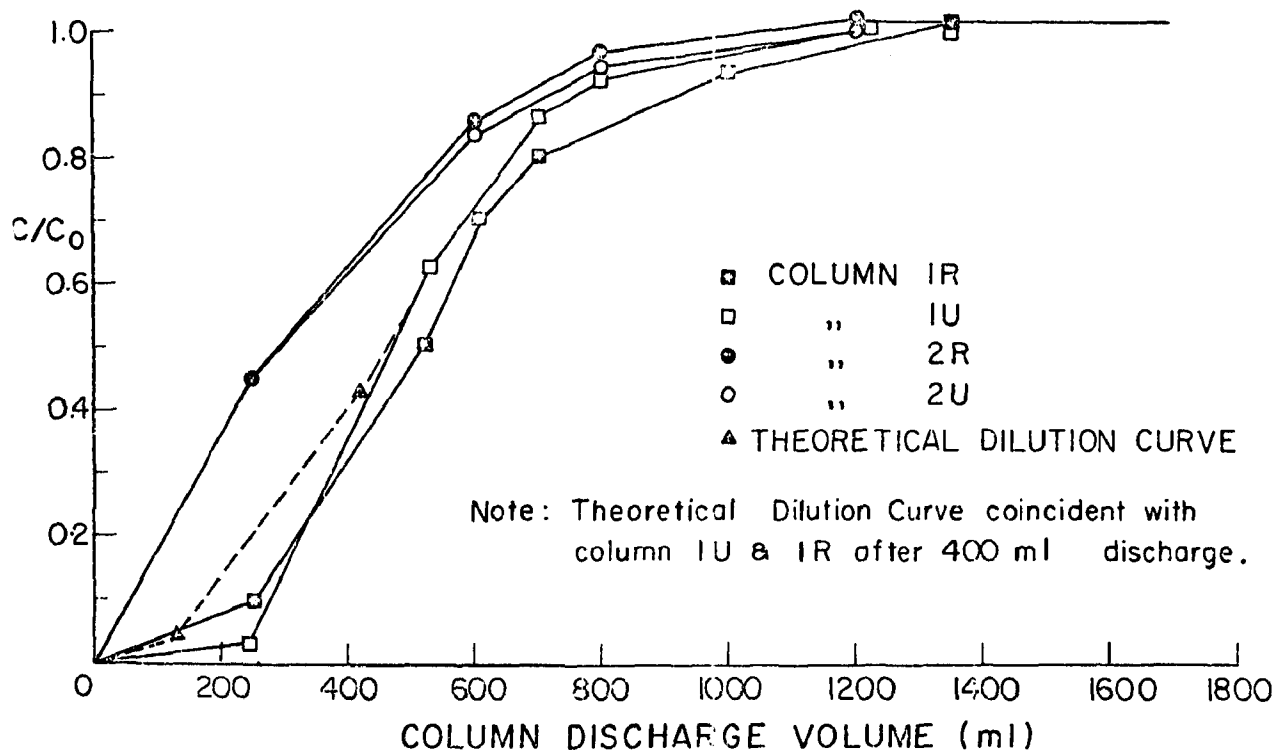
Figure 5. Comparison between undisturbed and remoulded soils. Breakthrough curves for sodium.
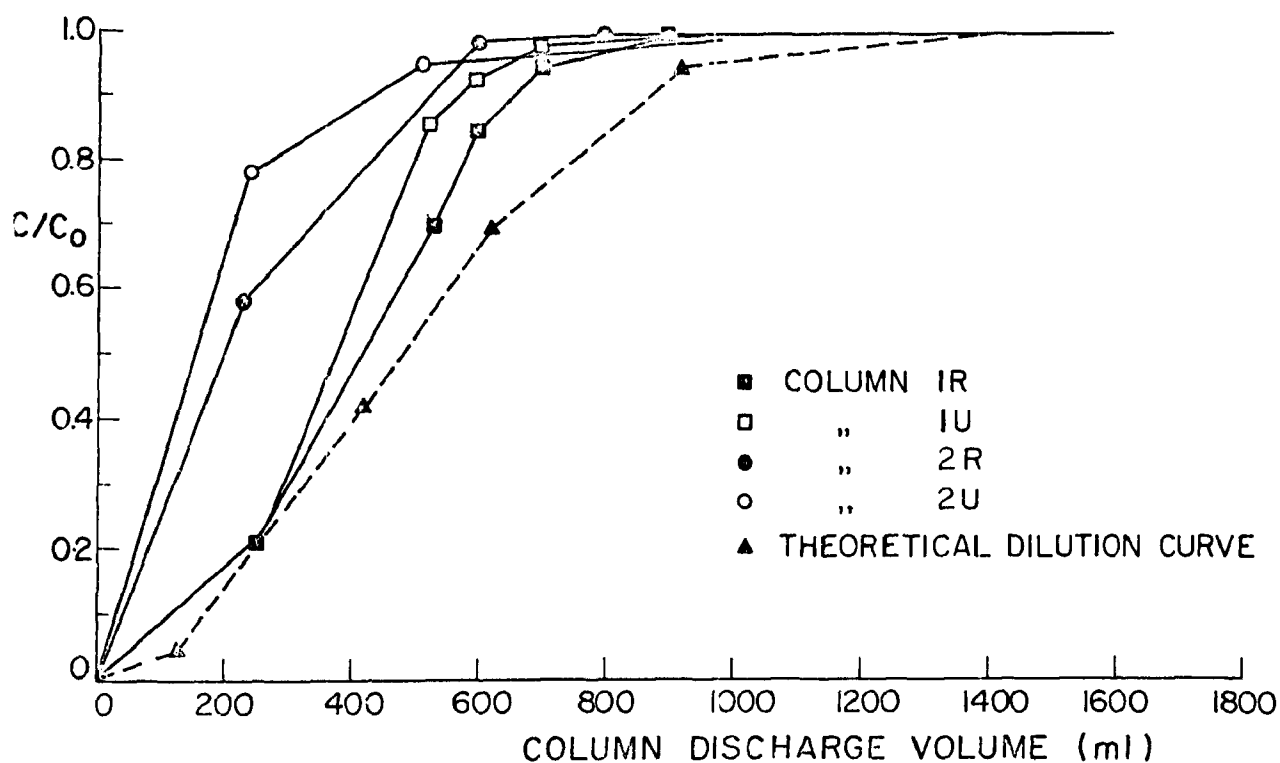
Figure 6. Comparison between undisturbed and remoulded soils. Breakthrough curves for ammonia nitrogen.

Figure 3 shows the chloride ion concentration in the effluents and the theoretical dilution curve (TDC). The TDC was generated by assuming a complete intermixing between the contaminants of the migrating liquid and those of the soil water. The ratio of the intermixing was calculated based on an estimate of porosity. The volume of migrating water used was calculated to be equal to the pore volume minus the volume of soil water. The TDC infers that all contaminants including chloride ion will be attenuated within the soil simply by the process of dilution due to the soil water. While a controversy exists on this subject, it is generally held that chloride ion does not interact with the soil during migration. Thus, deviations between the chloride breakthrough curve and the TDC can be attributed to a combination of incomplete contaminant intermixing with the soil water and axial diffusion. The contribution of either is difficult to determine, but it is believed that the influence of axial diffusion would be small by comparison.

The data in Figure 3 show that while intermixing of chloride ion did occur, it was not complete particularly in soil 2. Soil 2 contained 9.9% particles in the clay and silt size range, while soil 1 contained 49.2%. The time required for the 50-ml leachate aliquots to flow through soil 2 was approximately 1 hr compared to several hours for soil 1. The diffusion of chloride ion from the leachate into the soil water is a time dependent process. Thus, the difference between the chloride ion breakthrough curves of soils 1 and 2 is assumed to reflect this dependence.

The reduction in ion concentration in the migrating leachate due to ion intermixing with the soil water is considered to represent a form of attenuation. From Figure 3, the amount of chloride ion attenuated is represented by the area above the breakthrough curve.

Table 4 represents the masses attenuated which were collected for an initial chloride ion concentration (C_0) of 928 mg/liter and the soil masses given in Table 3.

Table 4. MASSES ATTENUATED*

Column number	Mass attenuated to breakthrough, mg	Mass attenuated to breakthrough per 1,000 g dry soil, mg/1000 g
1U	326.00	110.84
1R	332.96	112.84
2U	89.24	27.89
2R	176.00	54.90

*Collected for an initial chloride ion concentraction (C_0) of 928 mg/ liter and the soil masses given in Table 3.

Breakthrough curves for potassium, sodium, and ammonia ions are shown in Figures 4 to 6, respectively. Masses of contaminant attenuated for several ions are shown in Table 5. The difference between the mass of a contaminant attenuated and the mass of chloride ion attenuated is believed to reflect the influence of processes other than that of dilution.

The data show that for the two soils used, the discharge characteristics of the undisturbed and remoulded conditions compared reasonably well. It is noted that, in general, for soil 1 the remoulded sample attenuated the contaminants to a slightly greater degree. This trend was not as obvious for soil 2 where the percent clay and silt content was considerably less. Upon this basis, the use of remoulded columns would appear to be justified.

The data also show that the mass of contaminant attenuated by soil 1 is consistently greater than

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Table 5. MASS ATTENUATED IN UNDISTURBED AND REMOULDED SOILS 1 AND 2

Chemical characteristics	Influent concentration, mg/l	App. mass applied mg/1000 g dry wt., soil	Mass attenuated, mg/1000 g soil, dry weight							
			Including dilution				Minus dilution			
			1U	1R	2U	2R	1U	1R	2U	2R
Phosphorous (P)	23	18.03	17.17	17.23	15.20	15.08	14.42	14.43	14.51	13.72
Calcium (Ca)	965	756.46	-388.15	-466.41	-392.04	-396.23	-503.41	-583.74	-421.05	-453.31
Magnesium (Mg)	829	649.77	421.63	432.70	380.22	383.89	322.61	331.90	355.31	334.25
Sodium (Na)	665	521.29	132.49	136.89	80.63	81.31	53.06	56.03	60.64	41.97
Potassium (K)	955	748.62	344.62	362.65	213.11	185.33	230.56	246.53	184.40	128.84
Chloride (Cl)	928	727.46	110.84	112.84	27.89	54.90	0.00	0.00	0.00	0.00
Manganese (Mn)	23	18.03	-8.73	-5.83	-10.09	-7.10	-11.47	-8.63	-10.79	-8.46
Iron (Fe)	240	188.14	188.52	175.36	173.89	148.29	159.86	146.17	166.68	134.09
Zinc (Zn)	5	3.92	3.16	2.67	2.51	2.83	2.56	2.07	2.36	2.54
Copper (Cu)	0.25	0.20	-0.03	0.03	-0.18	-0.10	-0.06	0.00	-0.19	-0.12
Nickel (Ni)	0.35	0.27	-0.29	-0.17	-0.24	-0.30	-0.33	-0.21	-0.25	-0.32
Ammonia Nitrogen as N	1200	940.68	422.55	446.35	229.54	250.41	279.23	300.44	193.47	179.42
Organic Nitrogen as N	600	470.34	285.85	273.66	264.57	256.94	214.18	200.70	246.54	221.44
		1411.02								

that by soil 2. This is attributed to the greater percentage of clay and silt size particles in soil 1. The data also show that the extent of attenuation is clearly a function of the particular contaminant involved.

REMOULDED VERSUS DISPERSED SOIL STUDIES

Study Methods

A column containing a remoulded sample of soil 3 was prepared to serve as a basis for comparison between column and dispersed soil behavior. The column was packed to a bulk density of 1.97 g/cc. The column contained 3,553 g of dry soil and 442 ml of soil water. Leachate in a volume of 2,757 ml was added to the top of this column and allowed to percolate. The effluent was collected four times and analysed for its chemical

composition. The data obtained are shown in Table 6.

The data were used to prepare breakthrough curves for chloride, ammonia, and potassium ions. In addition, a TDC was calculated and plotted for comparison.

Dispersed soil experiments were conducted in reactors. Ten of these were filled with 300 g dry weight of soil 3. This soil contained 18.8 ml of soil water. To the first reactor, 250 ml of leachate was added and shaken for 1.5 hr on an Eberbach reciprocal shaker at 180 oscillations/min. The shaker was fitted with cradles into which the reactors were fastened. After 1.5 hr of shaking, the reactor was fitted to the filtration head and the liquid allowed to drain from the soil. The volume of liquid draining was measured and analysed for its chemical characteristics. Experi-

Table 6. CHEMICAL COMPOSITION OF COLUMN EFFLUENT--SOIL 3*

Chemical characteristic	Effluent volume, incremental and accumulative, ml				Influent concentration, mg/liter
	620 620	659 1279	641 1920	765 2685	
COD	14,483	22,717	24,602	26,605	31,489
Chloride as Cl	375	625	650	700	819
Total iron as Fe	4.36	17.20	21.00	24.80	71
Copper as Cu	0.050	0.075	0.075	0.075	0.26
Potassium as K	59	250	365	415	559
Sodium as Na	248	419	477	460	511
Manganese as Mn	4.25	14.25	17.50	17.00	27
Zinc as Zn	0.18	0.35	0.33	0.27	21
Magnesium as Mg	230	225	225	220	188
Calcium as Ca	680	860	970	940	630
Hardness as $CaCO_3$	2645	3073	3348	3253	2346
Organic nitrogen as N	107	140	126	126	108
Ammonia nitrogen as N	98	448	588	686	914

*The chemical composition is representative of the average effluent concentration of the accumulative collected effluent volume.

ments were performed in which the shaking time was varied. It was found that the chemical characteristics of the liquid changed very little after 1.5 hr.

Exposure to the atmosphere was prevented in both the reactors and the columns. Both were sealed and purged with nitrogen gas before experimentation. During the filtration of the dispersed soil reactors, a nitrogen environment was retained, as it was during the collection and storage of liquids.

After contact in the first reactor, a portion of the collected liquid was taken for analysis. The remaining liquid was then added to the next reactor in series. To ensure that an adequate supply of liquid would exist for all 10 reactors, the liquid was brought back to a 250-ml volume through the addition of water. This created an artifical dilution of the contaminant concentrations. The procedure was repeated for all 10 reactors.

Of the 250 ml of liquid added to each of the 10 reactors, the

volume recovered ranged from 215 ml to 236 ml. Thus, since the soil was not at field capacity, a portion ranging from 14 ml to 35 ml of the applied liquid remained in the soil.

Data Analysis and Interpretation

The data produced from this experimental programme consisted of concentrations of various contaminants remaining in the liquids after contact in each of the 10 reactors. The analyses included chloride, sodium, potassium, calcium, magnesium, total iron, manganese, copper, zinc, COD, ammonia, and organic nitrogen. These concentrations decreased with contact in successive reactors due to the combined influence of dilution and other attenuating processes. It was intended that the data be used to prepare contaminant removal isotherms for use in constructing breakthrough curves to simulate column behaviour.

Table 1 showed that the leachate used in this study consists of a complex combination of chemical components. This is to be expected for all leachates obtained from municipal refuse. It can be assumed, therefore, that the attenuation of a specific component will be influenced by the presences of others. This will be particularly true where sorption processes are involved. Thus, some components will experience retarded removal because of selectivity phenomenon. The prediction of this influence in a field situation would be an arduous task. Yet some account must be taken of it. The approach used in this study attempts to accomplish this accounting.

Removal isotherms could have been prepared by creating serial dilutions of the raw leachate and contacting these in the dispersed soil reactors. This would have generated data on contaminant removal as a function of concentration but would not have taken into account the existence of retarded removal of some ions. Consequently, the technique of passing leachate from

one reactor to the next was developed. In this way, the influence of retarded removal could be felt.

Removal isotherms were prepared for the ammonia nitrogen and potassium ion. These are shown in Figures 7 and 8.
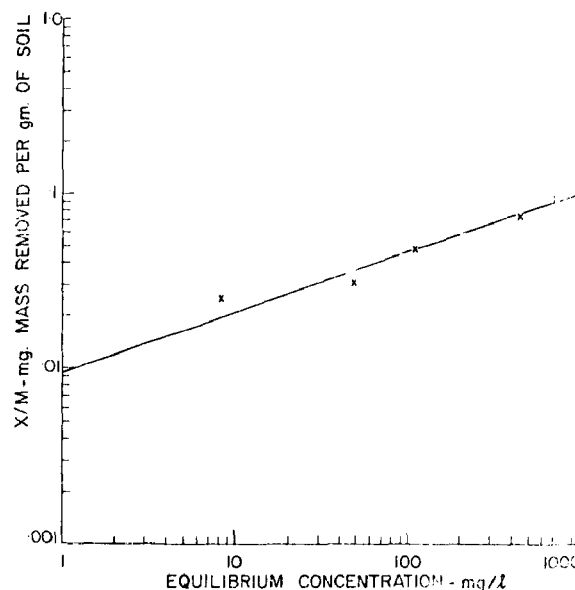
Figure 7. Removal isotherm for ammonia nitrogen.
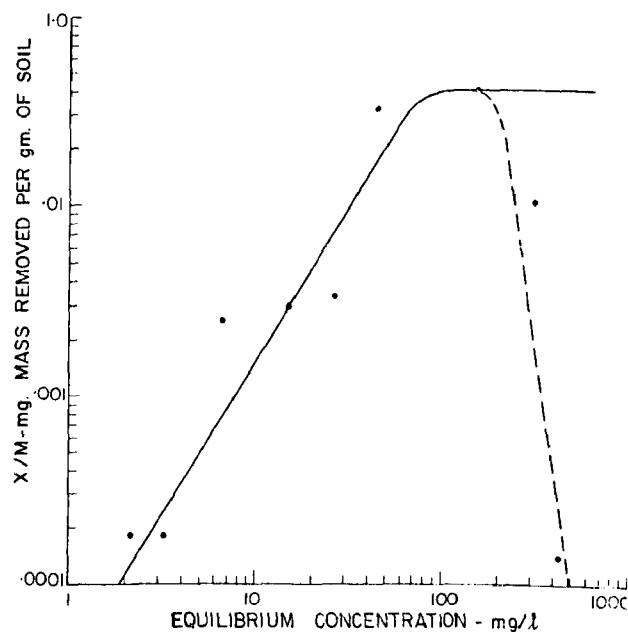
Figure 8. Removal isotherm for potassium.

The concentration, C, on the abscissa are those measured in the liquids collected from the reactors. These were taken as equilibrium concentrations for the removal processes. However, the mass of the particular component removed was calculated as the mass added to the reactor minus [the mass discharged from the reactor plus (the mass remaining in the soil liquid minus the mass in the soil liquid prior to contact)]. The mass removed was divided by the dry weight of soil within the reactor, expressed as mass removed/1000 g dry soil (X/M) and plotted as the ordinants of the isotherms.

The isotherms for ammonia nitrogen shown in Figure 8 exhibit linearity indicative of sorption. However, the non-linearity of the potassium isotherm shown in Figure 9 retarded removal at higher concentrations due to competition with other ions. Within the scope of all components analysed, ammonia nitrogen gave the most desirable isotherm configuration and potassium ion the least. Predictions were undertaken for both ions.
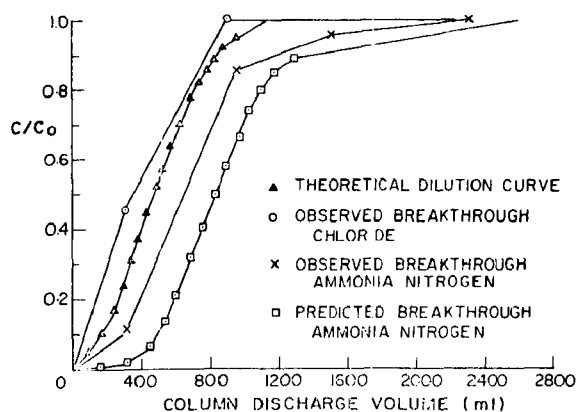
Figure 9. Observed and predicted breakthrough curves for chloride and ammonia nitrogen.

The isotherms were used to predict concentration breakthrough curves for the column described previously. These would be used in comparison with the actual

breakthrough curves of the column. Prediction of the column breakthrough curves took into account the dilution provided by the soil water within the column by assuming a complete intermixing of chemical components of the water and the migrating liquid. An incremental analysis was used by considering units of 147.3 ml of soil water and the corresponding 1184.3 g of dry soil. The estimated porosity of 0.45 was used to determine the amount of liquid to be used in the intermixing calculations. These were 124 ml initially and 100 ml after the soil reached field capacity. Iteration was necessary to determine the operating point on the removal isotherm.

The observed and predicted breakthrough curves for chloride ion and ammonia nitrogen of the soil 3 column are shown in Figure 9. The predicted chloride ion breakthrough curve is in fact the theoretical dilution curve. The mass attenuation to breakthrough including dilution by the soil water for the chloride and ammonia nitrogen is given below:

Item:	Mass attenuated, mg/1000 g dry soil
Observed chloride	88.17
Predicted chloride	110.42
Observed ammonia	178.30
Predicted ammonia	233.00

Comparison of the observed and predicted data exhibits fair agreement. As discussed previously, the lack of fit has been attributed to incomplete intermixing of the components of the leachate and the soil water.

It has been assumed that the ratio of the observed chloride attenuation to the predicted chloride

attenuation would describe the degree of incomplete intermixing. This assumption postulates that chloride is not attenuated by the soil by mechanisms other than dilution.

Therefore, it should be possible to use the above chloride ratio as a correction factor on the predicted ammonia breakthrough curve. In fact, the observed chloride to predicted chloride mass attenuation is 0.798 whereas the observed ammonia to predicted ammonia mass attenuation is 0.765. Therefore, the predicted ammonia curve corrected by a factor of 0.798 would result in a good approximation of the observed ammonia breakthrough curve.

The observed and predicted potassium ion breakthrough curves in the column are shown in Figure 10, along with the theoretical dilution curve. The fit in this case is poor. This is because the sequential removal of potassium ion and the probable desorption of this ion were not accounted for accurately in the calculations. The mass attenuation to breakthrough including dilution by the soil water for the chloride and potassium ion is given below:

Item:	Mass attenuated, mg/1000 g dry soil
Observed chloride	88.17
Predicted chloride	110.42
Observed potassium	402.00
Predicted potassium	303.00

In the case of potassium, the correction of the predicted potassium breakthrough curve cannot be accomplished by the use of the chloride ratio 0.798, which describes the degree of incomplete intermixing.
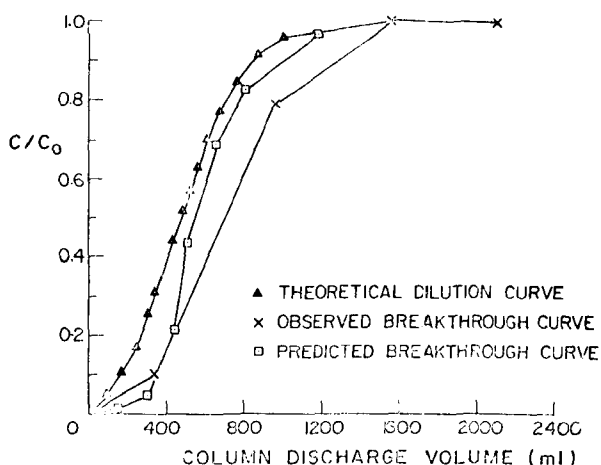
Figure 10. Observed and predicted breakthrough curves for potassium.

The results of the above calculations show that reasonable capability to predict column breakthrough curves from batch removal isotherm data exists provided that the isotherms are linear and that the degree of intermixing of leachate and soil water can be estimated. Additional research in this regard is continuing.

An alternative means of comparing column and dispersed soil experiments was sought to overcome the uncertainty associated with the interpretation of the nonlinear removal isotherm data. In this case, the ratio of discharge concentration (C) to the leachate concentration (C_0) was plotted against the dry soil weight to leachate volume ratio. An accounting was made of the original leachate contaminant mass and volume during movement through the column and through the sequence of dispersed soil reactors. At each point in the incremented analysis, the residual leachate contaminant mass and volume were calculated. This was done by considering the influence of dilution due to the intermixing of contaminants with the soil water, the retention of liquid within the soil, and the addition of water to retain sufficient liquid volume in the dispersed soil reactors.

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The data generated represented the change in contaminant concentrations in the original leachate due only to interaction with the soil as a function of the dry soil weight to leachate volume ratio. Such calculations were prepared for chloride, ammonia nitrogen, total iron, and COD. The data are shown in Figures 11 through 14, inclusive.
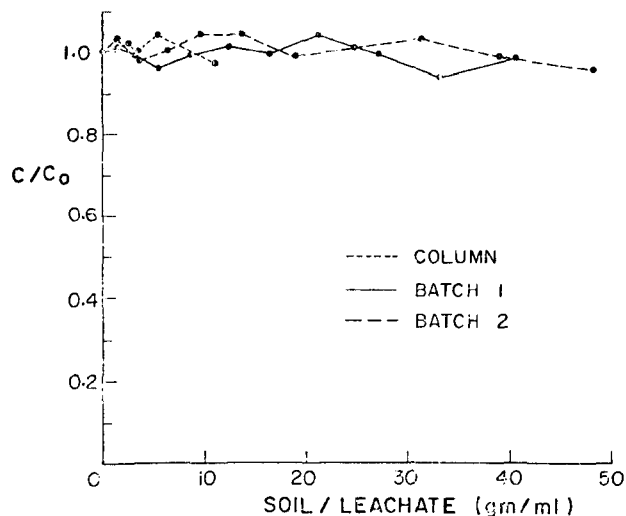
Figure 11. Discharge concentration vs soil to leachate ratio. Chloride.

Note: Batch 1 and 2 curves are coincident

Figure 12. Discharge concentration vs soil to leachate ratio. Ammonia nitrogen.
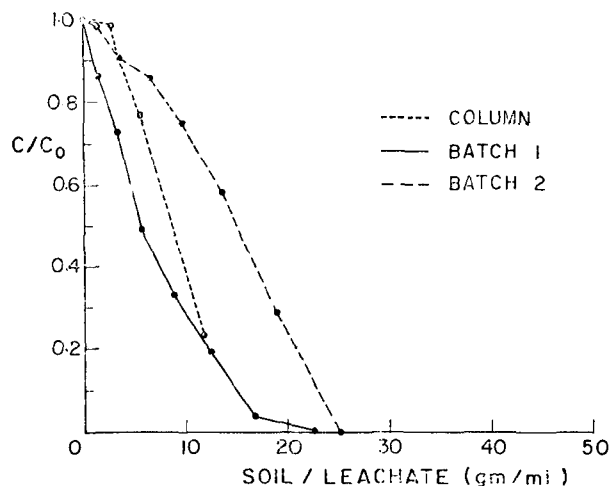
Figure 13. Discharge concentration vs soil to leachate ratio. Total iron.
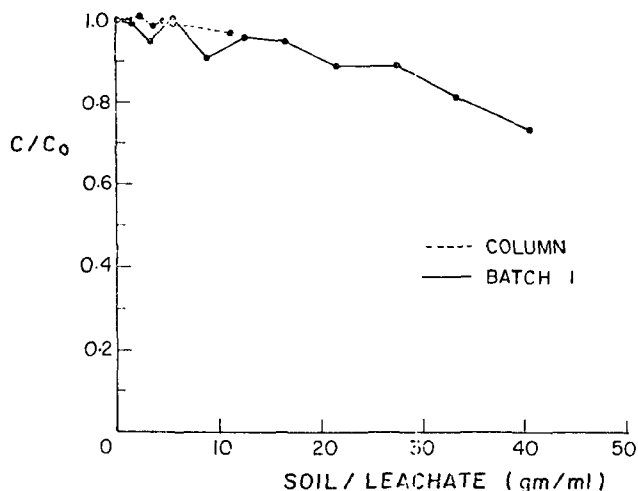
Figure 14. Discharge concentration vs soil to leachate ratio. Chemical oxygen demand.

Figure 11 gives a comparison between the column behaviour and the results of two dispersed soil experiments for chloride. Batch 1 refers to the first dispersed soil experiment in which a slug of leachate was passed sequentially through a series of 10 dispersed soil reactors. Batch 2 refers to a second dispersed soil experiment in which the same 10 reactors were exposed to a second slug of leachate. As expected, the data show that,

in the absence of dilution, C/C_O for chloride remained constant at 1.0 independent of the soil to leachate (g/ml) ratio. Good agreement between column and dispersed soil behaviour is evident.

The interaction of ammonia nitrogen and the soil is shown in Figure 12. Reasonable agreement between the column and Batch 1 is exhibited. The column concentrations are somewhat greater than those of Batch 1. The reason for this difference is not apparent at this time. The degree of intermixing between the constituent of the soil water and migrating liquid in the column was shown to be incomplete upon consideration of the theoretical dilution. Thus the average concentration of ammonia in the soil water would be less than that of the migrating liquid and according to the removal isotherm in Figure 7 this could result in reduced ammonia removal and increased values of C/C_O. In contrast, it is likely that, in the dispersed soil experiments, the degree of component intermixing would be greater than that of the column because of the dispersed condition. Some of the difference may be attributable to this.

The data in Figure 12 show that significant removal of ammonia occurs to the point of complete removal as the ratio of soil to leachate increases. In reality, the concentrations reduce more quickly by virtue of contaminant intermixing with the soil water, a condition which has been removed from the calculations used to prepare Figures 11 to 14. Figure 12 shows that ammonia removal also occurred in batch 2 where the soil in the dispersed reactor was exposed to a second slug of leachate. However, the extent of the removal at a given soil to leachate ratio was less than that in batch 1 due presumably to a reduction in removal capacity with increasing amounts removed.

A somewhat similar situation exists for iron removal as shown in Figure 13. However, a difference

in removal between batches 1 and 2 did not exist. This may be because the removal mechanism included precipitation as opposed to simply sorption, the probable mechanism for ammonia removal.

The data in Figure 14 show reasonable agreement between COD removal in the column and batch experiments. However, very little removal occurs in either case. The flow-through times in the column were short, only a matter of hours. Thus, the opportunity for establishing an active microbial community suitable for the decomposition of migrating organic matter would be small. However, in a field situation with longer contact times, microbial activity could be significant. In such cases, the dispersed soil reactors would not provide adequate information for the removal of organics.

With the exception of being unable to account for microbial activity, the results of this analysis show that dispersed soil experiments can be used to provide an approximate estimate of column behaviour. Based on the comparisons between the undisturbed and remoulded soil column results discussed earlier and by including in the analysis the influence of constituent intermixing between leachate and soil water, it would appear that the dispersed soil experiments can be used to approximate the behaviour of contaminants in the soil.

SUMMARY AND CONCLUSIONS

The potential to pollute the environment does exist with the disposal of waste on the land. At present, it is not possible to quantitatively estimate the pollution potential. To do this it would be necessary to predict the behaviour patterns of contaminant migration in the soil. The purpose of this paper was to develop and evaluate technology suitable for quantitative estimation of contaminant migration.

The major constraints on a prediction model were considered to be the difficulty in simulating flow conditions similar to those in the field and the time needed to generate the necessary attenuation information. To this end, a series of laboratory experiments were designed to evaluate a suitable technological approach.

The experiments have been described in the paper.

The following observations and conclusions were formulated:

1. A significant degree of attenuation by dilution is provided by the water retained in the soil below the field capacity. The theoretical dilution provided by soils 1 to 3 were calculated to be 151.76, 139.23, and 110.42 mg/1000 g dry soil, respectively. The measured dilution provided was observed to be 110.84 and 112.84 mg/1000 g for the undisturbed and remoulded soil 1, respectively; 27.89 and 54.90 mg/1000 g for the undisturbed and remoulded soil 2, respectively; and 88.17 mg/1000 g for the remoulded soil 3.

   It was observed that the remoulded soils provided more attenuation by dilution than did the undisturbed soils and that the soils with the greater content in soil particles of size .074 mm provided the highest ratio of observed to theoretical dilution.

2. When flow conditions are intergranular, the use of remoulded soil columns to estimate attenuation appears to be acceptable. Therefore, the estimation of attenuation using dispersed soil reactors corrected for the degree of dilution provided as calculated from the remoulded soil column would be acceptable. This was observed to be so when the ratio C/C_0 was plotted against the ratio of dry soil weight to leachate volume for the dispersed soil and remoulded column studies. In this case, good agreement was observed for chloride, ammonia nitrogen, chemical demand, and iron.

3. Removal isotherms constructed from the dispersed soil studies can be used to predict the breakthrough curves for some contaminants resulting from remoulded soil column experiments. At present, it appears this can be accomplished for contaminants where the isotherms constructed from the dispersed soil studies are linear, such as ammonium ion. However, more study is needed before nonlinear isotherms, which describe retarded removal of a contaminant such as potassium ion, can be used with any degree of accuracy for prediction.

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REFERENCES

1. Matthess, G. "Hydrogeologic Criteria for the Self-purification of Polluted Groundwater," Int. Geol. Congr., 24th Sess., Sect. 2, Hydrogeology, Canada, pp. 296-303, 1972.

2. Farquhar, G. J., F. A. Rovers, R. N. Farvolden, and H. M. Hill. "Sanitary Landfill Study, Final Report Vol. I, Field Studies on Groundwater Contamination from Sanitary Landfills," University of Waterloo Research Institute, 1972.

3. Hughes, G. M., R. A. Landon, and R. N. Farvolden. "Hydrogeology of Solid Waste Disposal Sites in Northeastern Illinois, A Final Report on a Solid Waste Demonstration Grant Project," Illinois State Geological Survey, Urbana, Ill., U.S. Environmental Protection Agency, 1971.

4. Rovers, F. A., G. J. Farquhar, and J. P. Nunan. "Landfill Contaminant Flux-Surface and Subsurface Behavior," 21st Industrial Waste Conference, Toronto, June 1973.

5. Personal Communication EPA, U.S.A., Sponsored Study, Illinois State Geological Survey, Urbana, Illinois.

6. Personal Communication EPA, U.S.A., Sponsored Study, University of Arizona, Arizona.

7. Personal Communication EPA, U.S.A., Sponsored Study, Dugway Proving Grounds, U.S.A.

8. Farquhar, G. J., and F. A. Rovers. "Industrial Waste Study," University of Waterloo.

9. Rovers, F. A., and G. J. Farquhar. "Contaminant Attenuation in Soil," University of Waterloo.

10. Drewry, W. A. "An Experimental and Theoretical Study of the Movement of Viruses in Groundwater," Ph.D. Dissertation, Stanford University, 1968.

11. Personal Communication with L. Webber, Professor, Department of Soil Science, University of Guelph, Guelph, Ontario.

12. LeGrand, H. E. "System for Evaluation of Contaminant Potential of Some Waste Disposal Sites," Journal AWWA, August 1964.

13. Apgar, M. A., and Langmuir, D. "Groundwater Pollution Potential of a Landfill Above the Water Table," Ground Water, 9, (6), 1971.

14. Bear, F. E. "Chemistry of the Soil," Reinhold Publishing Corporation, 1964.

15. Alexander, Martin. "Microbial Ecology," John Wiley & Sons, Inc., 1971.

VARIATIONS IN GAS AND LEACHATE PRODUCTION FROM BALED AND NON-BALED MUNICIPAL REFUSE

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OBJECTIVES OF THE STUDY EFFORT

Disposal of refuse in a landfill leads to a number of problems related to the design and operation of the landfill site. The major problems related to landfill situations are basically operational. In addition to these, however, there are two other problems which must be addressed to properly maintain and understand the situation that is occurring in the landfill itself. These are gas generation and leachate production. Solid waste, in a landfill consisting of primarily municipal solid waste, decomposes under initially aerobic conditions and then under anaerobic conditions as the oxygen is exhausted. This process yields gas in the form of methane, carbon dioxide, and other gases, such as hydrogen sulfide; and it produces leachate, a complex aqueous solution of dissolved and suspended organic and inorganic compounds and microorganisms.

The gases generated are a problem in the immediate vicinity of the landfill primarily because of the methane content. Methane, in concentrations of 5% to 15% with oxygen in the air, forms an explosive combination. Thus, it is of some interest to determine the amount of methane produced and the rate at which it is produced. These data are needed to predict its migration through soils. To do this, however, it is also necessary to know the quantitative and qualitative production rates of the other gaseous constituents.

Leachate is, similarly, an environmental threat associated with landfills. As a liquid, it can travel through or over the soils and contaminate both surface and groundwaters.

The goals of the program I am describing to you today are (1) to determine the production rate of gas from a landfill in a qualitative sense (that is, identify what gases are present) and in a quantitative sense (that is, determine the gas production rate); and (2) to characterize leachate produced from simulated landfills during the course of the program in terms of its quality (what constituents are present) and the quantity (how much is produced). Ultimately, the data on gas generation will be used in models for predicting landfill gas migration--models that can be used to predict the hazards associated with landfill gas production.

The method to be used for accomplishing these goals was to design and construct five landfill simulators, fill with municipal refuse, and then monitor for temperature, moisture, pressure, leachate, and gas production. The test cells contain unprocessed raw refuse, processed raw refuse, baled unprocessed raw refuse, baled processed raw refuse, and baled unprocessed raw refuse in a saturated environment. The use of all forms

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of landfilled solid waste will provide a basis for comparing the gas and leachate production rates under these various conditions.

During the course of the effort, data will be collected on the various parameters associated with the test cells and analyzed on an on-going basis. At the end of the program, the data will be compiled and analyzed to determine trends, to compare the simulated landfilling methods, to define the qualitative production of gases and leachate, and to make recommendations based on the analysis of the data.

To this date, the solid waste has been placed in the test cells and the monitoring of the gas and leachate has begun. In the following sections of this paper, the design

and construction phase, the cell loading phase, and the initial data from the monitoring of the temperature, gas, and leachate systems are described.

FACILITY DESIGN AND CONSTRUCTION PHASE

The basic design criteria were to provide a test facility that would permit the simultaneous study of the quality and quantity of gas and leachate generation from five different forms of municipal solid waste in an environment simulating a sanitary landfill. To accomplish this goal, a facility consisting of five identical test cells and an instrumentation cell was designed. The facility layout (Figure 1) illustrates the cell arrangement.
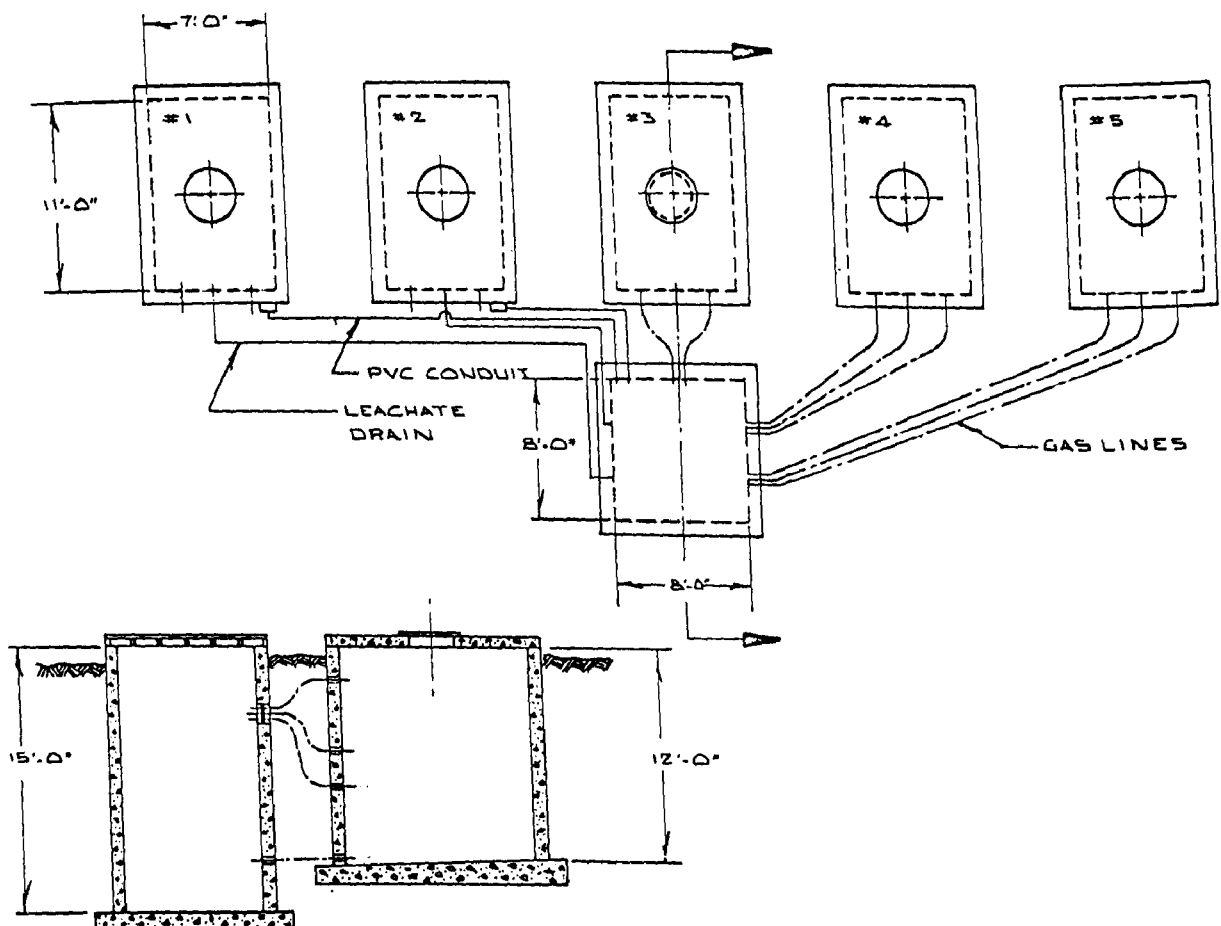
Figure 1. Facility layout.
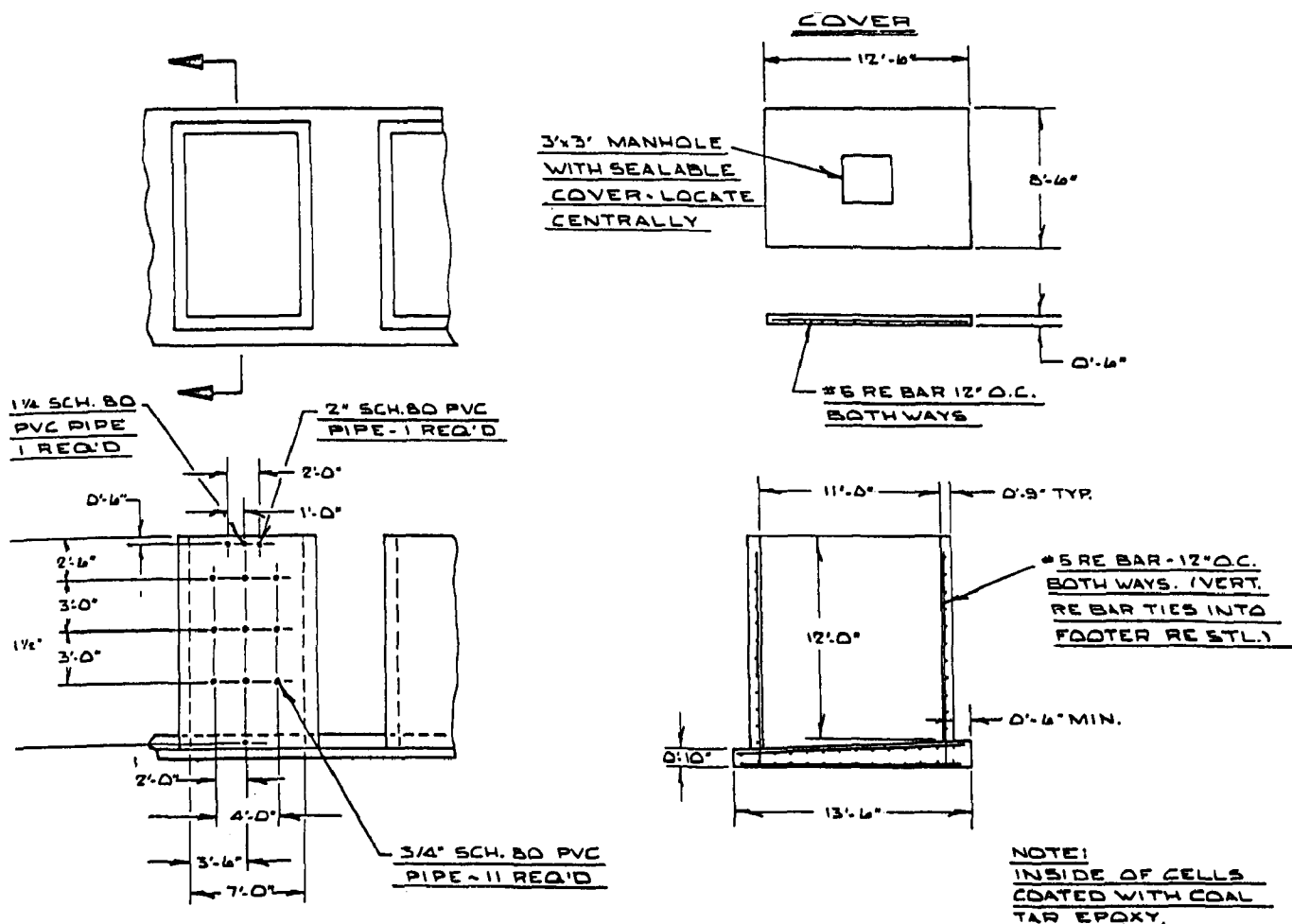
Figure 2. Cell design.

This arrangement provides for good cell accessibility without creating instrumentation difficulties or causing temperature influence problems between cells. The design of the test cells (Figure 2) was determined by both the size of the baled refuse as well as the requirements for compacting refuse in a test cell. Instrumentation access was provided by casting sleeves into the cell walls and then installing bulkhead fittings. The test cells were constructed of reinforced concrete and have inside dimensions of 7 X 11 X 12 ft (2.1 X 3.4 X 3.7 m). They have 8-in. (20.3-cm) thick walls and they are set in a straight line with a 3-ft 8-in. (1.1-m) clearance between each cell for a total overall length of approximately 55 ft (17 m).

The instrumentation and data collection cell, which is centrally located to the test cells, was designed to contain the terminals and collection ports for all the gas, leachate, temperature, and moisture measuring equipment. It was designed to permit all data collection in a central facility rather than several smaller facilities. The cell was constructed similarly to the test cells with the major variation being that the inside dimensions are 8 X 8 X 15 ft deep (2.4 X 2.4 X 4.6 m). The instrumentation cell location is shown on Figure 1, and the general instrumentation layout is shown in Figure 3.

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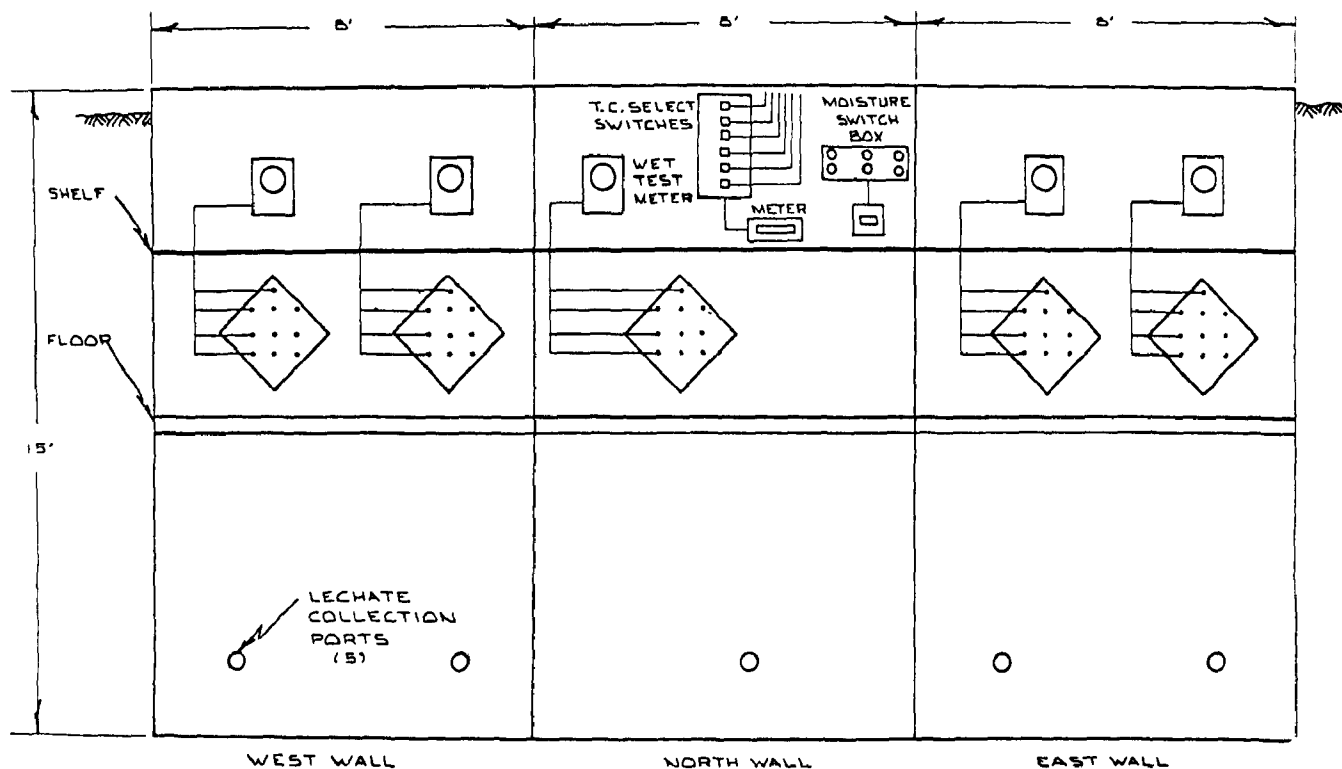
Figure 3. Instrumentation cell layout.

The materials used in the development of the test cells were intended to typify a sanitary landfill and still permit the required sampling and monitoring. The contents of the charged cell, as illustrated in Figure 4, consisted of: a 6-in. (15.2-cm) base of nonreactive gravel; three layers of 9-ft (2.7-m) baled, compacted refuse; 12 in. (30.5 cn) of compacted clay; 12 in. (30.5 cm) of pea gravel; and 6 in. (15.2 cm) of freeboard.

This simulation typifies a sanitary landfill environment in that it does contain the compacted refuse with soil cover and a water source (a water injection rake buried in the pea gravel) but differs because of its small size and sealed environment. The use of pea and nonreactive gravel facilitates testing without affecting the analysis of the data.

The on-site instrumentation for this facility consists of temperature, moisture, and gas monitoring equipment. The temperature monitoring equipment consists of a total of 120 copper-constant thermocouples with each cell having 24 probes distributed throughout the refuse. The extension leads of each cell's thermocouples are run through PVC conduit to each of five separate 24-position selector switches located in the instrumentation cell. The output of each of these five selector switches is connected to another selector switch whose positions correspond to the five test cells. The output of this selector switch is then connected to a digital readout temperature indicator. The wiring and switching arrangement is schematically shown in Figure 5. The thermocouple design is illustrated in Figure 6. The distribution of the thermocouples is shown in Figure 7.

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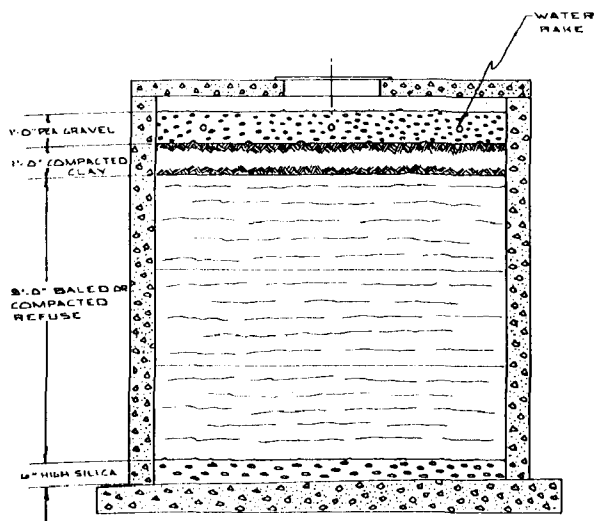
Figure 4. Test cell covers.

WATER RAKE

1'-0" PEA GRAVEL
1'-0" COMPACTED CLAY
8'-0" BALED OR COMPACTED REFUSE
6" HIGH SILICA

CELL #1 CELL #2 CELL #3 CELL #4 CELL #5

24 WIRE BUNDLE

INST. CELL SW. 1 INST. CELL SW. 2 INST. CELL SW. 3 INST. CELL SW. 4 INST. CELL SW. 5 CELL SELECT SW. 6 POS.

DIGITAL READOUT °C
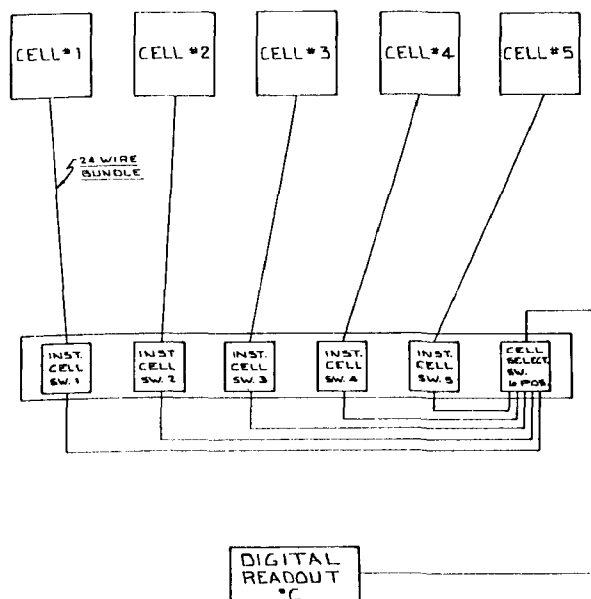
NOTE.
ALL INST. CELL SWITCHS
24 POSITION.

Figure 5. Thermocouple wiring.

2 COND. TYPE "T" THERMOCOUPLE WIRE
CRIMP
BEAD CONN.
FILL WITH HIGH TEMP. EPOXY
STNLS STL. CASING
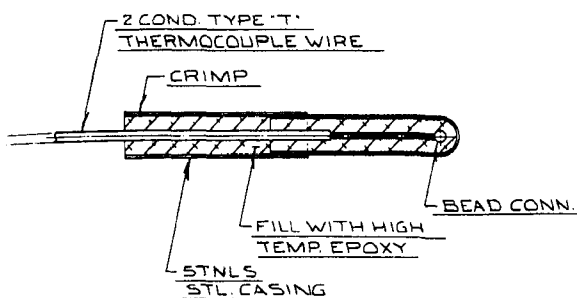
Figure 6. Thermocouple design.

2'-6" 1'-2"
1-2
5-6 7
3-4

1'-6"
1-3 5
2-4 6 7

2'-6"
1'-6"
8-9 14
10-11 15 17
12-13 16

1'-6"
8-10-12 14-15-16
9-11-13 17

1'-3"
2'-6"
19-20
18 23 24
21-22

1'-6"
19-21 23
18 20-22 24
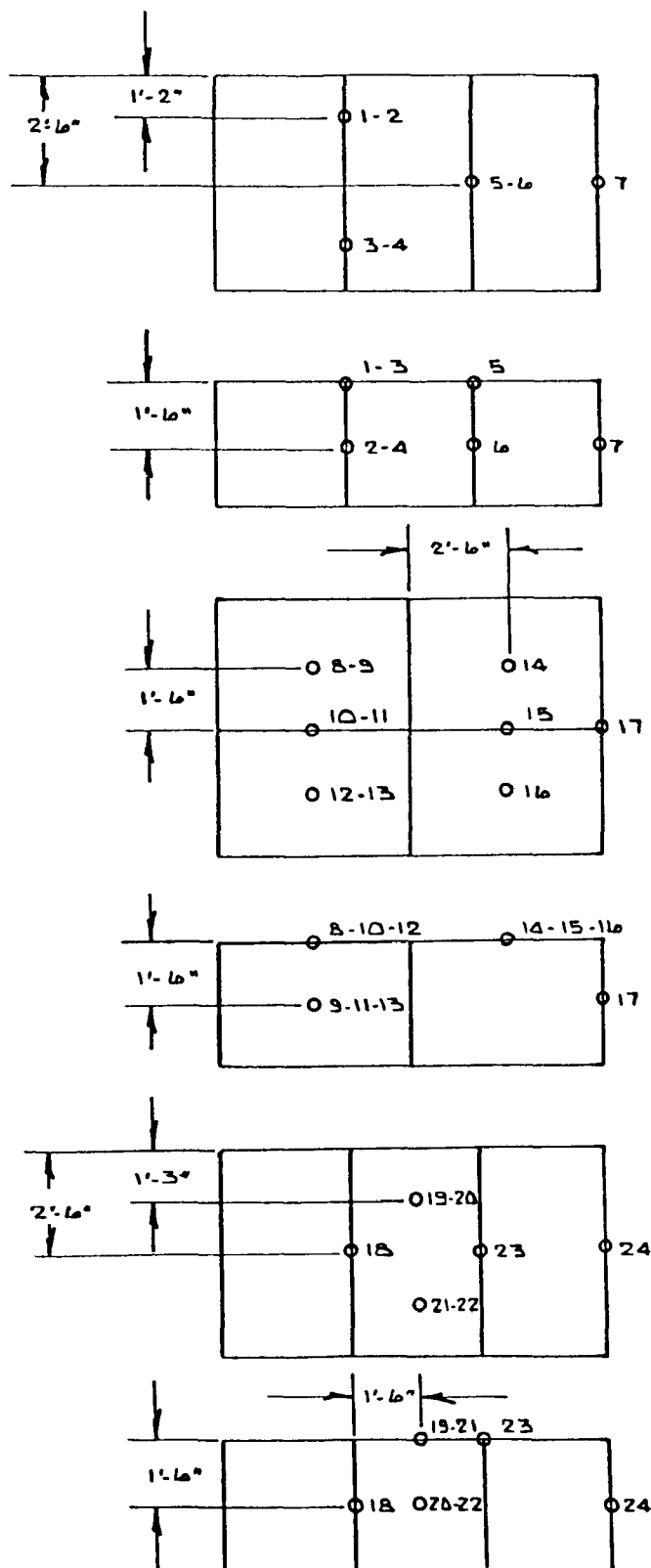
Figure 7. Distribution of the thermocuples.

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Figure 8. Moisture probe location.

The moisture monitoring instrumentation consists of both gypsum soil blocks and porous cup tensiometers. The gypsum soil blocks operate on the principal that the moisture in the soil is absorbed by the gypsum, which separates two electrodes, and causes the resistance across these electrodes to change. This change in resistance is measured on an ohm meter with a special calibration. Each cell contains nine of these probes, and their location in the refuse is shown in Figure 8. The extension leads for these probes are run through the PVC conduit used for the thermocouples and are connected to a switching unit similar to that described for the thermocouples.

The porous cup tensiometer is a unit that measures moisture availability in the soil to a pressure of 100 centibars. The unit consists of an air-tight, water-filled tube connected to a porous cup probe. The porous ceramic probe permits the water in the tube to flow in and out depending on the moisture content of the soil. The moisture content is measured on a vacuum gage connected to the water-filled tube. Two of these units are used in each test cell.

The gas monitoring system consists of 10 collection probes for each cell that is connected to a manifold and valve arrangement inside the instrumentation cell and then connected to a precision wet test gas meter. The gas collection piping in the test cells consists of: three rows of three perforated plastic pipes in the refuse and one collection port in the test cell freeboard. The manifolding is designed to permit gas collection from any of the three levels of perforated pipes, the freeboard area, or any combination of these two regions. Figure 9 schematically presents this piping and instrumentation arrangement.

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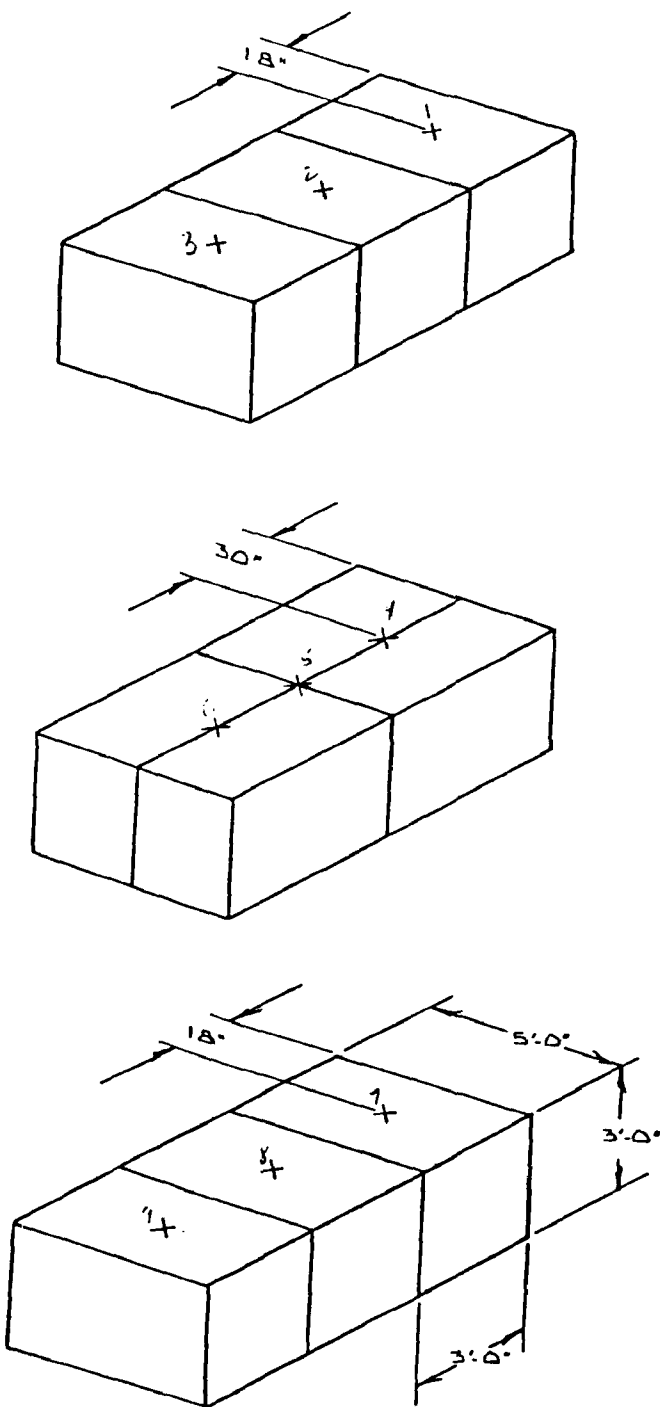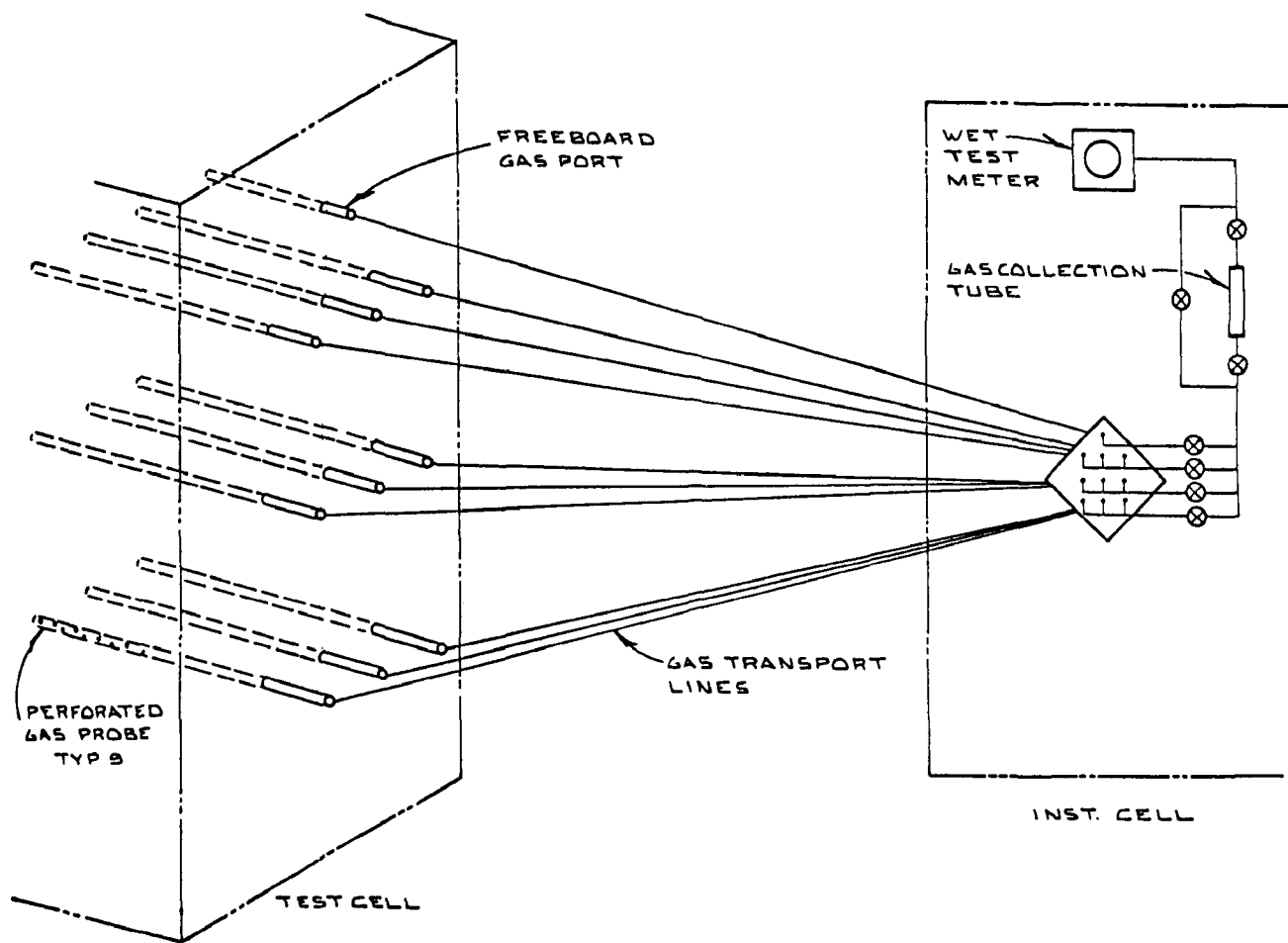
Figure 9. Gas collection schematic.

CELL LOADING PHASE

Introduction

The effort called for loading solid waste into five test cells. The solid waste to be placed in each of the cells was defined previously but will again be identified here: cell 1, baled shredded refuse; cell 2, baled unshredded refuse; cell 3, baled unshredded refuse (saturated); cell 4, shredded raw refuse; and cell 5, unshredded raw refuse.

Initially, all the solid waste was to be obtained from the City of Oakwood, Ohio, a suburb of Dayton. However, during the initial phase of the effort, it became apparent that the baler proposed to be used for baling the solid waste could not achieve the 1500 lb/cu yd compaction density required by the contract. Hence, a baler had to be located that would provide this compaction density.

During this search for the baler, only two were located that we believed would provide this compaction density. One was in Massachusetts and the other in Georgia. The Massachusetts baler facility was experiencing some legal and operational difficulties and, hence, it was decided not to use this facility. A visit to the Cobb County, Georgia, Baler facility showed that this facility would be the most desirable one to use

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for this study. This baler produced a baled size of approximately 3 X 3 X 5 ft at a compaction density of 1600 lb/cu yd.

The unbaled solid waste, both shredded and unshredded, was obtained from the City of Oakwood, Ohio, as was initially planned. A shredder at the Montgomery County Incinerator, Dayton, Ohio, was used to shred the raw refuse for cell 4. The contract required a compaction density of approximately 850 lb/cu yd for the shredded and unshredded raw refuse test cells.

To successfully load test cells with solid waste, it is necessary to develop detailed logistics plans. These plans included a material requirement plan, an equipment requirement plan, and a personnel requirement plan. The material section of the logistics plan defines the total quantities of solid waste, clay, pea gravel, etc., required to initiate the complete loading sequence and to specify sources for each of the materials required. The equipment section of the logistics plan identifies the type, source, and contact for each item of equipment required for the loading sequence. This includes hand tools, rental equipment, and contract equipment for work performed both at Dayton, Ohio, and Atlanta, Georgia. The manpower section identifies the various tasks involved in the total operation, identifies the type of labor required to perform the task, and the personnel required to supervise each group of tasks. Detailed outlines were prepared for the work effort at both Dayton and Atlanta.

Another requirement of the contract called for characterizing the solid waste at the different facilities. Hence, provisions had to be made for hand sorting approximately 350 to 400 lb of the solid waste at each facility from where waste was obtained. Several of these sorts were performed to better categorize the solid waste used in this study.

Since the baler facility in Atlanta, Georgia, was not operational until the middle to end of November 1974, cells 4 and 5 containing the shredded and unshredded raw refuse obtained from Oakwood, Ohio, were loaded first. After the loading of these two cells, plans were then made to load the remaining three cells with the baled solid waste from Atlanta, Georgia. In the following sections, the loading of the test cells, located in Franklin, Ohio, are described.

Loading of Shredded and Unshredded Raw Refuse (Cells 4 and 5)

The refuse from the City of Oakwood was delivered to the Montgomery County South Incinerator by the City of Oakwood collection vehicles. Plans were made to shred the material at the South Incinerator, perform the 350- to 400-lb hand sorts for waste characterization, and truck the shredded refuse to Franklin for loading into cell 4.

The first cell to be loaded was the shredded solid waste for cell 4. However, during the initial shredding operation at the incinerator facility, a malfunction of the shredder caused us to abort trying to load the shredded refuse and plans were immediately changed to load the unshredded cell, cell 5. The solid waste arrived at the incinerator facility about 1:00 p.m. and cell 5 was loaded by 1:00 a.m. The loading sequence used was as follows:

1. The solid waste was dumped on the tipping floor at the Montgomery County South Incinerator, mixed, and a 350- to 400-lb sample was removed. This material was then hand sorted for characterization.

2. The material was loaded on packer trucks and transported to Franklin, Ohio, where it was dumped on a concrete pad. The

vehicles were weighed
before and after leaving
the Montgomery County
Incinerator so that the
weight of the refuse would
be known.

3. A front-end loader at
    Franklin placed the refuse
    in the cells in ap
    proximately 3- to 4-ft
    layers. Sample buckets
    of the refuse as picked
    up by the front-end loader
    were weighed; the
    approximate weight of each
    load being placed in the
    test cell was needed so
    that the desired compaction
    density could be achieved.

4. After the material was
    placed in the cell, the
    material was compacted
    by using a crane and a
    wrecking ball modified
    with a rectangular plate.
    The crane would continually
    pick up the wrecking ball
    and drop it on the refuse
    for compaction.

5. At the required intervals,
    as identified in Section
    2, instrumentation in the
    form of gas probes, ther-
    mocouples, and moisture
    probes would be placed
    in the cells. As soon
    as thermocouples were
    placed in the cells,
    readings would be taken.

6. The above steps continued
    until the refuse was
    completely placed in the
    cells and the desired
    compaction density achieved.
    The material left on the
    pad was weighed so that
    the exact weights of the
    refuse in the cells could
    be obtained.

7. When the solid waste had
    reached the desired level
    in the test cell, a 1-ft
    layer of soil was placed
    on the refuse. This soil

was compacted by using
construction-industry-type
road tampers. A water
distribution system was
then installed at the top
surface of the soil layer,
and a 12-in. gravel layer
was then placed on top
of the soil and water rig.

8. A concrete lid was then
    placed on the cell to seal
    it.

The only additional steps
needed to load the shredded solid
waste from the City of Oakwood were
that the material had to be shredded
first at the Montgomery County
Incinerator and then loaded on
packer trucks for transportation
to Franklin, Ohio. The test cell
loading sequence was identical.
A compaction density of 840 lb/cu
yd was achieved for the raw refuse
test cell, and a compaction density
of 929 lb/cu yd was achieved on
the shredded test cell.

Loading of Cells 1, 2, and 3

The loading of cells 1, 2,
and 3 presented a few more challanges
than did cells 4 and 5. The main
reason for this is that the refuse
had to be trucked from Atlanta,
Georgia, to Franklin, Ohio. Detailed
plans had to be prepared to ensure
that the refuse could be baled and
delivered as quickly as possible.
To provide some insight as to the
logistics problems involved in this
particular effort, the following
discussion is presented.

The baler facility was located
in Marietta, Georgia, a city
northwest of Atlanta, and the
shredder was located southeast of
Atlanta. Provisions had to be made
to haul the shredded refuse to the
baler facility for baling. In
addition, the bales were banded
to ensure that they would be
delivered to Franklin in a baled
condition. A local Atlanta
contractor was retained to band
the bales. So the bales could be
handled easily at Franklin during
the loading of the test cells, a

3-in. I-beam was banded into the bale; the bale could then be moved using the I-beam and a crane.

The hauler of the bales to Franklin reported to the baler facility for the loading of the bales and then immediately took the bales to the banding subcontractor approximately 20 miles away. So you can see a good detailed logistics had to be developed to ensure the timely removal, baling, shredding, and delivery of the solid waste to Franklin, Ohio, for loading into the test cells.

As discussed previously for the loading of cells 4 and 5, 300- to 350-lb hand sorts of the solid waste had to be performed at both facilities (shredding and baling). The sorts were performed using local labor but with Systech's supervision.

The baling and shredding of the refuse in Atlanta started on December 16, 1974, and the cells were loaded, instrumented, and sealed by December 20, 1974. The bales were received in Franklin on December 19 and 2 days were required to completely load and seal the cells. The bales remained together during the shipment from Atlanta, and as bales were placed in the cells, the bands were removed. The loading and instrumentation procedures were identical to those described in the previous section.

SUMMARY OF DATA GATHERED THUS FAR

To this date, little gas has been generated, and in fact, insufficient gas has been generated for analysis. Leachate has been gathered several times. It is believed that the first leachate collected is the squeezings from the solid waste during loading. Further leachates gathered reflect leachate generated from moisture additions. Twenty-four inches of water will be added to the cells annually. The following tables and figure contain some of the initial data (Tables 1 through 3 and Figures 10 through 12). They are inclusive at this time, and no conclusions are drawn because of the limited amount of data gathered thus far. They are presented here for your perusal and illustrate the type of data gathering and analysis that will be performed on the gas and leachate produced from each of the five test cells.

Table 1. WEIGHT OF MATERIALS PLACED IN TEST CELLS AND DENSITY OF REFUSE

Categories		Cell 1	Cell 2	Cell 3	Cell 4	Cell 5
Silica gravel	(lb)	5,120	5,120	5,120	5,120	5,120
	(kg)	2,324	2,324	2,324	2,324	2,324
Refuse	(lb)	25,600	25,600	25,600	23,840	21,400
	(kg)	11,662	11,662	11,662	10,832	9,716
Clay backfill	(lb)	30,923	30,923	30,923	--	--
around bales	(kg)	14,039	14,039	14,039	--	--
0.3 clay cover	(lb)	7,161	7,161	7,161	7,161	7,161
	(kg)	3,251	3,251	3,251	3,251	3,251
Pea gravel	(lb)	4,197	4,197	4,197	8,393	8,393
	(kg)	1,639	1,639	1,639	3,863	3,863
Total	(lb)	73,001	73,001	73,001	44,514	42,074
	(kg)	33,142	33,142	33,142	20,270	19,154
Refuse	(lb/cu yd)	1,452	1,452	1,452	929	840
density	(kg/cu m)	853	853	853	554	496

Table 2. CHEMICAL ANALYSIS OF LEACHATE*

Cell Number	Color	pH, S.U.	Conductivity, μ mhos	COD, mg/ℓ	Hardness, mg/liter, as CaCo	TOC, mg/ℓ	Volume, ℓ
1	Clear	5.7	4,100	455	232	74	1
2	Clear	6.4	1,800	783	1,016	360	4
3	Reddish-yellow	6.4	3,000	957	1,320	410	1
4	Clear	5.7	1,500	164	588	60	1
5	Very light yellow	5.8	3,600	197	480	56	1

*Odor was not detectable for any of the samples.

Table 3. METAL ANALYSIS OF LEACHATE

Cell Number	Iron, mg/ℓ	Copper, mg/ℓ	Zinc, mg/ℓ	Nickel, mg/ℓ
1*				
2	14	1.0	0.42	0.25
3	50	1.0	.20	.25
4	0.5	1.0	.22	.25
5	0.3	1.0	.14	.25

*Insufficient sample for analysis. Cadmium and lead samples were preserved for later analysis.
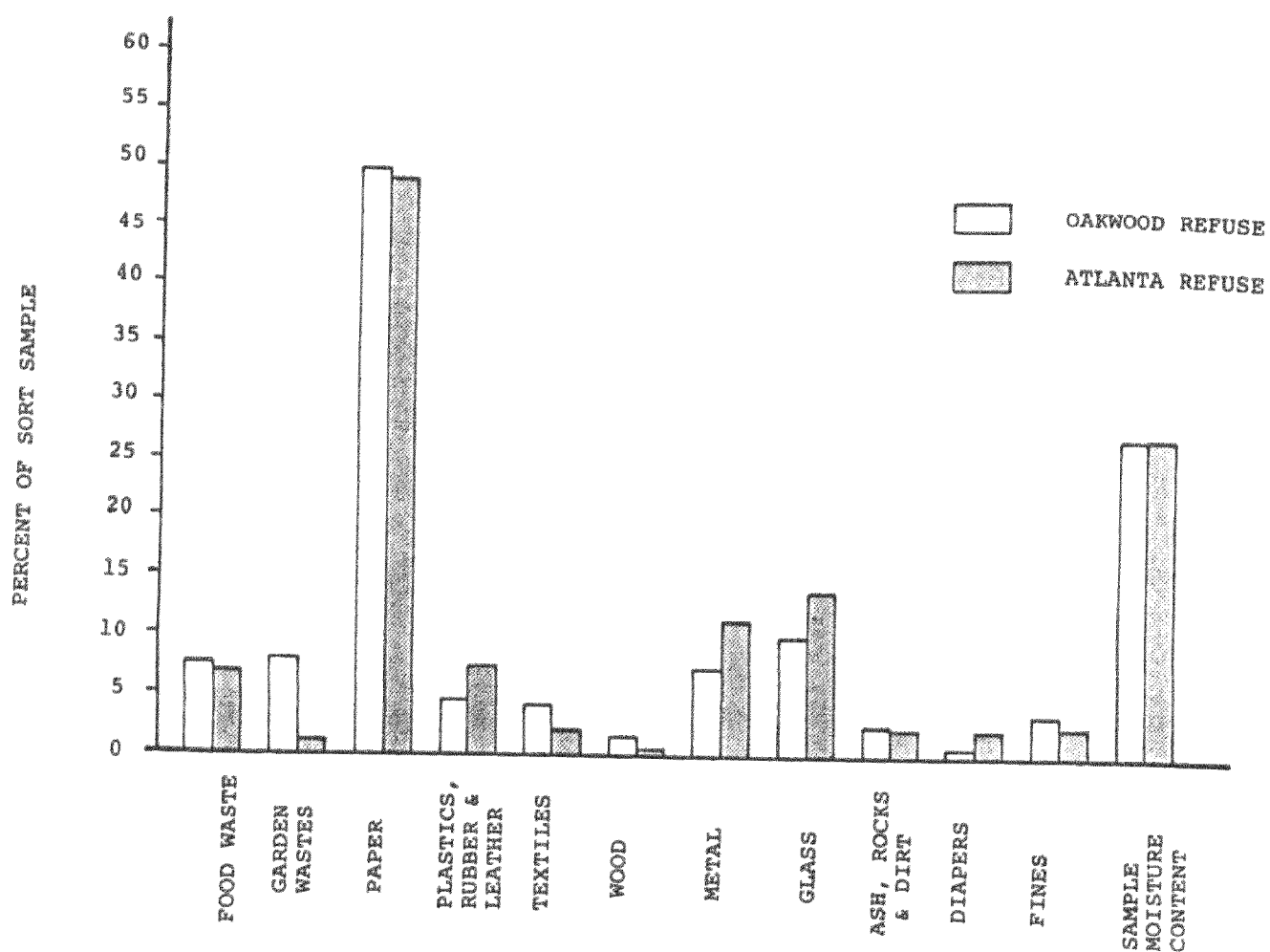
Figure 10. Refuse composition comparison. Average of three sorts for both Oakwood and Atlanta refuse.

GAS AND LEACHATE GENERATION IN VARIOUS SOLID WASTE ENVIRONMENTS

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and
D. R. Streng
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Dayton, Ohio

INTRODUCTION

The work being performed on this program consists of a detailed pilot scale landfill simulation being performed at the Center Hill, Cincinnati, Ohio, facilities of the U.S. Environmental Protection Agency (EPA). The program is divided into two phases: Phase I consists of preparation of 15 test cells and filling these test cells with solid waste and selected other wastes; Phase II consists of collecting and analyzing data generated by these cells.

Broadly speaking, the objective of the program is to study solid waste decomposition in sanitary landfills. More specifically, the objectives are to determine:

1. the effect on gas and leachate production by varying the moisture regimen,

2. the influence of temperature on gas and leachate production,

3. the effect of wastewater treatment plant sludges on solid waste decomposition,

4. the effect of pH control on solid waste decomposition,

5. the effect of premature wetting on solid waste decomposition,

6. the effect of hazardous liquid and sludge wastes on solid waste decomposition and the fate of the hazardous waste,

7. survivability of poliovirus, and

8. settlement rates for a variety of environmental and operational conditions.

Because of the variety of objectives and the number of cells, the design of this experiment is somewhat difficult to grasp quickly. To assist in clarifying the purpose of each cell in meeting the objectives presented above, Table 1 shows the important parameters of each of the test cells.

Cells 1 through 4 satisfy the varying rainfall regimen (objective 1); cells 5, 6, and 7 contain sewage sludge mixed with the solid waste (objective 3); cell 8 has calcium carbonate added to the solid waste (objective 4); cell 11 received enough dechlorinated water to simulate 75% of the field capacity (objective 5); cells 12 through 14, and 17 received hazardous wastes (objective 6); cells 16 and 17 are located inside the high bay area of the Center Hill facility where

the environment can be maintained at room temperature. These two cells provide the basis for comparison to determine the effect of temperature (objective 2). Cell 15 contained poliovirus inserted into the solid waste in the cell (objective 7). All cells have settlement devices for tracking the change in height of the solid waste over the period of the project (objective 8).

As indicated by the table, cells 9 and 10 contain no solid waste. These cells are reserved for two additional hazardous waste streams, which, at the time these cells were filled, were unavailable. A second loading sequence will begin on April 1, 1975, during which time these cells along with several others will be charged.

The majority of the cells (1-15) are located outside, south of the laboratory building. Their arrangement is a U-shape to facilitate loading and optimize instrumentation connections to the test shed containing temperature and gas measuring equipment. Cells 16 and 17 are located inside the high bay area of the laboratory where the ambient temperature is maintained between 65 and 75 F.

DETAILS OF CELL DESIGN

The test cells are 1.8 m (6 ft) in diameter and 3.6 m (12 ft) deep. They are made of 4.76-mm (3/16-in.) steel covered with coal tar epoxy. The cells outside have poured concrete bottoms. The two interior cells have steel bottoms (which were welded onto the main tube of the cell). Provisions for draining leachate have been included in the bottom of each of the cells. A depression in the concrete base contains a 76-mm tube that connects the cell to the central observation well. Each of the leachate collection lines was secured with ball valves. All leachate is collected anaerobically by incorporating a valving system that allows purging of the piping involved

Table 1. PARAMETERS OF EACH TEST CELL

CELL NO	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
WASTE STREAM	—	—		—	SEWAGE SLUDGE	SEWAGE SLUDGE	SEWAGE SLUDGE	CaCO₃	PETROLEUM SLUDGE	BATTERY WASTE	H₂O	ELECTROPLATING WASTE	INORGANIC CEMENT WASTE	CHLORINE PROD BRINE SLUDGE	POLIO VIRUS		SOLVENT BASED PAINT SLUDGE	—	—
AMT POUND/KG					150/68	450/204	1500/680	200/90.7	3347/1518	2847/1291	2851/12932	2624.9/1190.4	3131.9/1420.5	4494.4/2038			3536/1604.0		
TOP TYPE*	OPEN/CLOSED	OPEN/CLOSED	SEALED	SEALED	OPEN/CLOSED	OPEN/CLOSED	OPEN/CLOSED	OPEN/CLOSED	OPEN	OPEN	OPEN/CLOSED	OPEN	OPEN	OPEN	OPEN/CLOSED	SEALED	SEALED	SEALED	SEALED
DEPTH IN/ANNUAL MOISTURE mm	8/203.2	18/406.4	24/6096	32/812.8	16/406.4	16/4064	16/4064	16/4064	16/406.4	16/406.4	16/4064	16/4064	16/4064	16/4064	16/4064	16/406.4	16/406.4	16/406.4	16/406.4
VOL GAL/LITER	1410/5338	282/1067.6	423.8/1604	564/2135.3	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676	2821/10676
TEMP PROBES	6	6	6	6	3	3	3	3	3	3	3	3	3	3	6	6	6	6	6
GAS PROBES	3 v	3 v	3 v	3 v	3 v	3 v	3 v	3 v	3 v	2	3 v	2	2	2	3 v	3 v	3 v	3 v	3 v
TOTAL MASS SOLID WASTE pound/kg	6668/3025	6590/2989	6630/3007	6618/3002	6616/3001	6435/2919	6535/2964	6601/2994	6617/3001	6610/2998	6446/2924	6720/3048	6626/3006	6646/3015	6635/3010	6604/2996	6610/2998	6613/3000	6640/3012

* OPEN TOP HAS NO COVER. OPEN/CLOSED HAS PERIODICALLY REMOVED COVER; SEALED HAS PERMANENT COVER

v GAS PRODUCTION VOLUME MEASURED BY WET TEST METER

with Argon before collection. Steel
lids with manhole covers are provided
for 12 of the cells. These are
bolted to the cells that are in
the ground or in the high bay by
means of tabs located on the lids
themselves. An airtight seal is
accomplished by caulking.

A gasketed manhole cover is
attached to the cell cover by means
of bolts and welded into the
periphery of the hold provided for
the manhole cover.

Figure 1 shows a cross section
of a test cell. The size of the
test cell and the details of the
cross section were determined by
EPA staff to be suitable for the
purposes of this research effort.
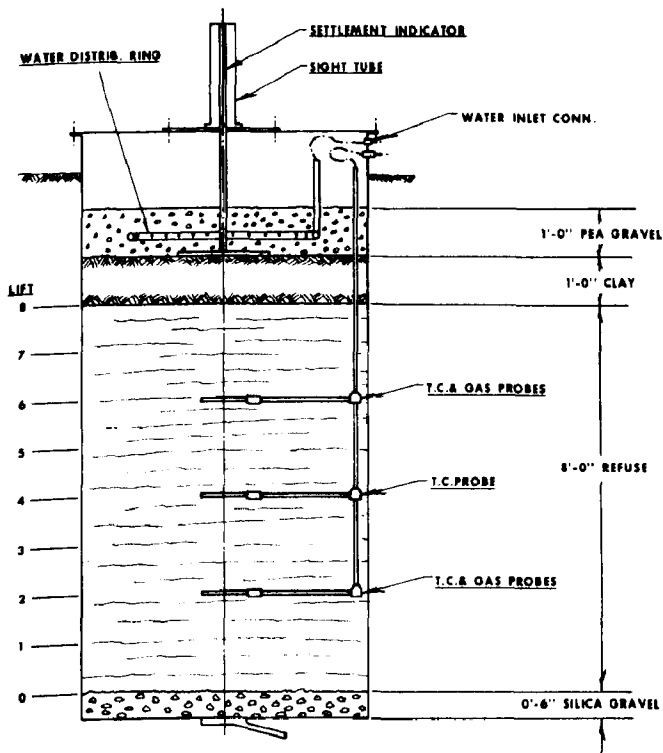
Figure 1. Cross section of test cell.

Starting from the bottom of
the cell, the first layer of material
is silica gravel. This serves as
the base for the solid waste and
allows leachate to flow through
the drain line. Silica gravel was
chosen to remove any interaction

between the leachate and the gravel
that would compromise the chemical
composition of the leachate.

Above the silica gravel are
eight lifts of solid waste, each
0.3 m (1 ft) in thickness.
Temperature probes are located at
the second, fourth, and sixth lifts,
and gas probes are located at the
second and sixth lifts and in the
pea gravel cover.

The cover for the solid waste
is clay, 0.3 m (1 ft) thick and
compacted to a predetermined density.
On top of the clay is 0.3 m of
washed pea gravel. Within the pea
gravel is the water distribution
system, which consists of a circular,
perforated polyethylene tube
connected to the outside through
a valve.

LOADING OF THE CELLS

To minimize the exposure time
of the solid waste, the cells were
loaded and sealed in 4 days. The
solid waste was placed in the cells
in eight, 0.3-m (1-ft) increments,
which approximated 407 kg (895 lb).
From this was removed an 11.4-kg
(25-lb) moisture sample and an 18.2-
kg (40-lb) separation sample.

Compaction was accomplished
using a 1,318-kg (2,900-lb) wrecking
ball supported by a 9-m (30-ft)
boom crane. Densities achieved
averaged 461 ± 12 kg/cu m (778 ±
20 lb/cu yd). The various materials
under scrutiny (i.e., sludge, etc.)
were added to all lifts except the
first. This was done to eliminate
premature leaching of the materials
present. All sludges were received
several weeks before the loading.
The sewage sludge was obtained on
a daily basis from the City of
Cincinnati. The poliovirus was
inoculated into eight expanded
aluminum baskets and into three
nylon bags containing solid waste
and added at each of three levels
within cell 15. Table 2 lists the
amounts of materials added to the

Table 2. MATERIALS ADDED TO TEST CELLS*

Material added†	Amount/lift (7-lift basis)				Total amount		Cell number
	kg	lb	liter	gal	kg	lb	
CPBS	291.3	642.1	171.3	45.26	2039.1	4494.4	14
SBPS	229.2	505.2	171.3	45.26	1604.4	3536.1	17
EW	170.1	374.9	171.3	45.26	1190.7	2624.4	12
IPW	202.9	447.4	171.3	45.26	1420.3	3131.9	13
Sewage sludge	9.7	21.4	--	--	67.97	150	5
Sewage sludge	29.2	64.3	--	--	204.4	450	6
Sewage sludge	97.2	214.3	--	--	630.4	1500	7
Calcium carbonate	12.9	28.5	--	--	90.3	200	8
Water	184.8	407.3	171.3	45.26	1293.6	2851.2	11

*Also to cell 15 were added eight aluminum baskets and three nylon packets of refuse inoculated with poliovirus at the 0.61-, 1.22-, and 1.83-m (2-, 4-, and 6-ft) level.
†CPBS=Chlorine production brine sludge; SBPS=solvent-based paint sludge; EW=electroplating waste; and IPW=inorganic paint waste.

various cells. All cells were weight and height normalized to ensure the final densities were approximately the same. Thermocouples on the outside cells and thermistors on the inside cells were installed in the appropriate lifts and monitoring of all probes was begun as soon as they were covered.

FILLING SEQUENCE

To accomplish filling of the cell in 4 days, a detailed sequence was devised that used two 454-kg (1/2-ton) pickup trucks, a special weighing platform, a sampling location, and an unusual compacting technique.

Use of two trucks allowed placing a lift every 15 to 20 minutes, or about 40 hours to fill all cells. On this basis, a double shift was planned to allow for delays such as breakdowns, rain, snow, and other potential delaying factors.

The filling operation itself began when a truckload of refuse was received at the facility and then was dumped onto the sort pad.

At this point the bags in the refuse were slit by hand, and the front-end loader mixed the refuse to provide relative uniformity of the sample. One of the pickup trucks was then driven onto a specially prepared weighing platform, consisting of four 908-kg (2,000-lb) capacity scales, one located at each wheel. A tare weight of the truck was taken and recorded. The front-end loader then placed enough refuse into the truck to equal 407 kg (895 lb). A gross weight on the truck was taken and recorded. If the net weight was within 4.5 kg (19 lb) of that needed, the truck was driven off to a sampling station and a second truck was cycled through.

At the sampling station, an 11.4-kg (25-lb) moisture sample and an 18.2-kg (40-lb) separation sample were removed. The weights were recorded, the bags were tagged, and removed for separation or storage at 4 C (41 F). All sample weights were within 0.45 kg (1 lb).

The truck was then driven to the cell area where the refuse was placed in the cell.

This was done by means of a

specially designed dumping device. Roller skate trays were anchored to the truck bed by tying one end to the bed. On top of the rollers were three plywood sheets hinged together to provide a flexible hard surface for a canvas cover. The cover was large enough to extend over the bed and the sides of the truck. The solid waste was loaded into the truck and was driven to the cell. At the cell, four men grabbed the canvas and pulled the load of solid waste out of the truck. After the refuse was in the cell, it was leveled out so that it would be reasonably flat for compacting.

CHARACTERIZING THE SOLID WASTE

Categorization of the solid waste was accomplished on each lift for every cell with the waste being separated into 11 categories. Table 3 shows the 11 sort categories and an average percent of each category present. The raw data are available upon request.

Microbiological analysis on both the solid waste and leachate produced to date is completed. To summarize the data from the

sorts, we have seen survival of total coliform, fecal coliform, and fecal streptococci in all of the sort categories. This is because of the inherent mixing of refuse that occurs both in the home and as the solid waste is transported. The largest concentration of total coliforms and fecal coliforms occurred in fines, garden waste, textiles, and paper, in that order.

Fecal streptococci predominated in ash, rock, dirt, fines, and garden waste. These results bear out the mixing theory quite well.

Microbiological assay of the leachate has generated some interesting data. The initial leachate collected indicated high levels (generally 100,000 colonies/100 ml) of fecal coliforms, and fecal streptococcus that varied tremendously. The initial leachate that was collected is believed to consist mainly of the squeezings from the compactive effort upon the refuse. Analysis of the leachates which followed have shown an increasing dieoff of coliforms in leachate as the cell ages. Streptococcus remained quite high for several months but is now beginning to indicate a decreasing number of colonies. A comparison

Table 3. CATEGORIZATION SUMMARY

Category	Average percent over fifteen cells by lift							
	1	2	3	4	5	6	7	8
Food	7.81	8.62	6.69	5.91	9.19	5.47	9.64	6.93
Garden	0.30	15.83	22.34	14.72	19.75	23.33	9.25	17.04
Paper	44.42	43.24	36.20	41.27	39.06	36.67	45.38	38.01
Plastic, rubber, leather	6.40	5.44	5.36	7.25	6.38	6.78	7.42	7.09
Textiles	2.77	2.91	3.10	5.83	3.28	3.04	5.18	7.38
Wood	0.65	0.85	0.71	0.54	0.89	0.79	1.60	0.84
Metal	10.12	8.01	9.04	7.54	6.95	8.99	7.77	7.91
Glass	6.79	6.85	11.39	5.77	7.02	7.37	5.88	8.28
Ash, rock, dirt	2.53	1.23	1.90	4.21	4.34	1.96	2.02	1.25
Diapers	1.89	0.97	1.14	4.66	1.23	1.25	1.99	1.09
Fines	4.25	2.06	2.04	2.28	1.91	4.34	3.33	4.16

of the most probable number (MPN) technique versus the membrane filtration technique shows much better recovery on the MPN. Recoveries have varied by as much as 100-fold. Unpublished reports have indicated increased recoveries by the addition of sodium ethylene-diaminetetracetrate (Na_2 EDTA) to the leachates. We have tried the recommended Na_2 EDTA along with Na_5EDTA and have seen no increase in recovery as yet.

As metals concentrations increase in the leachates, however, the EDTA may aid our recoveries. Additional bacteriological identification of the leachates has shown the presence of Pseudomonas, Enterobacter, Proteus, and various molds. An assay for salmonella has proven negative. No antagonistic effects can be seen from the hazardous waste cells.

A complete listing of chemical analyses is shown in Table 4.

A complete chemical analysis of the hazardous waste is shown in Table 5.

SOME INITIAL DATA

Since the cells have been filled for less than 4 months, data from the experiment are limited. Temperature data on cells 1-8 and 11-15 have been plotted, and leachate and gas produced thus far have been analyzed.

The cells are in the early stages of methane generation, having just completed the change from aerobic to anaerobic condition.

TEMPERATURE DATA

Figure 2 is a plot of temperature versus time. All temperature plots are illustrated as functions of hours after placement of lift number 1. The data for cells 1-4 indicate that the maximum

temperature was reached about 45 to 60 hr after placement. Cells 1-4 and 15 received solid waste only, and all exhibit comparable behavior. The temperature increases as:

$$T (t)/_{t>p} \alpha \ T_0 \ exp \ [at \ exp \ (At)]$$

After peaking, the temperature decreases as:

$$T (t)/_{t>p} \alpha \ T \ [\ 1-exp \ (-bt)]$$

or approximately in a cooling curve of a hot body dissipating heat energy to a heat sink.

Cells 5, 6, and 7 received sewage sludge in increasing amounts (See tables). Cells 5 and 6 have similar behavior, reaching a peak temperature at about 50 to 55 hr. After peaking, the temperature dips, rises slightly, then slowly decreases. Cell 7, on the other hand, does not reach a peak temperature until about 95 hr, after which there is a slow decrease in temperature.

Reasons for this, in terms of the conditions in the cells, are not clear at this time. Since cell 7 received the largest amount of sludge, the conditions for reaching maximum were slowed by the presence of 680 kg of sludge. Detailed analysis is underway.

Temperature data from cells 8 and 11-14 indicate the curves suggested by the data points appear to be very similar. Peak temperature was reached at about 95 hr in 8 and 11. Cell 12 peaked at 85 hr; cell 13, at 80 hr; and cell 14, at 50-60 hr. Differences in cells 12-14 clearly are a result of the various materials added, whereas cells 8 and 11 are not sensitive to the materials.

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Table 4. ANALYTICAL DATA

Material*	Analysis	Cells analyzed	Type of analysis
SW, HW, SS	pH	All	Electrometric
SW, HW, SS	Conductivity	All	Conductivity bridge
L, SW, HW, SS	Acidity	All	Potentiometric titration
L, SW, HW, SS	Alkalinity	All	Potentiometric titration
SW, HW, SS	COD†	All	Reflux/titration
SW, HW, SS	Hardness	All	EDTA titration
SW, HW, SS	Total $PO_4^=$	All	Persulfate digestion/ ascorbic acid titration
SW, HW, SS	NH_3- nitrogen	All	Specific ion electrode
SW, HW, SS	Organic nitrogen	All	Kjeldahl digestion
SW, HW, SS	Nitrate nitrogen	All	Specific ion electrode
SW, HW, SS	Organic acids∇	All	Partition chromatography
SW, HW, SS	Total solids	All	Gravimetric
SW, HW, SS	Dissolved solids	All	Filtration/gravimetric
SW, HW, SS	Sulfate	All	Gravimetric
SW, HW, SS	Chloride	All	Specific ion electrode
SW, HW, SS	Sulfide	All	Specific ion electrode
SW, HW, SS	Cu, Zn, Ni, Fe	All	Atomic absorption
SW, HW, SS	Ca, Mg, Mn, K, Na	All	Atomic absorption
L, SW, SS	BOD	All	D.O. probe/incubation
L, HW	Pb, Hg, Be, Se, Cr	11-14, 17+4	Atomic absorption
L, HW	CN	11-14, 17+4	Distillation/titration
L, HW	Asbestos	4,12,14	Electron microscope/x-ray
L, HW, SS	Chlorinated hydro-carbons	12-14 + 4	GC/electron capture
SW, SS	Moisture	NA	Drying oven
SW	Ash		Muffle furnace
SW, SS	Carbon		Gravimetric
SW	Lipids		Soxhlet extraction
SW	Sugar		Reflux/titration
SW	Starch		Anthrone/spectrometric
SW	Protein		Calculation
SW	Crude fiber		Ether extraction/gravi-metric
HW, SW, SS	Water solubles		Gravimetric

*L=leachate; SW=solid waste; HW=hazardous waste; SS=sewage sludge.

†COD analyses will be done on both over-dried and air-dried solid waste samples.

∇Organic acids may be cancelled on solid waste samples.

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Table 5. HAZARDOUS WASTE ANALYTICAL DATA

| Measurement	Solvent based paint sludge, cell 17	Chlorine production brine sludge, cell 14	Electro- plating waste, cell 12	Inorganic pigment waste, cell 13	Petroleum waste
Total solids, %	75.25	75.89	20.47	48.25	21.00
Total volatile solids, %	55.31	1.17	8.98	25.25	31.00
Moisture, %	24.75	24.11	79.53	51.75	79.00
Cr, mg/liter	75	5.0	1.56	0.5	125
Ni, mg/liter	0.5	65	35	10	23
Cu, mg/liter	2.0	125	100	110	3500
Fe, mg/liter	150	2000	1.37	1000	5563
As, mg/liter	12.8	14.5	460	3.4	1.0
Be, mg/liter	0.0	0.0	0.25	20.5	4.8
Se, mg/liter	7.6	16.5	4.5	16.0	26.0
Cd, mg/liter	0.5	0.7	38.5	10.5	0.5
Cyanide, mg/liter	12.8	14.5	460	3.4	1.0
Pb, mg/liter	*	*	*	*	182
Cl, mg/liter	7500	20.0	1.35	10.0	2.35
Asbestos, fibers/100 g	9	110	23	45	3

*Not completed yet.
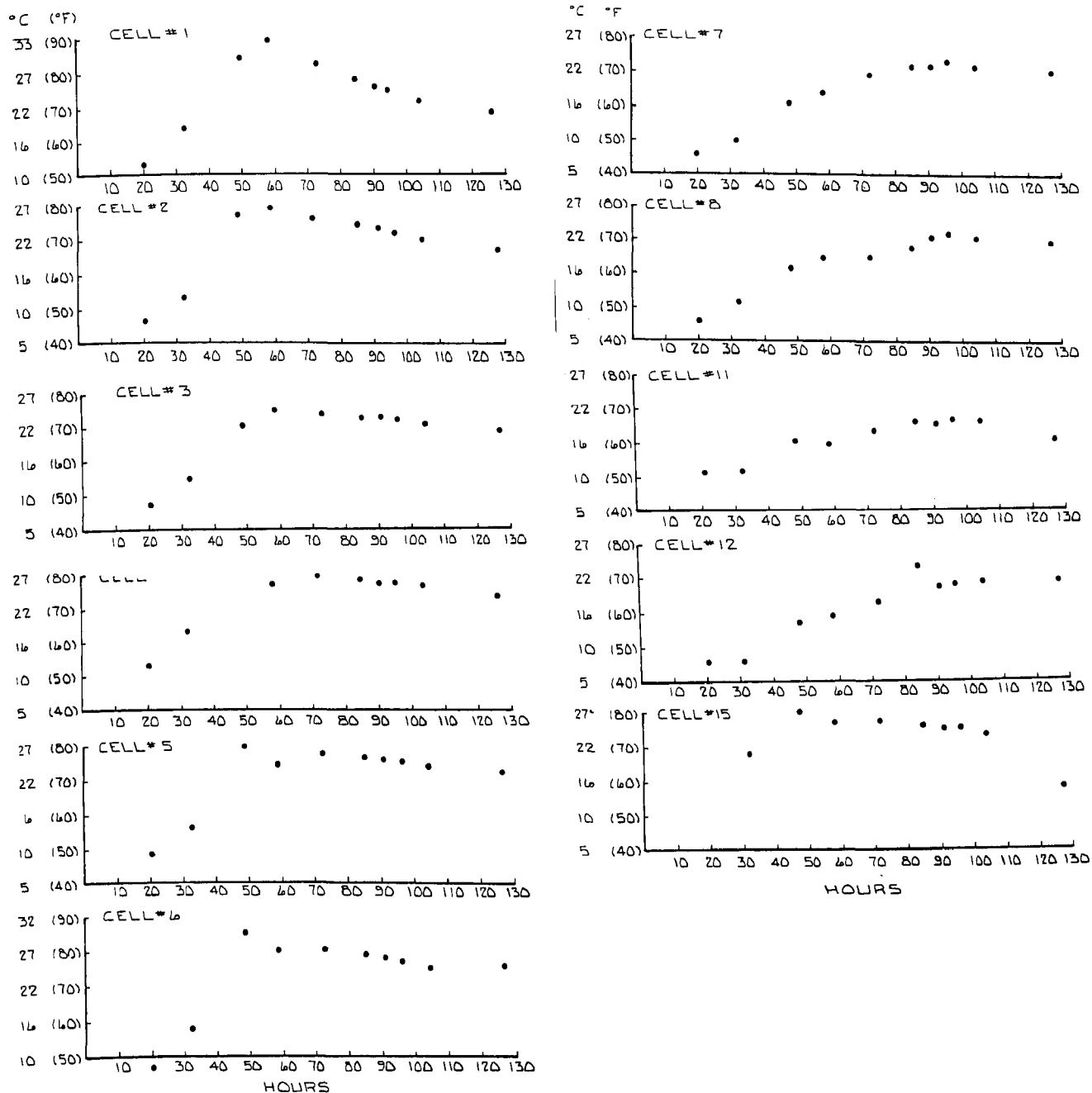
Figure 2. Temperature of lowest test cell versus time elapsed after refuse placed in cell.

LEACHATE MIGRATION THROUGH SELECTED CLAYS

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and
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INTRODUCTION

An investigation of the use of clay minerals to limit the pollution of waters by landfill leachates is being conducted at the Illinois State Geological Survey; the study is being supported in part by a U.S. Environmental Protection Agency (EPA) contract. The goal of the project is to evaluate the potential use of clay minerals as liners for sanitary landfills to prevent or mitigate pollution of ground and surface waters by liquid effluents from solid wastes. The results of this investigation will also find application in the land disposal of industrial and power plant wastes.

The research is being conducted in the Environmental Geology Laboratory of the Illinois Geological Survey. The laboratory apparatus consists of 44 laboratory columns containing clay minerals and mixtures of clay minerals through which leachate was passed. The three clay minerals used in this study are kaolinite, montmorillonite, and illite. The columns were constructed to simulate the slow, saturated, anaerobic flow of refuse effluent as it is thought to occur at the bottom of a landfill disposal site.

The refuse leachate used in this study was collected from the Du Page County sanitary landfill near Chicago. The leachate was passed through two series of columns.

The first series was leached with sterilized effluent and the second with natural (microbiologically active) leachate. The mechanisms involved in attenuating pollutants, including microbial activity, by the three clay minerals were evaluated. The first step in the evaluation was a careful chemical and physical characterization of the original column contents and the influent leachate. Analyses were made for the following: Na, K, Ca, Mg, Al, Zn, Pb, Cd, Hg, Fe, Mn, NH_4, B, Si, Cl, chemical oxygen demand (COD), pH, Eh, and permeability. The original leachate characterization also included analysis for Cu, Ni, Cr, As, S, PO_4, organic acids, carbonyls, and carbohydrates. The clays were characterized for surface area, cation exchange capacity (CEC), exchangeable cations, total elemental content, and bulk density.

The second step was to pass the leachate through the columns for periods of time up to 10 months. During this time effluents from each column were periodically collected and measurements were made for the following: Na, K, Ca, Mg, Al, Zn, Pb, Cd, Hg, Fe, Mn, NH_4, B, Si, Cl, COD, Eh, pH, and permeability.

Finally, after leaching was completed, the columns were sectioned and the contents analyzed to determine the vertical distribution of chemical constituents and particle sizes in each column.

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All of the leachate data are being statistically evaluated, and predictive equations are being constructed for estimating the capacity of earth materials of known clay mineral composition to attenuate pollutants. In addition, a series of separate studies on the capacity of clays to adsorb eight hazardous elements (Pb, Cd, Zn, Cu, Cr, As, Se, and Hg) is being performed. From these studies, adsorption isotherms for kaolinite and montmorillonite are being constructed to obtain maximum adsorption capacities under various pH and ionic competition conditions.

RESULTS OBTAINED TO DATE FROM COLUMN-LEACHING STUDY

Permeability Measurements

Hydrologic gradient and flow-rate readings were collected from the columns for the 10-month period from February 4, 1974, to December 20, 1974. The data have been statistically evaluated, and they indicate that a significant ($.01$ level) decrease in the permeability of the columns occurred during the experiment. Microbiologically active columns had significantly larger ($.05$ level) permeability reductions than did sterile columns.

Further statistical evaluation is at present being carried out to determine the effect of clay type, clay percentage, and amount of clay migration on the observed permeability reductions.

Chemical Data

Chemical analysis of the soluble fraction of the column effluents has been completed. It allows us to make some tentative conclusions and attenuation rankings of the clays even though final conclusions must await completion of analysis of the "suspended" fraction of effluent and the column section samples.

The three clay minerals can be ranked according to their attenuating capacity:

montmorillonite > illite > kaolinite.

Montmorillonite attenuates pollutants approximately four times better than illite and five times better than kaolinite. These ratios are nearly identical with the cation exchange capacity ratios for the three clays. The ratios of the surface area of montmorillonite to the surface areas of illite and kaolinite are 1.3 and 2.5, respectively. These data suggest that surface area is not the property of the clays that is responsible for attenuation but rather that the cation exchange capacity is probably the principal attenuating property.

These individual chemical constituents can be ranked according to their relative degree of attenuation by the three clays as follows:

$Cl < COD < Na < NH_4 < K < Mg < Si < Cd < Zn < Pb \approx Hg$.

The elements Ca, Fe, and Mn were not attenuated by the clays, but rather were found in substantially higher concentrations in the column effluents than in the influent leachate. The clays used in the experiment were substantially Ca-saturated with respect to their exchangeable cations. It is presumed that the observed flush of Ca was due to exchange with the high concentrations of other cations found in the leachate. The Fe and Mn flush is presumed to be due to the reduction of oxidized Fe and Mn compounds on the clay surfaces by the anaerobic refuse leachate to more soluble reduced species. It should be noted that an increase in the concentrations of these three elements has been observed in the field during monitoring of the groundwater chemistry around landfill sites, and this increase has been dubbed the "hardness halo".

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The constituents Al, Cu, Ni, Cr, As, S, and PO were found in such low concentrations in the Du Page leachate that no attenuation order could be determined.

The elements for which an attenuation order could be determined can also be ranked by relative degree of attenuation as follows:

High   Hg, Pb, Zn, Cd
Moderate   Si, Mg, K, NH4
Low   Na, COD, Cl
No attenuation Ca, Fe, Mn

Measurement of the effluent concentrations from sand columns, which contain no measurable cation exchange capacity, indicates no Pb or Hg and markedly reduced concentrations of Zn and Cd eluted from the columns. These data indicate that precipitation of heavy metal hydroxides and carbonates is an important attenuation mechanism. This conclusion is further verified by studies of the effect of pH on adsorption.

A tentative conclusion from the column-leaching study thus far is that the principal mechanisms affecting pollutant attenuation by clay minerals in landfill leachates are: a) microbial reduction of permeability, b) the cation exchange capacity of the clay, and c) the effect of pH on the formation of heavy metal hydroxide and carbonate precipitates.

HAZARDOUS ELEMENTS ADSORPTION STUDIES

A hazardous elements adsorption project, partly supported by the EPA as an extension of the landfill leachate project, is also being conducted at the Survey. The goal of the second project is to determine the adsorption capacity of the clay minerals kaolinite and montmorillonite for eight hazardous elements-- Hg, Pb, Cd, Zn, Cr, Cu, As, and Se. The adsorption properties of the clays are being studied over

a wide range of solution concentrations, competing ion matrices, and pH values.

The adsorption of Pb, Zn, Cr^{+3}, Cd, Cr^{+6}, and Se from landfill leachate by kaolinite and montmorillonite clay at pH 5.0 and 25 C has been studied. Characterization of As and Hg adsorption, as well as further studies of the above elements, is currently in progress.

Element adsorption is measured from three solution matrices: pure water, inorganic salt solutions similar to leachate in ionic composition, and landfill leachate from Du Page County. In addition, Pb adsorption has also been studied from Blackwell Forest Preserve leachate. The nitrate salt of a metal is added in various concentrations to each of the three solution matrices, and the amounts adsorbed are measured. The effect of varying pH on the amounts of Pb, Zn, Cu, Se, and Cr^{+6} adsorbed has also been studied.

The inorganic leachate and the natural leachate contain all of the same major cations and anions, and in approximately the same concentrations. Any observed differences between the adsorption of the metals from the inorganic leachate solution and that from the natural leachate solution are attributed to the presence of the organic component of the natural leachate. The amount of metal ion adsorbed from the pure water solution is considered to be the maximum that can be adsorbed under these experimental conditions.

RESULTS OBTAINED TO DATE FROM ADSORPTION STUDIES

The adsorption characteristics of the seven elements investigated to date are quite similar in many respects and can be summarized by the following description. The amount of metal ion adsorbed by the clay was about 70% to 80% less

from leachate than from pure aqueous solutions. (It is also interesting to note here that only about half as much Pb was adsorbed from Blackwell leachate as from Du Page leachate.) This decrease in the amount of heavy metal sorption has the environmental consequence that the metal ions will migrate farther in landfill leachate than in pure water solutions of the metal. The results further show that metal ions can be expected to migrate about twice as far in Blackwell leachate as they would in Du Page leachate.

The most important factor affecting the amount of the metal removed from solution was the pH of the solution. The five cations Cr, Cu, Pb, Cd, and Zn showed a marked increase in adsorption with increasing pH in the range from pH 2 to about pH 6. This increase in adsorption is consistent with the increase in the pH-dependent cation exchange capacity of the clays and with the formation of metal-hydroxyl complex ions known to occur in this pH range. Blank (no clay) solutions carried through the experiments indicated that the formation of insoluble carbonate and hydroxide compounds was initiated between pH values of 5.5 to 7.5, depending on the element and its concentration.

The metals Se and Cr^{+6} followed a reverse trend with respect to pH. Their adsorption increased as the pH was lowered. Since Se is known to exist in solution as the SeO_4^{-2} anion and Cr^{+6} as the $Cr_2O_7^{-2}$ anion at low pH values, this behavior is consistent with an anion exchange mechanism.

The adsorption maximums estimated from the isotherms of the five metal cations adsorbed from solutions of the pure nitrate salt of the metal were found to correspond closely to the cation exchange capacity of the clay being used as the adsorbent. This close correspondence is strong evidence that cation exchange is the principal attenuation mechanism at pH values that preclude precipitation.

Whereas the metal cations adsorbed to a maximum value that could be estimated by the CEC of the clay in pure solutions, the amounts adsorbed from landfill leachate, at constant pH, varied widely. This wide variation is presumed to be due to the relative affinity of each metal ion for exchange sites when competing with the high concentrations of other cations present in leachate. A tentative ranking to indicate the relative adsorption affinity for kaolinite at pH 5 of each of the seven elements studied from Du Page leachate is given as follows:

$$Cr^{+3} > Cu \approx Pb > Cd > Zn > Cr^{+6} > Se.$$

The adsorption isotherm data allow us to fit to the equations that yield reliable predictions of the amount of metal ion removed from a solution of known concentration and pH. A computer simulation model has been written to predict metal ion migration through clay columns. The model is at present being tested on Pb migration through clay-sand columns.

The tentative conclusion of the adsorption study is that the most important factors affecting the prediction of a given metal ion's migration under a solid waste disposal site are the pH of the solution, the CEC of the clay, and the ionic composition of the solution matrix.

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ORGANIC POLLUTANTS CONTRIBUTED TO GROUNDWATER BY A LANDFILL

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ABSTRACT

Organic compounds contributed to groundwater by a landfill containing refuse deposited below or near the water were investigated. Groundwater from a well within the landfill and a control well was sampled by modified low-flow carbon adsorption procedures incorporating all-glass/Teflon systems to preclude introduction of extraneous organics. Column chromatography, solubility separation, and gas chromatography/mass spectrometry were employed for separation, identification, and quantitation of individual compounds in organic extracts. The groundwater was shown to contain low levels of many undersirable organic chemicals leached from the landfill. More than 40 compounds were identified, most of which were chemicals commonly employed in industry for manufacturing many domestic and commercial products. The source of these compounds was apparently manufactured products discarded in the landfill, since it had not received appreciable wastes from industrial operations. The compounds identified were believed to be substances leached very slowly from refuse and/or transported away from the landfill very slowly because of adsorption on aquifer solids. Potential long-term pollution of groundwater by industrial organic chemicals from landfills may be indicated by this work.

INTRODUCTION

Countless tons of solid waste have been deposited within the upper layers of the earth's crust at land disposal sites throughout the United States. Within recent years, it has become increasingly apparent that this waste poses a potentially serious threat to the quality of the Nation's groundwater. This is especially so because of the past tendency to locate dumps and landfills in low-lying areas where the waste is in contact with or in close proximity to groundwater. In a recent study of groundwater problems in 11 northeastern states, Miller et al. (1) presented information on 60 cases in which landfills were pinpointed as sources of groundwater pollution and noted that numerous additional cases of a similar nature were probably present in the region. That this situation is not unique to the northeast is indicated by a number of reports from other regions, including those of Walker (2), Anderson and Dornbush (3), Fuhriman and Barton (4), Scalf et al. (5), and the California State Department of Water Resources (6).

Among the many substances that might possibily enter groundwater

by leaching of solid waste are
potentially hazardous organic
compounds, particularly synthetic
organics that may decompose slowly
or be essentially nondegradable
in the subsurface environment.
Until recently, land disposal sites,
regardless of location and design,
have been used for deposition of
practically every kind of solid
waste, including hazardous
industrial, hospital, and agri-
cultural wastes--such as solvents,
plasticizers, phenolic compounds,
and pesticides. Also, the bulk
of waste from great quantities of
products manufactured for domestic
and commercial use has been and
continues to be placed in land
disposal sites, and these products
may contain or have been produced
from a vast array of potentially
hazardous organic chemicals. That
pollution of groundwater by organic
matter leached from solid waste
in land disposal sites can and does
occur has been well documented (3,
6, 7), but practically no information
has previously been developed
concerning the nature of the organic
pollutants involved. Clearly, such
information is required for realistic
and comprehensive evaluation of
the threat to groundwater quality
posed by land disposal of solid
waste. The investigation reported
in this paper comprised an effort
to provide such information by
identifying specific organic pol-
lutants contributed to groundwater
by a landfill.

METHODS AND RESULTS

Site of the Study

The landfill chosen for this
study was located at a land disposal
site approximately 1 mile south
of Norman, Oklahoma. This site,
as shown in Figure 1, lies on the
north bank of the South Canadian
River in an area of moderately to
highly permeable soil consisting
of quaternary recent alluvium com-
posed of silt, sand, clay, gravel,
and dune sand.
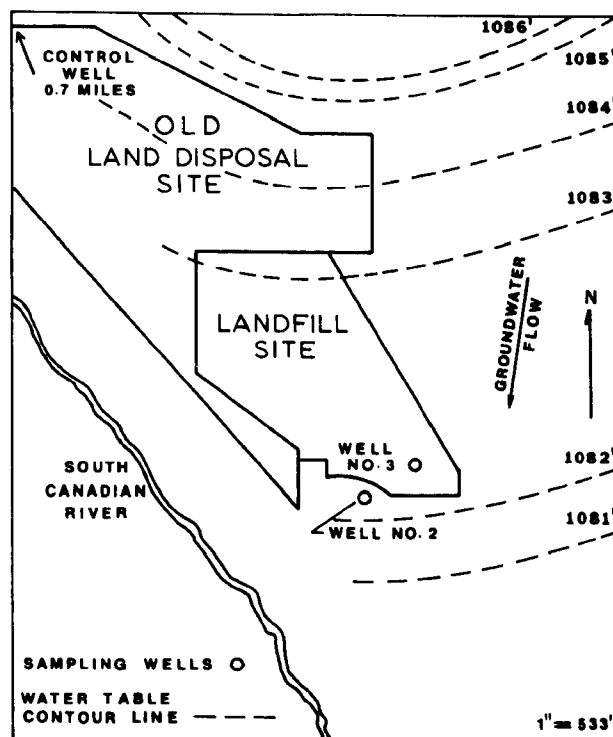
Figure 1. Land disposal site near
Norman, Oklahoma.

The depth of the alluvium, which
lies over a 300-ft impervious layer
of dense clay and chert gravel known
locally as the "red bed," varies
from 35 to 40 ft. The water table
throughout the site is normally
quite high, averaging from 2 to
5 ft below the original land surface
in areas adjacent to the river.
The direction of groundwater flow
was previously determined to be
approximately 7 degrees west of
south, essentially normal to the
water table contours (8). The flow
of the river in the area is very
low except during periods of high
rainfall.

For 38 years, from 1922 to
1960, the City of Norman operated
the site as a dump in which there
were no restrictions concerning
the type of material accepted and
in which open burning was practiced.
In 1960, a trench-type operation
was begun in the area designed as
"Landfill Site" in Figure 1. Layers

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of refuse were bulldozed into trenches which had been dug by a dragline during commercial sand production. Because of the shallow depth of groundwater in the area, large quantities of refuse were placed below the water table in most of these trenches. The deposited refuse was eventually covered with approximately 6 in. of relatively permeable fine sand obtained in the area. In 1972, because of new state solid waste legislation, a modified area fill operation was initiated in which solid waste was deposited at least 2 ft above the water table and covered at least weekly. At present, deposited waste is covered almost daily and the site is classified as a sanitary landfill.

Sampling of Organics in Groundwater

Groundwaters from a well located in the southeast part of the landfill (designated well No. 3, Figure 1) and from a control well (located approximately 0.7 mile from the upstream edge of the landfill and 1.3 miles northwest of well No. 3) were the waters subjected to most intensive analysis in this investigation. Well No. 3, drilled in November 1972, was cased to a depth of 32 ft and perforated in the lower 12 ft of casing. It contained 17 ft of standing water at the time of sampling and was expected to yield groundwater contaminated by solid waste since it passed through a layer of refuse 20 to 22 ft thick. The control well, drilled in October 1973, was cased to a depth of 42 ft and perforated in the lower 12 ft. It contained 37 ft of standing water and was expected to yield water unaffected by the landfill because of its location. Both wells were carefully drilled and cased and were thoroughly bailed and pumped after construction. The wells were also thoroughly pumped before sampling to remove standing water and allow fresh formation water to enter the well bores.

The two wells were sampled simultaneously by identical procedures so that comparison of organic matter from the control and landfill wells would clearly reveal the extent and nature of organic contamination of groundwater by the landfill and provide a guide for selection of compounds that should receive priority attention in identification studies. The sampling procedures employed during this work incorporated a modified version of the low-flow carbon adsorption method (9, 10), with the groundwater being pumped from the saturated zone directly through columns containing activated carbon to adsorb and recover the organic compounds. Single-piece all-glass columns fabricated from 3-in.-diameter borosilicate tubing were used. They were packed with 18 in. of 30-mesh activated carbon (Nuchar C-190, Plus 30, Hebert Chemical Co., St. Bernard, Ohio) held in place by solvent-washed glass-wool plugs. For sampling, a packed column was placed in a vertical position at the top of the casing of each well. Suitable lengths of Teflon tubing were attached to the bottom inlets of the columns and extended down the well shafts into the saturated zone. The groundwater was then pumped up the tubing and through the carbon columns by variable-speed peristaltic-type pumps ("Masterflex" 7545 Variable Speed Drive with 7014 pump head, Cole-Parmer Instrument Co., Chicago, Ill.) attached by Teflon tubing to the outlet (downstream) ends of the columns. Power was provided to the pumps at the field sites by portable gasoline-operated 1500-watt AC generators. The variable speed pumps permitted sustained pumping of groundwater from the water table through the carbon adsorption columns at accurately controlled, constant, low-flow rates. Flow rates and quantities of water sampled were verified by collecting discharge water from the pumps and measuring the volumes. A sampling system in operation is shown graphically in Figure 2.
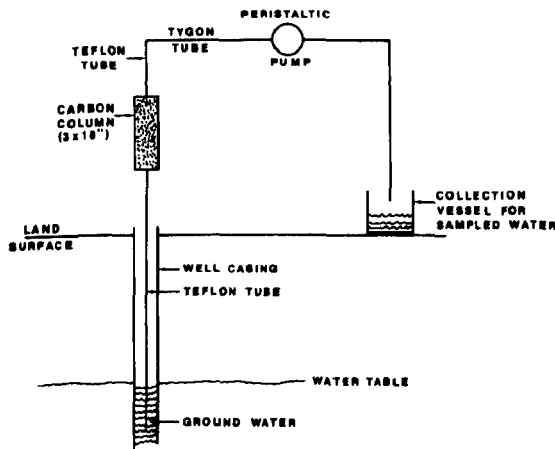
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Figure 2. Groundwater sampling
system.

Groundwater was pumped for
each of the two wells through
essentially identical carbon columns
for 126 hr at a rate of 100 ml/min.
In this manner, 200 gal (757 liters)
of water was sampled at each
location. Use of the sampling
systems consisting only of glass
and Teflon from the saturated zone
to the outlet of the carbon
adsorption column and placement
of the pump on the downstream side
of the column virtually precluded
introduction of organic contaminants
during the sampling operation.

Desorption of the Sampled Organic Material

Upon completion of sampling,
the carbon columns containing the
adsorbed organics from the water
from well No. 3 and the control
well were drained to remove excess
water, sealed with solvent-washed
aluminum foil, and transported
immediately to the laboratory for
processing. A third carbon column,
which had been prepared identically
and at the same time as the columns
used for sampling but which did
not have any water passed through
it, was processed with the sampling
columns to serve as a blank.

The glass columns were scored
with a glass saw and then were
carefully broken open to permit
removal of the carbon in a special
carbon-handling room designed to
minimize the potential for
contamination of the carbon during
processing. The carbon was carefully
transferred to Pyrex glass dishes
and dried at approximately 40 C
for 48 hr under a gentle flow of
clean air in a Precision-Freas
mechanical convection oven (Model
845, Precision Scientific Company,
Chicago, Ill.). The air inlet of
the oven was equipped with a carbon
filter to prevent contamination
from the atmosphere.

The dried carbon was transferred
to 2200-ml modified Soxhlet
extractors and extracted for 48
hr with chloroform. The carbon
chloroform extracts obtained from
the blank carbon and the carbon
employed in sampling well no. 3
and the control well were designated
CCEB, CCE3II, and CCEC, respectively.
These extracts were filtered through
solvent-extracted glass-fiber filters
to remove carbon fines and then
vacuum concentrated in rotary
evaporators at temperatures not
exceeding 27 C to a final volume
of 3 ml each.

The chloroform-extracted carbon
samples were dried in the Soxhlet
extractors by passing a gentle
stream of warm, dry air through
the extractions chambers via the
siphon tubes for 20 hr. The carbon
was then extracted for 32 hr with
pure ethanol, and the carbon alcohol
extracts (CAE's) were filtered and
concentrated in the same manner
as the CCE's. However, it was
necessary to filter the CAE's through
extracted glass-fiber filters when
volumes of about 10 ml had been
attained to remove precipitated
material. These precipitates,
together with solid material that
had precipitated on the flask walls
during evaporation, were dried and
weighed. The filtered CAE's were
then further evaporated to the
following final volumes: 4.0 ml
for CAE3II, from well No. 3; 2.0
ml for CAEC, from the control well;
and, 1.0 ml for CAEB, from the
blank.

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Table 1. WEIGHTS OF CARBON CHLOROFORM AND CARBON ALCOHOL EXTRACTS

Source	Weight of CCE Total, mg	mg/ℓ	Weight of CAE Total, mg	mg/ℓ	Weight of CCE + CAE Total, mg	mg/ℓ
Well No. 3	304.5	0.402	1219.1	1.610	1523.6	2.013
Control well	11.9	0.016	314.2	0.415	326.1	0.431
Control blank	2.6	0.003*	262.5	0.347*	264.8	0.350*

*Calculated as if this carbon had actually been employed for sampling of 757 of water.

Comparison of CCE's and CAE's

Visual comparison of the various CCE's and CAE's showed CCE3II, prepared from groundwater from well No. 3, to be deep yellow, whereas CCEC, from the control well water, was light yellow, and CCEB, from the carbon blank, was practically colorless. Similarly, CAE3II was deep yellow-orange, CAEC was yellow, and CAEB was pale yellow. Both CCE3II and CAE3II were very odorous, and the control and blank CCE's and CAE's were essentially odorless.

Aliquots of each of the concentrated CCE's and CAE's were carefully evaporated to dryness in tared foil cups to determine the weights of soluble material dissolved in the concentrates. Total weights of the CCE's and CAE's were calculated from these weights and the weights of material that had precipitated during preparation of the concentrated extracts. The data obtained, presented in Table 1 both in terms of total weights and weights per liter of sampled water, showed CCE3II to contain approximately 25 times as much material as CCEC and more than 100 times the weight of material in CCEB; these data also revealed CAE3II to contain about four times the material contained by CAEC, the control, and about six times the material contained by CAEB,

the blank. Hence, the presence of much greater quantities of organic constituents in the groundwater in the locale of the Norman landfill than in groundwater from the same aquifer approximately 1 mile upstream from the landfill perimeter was clearly indicated.

The various CCE's and CAE's were next compared by gas liquid chromatography. Figures 3 through 6 show chromatograms obtained by chromatographing, under identical conditions, aliquots of CCE3II and CCEC representing 190 ml of groundwater from well no. 3 and the control well, respectively, and aliquots of CAE3II and CAEC representing 380 ml of groundwater from these wells. Analogous chromatography of suitable aliquots of CCEB and CAEB from the carbon blank produced chromatograms (not shown here) that were very similar to those obtained for CCEC and CAEB. These chromatographic comparisons revealed that the groundwater from the landfill contained a complex array of organic compounds that were readily amenable to gas chromatography and that were either not present or were present in very much less quantity in groundwater not subject to the influence of the landfill. It was obvious that practically all of the major organic components of CCE3II and CAE3II, which were sufficiently volatile
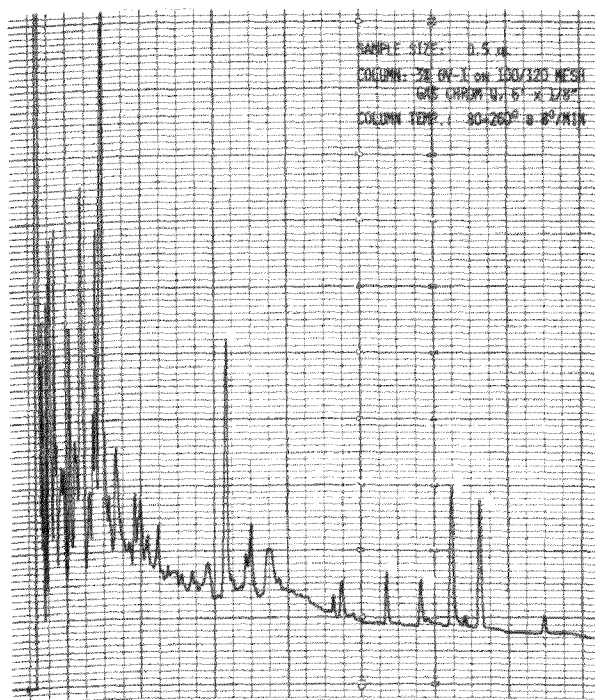
Figure 3. Gas chromatogram of CCE3II, landfill well.
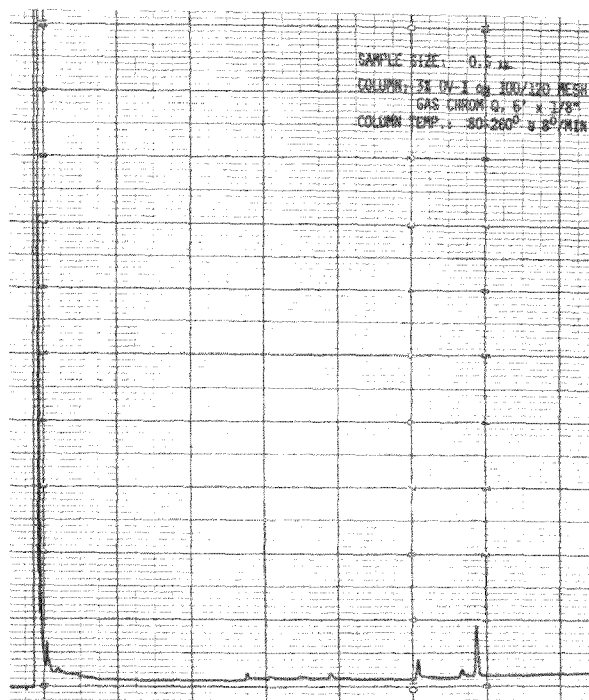

Figure 4. Gas chromatogram of CCEC, control well.
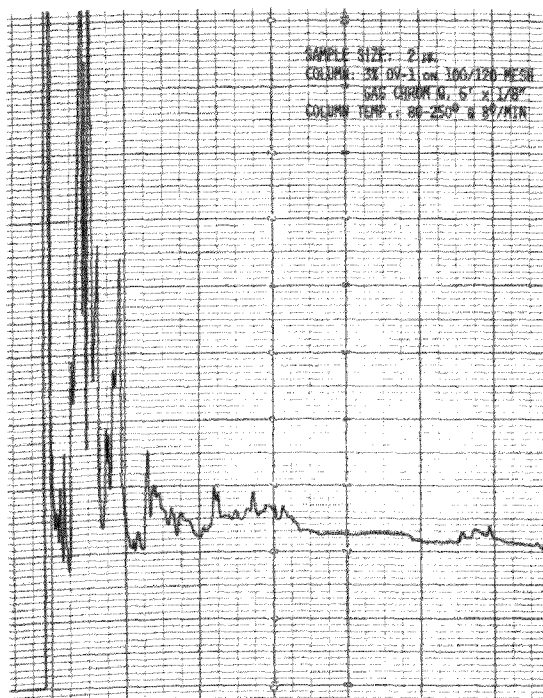
Figure 5. Gas chromatogram of CAE3II, landfill well.
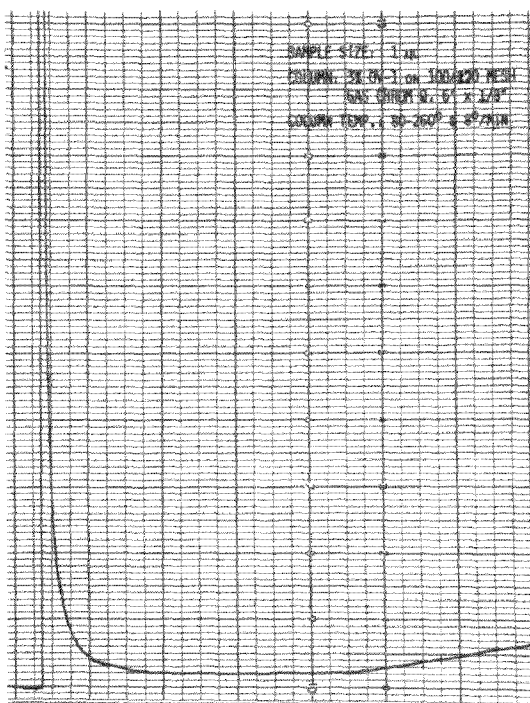
Figure 6. Gas chromatogram of CAEC, control well.

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for gas chromatography, had been contributed to the groundwater by the landfill, and identification of any of these compounds would, therefore, be helpful in elucidating the effect of the landfill on groundwater.

Further Fractionation of the CCE and CAE from the Landfill Well

CCE3II was subjected to liquid-solid chromatography on micro silica gel columns to obtain fractions of less complexity for further study. Initially, a 7- by 76-mm column of 100/200 mesh Silicar CC-7 (Mallinckrodt Chemical Works, St. Louis, Mo.) was prepared by dry packing and subsequent washing with hexane to wet the column and determine the void volume. A 1.5-ml aliquot of CCE3II was charged to the column by carefully adsorbing it on a small portion of Silicar CC-7, suspending the Silicar containing the adsorbed CCE in a small quantity of hexane, and carefully placing this suspension on top of the column. The column was eluted successively with 8 ml each of hexane, benzene, and chloroform-methanol (1:1), followed by 5 ml of methanol. The fractions of 2 or 3 ml each were collected after a volume of hexane equivalent to the void volume had been eluted.

These fractions were designated CCE3II-SG1 through 10, as shown in Table 2.

The 10 fractions from the silica gel column were each carefully concentrated to a volume of 1 ml in a rotary evaporator. The concentrated fractions, together with the hexane representing the void volume of the column, were then examined by gas chromatography. Fractions CCE3II-SG4, 5, 6, and 7 were found to contain sufficient quantities of organic compounds to warrant further study, with fractions SG4, 5, and 6 appearing amenable to conclusive investigation without additional purification. Fraction CCE3II-SG7, which contained a very significant portion of the organic compounds of the parent CCE, was quite complex and hence was rechromatographed on a fresh 7 by 76-mm column of Silicar CC-7. Elution was accomplished with a total volume of 370 ml of solvent ranging in polarity from hexane to methanol, with 26 fractions being collected as shown in Table 3. Collection of fractions and the sequence of eluting solvents were based partially on the movement from the column of fluorescent zones, which were detected under 366 nm uv light. The fractions were examined by gas chromatography,

Table 2. FRACTIONS PREPARED FROM CCE3II BY SILICA GEL COLUMN CHROMATOGRAPHY

Fraction	Total volume	Eluting solvents
CCE3II-SG1	2 ml	Hexane
CCE3II-SG2	3 ml	Hexane
CCE3II-SG3	3 ml	Hexane
CCE3II-SG4	2 ml	Benzene
CCE3II-SG5	3 ml	Benzene
CCE3II-SG6	3 ml	Benzene
CCE3II-SG7	2 ml	Chloroform-Methanol (1:1)
CCE3II-SG8	3 ml	Chloroform-Methanol (1:1)
CCE3II-SG9	3 ml	Chloroform-Methanol (1:1)
CCE3II-SG10	5 ml	Methanol

Table 3. FRACTIONS PREPARED FROM CCE3II-SG7
BY SILICA GEL COLUMN CHROMATOGRAPHY

Fraction	Total volume, ml	Eluting solvents
CCE3II-SG7-SG1*	10	Hexane
" SG2*	10	Hexane
" SG3*	10	Hexane
" SG4†	10	Hexane
" SG5†	15	Hexane, 10 ml Hexane-Benzene(1:1), 5 ml
" SG6†	15	Hexane-Benzene (1:1)
" SG7	15	Hexane-Benzene (1:1)
" SG8	15	Hexane-Benzene (1:1), 5 ml Hexane-Benzene (1:3), 10 ml
" SG9Ψ	15	Hexane-Benzene (1:3), 10 ml Benzene, 5 ml
" SG10Ψ	15	Benzene
" SG11Ψ	15	Benzene
" SG12	15	Benzene
" SG13	15	Benzene-Chloroform (3:1)
" SG14δ	15	Benzene-Chloroform (3:1)
" SG15δ	15	Benzene-Chloroform (3:1), 10 ml Benzene-Choloroform (1:1), 5 ml
" SG16δ	15	Benzene-Chloroform (1:1)
" SG17δ	15	Benzene-Chloroform (1:1)
" SG18δ	15	Benzene-Chloroform (1:1), 5 ml Benzene-Chloroform (1:3), 10 ml
" SG19δ	15	Benzene-Chloroform (1:3)
" SG20δ	15	Benzene-Chloroform (1:3)
" SG21	15	Chloroform
" SG22	15	Chloroform
" SG23∇	15	Chloroform, 5 ml Chloroform-Methanol (1:1), 10 ml
" SG24∇	15	Chloroform-Methanol (1:1)
" SG25∇	15	Methanol
" SG26∇	15	Methanol

* Recombined as CCE3II-SG7-SG1→3.
† Recombined as CCE3II-SG7-SG4→6.
Ψ Recombined as CCE3II-SG7-SG9→11.
δ Recombined as CCE3II-SG7-SG14→20.
∇ Recombined as CCE3II-SG7-SG23→26.

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and several groups of fractions that appeared to contain very little organic matter or low levels of essentially the same components were recombined, as indicated in Table 3, prior to attempting to identify individual compounds.

CAE3II, the carbon alcohol extract from well no. 3, was separated into fractions of less complexity by classical solubility separation procedures (11, 12). A 1.5-ml aliquot of the concentrated CAE (total volume 4 ml) was dissolved in 30 ml of diethyl ether, filtered, and extracted successively with water, dilute hydrochloric acid, and dilute sodium hydroxide, as shown in Figure 7. Five fractions, namely ether insolubles, water solubles, bases, acids, and neutrals, were obtained.

The ether insolubles fraction contained an appreciable quantity of material (0.3 g), whereas the water solubles fraction was of lesser, but still significant, weight (0.05 g). However, because of the polarity and probable complexity of these fractions and time limitations on this investigation, no further studies of the ether insolubles and water solubles were conducted.

The bases, acids, and neutrals fractions were dried on anhydrous sodium sulfate columns and concentrated to 1 to 2 ml each in rotary evaporators. Examination of these fractions by gas chromatography indicated that the acids fraction, designated CAE3II Acids, contained a total quantity of organic compounds several orders of magnitude greater than that present in the other fractions. Hence, the acids fraction was selected for further study.

Identification of Compounds in the CCE and CAE from the Landfill Well

The various fractions obtained by silica gel column chromatography of CCE3II and the acids fraction obtained by solubility separation of CAE3II were analyzed by combined
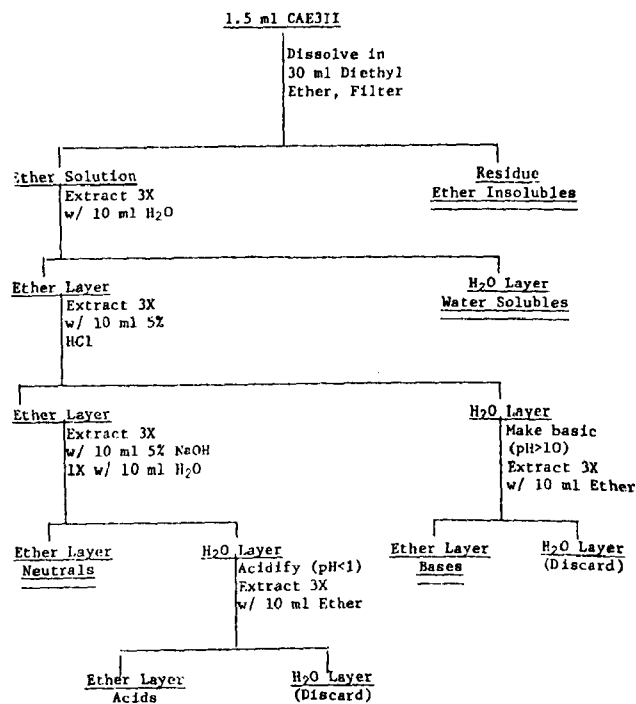
Figure 7. Solubility separation of CAE3II.

gas chromatography/mass spectrometry (GC/MS), employing computerized data acquisition and processing, to achieve identification of individual compounds therein. A Finnigan Model 10150 gas chromatograph/mass spectrometer with both electron impact and chemical ionization sources and System Industries System 150 data system were used for this work. Gas chromatography columns packed with 3% OV-1 on 100/120 mesh Gas Chrom Q or 3% Carbowax 20M on 80/100 mesh Gas Chrom Q (applied Science Laboratories, State College, Pa.) proved to be most effective for separation of the CCE3II fractions, and a column packed with 10% SP 2100/1% H_3PO_4 on 80/100 mesh Chromosorb W AW (Supeloco, Inc., Bellefonte, Pa.) was most useful for the CAE3II acids fraction.

Mass spectra were examined by standard interpretive procedures based on mass spectral fragmentation theory and by computerized spectral matching programs that permitted comparison of the unknown spectra with the contents of libraries of

mass spectra at the National Institutes of Health, Bethesda, Maryland, and Battelle Memorial Institute, Columbus, Ohio (13). Additional corroborative evidence for the structures of compounds identified on the basis of their spectra was obtained by direct comparison with standard compounds whenever such standards were available. When possible, the quantities of the identified compounds in the various fractions were estimated by comparing their peak heights produced by known quantities of standard compounds chromatographed under identical conditions. These data were then used to calculate estimated concentrations of the identified organic compounds in the sampled groundwater.

Table 4 presents a tabulation of those compounds which were identified in the carbon chloroform and carbon alcohol extracts prepared from groundwater from well no. 3. The CCE and/or CAE fraction(s) in which the compound was identified and additional pertinent data such as industrial uses and toxicity information are presented for each compound. Also, estimates of the quantities present in the sampled water are given for those compounds for which quantitative evaluations were achieved. The general structures of all the compounds listed in Table 4 were established beyond reasonable doubt. However, it should be noted that exact positions of substituent attachment and chain branching were not achieved for a few compounds, such as the two C_3 alkylbenzenes in CCE3II-SG7-SG9 and come of the C_7, C_8, and C_9 isomeric acids, because of unavailability of required standards or failure of GC columns to separate closely related compounds.

There was strong evidence for the presence in the various fractions of several compounds in addition to those listed in Table 4, but their structures were not considered sufficiently confirmed for inclusion in this Table. These "possible" compounds and the fractions in which

they were found were: a C_8 ketone, CCE3II-SG6; a glycol ether, CCE3II-SG7-SG12; triethyleneglycol ether and a diester of adipic acid, CCE3II-SG7-SG13; and benzoic and nonanoic acids, CAE3II Acids. Also, gas chromatography of a small aliquot of the CAE3II Acids fraction after it was reacted with 14% boron trifluoride in methanol to esterify carboxylic acids (14) indicated the presence of relatively low quantities of C_{12}, C_{14}, C_{16}, and C_{18} fatty acids in this fraction. This was not confirmed by mass spectrometry, however, due to time limitations.

DISCUSSION

The data presented in Table 4 clearly show that low levels of many potentially undersirable organic compounds were being contributed to groundwater within and immediately under the Norman landfill by solid waste deposited in this landfill.

A few of the compounds identified in this study could be leachates of natural products, including foods, or possibly end products of microbial metabolism. For example: the short chain, normal carboxylic acids are constituents of many foods and plants; acetic and butyric acids are common end products of anaerobic metabolism of carbohydrates; and, phenolic substances are ubiquitous phytochemicals. However, most of the identified compounds are chemicals commonly employed in industrial operations (Table 4) and, therefore, usually associated with industrial waste. Since available information indicates that the Norman landfill has never received appreciable quantities of solid waste from industrial operations, there is an obvious question concerning the source of the industrial organic chemicals leached from this landfill. It should be noted, however, that most of the compounds listed in Table 4 are used in the manufacture of

Table 4. COMPOUNDS IDENTIFIED IN GROUNDWATER FROM LANDFILL WELL

Compound	Estimated concentration, µg/l	CCE and/or CAE fraction(s)	Uses, sources, toxicity
Fenchone	0.2	CCE3II-SG4	Flavoring.[15] Present in paper mill's raw waste.[16]
Camphor	0.9	CCE3II-SG4	Plasticizer for cellulose esters and ethers, moth and mildew preventive, flavoring.[15] Neoplastic effects produced in rats.[17]
2,6-Di-t-butylbenzoquinone	--	CCE3II-SG4	Polymerization catalyst.[15]
Diethyl phthalate	4.1	CCE3II-SG4	Plasticizer, solvent for cellulose acetate, camphor substitute, perfume fixative, wetting agent.[15] Moderately toxic when ingested.[17]
2,6-Di-t-amylbenzoquinone	--	CCE3II-SG4	Polymerization catalyst.[15]
Diisobutyl phthalate	0.1	CCE3II-SG4 and CCE3II-SG7-SG4+6	Plasticizer.[15]
Di-n-butyl phthalate	--	CCE3II-SG4	Plasticizer, polymerization catalyst, oxidant.[15] Produces central nervous system effects.[17]
Butycarbobutoxymethyl phthalate	--	CCE3II-SG4	Plasticizer.[15]
Butylbenzyl phthalate	--	CCE3II-SG4	Plasticizer for polyvinyl and cellulose resins.[15]
Dicyclohexyl phthalate	0.2	CCE3II-SG4	Plasticizer for rubber, polyvinyl chlorides, and other polymers.[15]
Dioctyl phthalate*	2.4	CCE3II-SG4	Plasticizer for polyvinyl chlorides, and other vinyls.[15]
p-Cresol	14.6	CCE3II-SG5 and CCE3II acids	Constituent of creosote.[15]
o-Xylene	0.6	CCE3II-SG7-SG7	Raw material for manufacture of phthalic anhydride, insecticides, motor fuels, dyes. Moderately toxic.[15]
p-Xylene	0.9	CCE3II-SG7-SG7	Source of terephthalic acid for polyester resin synthesis; manufacturing of insecticides. Moderately toxic.[15]
Cyclohexanol	1.0	CCE3II-SG7-SG7	Manufacture of phenolic insecticides, lacquer polishes, plastics, germicides, nylon.[15]
N-Ethyl-p-toluenesulfonamide	0.1†	CCE3II-SG7-SG8	Plasticizer. Moderately toxic.[15]
N-Ethyl-o-toluenesulfonamide	--	CCE3II-SG7-SG8	Plasticizer. Moderately toxic.[15]
C3 Alkylbenzenes* (2 compounds)	--	CCE3II-SG7-SG9+11	American petroleum.[18]
Diacetone alcohol	10.9	CCE3II-SG7-SG9+11	Solvent for cellulose acetate, various oils, resins, dyes, tars, and waxes. Hydraulic compression fluids, wood preservatives, and metal cleaning.[15]
Butoxyethanol	--	CCE3II-SG7-SG9+11	Solvent for nitrocellulose resins, sprays, lacquers, and enamels.[15]
Ethyl carbamate	--	CCE3II-SG7-SG9+11	Solvent for various organics; solubilizer and co-solvent for pesticides, fumigants, and cosmetics.[18] Carcinogenic.[17]
Tri-n-butyl phosphate	1.7	CCE3II-SG7-SG9+11	Plasticizer, antifoam agent, solvent for nitrocellulose and cellulose acetate, heat exchange medium, dielectric. Moderately toxic by ingestion.[15]
p-Toluenesulfonamide	--	CCE3II-SG7-SG12	Plasticizer, fungicide, mildewcide in paints, resin synthesis.
Methylpyridine	--	CCE3II-SG7-SG12	Insecticide manufacture, dyes, rubber, production of vinyl pyridine.[15] Moderately toxic.[15]
N,N-diethylformamide	--	CCE3II-SG7-SG12	Used in rubber manufacture.[16]
Triethyl phosphate	0.3	CCE3II-SG7-SG12	Plasticizer for resins, plastics, gums; pesticide manufacture, catalyst, solvent, lacquer remover.[15] Highly toxic.[18]
bis-2-Hydroxypropyl ether	--	CCE3II-SG7-SG21	A dimerization product of propylene glycol, a non-toxic antifreeze used in dairies and breweries, production of synthetic resins.[18]
3-Methylcyclopentane-1,2-diol	--	CCE3II-SG7-SG21	--
Acetic acid	--	CAE3II acids	Production of plastics, insecticides, vinyl acetate, and photographic chemicals; oil well acidizing and food additive.[18]
Isobutyric acid	48.7	CAE3II acids	Manufacture of esters for solvents, flavors, and perfumes; disinfection, tanning agent, deliming hides.[15] Mild irritant.[18] Detection of odor in water at 8.1 mg/l and of taset in water at 1.6 mg/l.[19]
Butyric acid	1.5	CAE3II acids	Emulsifying agent, disinfectant, gasoline sweetner; perfume ester preparation, and deliming agent.[15] Neoplastic effects produced in rats.[17]
Isovaleric acid	0.7	CAE3II acids	Used in flavors, perfumes, and manufacture of seda-tives.[18] Occurs in tobacco and several other plants. Detection of odor in water at 0.7 mg/l.[19]
Valeric acid	1.1	CAE3II acids	Used as an intermediate in perfummery.[18]
2-Ethylhexanoic acid	4.2	CAE3II acids	Typical low to medium weight isoacid. Plasticizer component of alkyd resins. Salts used for varnish driers, heat stabilizers for vinyl resins, greases, thickening agents in certain lacquers and paints, sludge, and varnish inhibitor in mineral oils.[20]
Isomeric C6 acid*	17.1▼	CAE3II acids	See 2-ethylhexanoic acid uses above.
Isomeric C6 acid*	0.2▼	CAE3II acids	See 2-ethylhexanoic acid uses above.
Isomeric C7 acid*	7.5▼	CAE3II acids	See 2-ethylhexanoic acid uses above.
Isomeric C8 acid*	--	CAE3II acids	See 2-ethylhexanoic acid uses above.
Cyclohexanecarboxylic acid	2.8	CAE3II acids	Insecticide formulations, stabilizer for vulcanized rubber, paints and varnishes, lubricating oils, dry cleaning soaps.[15]
Caprylic acid	0.6	CAE3II acids	Manufacture of esters used in perfummery, dye production.[18]
Caproic acid	1.1	CAE3II acids	Manufacture of esters for artificial flavors, hexyl-phenols, hexylresorcinol.[18]
Heptanoic acid	1.0	CAE3II acids	Found in fusel oils and rancid oils.[18]

*General structure confirmed beyond reasonable doubt, but position of substitution or chain branching not determined because necessary standards were unavailable or compounds were not separated by G.C. columns employed.
†Determined as N,N-dimethyl-p-toluenesulfonamide.
▼Determined as 2-ethylbutyric acid, but probably is not this compound.
qDetermined as n-heptanoic acid.

a wide array of finished products for domestic and commercial use that ultimately will find their way into most landfills. For example: the phthalic acid esters are used very extensively for production of polymers employed in such diverse products as food wrap film, garden hose, upholstery, electrical insulation, and clothing. The decomposition and/or leaching of such manufactured products deposited in the Norman landfill would appear most likely to account for the introduction of industrial organic pollutants into groundwater in and near this landfill, even though it had received essentially no industrial solid waste, per se.

Those compounds for which quantitative data were obtained during this investigation appeared to be present in groundwater from well no. 3, the landfill well, only in low concentrations. However, the quantitative data resulting from this work must be considered as minimum values because of quantitative inadequacies of the procedures used, particularly the carbon adsorption method. These inadequacies result principally because: activated carbon may fail to quantitatively adsorb dissolved organic compounds from sampled water; complete recovery of adsorbed compounds from the activated carbon may not be accomplished during extraction; and, volatile sample components may be lost during drying of the activiated carbon and evaporation of extracting solvents. This is illustrated by comparing the total combined weight of the landfill well CCE and CAE, 2.013 mg/l, with the average total organic carbon content of 13.4 mg/l of water from this well. If the organic equivalent of 13.4 mg carbon/l is considered, it becomes apparent that less than 10% of the organic matter present in the sampled groundwater was recovered in the combined CCE and CAE.

The history of the Norman landfill and dates of newspapers recovered from well no. 3 during drilling indicated that the refuse in the area of the fill near this well had been in place at least 3 yr at the time this investigation was conducted. Based on this information, as well as the relatively low concentration of organic carbon (13.4 mg/l) in groundwater from well no. 3, it appears likely that most of the readily leachable organic matter had already been removed from refuse near the test when sampling of groundwater for organic pollutants was accomplished. It is probable, therefore, that most of the compounds identified in this study are substances that were leached very slowly from the refuse in the landfill and/or substances that persisted for considerable periods of time in the aquifer in the vicinity of the refuse from which they were leached because of sorption on the earth solids comprising the aquifer. This observation implies the potential for long-term insidious pollution of groundwater by undesirable organic chemicals from landfills. Slowly decaying domestic and commercial products in landfills would appear likely to serve as reservoirs feeding low levels of industrial organic pollutants into aquifers for many years after the landfills have been closed and forgotten. Even those substances that are sorbed relatively strongly on aquifer solids could ultimately pose a pollution threat if they were resistant to biochemical and abiotic degradation in the groundwater environment. Such compounds could move as zones by slow, "chromatographic" migration to finally reach wells providing water for consumption by humans or domestic animals. Because of the low levels of pollutants likely to be involved, physical properties of the polluted groundwater would probably not be altered sufficiently to indicate the presence of the offending compounds. This presence could be a matter of considerable concern, however, since the health effects of chronic ingestion through water of even very low levels of compounds such as those identified in this study are largely unknown. This, coupled with the great

difficulty involved in removing pollutants, particularly those which tend to adsorb significantly on aquifer solids, from a polluted aquifer, dictates the need for further investigation of this potential problem. In particular, information concerning the mobility and longevity in the groundwater environment of compounds such as those in Table 4 are needed. A limited and less rigorous study of organic compounds in groundwater from well No. 2, outside the landfill (Figure 1), indicated the probable presence of the same phthalic acid esters as those identified in groundwater from well No. 3, thus suggesting that these compounds were moving through the aquifer (21). In general, however, information of this type is very scarce.

In assessing the results of this investigation, it should be clearly noted that the compounds identified included only substances readily amenable to gas chromatography and represented probably less than 10% of the combined weights of the carbon chloroform and alcohol extracts. Most of the missing material was probably composed of compounds too polar and/or too high in molecular weight to yield readily to gas chromatography procedures. Characterization of this material would undoubtedly have yielded much additional information concerning the organic pollutants contributed to groundwater by the Norman landfill, but the necessary analytical effort for such characterization during this study was precluded by time limitations.

CONCLUSIONS

On the basis of information developed by this investigation, several conclusions may be proposed, as noted below.

1. Landfills in which refuse is deposited in or near the water table are likely to contribute many undesirable organic chemicals to groundwater in their proximity.

2. Even those landfills that do not receive appreciable quantities of solid wastes from industrial operations may pollute groundwaters with industrial organic compounds, probably by leaching of such substances from finished products manufactured for domestic and commercial use that ultimately are deposited in landfills.

3. The potential exists for long-term pollution of groundwater by industrial organic chemicals from landfills in contact with the water table. Such pollution could persist for many years after closing of landfills because of: slow leaching of organic compounds from discarded manufactured products that serve as reservoirs of these compounds; and/or slow "chromatographic" movement of adsorbed, intractable compounds away from the landfill site.

4. The potential for long-term, perhaps insidious, pollution of groundwater by industrial organic chemicals from landfills emphasizes the need for information concerning the health effects of long-term ingestion of water containing low levels of such compounds.

5. Additional information concerning the generation and/or release of organic compounds from refuse, the persistence of such compounds in saturated and unsaturated subsurface environments, and the mobility of recalcitrant

organic compounds in groundwater aquifers, as well as health effects data, is needed for realistic evaluation of the problem of groundwater pollution by organic compounds leached from landfills.

REFERENCES

1. Miller, D. W., F. A. De Luca, and T. L. Tessier. Ground Water Contamination in the Northeastern States. U. S. Environmental Protection Agency, Washington, D. C. Report Number EPA-660/2-74-056. June 1974. p. 211-216.

2. Walker, W. H. Illinois Ground Water Pollution. J. Amer. Water Works Assn. 61:31, 1969.

3. Anderson, J. R., and J. N. Dornbush. Influence of Sanitary Landfill on Ground Water Quality. J. Amer. Water Works Assn. 59:457-470, 1967.

4. Fuhriman, D. K., and J. R. Barton. Ground Water Pollution in Arizona, California, Nevada, and Utah. U. S. Environmental Protection Agency, Washington, D. C. Report Number 16060 ERU 12/71. December 1971. p. 92-94.

5. Scalf, M. R., J. W. Keeley, and C. J. LaFevers. Ground Water Pollution in the South Central States. U. S. Environmental Protection Agency, Corvallis, OR. Report Number EPA-R2-73-268. June 1973. p. 100-102.

6. Sanitary Landfill Studies, Appendix A: Summary of Selected Previous Investigations. California State Department of Natural Resources, Sacramento, CA. Bulletin Number 147-5. July 1969.

7. Hughes, G. M. R. A. Landon, and R. W. Farvolden. Hydrology of Solid Waste Disposal Sites in Northeastern Illinois. Illinois State Geological Survey, Environmental Geology Notes, Urbana, Il. Publication Number 45. April 1971. 25 p.

8. Garbutt, G.H. Report of the Preliminary Study of a Landfill in McClain County, Oklahoma. University of Oklahoma. (Unpublished special project report. Norman, OK. June 1972.) 16 p.

9. Breidenbach, A. W., J. J. Lichtenberg, C. G. Henke, D. J. Smith, J. W. Eichelberg, Jr., and H. Stierle. The Identification and Measurement of Chlorinated Hydrocarbon Pesticides in Surface Waters. U. S. Department of the Interior, Federal Water Pollution Control Administration, Washington, D. C. 1966. p. 44-50.

10. Booth, R. L., J. N. English, and G. N. McDermott. Evaluation of Sampling Conditions in the Carbon Adsorption Method. J. Amer. Water Works Assn. 57:215-220, 1965.

11. Shriner, R. L., R. C. Fuson, and D. Y. Curtin. The Systematic Identification of Organic Compounds, Fifth Edition. New York, NY. John Wiley and Sons, Inc., 1965. p. 67-107.

12. Breidenbach, A. W., J. J. Lichtenberg, C. F. Henke, D. J. Smith, J. W. Eichelberger, Jr., and H. Stierle. The Identification and Measurement of Chlorinated Hydrocarbon Pesticides in Surface Waters. U. S. Department of the Interior, Federal Water Pollution Control Administration, Washington, D. C. 1966. p. 12-14.

13. Webb, R. G., A. W. Garrison, L. H. Keith, and J. H. McGuire. Current Practice in GC-MS Analysis of Organics in Water. Environmental Protection Agency, Corvallis, OR. Report Number EPA-R2-73-277. 1973. p. 37-41.

14. Metcalfe, L. D., and A. A. Schmitz. The Rapid Preparation of Fatty Acid Esters for Gas Chromatographic Analysis. Anal. Chem. 33:363-364, 1961.

15. Hawley, G. G., Ed. The Condensed Chemical Dictionary, Eighth Edition. New York, NY. Van Nostrand Reinhold Co., 1971. 971 p.

16. Webb, R. G., A. W. Garrison, L. H. Keith, and J. H. McGuire. Current Practice in GC-MS Analysis of Organics in Water. U. S. Environmental Protection Agency, Corvallis, OR. Report Number EPA-R2-73.-277. 1973. p. 62-87.

17. Christenson, H. E., Ed. The Toxic Substances List. U.S. Department of Health, Education, Welfare. National Institute for Occupational Safety and Health, Rockville, MD. Publication Number HSM 72-10265. June 1972. 563 p.

18. Stecher, P. G. The Merck Index. Rahway, NJ. Merck and Co. Inc., 1968. 1713 p.

19. Stahl, W. H., Ed. Compilation of Odor and Taste Threshold Values Data. American Society for Testing and Materials, Philadelphia, PA. 1973.

20. Mark, H. F., Ed. Encyclopedia of Chemical Technology. New York, NY. John Wiley and Sons, Inc., 1970. Vol. 8. p. 849-850.

21. Robertson, J. M., C. R. Toussaint, and M. A. Jorque. Organic Compounds Entering Ground Water From a Landfill. U. S. Environmental Protection Agency, Washington, D. C. Report Number EPA-660/2-74-077. September 1974. p. 23-24.

ATTENUATION MECHANISMS OF POLLUTANTS THROUGH SOILS*

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INTRODUCTION

The title implies far more than we or anyone can deliver in the short time allotted to the subject at this symposium. Despite the complicated and interrelated subject of attenuation in soils, we believe that, from the great volume of material, something can be extracted and refined that can be useful as a basis for disposal-site selection and management.

As a necessary beginning, the meaning of two words, attenuation and mechanism, requires some explaining to develop common ground for their use in this discussion.

Attenuation

Attenuation is defined here by looking at the movement of a pulse of a solute through a soil. As the pulse migrates, the maximum concentration decreases. Attenuation can then be defined as the decrease of the maximum concentration for some fixed time or distance traveled.

Mechanisms

The word mechanism cannot be defined precisely. Two choices in the latest edition of Webster's Dictionary are "A system whose parts work together like those of a machine," or "any system or means for doing something." What most often are described as "mechanisms" by the various pollution control communication media are, in reality, a series of ill-defined processes that relate to or correlate with some measurable parameter of the microhabitat of those reactions. For example, Eh (red/ox) has been called a mechanism. Red/ox, though, is not a mechanism by which soils retain trace elements of leachates or other wastewaters. Red/ox potential of a soil habitat, however, can be significantly related to a host of electron transfers among elements that influence their solubility in the displacing soil solution. Mechanism as used here will relate to the more specific chemical reactions that can be identified.

For a lack of better terminology, the phrase "factor(s) of attenuation" will be used in identifying broad classes of reactions that appear to control migration rates. They may be grouped because of trends in a pollutant's solubility, which is controlled by an identifiable chemical or physical condition and which has a standard means of measurement. For example, pH value may be expected to initiate or limit the solubility of certain substances through $[H]^+$ and $[OH]^-$ activity levels. The pH values of a solution or suspension habitat can be readily monitored by a pH meter.

A definition of the term "trace element(s)" also will help clarify this communication. The term is used here to identify those selected, potentially hazardous pollutants (As, Be, Cd, Cr, Cu, Ni, Hg, Pb, Se, V, and Zn) and other elements that may be found in biological tissues or cells in amounts usually considered to be trace as opposed to the more macro-levels of N, P, K, H, O, C, Ca, Na, etc.

Microhabitat of the Soil

Before discussing specific reactions influencing mobility and immobility of trace elements in soils, it may be well to review some of the broad alterable factors of the soil habitat that can affect trace-element mobility. Disposal sites of wastes usually are located in subsurface soils, sand and gravel excavations, geologic materials, and shrouded in disturbed soils. The waste is left to sour in the bowels of anaerobosis. Thus, disposal management must consider the unusual soil condition. Leachates, themselves, also will alter the natural soil environment.

Some important characteristics of the microhabitat of the soil that may be altered and, in turn, may alter trace-element movement, as it might occur in the usual aerated soil, are:

1. aeration (anoxic, waterlogged, swampy, reducing conditions, etc.)

2. particle size distribution, texture, or clay content

3. permeability or pore size distribution as it may influence flux of the soil solution

4. pH values (either high degree of acidity or alkalinity may develop from waste disposal and acid status of leachate and waste streams may influence the solubility of complexes of potential pollutants)

5. lime (free soil lime, caliche, agricultural limestone, and commercial limes)

6. iron, aluminum, and manganese hydroxy oxides in unusually high concentration or state of reactivity or solubility

7. organic matter and organic soils (sequestering of heavy metals with organic complexes alters solubility and mobility as chelates and certain chemical unions can form immobile organometallic complexes)

8. high specific salt concentrations, where trace element reactions become salt dependent.

This brief listing over simplifies the factors in the microhabitat that influence trace element mobility in disposal-site soils. For example, organic matter can attenuate heavy metals by combining with them to form very slowly soluble complexes or increase their mobility by forming highly soluble organo-metal-ion complexes, which differ greatly in degree of

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attenuation. Nevertheless, it is well to make a beginning in a very general way, recognizing that this approach is necessary for practical problem solving.

ATTENUATION AS REVEALED BY SOIL RESEARCH

First let us define the system and habitat that concerns us in the soil trace element attenuation program. The system is a municipal solid-waste, sanitary-landfill leachate generator. The habitat is an anaerobic fermentation process that accumulates aqueous acid leachate containing various levels of potentially hazardous trace-element pollutants. The amount of leachate generated depends on the disposal site rainfall pattern. The concentration of trace element appearing in the leachate depends on the kind of industry dumping into the municipal landfill. The soil habitat also is anaerobic in the landfill where leachate collects.

Experimental Procedures at The University of Arizona

A 1000-gal leachate generator was constructed, packed with representative municipal refuse, filled to brimming with water, and allowed to ferment for 6 warm-season months. The leachate was drawn off under high CO_2 pressure in absence of atmospheric O_2 and displaced through 10 soils from 7 major orders (Table 1), again in the absence of atmospheric O_2 and in the presence of CO_2. The soil columns were cylinders measuring 10 x 22 cm and 5 x 10 cm, packed to known densities. Water, natural leachate, and trace-element "spiked" leachate were passed through the soil. The effluent was collected in increments of 0.5 and 1.0 pore-space volumes (depending on column size) in a 24-hr period under CO_2 in the exclusion of atmospheric O_2. The concentration of certain elements in the influent and effluent was monitored and data were collected.

Table 1. CHARACTERISTICS OF THE SOILS USED

Soil series	Soil order	pH	Cation exch. capac. meq/100 g	Elec. cond. of extract, μmhos/cm	Column bulk density, g/cm³	Sand, %	Silt, %	Clay, %	Texture class	Predominant clay
Anthony	Entisol	7.8	6	328	2.07	71	14	15	Sandy loam	Montmorillonite, mica
Ava	Alfisol	4.5	19	157	1.45	10	60	31	Silty clay loam	Vermiculite, kaolinite
Chalmers	Mollisol	6.6	26	288	1.60	7	58	35	Silty clay loam	Montmorillonite, vermiculite
Davidson	Ultisol	6.2	9	169	1.89	19	20	61	Clay	Kaolinite
Fanno	Alfisol	7.0	33	392	1.48	35	19	46	Clay	Montmorillonite, mica
Kalkaska	Spodosol	4.7	10	237	1.53	91	4	5	Sand	Chlorite, kaolinite
Mohave	Aridisol	7.3	10	615	1.78	52	37	11	Sandy loam	Mica, kaolinite
Mohave$_{Ca}$	Aridisol	7.8	12	510	1.54	32	28	40	Silty clay loam	Mica, montmorillonite
Molokai	Oxisol	6.2	14	1262	1.44	23	25	52	Clay	Kaolinite, gibbsite
Nicholson	Alfisol	6.7	37	176	1.53	3	47	49	Silty clay	Vermiculite
Wagram	Ultisol	4.2	2.1	225	1.89	88	8	4	Loamy sand	Kaolinite, chlorite

*Listed in order of importance.

The data reported here were collected from experiments (1) where columns were leached with a landfill leachate individually spiked with selected potentially hazardous elements (As, Be, Ce, Cr, Cu, Hg, Ni, Pb, Se, V, and Zn). In each instance, the leachate was acidified to pH 5.0 with HCl and spiked with 70 to 120 ppm of the element of interest. The leachate was passed through soil columns (5-x 10-cm) and adjusted to maintain a steady solution flux of one pore-space displacement in 24 hr. The leaching was continued until (a) breakthrough (effluent concentration = influent concentration), (b) steady state, or (c) continued absence of the element in the effluent. In all cases, the columns were cut into 1-cm segments and the element extracted by H_2O and dilute HCl.

Classification of Factors in Attenuation

Faced with a paucity of critical soils research data, speculation dominates both the literature and our thinking concerning the levels of importance of the biological, physical, and chemical factors influencing attenuation in soils of trace elements in landfill leachates. It seems necessary, therefore, to divide this soils presentation into at least three parts to distinguish among those factors that have received (a) research attention as being important in attenuation, (b) research attention as being unimportant in attenuation, and (c) little or no research attention as to their importance in attenuation. These groupings find reality in unpublished

Table 2. TOTAL ANALYSIS OF SOILS FOR TRACE METALS AND FREE IRON OXIDES

Soil	Mn, g/g	Co, g/g	Zn, g/g	Ni, g/g	Cu, g/g	Cr, g/g	Fe oxides, %
Anthony	275	50	55	80	200	25	1.8
Ava	360	50	77	110	80	55	4.0
Chalmers	330	60	100	130	83	68	3.1
Davidson	4100	120	110	120	160	90	17.0
Fanno	280	45	70	100	60	38	3.7
Kalkaska	80	25	45	50	46	15	1.8
Mohave	825	50	85	100	265	18	1.7
Molokai	7400	310	320	600	260	410	23.0
Nicholson	950	50	130	135	65	68	5.6
Wagram	50	--	40	80	62	--	0.6
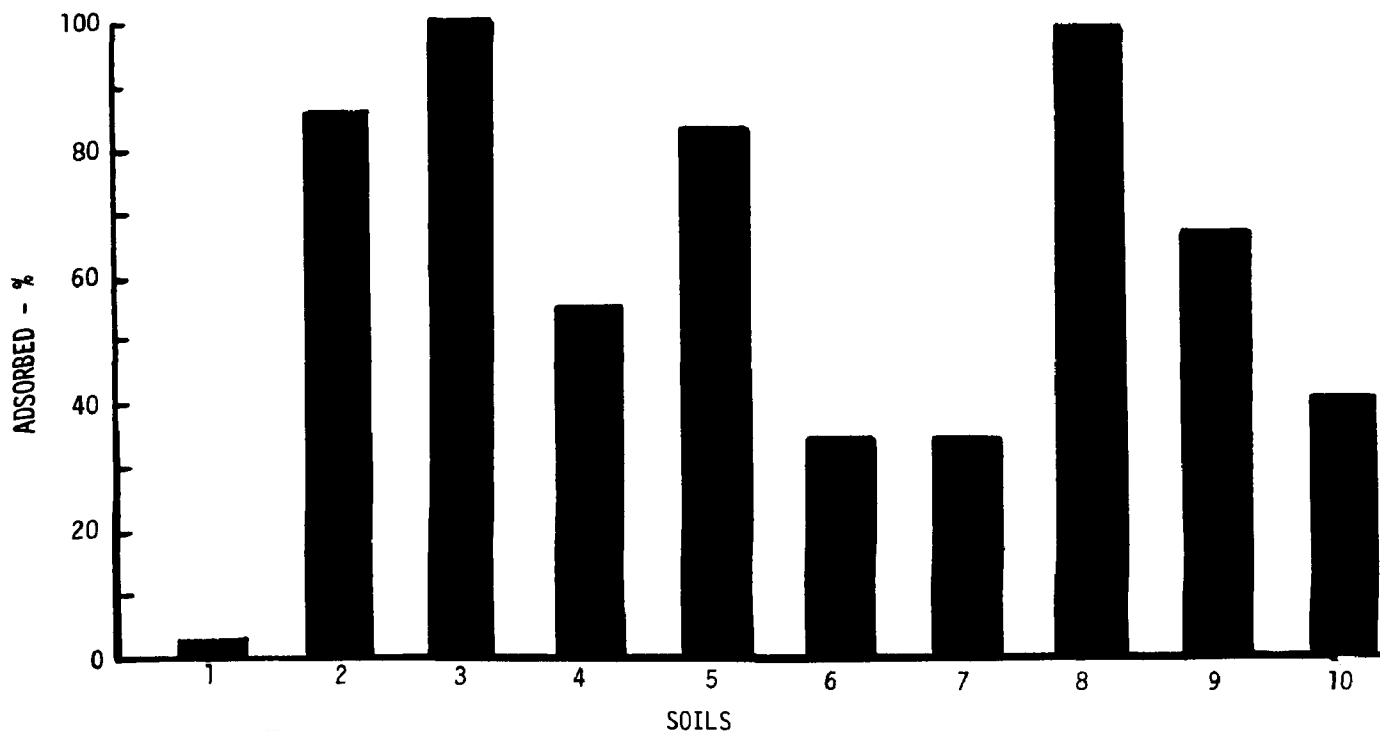
Figure 1. Percent of chromium VI adsorbed by 10 cm of soil after 14 pore-space displacements. 1, Anthony; 2, Ava; 3, Davidson; 4, Fanno; 5, Kalkaska; 6, Mohave; 7, Mohave(Ca); 8, Molokai; 9, Nicholson; and 10, Wagram.
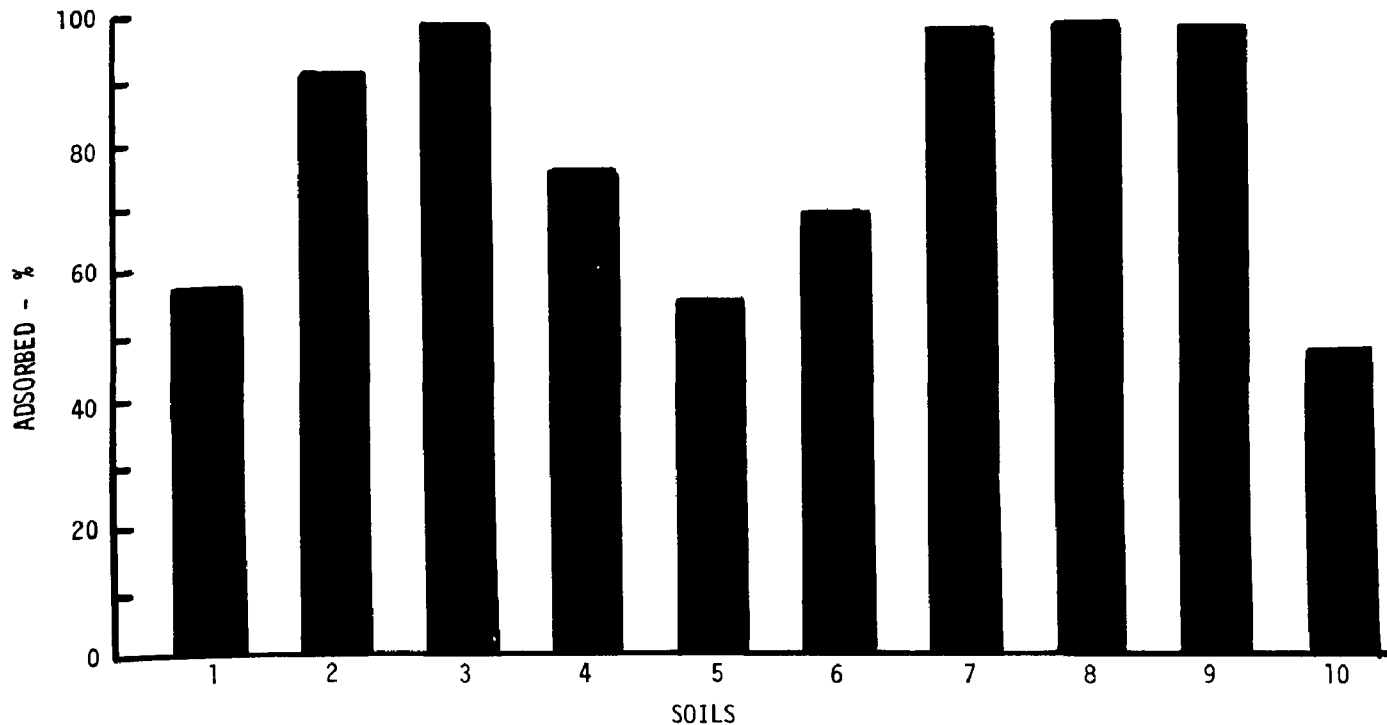
Figure 2. Percent of arsenic adsorbed by 10 cm of soil after nine pore-space displacements. 1, Anthony; 2, Ava; 3, Davidson; 4, Fanno, 5, Kalkaska; 6, Mohave; 7, Mohave(Ca); 8, Molokai; 9, Nicholson; and 10, Wagram.

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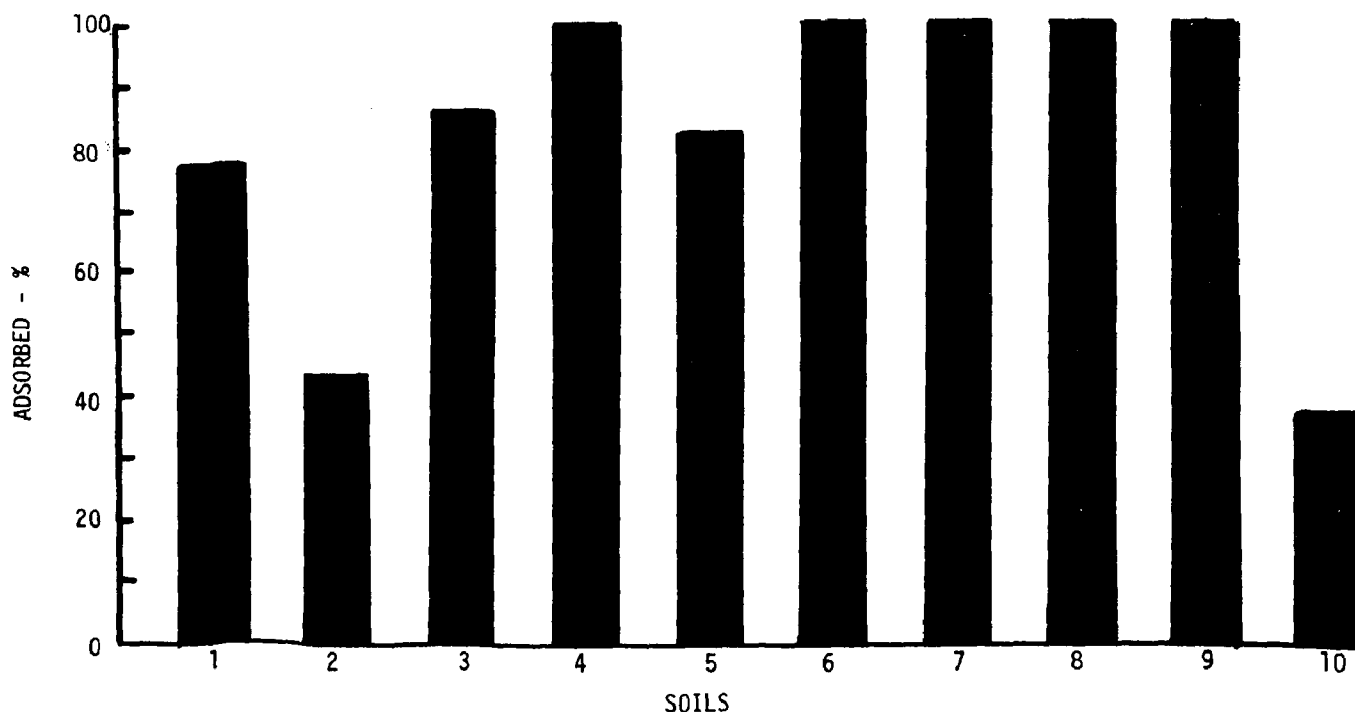
Figure 3. Percentage of cadmium adsorbed by 10 cm of soil after 1.05 pore-space displacements. 1, Anthony; 2, Ava; 3, Davidson; 4, Fanno; 5, Kalkaska; 6, Mohave; 7, Mohave(Ca); 8, Molokai; 9, Nicholson; and 10, Wagram.

data similar to those respresented in Tables 1 and 2 and Figures 1, 2 and 3. By comparing the soil parameters reported in Tables 1 and 2 with the migration of the elements depicted in Table 3 and the three figures, certain soil characteristics stand out more clearly as factors of attenuation than others.

The figures show that particle size distribution (texture) plays the dominant role in attenuation with the partial exception of chromium (Figure 1) where pH is also important. In general, those soils highest in clay-sized particles and free iron oxides (Molokai, Davidson, and Nicholson) are found to attenuate the trace elements to the greatest extent (Figures 2 and 3). Mohave is high in lime and as such is also highly effective in immobilizing trace elements. The negative effect of pH on chromium migration is illustrated in Figure 1 by chromium's mobility in Mohave.

The usual behavior for this soil is displayed in Figures 2 and 3.

Important Factors in Attenuation

Those factors found to be the most important in attenuation in soils by the research reported in Table 3 and Figures 1, 2, and 3 are:

1. size of the soil particles (or total clay content)

2. free "iron oxide" content of the soil (other hydrous oxides have yet to be confirmed)

3. soil pH value, and

4. solution flux through soil.

Size of the soil particle: Particle-size distribution was highly variable in the 10 soils studies, ranging from sand to clay (Table 1). In fact, it was believed

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Table 3. THE PORE VOLUME IN WHICH THE ELEMENT FIRST APPEARED IN THE SOIL-COLUMN EFFLUENT

	pH	As	Be	Cd	Cr	Cu	Pb	Se	An	Hg
Wagram	4.2	1	1	1	1	20*	1	4	1	1
Ava	4.5	5	3	1	7	1	10	21	1	1
Kalkaska	4.7	1	4	2	1	19	17	10	1	1
Davidson	6.2	13	6	3	17	27	26	17	5	1
Molokai	6.2	13	12	30	19	13	22	14	27	7
Nicholson	6.7	12	5	13	5	12	16	8	7	1
Fanno	7.0	1	4	14	1	15	16	3	13	4
Mohave	7.3	1	13	13	1	19	16	2	17	5
Mohave Ca	7.8	10	6	8	1	15	23	6	11	6
Anthony	7.8	1	5	2	1	15	17	2	2	1

*Indicates that none of the elements appeared in the effluent for the listed number of pore volumes.

early in the research program that soil texture was so highly variable it would dominate to the exclusion of the other soil parameters. This was not the case. Although when the data was subjected to statistical analyses, a positive correlation between percentage of the element adsorbed and total clay content, and a negative correlation to sand were shown. Not all correlation coefficients, however, were found to be significant, since some other factors contribute to attenuation and/or element mobility.

Hydrous oxides: In general, the most significant correlations occurred between percentage trace element retained and "free iron oxide" as determined in the soil by the method of Kilmer (4). The Davidson and Molokai soils are highest in extractable free iron oxide (Table 2). These soils attenuate most trace elements more strongly than other soils (Table 3). The correlation between free iron oxide and total soil Mn is highly significant. Thus, total Mn also is closely correlated with trace element migration. The importance of total Mn, at first analyses appears to be less significant than free iron oxide. These practical soil research findings agree with the pure-system chemical research of Jenne (3) and Gadde and Laitinen (2).

Soil pH Value: Although pH value is one of the most important factors in attenuation, it is less significant than the first two factors. Using ground agricultural limestone from Kentucky in soil columns, the presence of a thin layer of lime slowed the migration rate of Cd and Ni (Tables 4 and 5, respectively). The lime effect is considered to be pH dominated in these instances. Considering total attenuation or percentage adsorption, the effect of lime is the least important for As, Cr, and Se (Tables 3 and 6).

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Table 4. THE EFFECT OF LIME ON ATTENUATION OF Cd IN WAGRAM SOIL*

Pore-space displacements	Soil alone, ppm	2 cm of lime ppm
1	15	0
2	42	0
3	46	0
4	47	0
5	37	0
6	42	0
7	56	0
8	62	0
10	75	0
11	80	0
13	86	0
15	100	T
23	--	14

*10-cm depth of Wagram soil; Cd in leachate =100ppm.

Table 5. THE EFFECT OF LIME ON ATTENUATION OF Ni IN WAGRAM SOIL*

Pore-space displacements	Soil + 2 cm limestone, ppm	Soil alone, ppm
0.8	1.5	65
2.0	9.0	90
4.0	48.0	--
10.0	66.0	100

*Each column held 10 cm (depth) of Wagram soil; Ni in leachate = 100ppm.

Table 6. THE EFFECT OF LIME AND HYDROUS OXIDES OF IRON ON ATTENUATION OF Cr IN WAGRAM AND ANTHONY SOILS

Pore-space displacements	Anthony			Wagram		
	Soil alone, ppm	2 cm lime, ppm	Fe oxide added, ppm	Soil alone, ppm	2 cm lime, ppm	Fe oxide added, ppm
0.5	--	--	--	4	10	0.3
1.0	36	34	1.8	--	--	--
1.5	--	--	--	25	30	0.35
2.0	72	75	2.0	35	45	1.0
3.0	75	73	7.5	75	78	5.0
4.0	--	--	--	--	--	--
4.5	75	75	24.0	--	--	--

*The columns held 10 cm depth of the two soils; Cr in leachate=75ppm.

These are the only selected trace elements that appear as anions in aqueous solutions. The relationships of pH to attenuations, which were found, may be summarized briefly:

1. Se and Cr show negative correlation to pH. All other trace elements are positively correlated. (See Tables 3 and 6).

2. Movement of Se and Cr relate more strongly to the soil hydrous oxide content than any other elements (Figure 1 and Table 2).

3. The initial appearance of the trace element in the solution displacements for soils with alkaline

pH values is slower than those low in pH values (Table 3) except for Cr and Se and maybe As (Figure 1 and 2).

4. Hg shows no strong trend to differences found at normal soil pH values (Table 3). Mercury migrated more rapidly when associated as an inorganic ion in water than when associated with landfill leachate (Figure 4). The presence of organic substances in the leachate appears to enhance Hg movement since concentration of other constiuents was very low.

Soil solution flux: Solution flux is one of the most important physical factors in attenuation in landfill leachate and wastewater disposal. The attenuation of trace elements in natural systems may occur during any number of possible flow regimes. By studying flux, we can describe migration under realistic field conditions. It provides another mechanism of control by which attenuation may be maximized. The soil parameter that most closely relates to flux is clay content. The importance of solution flux appears repeatedly in the research, and at times is expressed in the literature (6), yet specific data only now are being generated at quantitative levels (1). The movement of Fe and Al, for example, was greatly retarded by slight changes in density of Nicholson soil (Table 7).

Factors Considered as Unimportant in Attenuation

Those factors found to be the least important in attenuation in soils by the research at The University of Arizona are:

1. sand

2. cation exchange capacity, and
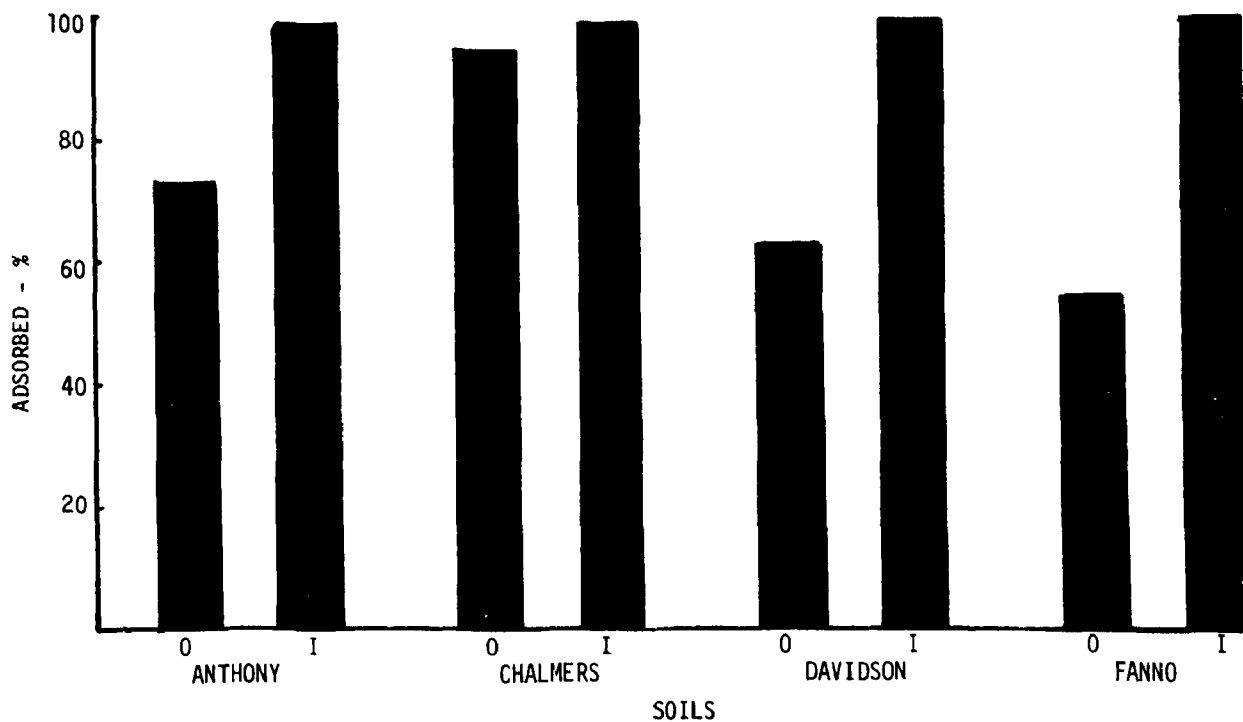
3. soil organic matter (except for Hg)

Figure 4. The absorption of mercury (HgCl) spiked into landfill leachate and water alone at pH 5.0 through four soils after 7.5 pore space displacements. 0 = Organic-Leachate; I = Inorganic-Water.

Sand: Sand is negatively correlated with attenuation. Those soils highest in quartz sand are the least retentive of trace elements. The influence of particle size distribution raises the question of the physical aspect of surface reaction and total surface area as a factor in mobility of trace elements.

Cation exchange capacity:
The cation exchange capacity (CEC) of the soils usually does not correlate significantly with trace element migration. Where CEC does seem to have some relationship, percentage clay, percentage FeO, and pH all provide a better correlation. CEC appears to be a transitory mechanism to retain ions for short periods of time. Those trace elements attached to the exchange position are clearly in a highly migratory state.

Soil organic matter: Soils having organic carbon compounds in abundance such as the Spodic

Kalkaska appeared to have little effect on attenuation except for Cu and Pb (Table 3). These two elements, however, were strongly retained by nearly all soils. The importance of soil organic matter in attenuation certainly needs further careful research; now it qualifies for a place in the next section as belonging to a group of little-evaluated or known importance. Mercury, however, appears to be more mobile in Anthony Chalmers, Davidson, and Fanno when "spiked" into landfill leachate where organic constituents are present than when in water where no organic matter exists (Figure 4).

Factors of Little Known Importance

Those factors of little known impact on attenuation appear in this group because they (a) have not received enough research attention to be evaluated, (b) have not appeared to be sufficiently dominant as to reveal a clear-cut effect, (c) appear in a state too

Table 7. A COMPARISON OF IRON AND ALUMINUM ATTENUATION AT TWO SOLUTION FLOW RATES THROUGH NICHOLSON SOIL*

Pore space displacement	Nicholson A				Nicholson B			
	Time of contact, days	Cumulate effluent volume, ml	Conc. in effluent Fe, ppm	Al, ppm	Time of contact, days	Cumulate effluent volume, ml	Conc. in effluent Fe, ppm	Al, ppm
0.5	1	330	0.2	0	1	340	0.2	0
1.0	2	690	0.2	0	4	620	0.4	0
1.5	3	1020	0.3	0	6	1040	0.5	0
2.0	4	1435	12	0	12	1435	2.6	0
2.5	5	1850	104	2	16	1725	1.6	0
2.0	6	2185	136	3	21	2115	0.3	0
3.5	7	2555	230	8	29	2685	0.7	0
4.0	8	2955	300	10	33	2960	2.2	0
4.5	9	3340	340	16	37	3235	1.8	0
5.0	10	3775	500	44	46	3805	1.6	0
5.5	11	4250	780	125	55	4420	1.1	0
6.0	12	4745	830	140	60	4750	2.0	0
6.5	13	5240	830	160	65	5070	110.0	1.0

*Soil columns were 10 cm diameter x 22 cm long. Column B was packed to slightly greater denisty than column A.

elusive to be measured quantitatively, (d) are truly marginal in effect, and/or (e) are highly specific for trace elements not included in the ones being considered now. Some of the factors that fall into this class are:

1. kind of clay mineral

2. concentration of total salts or total dissolved solids (TDS)

3. specific ion effect and ion interaction effects

4. biological mineralization and immobilization

5. reactions with organic constituents (chelation and other complexing as an example)

6. precipitation in a highly mixed and heterogeneous medium as soils

7. ion exchange reactions, and

8. physical reactions involving pore-size distribution, surface reactions, physiochemical adsorption, time, and temperature effects.

RELATIVE MOBILITY IN LANDFILL LEACHATE

Rank of Trace Elements

The relative mobility of 11 trace elements in landfill leachate through 11 soils representing 7 soil orders may be oriented as follows:

1. most generally mobile-- Cr, Hg, Ni

2. least generally mobile-- Pb, Cu

3. mobility varies with conditions--As, Be, Cd, Se, V, and Zn.

Rank of Soils

The soil that attenuates most effectively is Molokai, an Oxisol from Hawaii (Table 3 and Figures 1, 2, and 3). Davidson, an Ultisol from North Carolina, ranks second in its effectiveness to immobilize trace elements. Both soils are high in clay and extractable "free iron oxides." The Wagram Ultisol, a daolinite, quartz sand (88%), retains trace elements least effectively. The other soils vary but are generally ranked according to amount of clay-sized particles.

REFERENCES

1. Fuller, W. H., et al., 1975. "Investigation of leachate pollutant attenuation in soils," Final Report for US EPA Contract No. 68-03-0208 (in press).

2. Gadde, R. Rao, and Herbert A. Laitinen, 1974. "Studies of heavy metal adsorption by hydrous iron and manganese oxides", Anal. Chem. 46:2002-2026.

3. Jenne, E. A., 1968. "Controls on Mn, Fe, Co, Ni, Cu, and Zn concentrations in soils and water: The significant role of hydrous Mn and Fe oxides," in "Trace Inorganics in Water" Advan. Chem. Ser. 73:337-387.

4. Kilmer, V. J., 1960. "The estimation of free iron oxides in soils," Soil Sci. Soc. Am. Proc. 24:420-421.

5. Korte, Nic, J. Skopp, E. E. Niebla, and W. H. Fuller, 1975. "A baseline study on trace metal elution from diverse soil types," (Submitted for publication. In press).

6. Lapidus, L., and N. R. Amundson, 1952. "Mathematics of adsorption in beds. VI. The effect of longitudinal diffusion in ion exchange and chromatographic columns," J. Phys. Chem. 56:984-988.

MONITORING TOXIC CHEMICALS IN LAND DISPOSAL SITES*

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INTRODUCTION

Existing air and surface-water pollution abatement regulations are forcing an increasing volume of hazardous chemical waste to be diverted to the land for ultimate disposal, particularly in heavily populated, industrial regions of the United States. Land disposal of potentially hazardous chemical, bacteriological, virological, and radiological waste is now being widely employed as being the most practical and economically feasible waste disposal means available. Justification for using the land for this purpose is based on the hypothesis that earth materials have the capability to precipitate, absorb, exchange, convert, decompose, or volatilize all kinds of hazardous material to harmless states.

This form of disposal has special appeal to municipalities throughout the country presently confronted with the dilemma of upgrading inadequate sewage treatment plants to meet existing Water Quality Effluent Standards. An increasing number of air, vegetation, and water pollution occurrences from such sources seems to suggest that some landfills are serving only as partial or temporary filtration-retention beds.

Practically all of the municipal waste treatment systems in the country recieve, treat, and eventually discard liquid waste streams. Most of these waste treatment plants were designed for an earlier time when there were fewer people and when contaminants were limited to bacteria and uncomplicated organic wastes. Water pollution control is becoming more complex because of a growing roster of bacteria, viruses, antibiotics, hormones, nutrients, weedkillers, fungicides, pesticides, and trace metals, and a large number of toxic chemical compounds. Any of these pollutants may be contained in wastewater streams dumped into municipal sewers.

However, adequate dilution does not always occur before or after the waste stream reaches the treatment plant. Also, treatment by the older methods generally does not remove or render harmless all of the hazardous materials contained in the waste streams entering the plant. In fact, some of these materials are in a more concentrated form when they leave the plant in effluent and sludge than they were upon entry.

Many of the hazardous wastes present in treatment plant discharges are difficult to detect and quantitatively evaluate by available equipment and methods. They are often not identified by prescribed waste stream analyses programs. Their existence in sewage-plant

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effluent and sludge may remain
unrevealed. It is this liquid and
semi-liquid waste material that
is now being dumped on the land.

Hazardous waste pollutants
may be returned to the environment
from land disposal sites by one
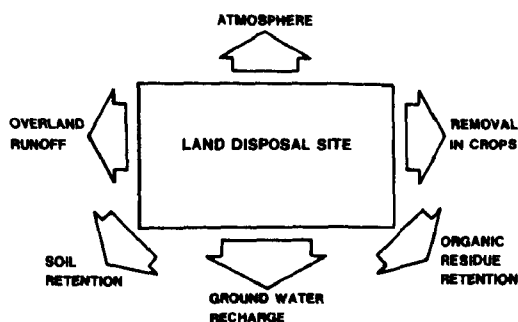or more of six avenues (Figure 1).

Figure 1. Possible avenues for
pollutants to reenter
the environment from
toxic waste land
disposal sites.

Some chemical compounds are
volatilized in the soil (for example,
hydrogen sulfide, methane, ammonia
and other nitrogen gases), and these
may enter the atmosphere and be
transported from the disposal area
in gaseous form. Other constituents,
such as phosphate, arsenic, iron,
zinc, chromium, mercury, and lead,
are retained in the soil for varying
lengths of time. Nitrate, chloride,
sulfate, boron, cyanide, and some
pesticides readily pass through
the soil to the groudwater reservoir.
Thus, many of the chemical compounds
commonly discarded on the land may
be taken up by vegetation growing
in the disposal area and returned
to the ecosystem. In fact, the
six dissipation routes shown in
Figure 1 are so interrelated that
pollutants are exchanged from one
to another.

MONITORING THE SITES

An effective monitoring system
for toxic waste disposal areas may
become a routine requirement. Such
a program would provide an evaluation
of immediate and potential long-
term pollution effects, including
total pollutant volumes retained
and those dissipated in each of
the regimes shown in Figure 1.
In practice, most monitoring
facilities are designed to detect
only surface water and groundwater
pollution. Little, if any, attention
is given to possible adverse effects
of banked pollutants reentering
the environment from soil or plant
storage areas. As a result, the
data obtained are often incomplete
and not representative at all of
the total air, plant, soil, and
water pollution that actually may
be originating from such sources.

For example, in the case of
surface water monitoring, little
if any actual measuring of overland
runoff is ever attempted. Even
if measurements are made of this
component, they usually consist
only of periodic water sampling
and analysis without corresponding
volume-of-flow measurements, which
must be collected concurrently if
an accurate quantitative evaluation
of this factor is to be obtained.
The same may be true of field
drainage tile monitoring designed
to detect pollution released from
surface soils and shallow water-
bearing formations. Also, in both
of these cases, the frequency of
water sampling generally is monthly
or bimonthly. Seldom are samples
taken during and immediately
following major precipitation runoff
periods when the major quantity
of toxic chemical removal by these
routes should be expected to occur.
This makes it unlikely that peak
or minimum pollutant concentrations
can be accurately defined.

The monitoring systems
themselves need to be studied and
updated. For instance, in fine-
grained, low permeability earth
materials generally considered to
be most desirable for land disposal
sites, a representative groundwater
sample may not be obtainable from
a properly placed monitoring well
for several weeks or even months
following installation because of
the low water-yielding

characteristics of these materials. Also, due to the usual variations in permeability of earth materials in the zone of saturation beneath landfill sites, observation wells in short vertical sections of the groundwater reservoir may not always provide representative sampling of groundwater flowing through the various aquifer segments at any given location and time.

In many existing land disposal sites, only the lower, more permeable parts of the unconsolidated earth material above bedrock are tapped by monitoring wells, even though overlying water-bearing zones are known to be present. Under such conditions (Figure 2), a major portion of groundwater pollution derived from the disposal sites may flow undetected through these overlying beds to some nearby natural or man-made drainage course.

Figure 2. Groundwater monitoring scheme using deep wells.
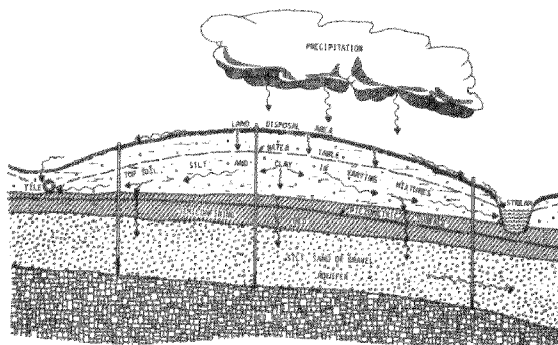
A similar situation can occur in areas underlaid by only one primary water-bearing zone if too few monitoring wells are installed, or if the wells installed tap only the uppermost part of the zone of saturation.

CORE SAMPLING

Recent studies suggest that observation of the well monitoring system may not be the most effective means to trace chemical pollutant flow paths or to determine groundwater chemical concentrations at any time or depth. Instead,

these studies show that chemical analyses of core samples from the underlying earth material profile permit a positive definition of any chemical constituent within the profile at any given location. This is true regardless of whether the chemicals are present in precipitated form in the zone of aeration, are held by retention on soil particles in the semisaturated fringe, or are dissolved in groundwater within the zone of saturation. In addition, the studies show that chemical analyses of soil core samples usually prove to be a much faster, easier, and more economical monitoring method for soil and groundwater pollution evaluation than are analyses of groundwater samples collected from observation wells.

Theoretically, the detection of pollution in any of the six dissipation regimes illustrated in Figure 1 requires only two samples--one collected prior to the beginning of the disposal operation, and another at some later time after a detectable concentration has occurred. However, a quantitative evaluation of the buildup of any given chemical constituent requires a much more elaborate sampling procedure over a longer period of time. For example, if the pollutant uptake in vegetation is to be defined, the vegetation at any given site must be sampled and analyzed before application commences, and at the end of each growing season afterward for as long as the site is used. In addition, if part of the vegetation is removed from the disposal area, samples of the harvested portion of the plant must be analyzed and representative samples of plant residue left in the field. The quantity of pollutant uptake in the water or soil at any given site may vary considerably with the season, with the rate and method of waste application, and with the total service life of the site. These factors, plus the possible toxic chemical interchange between the various regimes, make frequent sampling of all of them

mandatory, perhaps for an extended period of time after the site has been abandoned as a waste disposal site.

SAMPLING POINTS

Accurate evaluation of overall pollutant buildup, migration patterns, and flow rates within and beneath the site and surrounding area requires several strategically located stations for samples to be concurrently collected and analyzed.

The number of sampling points required is primarily controlled by the expected variability of each parameter and the degree of mon- the degree of monitoring accuracy desired. Sampling-point distribution and monitoring procedures are dictated by geologic, hydrologic, and chemical complexities likely to be encountered. Under ideal conditions, where the underlying earth materials are fairly homogeneous, impermeable, and uniformly sloping on one direction, only three sampling points should be required. These three points should be equally spaced on a line through the center of the disposal area and extending from the area of highest water table to lowest elevations on the property. In such an arrangement, the direction of groundwater flow and its chemical concentration change with distance of travel should be readily discernible if groundwater flow is uniform throughout the zone of saturation. However, earth materials may not be homogeneous, and the flow paths of groundwater through any given profile may be complex. The upper and basal surfaces and intervening strata may be warped in a manner that will influence the direction of groundwater flow and resultant horizontal and vertical migration patterns of toxic chemical movement. If more than one water-carrying stratum is present in the underlying earth material section, each water-bearing unit may have to be monitored by properly spaced, 3-station lines.

In systems such as these, the preferable pattern of sampling stations from a mathematical standpoint is a square-grid network uniformly distributed throughout the entire disposal area and the downgradient lands likely to be adversely affected. The number and spacing of sampling stations suggested is illustrated in Figure 3. Within any 10-ft by 10-ft sampling station shown in this figure, room is provided for a minimum of 25 different core-sample monitoring probes without destroying the effectiveness of that site during the life of the monitoring program.

USE OF DATA

All analyses of plant, soil, and water samples from hazardous waste land disposal monitoring systems should be made under accurate, standard-method procedures by an accredited laboratory. Also, considering the value of such data in determining the effectiveness of a disposal site's pollutant removal capability and the usefulness of these data in the design and operation of other disposal sites, it is recommended that a copy of all data collected be filed at a state or Federal scientific agency.

Background data on all potential pollution-dissipation regimes should be obtained at all sample stations just prior to the first application of waste material on the disposal land. Then after the site has been placed in service, subsequent sampling should be on a frequency and in a manner described in the following discussion on monitoring the various regimes.

Atmospheric dissipation of volatilized chemical compounds from toxic waste disposal lands generally is limited to gases such as hydrogen sulfide, ammonia, and methane. Under prevailing land disposal prac- tices, none of these ingredients are apt to be present in high enough concentrations to cause a major
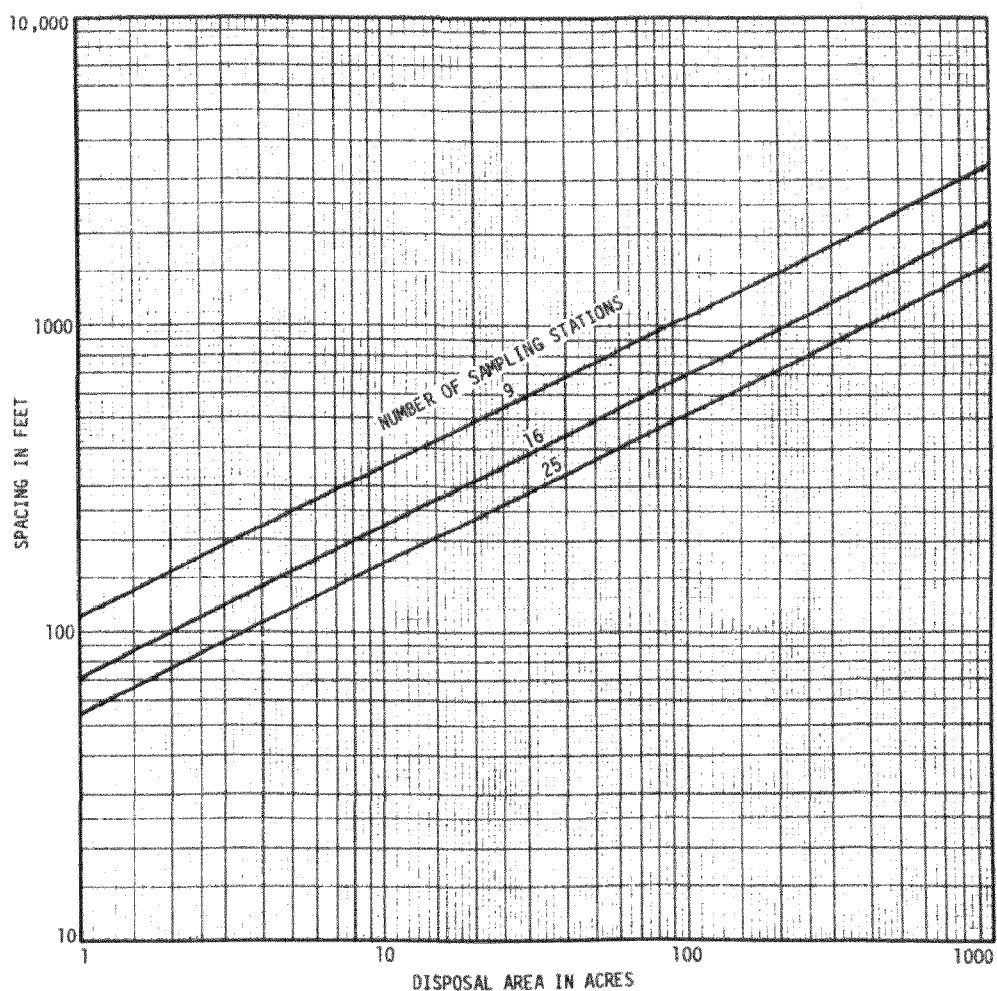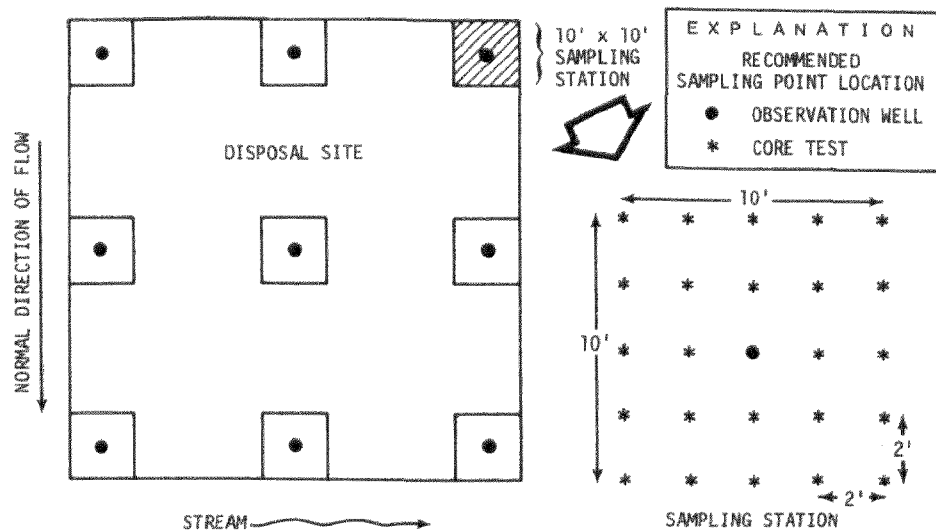
Figure 3. Recommended positioning and spacing of sample stations for toxic waste land disposal sites.

hazard to public health or pose a serious pollution threat to other parts of the environment. Specialized monitoring facilities normally should not be necessary for this particular regime.

Plant uptake of toxicants can be evaluated by using chemical analysis data obtained from a select number of composite samples of vegetation collected periodically from the sampling stations situated throughout the disposal area. The reason for this sampling is to ascertain the quantity of pollutants leaving the field in harvested portions of crops, or being left on the field as unharvested plant residue. It seems advisable to collect plant samples just at harvest time, or if the entire plant is left in the field, after the plant becomes dormant in the late fall.

Soil retention can be monitored using chemical analyses of soil core samples obtained from the entire vertical column of earth material within the disposal area and contiguous lands. A minimum of 9 and no more than 25 core test stations as illustrated in Figure 3 should be established on every individual parcel of land receiving hazardous waste material. At each of these stations core samples of the entire earth material profile should be collected twice yearly, one just after the spring groundwater recharge season ends, and the other approximately 6 months later after crops have been harvested but before the late-fall groundwater recharge period begins. In the humid northeastern part of the United States, these most desirable sampling periods occur each year between about May 1 and June 15 in the spring and from about September 15 to November 1 in the fall.

Initial soil core test probes made to obtain background data should be placed at the center of each sampling station. Subsequent cores may be taken at any of the 2-ft grid intersection points within the station. In every case, the core hole should be kept open for approximately 24 hr and then a water-level reading obtained to provide water-table gradient information. Then the hole should be refilled to the surface with compacted clay or dry bentonite. This is necessary to prevent later entry of pollution from the surface. All core tests should extend into dense impermeable clay or similar material proved by test drilling or coring to underlie the entire disposal-land area. Such an impermeable barrier to downward flow must lie at a depth everywhere greater than the lowest water-table elevation.

Since the primary reason for this sampling procedure is to determine both horizontal and vertical movements of toxicants in the earth profile, analyses should be made of at least every 5-ft interval of the core or at closer intervals if certain thinner zones prove to be carrying much of the total pollutant load.

Overland runoff from the disposal area may not ever occur. However, if it does happen frequently and in appreciable amounts, monitoring facilities must be provided to obtain representative water samples and corresponding volume-of-flow measurements. Sampling should be done throughout the entire period when overland runoff is occurring. The data can be used to plot chemical concentrations and volume-of-flow graphs. A comparable monitoring procedure can be followed if drainage-tile discharge is associated with a land-disposal project.

Groundwater toxic chemical pollution from hazardous waste land disposal sites should be insignificantly minimal if the site and contiguous land is underlaid by only dense, impermeable clay or shale deposits. In such earth materials, where the rate of groundwater movement may be less than 1 ft/yr, toxic chemical buildup and the movement of toxicants beneath affected lands can be readily and accurately defined by chemical

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analyses of earth material samples obtained from only a few strategically placed core tests as previously discussed. However, if the unconsolidated earth material above impermeable bedrock contains extensive water-bearing stringers or beds of silt, sand, or gravel, core test data may have to be supplemented by chemical analyses of groundwater samples obtained from properly placed observation wells in each water-bearing unit. Each observation well must be screened opposite only that layer under observation and isolated from all others with casing and cement grout or bentonite, from the top of the screen to land surface. All observation wells required in such cases are best placed in a cluster near the center of each sampling station. The wells installed can be measured and sampled on a routine basis so that accurate hydrographs, water table/ piezometric surface maps, and chemical constituent fluctuation graphs may be constructed from the data.

ASSESSING SYNTHETIC AND ADMIXED MATERIALS FOR LINING LANDFILLS

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INTRODUCTION

The need to develop practical and effective methods for controlling the leachate generated by water percolating through a landfill and entering and polluting the nearby groundwater system has become increasingly apparent (1, 2). Lining the landfill with an impervious barrier and diverting the leachate for final disposal is a potential method of controlling leachate. A large number of materials from other technologies are available which have been used to prevent seepage of water and various liquid wastes in pits, lagoons, reservoirs, canals, etc. (2-23). In spite of these wide uses, there is almost no information with respect to the effects of landfill leachate upon the properties and performance of these liner materials. Certainly there is no comparative information or information that can be used in selecting specific materials for use in a landfill application. It is to fill this need that the U.S. Environmental Protection Agency's Solid and Hazardous Waste Research Laboratory in Cincinnati is sponsoring this experimental study to assess 12 specific liner materials under exposure to leachate in simulated landfills.

The liner materials in this test program are now being exposed to landfill leachate. In November 1975, after 12 mo exposure, a set of these materials will be recovered and tested. A duplicate set will be recovered and tested in November 1976, after 2 yr exposure.

This paper is a preliminary report of the work carried out so far on this project. The approach and methodology to assess the materials are outlined. The experimental work performed, the significant results obtained, the observations, and the type of information that should be generated over the next 2 yr are presented.

OBJECTIVES OF STUDY

The overall objectives of this project are:

• To estimate the effective lives of 12 liner materials exposed to prolonged contact with landfill leachate under conditions comparable to those encountered in a sanitary landfill. The materials were specifically selected as being useful for lining sanitary landfills.

• To determine the effects of sanitary landfill leachate on the physical properties of liner materials after their exposure for 12 and 24 mo. Twelve liner materials (six membrane and six admix) are mounted in the bases of 24 simulated

sanitary landfills; 42 smaller specimens (of 20 additional membranes) are buried in the sand placed above the mounted liners.

• To estimate and compare the relative long-range costs of these materials as sanitary landfill liners, including materials and installation costs, and the cost benefits of better performance and longer durability of the liners.

BACKGROUND

Leachate Generation in a Sanitary Landfill

The generation of leachate in a landfill is the result of water entering the fill, percolating through it, and picking up many soluble materials and soluble products of chemical and biological reactions, which could pollute the groundwater. Water can enter a fill by such means as precipitation, a spring or groundwater draining in the fill, or accidental flooding. Sites are selected and sanitary landfills are designed and con- structed to avoid the intrusion of water (1). Leachate is generated where these conditions cannot always be met.

The composition of landfill leachate varies widely and depends on many factors, e.g., the com- position and age of the refuse, temperature, amount of available oxygen, etc. It contains inorganic and organic constituents. Table 1 shows the range of compositions of leachates from sanitary landfills. Constituents such as those given in the table should be intercepted and prevented from entering and polluting groundwater. Use of an impervious liner at the base of a landfill could prevent this by intercepting the leachate and allowing it to drain to a point

Table 1. COMPOSITION OF TYPICAL LEACHATES FROM SANITARY LANDFILLS*

Constituent	Concentration range+
Iron	200 - 1700
Zinc	1 - 135
Phosphate	5 - 130
Sulfate	25 - 500
Chloride	100 - 2400
Sodium	100 - 3800
Nitrogen	20 - 500
Hardness (as $CaCO_3$)	200 - 5250
COD	100 - 51,000
Total residue	1000 - 45,000
Nickel	0.01 - 0.8
Copper	0.10 - 9.0
pH	4.00 - 8.5

*Reference 23.
+All values except that for pH are in mg/l (ppm).

where it can be disposed of in a satisfactory manner.

Concept of Using Impervious Barriers To Control Leachate Generated in Landfills

The concept of using an imper- vious barrier as a liner for a landfill is illustrated in Figure 1 (adapted from Figure 14 in Ref. 1). An impervious material is placed upon a properly prepared surface that is graded for drainage. The amount of surface preparation depends on the specific type of liner material being installed and on the soil base on which the liner is being placed. This base must be free of stumps and rocks and should be compacted. The liner material can be compacted native soil, asphaltic concrete, polymer membranes, or other artificial barriers. Above the barrier is placed a porous soil, on which is placed the compacted refuse in the manner normally used in sanitary landfills. Leachate generated by water percolating through the refuse
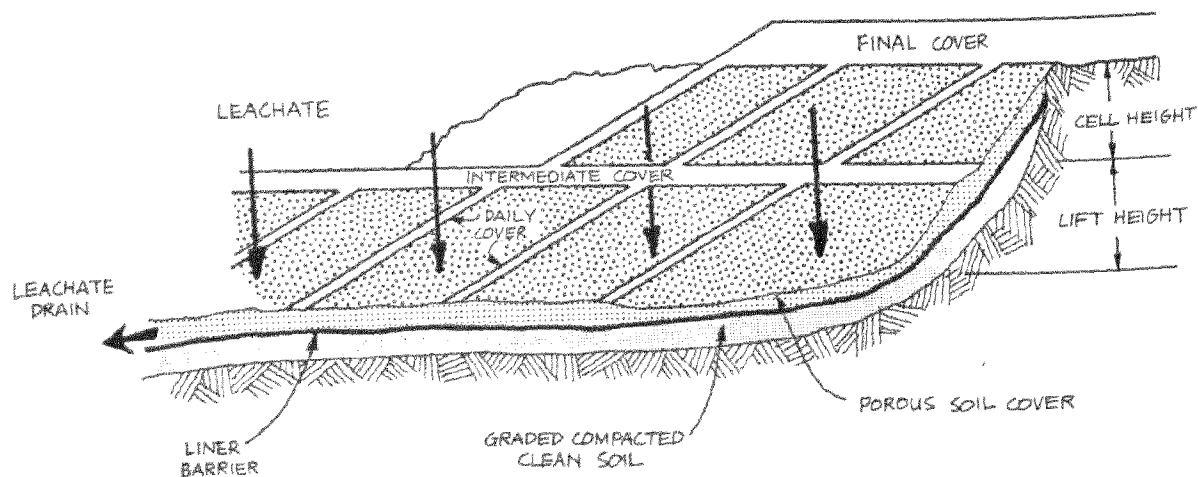
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Figure 1. Concept of lining a sanitary landfill.

will be intercepted by the barrier, drain through the porous soil, and be collected for ultimate disposal in a sanitary sewer system or a leachate treatment system, or be recycled through a landfill to hasten stabilization of the fill.

Environment of a Barrier in a
Sanitary Landfill

The environment in which an impervious barrier must exist will ultimately determine how well it can service and function for long periods of time. Some of the environmental conditions at the base of a landfill should have no adverse effect on life expectancy of a given material, whereas other conditions could be quite deleterious. Some important conditions that contribute to the life of a barrier are:

1. The barrier is placed on a prepared surface that has been graded, to allow drainage, and compacted and is presumably free of rocks, stumps, etc., but that may settle to cause cracking of hard liners. A brittle or weak material would fail.

2. Anaerobic condition with no oxygen to cause oxidation.

3. No light, which normally degrades many polymeric materials.

4. Generally wet-humid conditions, particularly if leachate is being generated regularly, that could result in the leaching of ingredients from a liner.

5. Cool temperatures of 40 to 70 F normally, although high temperatures can be generated within the fill if aerobic decomposition takes place.

6. Generally acidic conditions from the leachate.

7. High concentration of ions in the leachate that may exchange with clay soil and reduce impermeability.

8. Considerable dissolved organic constituents in the leachate that may degrade some of the organic material liners.

9. Only modest head pressure, since drainage above the liner is designed to take place continually. A porous soil is placed on top of the liner before

refuse is placed. Such a condition may allow less impervious liners if good drainage can be maintained.

The effects of these environmental conditions will differ on the various barrier materials. However, it appears at present that mechanical failure during installation or

during operation of the fill due to settling of the soil may be the most significant source of failure of a liner.

Potential Materials for Lining Landfills

Typical of the wide range of materials that have been or are being used as barriers to the seepage of water and hazardous toxic wastes in holding ponds, pits, lagoons, canals, reservoirs, etc., are those listed in Table 2. Selection of liner materials for a specific job depends upon the type of fluid or waste being confined, the types of materials that can perform, the lifetime needed, and economics. Often several materials can be used and the choice then becomes one of economics and the length of time which the liner should function. At times it may be desirable to use combinations of materials.

Table 2. POTENTIAL MATERIALS FOR LINING SANITARY LANDFILLS

Compacted native fine grain soils

Bentonite and other clay sealants
- Bentonite-polymer sealants

Asphaltic compositions
- Asphalt concrete
- Hydraulic asphalt concrete
- Preformed asphalt panels laid on concrete surfaces
- Catalytically blown asphalt sprayed on soil
- Emulsified asphalt sprayed on soil or on fabric matting
- Soil asphalt
- Asphalt seals

Portland cement compositions
- Concrete with asphalt seals
- Soil cement with asphalt seals

Soil sealants
- Chemical
- Lime
- Rubber and plastic latexes
- Penetrating polymeric emulsions

Liquid rubbers sprayed
- Rubber and plastic latexes
- Polyurethanes

Synthetic polymeric membranes
- Butyl rubber
- Ethylene propylene rubber (EPDM)
- Chlorosulfonated polyethylene (Hypalon)
- Chlorinated polyethylene (CPE)
- Polyvinyl chloride (PVC)
- Polyethylene (PE)

GENERAL APPROACH TO EVALUATION OF LINER MATERIALS

Taking into account the wide diversity in the types of materials that are being considered for lining landfills and the urgent need for information regarding the relative merits of the various liners and their expected lifetimes in a landfill environment, the following overall approach is being taken:

1. Select for exposure testing 12 specific liner materials from the various types of liner materials that have been successfully used in lining pits, ponds, lagoons, canals, etc., to prevent seepage of water or various wastes and that appear suitable for lining sanitary landfills. Cost factors and performance requirements suggest thin liners, which also should accelerate the effects of leachate.

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2. Expose liner specimens individually to leachate under laboratory conditions that simulate as closely as possible those conditions a liner would encounter at the bottom of a real landfill. The simulated sanitary landfills should be so designed and constructed as to ensure anaerobic conditions, and the leachate generated should be representative of the leachate generated in sanitary landfills.

3. Expose specimens of sufficient size so that physical tests can be made to measure the effects of exposure to leachate and, if appropriate, a typical seam can be incorporated for testing.

4. Subject the liner specimens to appropriate tests for the specific type of liner. Properties would be measured that could be expected to reflect on the performance of the liner in sanitary landfills. The tests shown in Table 3 are those used in evaluating membrane liners. The tests of the admix liners are given in Table 4.

5. Measure the properties of the specimens before exposure and after 1 and 2 yr exposure to leachate. Assess the performance of the respective liners, and from the changes in their properties, estimate their respective lifetimes in the landfill environment.

A primary objective of this project is to make an estimate of the respective lives of the 12 liner materials in the landfill environment. To make such an estimate, it is necessary to determine what

Table 3. TESTING OF POLYMERIC MEMBRANE LINERS

- Water permeability, ASTM E96.

- Thickness.

- Tensile strength and elongation at break, ASTM D412.

- Hardness, ASTM D2240.

- Tear strength, ASTM D624, Die C.

- Creep, ASTM D674.

- Water absorption or extraction at RT and 70 C, ASTM D570.

- Splice strength, in peel and in sheer, ASTM 413.

- Puncture resistance--FED. Test Method Std. No. 101B, Method 2065.

- Density, ash, extractables to assess composition.

Table 4. TESTING OF ADMIXED LINER MATERIALS

Permeability	Back pressure permeameter (Ref. 24)
Density and voids	ASTM D1184 and D2041
Water swell	California Division of Highways 305
Compressive strength	ASTM D1074
Viscosity, sliding plate of asphalts	California Division of Highways 348
Microductility of asphalts	California Division of Highways 349

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constitutes the point of
failure of the liner.
The function of a liner
is to reduce or prevent
leakage of leachate. When
seepage or leakage through
a liner reaches an "unsat-
isfactory" level, then
the liner has failed.
Such failures could arise
from:

-- Cracking, breaking,
or tearing of the liner
due to ground settling,
rocks, etc.

-- Puncturing due to rocks,
stumps, plant growth, etc.

-- Failure of the seams

-- Disintegration of the
liner due to solution by
or reaction with the
leachate.

The ability of the liner
material to maintain its
integrity in the environment
will be measured by
observing the seepage
through the liner and the
changes in properties
during exposure to leachate.

6. Seal the liner specimens
in individual simulated
landfills so that whatever
seepage might come through
can be collected and tested.
This required special
efforts to avoid leachate
by-passing the liner or
channelling through the
liner, particularly in
the cases of soil cement
and soil asphalt liners.

7. Create equal conditions
in all simulated fills,
so that valid comparison
between liners can be made.
To accomplish this, fill
the simulated landfills
with well-compacted,
shredded municipal refuse.
Compaction, composition,
and amount of refuse should
be as equal as possible

in each of the 24 cells
so that a relatively highly
concentrated and equal
leachate is generated in
all the cells.

8. Determine the composition
of the shredded refuse
from a blend of grab samples
taken during the loading
of the cells.

9. After the refuse in the
cells is saturated, i.e.,
brought to "field capacity,"
generate leachate by adding
1 in. of tap water every
2 wk (26 in./yr) and allow
leachate to pond on the
liner at a depth of about
1 ft by draining and
collecting leachate every
other week.

10. Monitor the simulated
landfills and characterize
the leachate during exposure
period.

Testing of the leachate has
been limited to determining
temperature, levels of leachate
in generators before collection,
amount of leachate collected, total
solids, pH, chemical oxygen demand
(COD), total volatile acids (as
acetic acid), acetic acid, propionic
acid, isobutyric acid, butyric acid,
isovaleric acid, valeric acid, and
caproic acid. These tests are
needed to characterize the leachate,
to test for the uniformity of the
leachate among the various
generators, and to measure some
of the organic constituents of the
leachate. Most of the liner
materials under test are organic
in composition. It can be expected
that some of these organic components
of the leachate could swell or
otherwise deteriorate the liner
materials. Of particular interest
is the butyric acid, which can
degrade many rubber liners used
in chemical equipment.

In addition to the six membrane
liner materials that are mounted
and being tested as barriers in
the bases of the simulated landfills,

approximately 20 additional membranes had undergone preliminary testing and were available for exposure testing. As these materials included variations in suppliers, thickness, composition, and use of fabric reinforcement, their exposure to leachate could give additional information as to the effects of these variables. Also, it would give us the opportunity to check various adhesion systems and to determine whether exposure to leachate on both sides of a test specimen would yield the same results as exposure to one side, as encountered with the barrier arrangement.

Consequently, strips (2.25 in. X 20 in.), most of which incorporated seams, were coiled and buried in the sand above the barrier specimens. Most of the tests to be performed on the larger barrier specimens can be performed on these strips.

SELECTION OF SPECIFIC MATERIALS TO TEST AS BARRIERS IN SIMULATED LANDFILLS

The selection of the specific specimens of liner materials for testing as barriers involved two major steps:

    1. selecting a broad class of membrane and admix types of material

    2. within each class, selecting a specific material or composition.

At the outset, the Solid and Hazardous Waste Research Laboratory specifically eliminated soils, as they were being tested in other studies. The following factors were considered in selecting the 12 classes of materials that would be included in the test program:

    1. inclusion of as broad a range of materials as possible

2. successful past use of material as barriers to prevent seepage of water or various wastes in pits, ponds, lagoons, canals, etc.

3. level of permeability offered by the material

4. anticipated ability of the material to resist changes in permeability and physical properties when exposed to leachate in landfill environments

5. compatibility of the materials system with landfill operations

6. costs of materials and installation.

After selecting the broad class of material, a specific material had to be selected. In making this final selection, the following factors were considered:

    1. thickness of each material selected would be typical of that normally employed, except that thin liners would be selected so as to accelerate the effects of leachate

    2. high quality compositions would be used; e.g., in selecting specific membranes from those available, the membrane of a given class with best physical properties was generally selected.

We did not attempt to get liner samples from all possible liner producers, but tried to select specific liners that were representative of the respective classes of materials.

The specific liners selected and mounted as barriers in the bases of the simulated landfills include six flexible synthetic polymeric membranes and six admix liner

materials. They are listed in Table 5 with their respective thicknesses.

Flexible Synthetic Polymeric Membranes

This group of liner materials is based on a series of synthetic rubbers and plastics produced from petrochemical sources. These polymeric materials are used in a wide range of products and represent considerable variation in chemical and physical properties. Each is generally compounded with other ingredients, e.g., fillers, plasticizers or oils, antidegradants, curatives, etc. Narrow sheeting or membranes are produced by calendering or other coating processes as unsupported film or sheeting, or with fabric reinforcement to increase tear strength, particularly for installation (25). These membranes or sheeting are seamed in a factory

Table 5. LINER MATERIALS SELECTED FOR LEACHATE EXPOSURE TESTS

Material	Thickness
Polymeric liner membranes	
Polyethlyene (PE)	10 mils
Polyvinyl chloride (PVC)	20 mils
Butyl rubber	63 mils
Chlorosulfonated polyethylene (Hypalon) nylon scrim reinforced	34 mils
Ethylene propylene rubber (EPDM)	51 mils
Chlorinated poly- ethylene (CPE)	32 mils
Admix materials:	
Paving asphalt concrete	2.2 in.
Hydraulic asphalt concrete	2.4 in.
Soil cement	4.5 in.
Soil asphalt	4.0 in.
Bituminous seal	0.3 in.
Emulsion asphalt on fabric	0.3 in.

or shop to make large prefabricated panels that are transported to the pit site and assembled in the pit to form the completed liner. The following operations, therefore, become involved in the ultimate installation of membrane liners:

1. production of the polymer

2. manufacture of the rubber and plastic sheeting or film

3. fabrication of prefabricated liners

4. field installation of the liners.

A single organization can be involved in two or more of these steps such as in the PVC liner industry.

The composition of the polymeric liners can vary considerably amoung the various producers and to some extent between quality lines of a given producer. Consequently, care must be taken in generalizing on the performance of a given polymer in membrane liners.

Comments on the specific polymeric liner materials used in the test are given below:

Polyethylene (PE). Films of PE have a very simple composition consisting primarily of the hydrocarbon polymer, polyethylene, plus a small amount (ca.1%) of an antidegradant and a few parts of a carbon black. Thus, the potentially extractable fraction of this film is very low. These membranes are relatively low in cost and have a great resistance to bacteriological deterioration. They can be heat sealed in the factory to fabricate large panels, but are usually seamed with gum tape in the field in assembling the final liner. PE liners must contain black to be resistant to light. Although PE is more impermeable to water than is plasticized PVC, the PE films normally available for agricultural

and industrial applications occasionally have pinholes and blisters and, therefore, in practice are not as watertight as the PVC films. Some pinholes were encountered in the film as received; however, the sections mounted as barriers are carefully selected so that no pinholes were present.

Plasticized Polyvinyl Chloride (PVC). Films of this composition are the most widely used flexible liners. They are available in 10- to 30-mil thickness; the bulk is used as unsupported film, and the remainder, with fabric reinforcement. The PVC compound contains 30% to 50% of one or more plasticizers to make the films flexible and rubber-like. It also contains 2% stabilizer or antidegradant and, at times, fillers. There is a wide choice of plasticizers that can be used with PVC, depending upon the application and service conditions under which the film will be used. PVC generally holds up well in burial tests; however, in some liner applications, PVC films have deteriorated, presumably due to the specific plasticizer. Some plasticizers can be degraded by microorganisms and are soluble to a limited extent in water. On exposure to weather with its wind, sunlight, and heat, PVC liner materials can deteriorate badly due to loss of plasticizer and to polymer degradation. Plasticized PVC films are quite resistant to puncture, relatively easy to splice, and available in wide sheets.

Butyl Rubber Sheeting. Butyl rubber is a copolymer of a major amount of isobutylene (97%) and a minor amount of isoprene to introduce unsaturation in the rubber as sites for vulcanization. A vulcanized butyl rubber compound is used in the manufacture of the sheeting, which is available in either unsupported or fabric-reinforced versions of 20- to 125-mil thickness. Butyl rubber has excellent resistance to permeation of water and swelling in water. The permeability factor for butyl rubber is 0.119 perms per mil.

Giving butyl a relative water permeability index of 1.0, PE rates 1.9 and PVC, 59. Butyl rubber has poor resistance to hydrocarbons, but is quite resistant to animal and vegetable oils and fats. The butyl rubber compounds have good resistance to water and contain low amounts of extractable material. Overall they age very well, although some compounds will ozone crack on long exposure. In outdoor exposure in water management use, butyl rubber sheeting has shown no degradation after 20 yr of service. Some of the recent compounds contain minor amounts of EPDM to improve ozone resistance. Obtaining good splices of butyl sheeting, particularly in the field, continues to be a problem.

Chlorosulfonated Polyethylene Sheeting (Hypalon). This synthetic rubber is made by the chloro-sulfonation of the plastic polyethylene. In liners, it is used in unvulcanized compounds containing at least 45% rubber. The other ingredients are predominantly fillers. Most of the Hypalon liner sheeting is made with fabric reinforcement (e.g., nylon scrim). It and has good puncture resistance, is easy to splice by cements, solvents, heat and/or mechanical "zipping," and has good characteristics with respect to aging, oil resistance, and bacterial resistance. It has been reported successfully used for lining holding pits and ponds in mining operations where highly acid-contaminated fluids are encountered. After PVC, it is the most used of the polymeric flexible liner materials.

Ethylene Propylene Rubber (EPDM). This synthetic rubber is a terpolymer of ethylene, propylene, and a diene monomer that introduces a small number of double bonds into the polymer chain, which are sites for vulcanization of the rubber. The unsaturation in the side chain of the polymer material and not in the main chain of the polymer imparts good ozone, chemical, and aging resistance. The rubber is

compatible with butyl and is often added to butyl to improve resistance of the latter to oxidation, ozone, and weathering. As it is a wholly hydrocarbon rubber like butyl, EPDM sheeting has excellent resistance to water absorption and permeation but has relatively poor resistance to some hydrocarbons. It is available in sheetings of 20-to 60-mil thickness, both unsupported and fabric reinforced. Special attention is required in splicing and seaming this material.

Chlorinated Polyethylene (CPE). This relatively recently developed polymer is an inherently flexible thermoplastic produced by chlorinating high density polyethylene. Sheeting of CPE makes durable linings for waste, water or chemical storage pits, ponds or reservoirs. CPE withstands ozone, weathering and ultraviolet and resists many corrosive chemicals, hydrocarbons, microbiological attack and burning. Compounds of CPE are serviceable at low temperatures and are nonvolatile.

Results of the laboratory testing of these six polymeric liner materials, as received from the respective suppliers and prior to exposure to leachate, are given in Appendix A.

Admix Liners

The admix or formed-in-place liner systems include hard surface linings and soil sealants. They are made by:

1. importing an admixed material, such as asphalt concrete, and placing it in thicknesses of 2 in. or more

2. mixing Portland cement or asphalt with the in-place soil (or sometimes with imported soil) to form a hard surface 4 to 6 in. thick

3. spreading on surface sealant materials, such as emulsion

seals, rubber latexes, resin solutions, expanding clays or various forms of asphalts.

The four hard surface liners being tested are asphalt concrete, hydraulic asphalt concrete, soil cement, and soil asphalt. Hot-sprayed canal lining asphalt and a cold-applied asphalt emulsion sprayed on fabric are the two soil sealants being tested.

Asphalt Concrete and Hydraulic Asphalt Concrete. Conventional asphalt concrete, hot-mixed and hot-laid, is widely used for paving and is readily available. Contractors are experienced in its placement and have the necessary equipment. It presents a hard surface resistant to traffic and impact forces. It is resistant to acids and to aging, especially in the absence of light and air. It is designed to have a voids content of about 5%, necessary for the stability required for pavements. It is, therefore, not completely impervious and may require a surface treatment in-situ to seal the voids. None was applied in the asphalt concrete being tested in this project.

Hydraulic Asphalt Concrete. This concrete, also hot-mixed and hot-laid, is specially designed to be impervious. Imperviousness is achieved by controlling the gradation of the aggregate and the asphalt content to obtain a virtually voidless structure after compaction. Because it is voidless, it is more susceptible to displacement and rutting under traffic than conventional asphalt concrete and, therefore, is not suitable for highway pavement. Its other properties are similar to asphalt concrete. Hydraulic asphalt concrete is mixed, laid, and compacted with the same equipment used for conventional asphalt concrete, but is more difficult to handle. Its cost is higher because of the extra work necessary in handling, the more stringent gradation requirements and the higher asphalt content.

Soil Cement. Soil cement is made by mixing the in-place soil with Portland cement and water, and compacting the mixture. As the Portland cement hydrates, the mixture becomes a hard, low-strength Portland cement concrete. Soil cement is sometimes used as a surface for pavements with low-traffic volume, and is extensively used for the lower layers of pavements, where it is called "cement-treated base." Strong soil cement can be constructed with many types of soil, but permeability varies with the nature of the soil: the more granular the soil, the higher the permeability. With fine-grained soils, soil cements with permeability coefficients of about 10^{-6} cm/sec are achievable. In practice, surface sealants are often applied to the soil cement to obtain a more waterproof structure. Aging characteristics of soil cement are good, especially under conditions where wet-dry and freeze-thaw cycling are minimal. Some degradation of the cement can be expected in an acid environment.

Soil from the Radum quarry near Pleasanton, California, which has been proposed as the site of a future landfill, was used for preparation of the soil cement, with Type 5 (sulfate-resistant) Portland cement. Since the fines content of the Radum soil was lower than optimum for soil cement, a few percent of nonswelling clay (kaolin) was added for some of the tests.

Soil Asphalt. Soil asphalt of mixed-in-place asphalt surfacing is made by mixing a liquid asphalt with the in-place soil or with imported aggregate. It is widely used for low-cost pavements for low volume traffic. Permeability characteristics can be controlled by the amount and type of asphalt added. Soil asphalt is more flexible and resistant to cracking than asphalt concrete or soil cement. It is resistant to acid and has good aging characteristics in the absence of light.

Liquid asphalt grade SC-800 was used to prepare the barrier specimens because it is essentially nonvolatile; MC-type liquid asphalts would be expected to leave voids when the kerosene diluent evaporated.

Laboratory specimens prepared using soil from three different locations showed that impermeable soil asphalt could be made from any of the three. Soil from the Radum quarry was selected because the location may become a landfill site.

Bituminous Seal--Catalytically Blown Asphalt. Bituminous seals of buried asphalt membranes have been used extensively as linings for canals and reservoirs and to seal off layers of expansive soils under pavements. This type of asphalt is produced by air-blowing in the presence of a catalyst (phosphorous pentoxide or ferric chloride), which produces an asphalt which has a high softening point, yet remains flexible at low temperatures. Membranes are applied to compacted, smooth soil surfaces by spraying the hot (200 to 220 C) asphalt in two successive applications to ensure a continuous film free of pinholes and holidays. Recommended application rates are 4.5 to 6.8 kg/sq m (1 to 1.5 gal/sq yd) to form a film 5- to 8-mm thick (3/16 to 5/16 in.). When cooled, the membrane is flexible, tough, and impervious to water. It is resistant to acids, but not to oily materials. Aging resistance is good when protected from light. It is usually covered with a protective layer of soil to prevent damage by traffic and deterioration by light.

Bituminous Seal--Fabric Plus Asphalt Emulsion. Emulsions of asphalt in water can be applied at temperatures above freezing. They form continuous films of asphalt after breaking of the emulsion and evaporation of the water. The films are less tough and have lower softening points than films of hot-applied, catalytically blown asphalt.

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Toughness and dimensional stability can be achieved by spraying asphalt emulsions onto a supporting fabric. Fabrics of woven jute, woven or nonwoven glass fiber, and nonwoven synthetic fibers have been used with various anionic or cationic asphalt emulsions to form linings for ponds and canals and as reinforcing patches under asphalt concrete overlays to prevent "reflection" of cracks in the old pavement beneath. Seams in the supporting fabric are often sewn with portable sewing machines after the fabric is placed. Nonwoven polypropylene fabric coated with asbestos-filled anionic asphalt emulsion was supplied already prepared for installation as barriers.

Physical test data and composition information on the admix liner specimens which were mounted as barriers in the simulated landfills are presented in Appendix B.

Seaming of Liner Specimens

Critical to the effective performance of polymeric membrane liners for ponds and sanitary landfills is the capability of making large impervious sheets of them. The liners are manufactured in relatively narrow widths of sheeting that must be spliced together either in the field or in the factory to make continuous, large, impervious sheets, sometimes many acres in area. Usually the panels are prefabricated from the narrow sheets in the factory or shop, brought to the site, and then spliced together in the field.

Therefore, in a normal field installation there are both factory and field splices. In the usual, favorable factory environment, more durable seams can be made using electronic sealing, "solvent welding," or possibly heat curing adhesives. Seaming in the field can pose many problems.

As seams may be the weak point in installed liners and the most

likely source of liner failure, it was decided to incorporate seams in all the test specimens being exposed. Either factory seams or the recommended practice for the specific membrane being installed were used. Adhesives are often designed for specific films, depending upon the polymer and even upon the specific compounding recipe used. It may not be possible to use a given adhesive designed for a given polymer for all sheetings made of that type of polymer.

Adhesives systems recommended by the liner suppliers for the various membranes are being tested. Test joints have been incorporated in the test strips buried in the sand above the barriers in the leachate generators.

CONSTRUCTION OF SIMULATED LANDFILLS AND INSTALLATION OF LINER SPECIMENS

Design and Construction of the Simulated Landfills

The design of the individual simulated landfills (Figure 2) has the following features:

1. It is made of two parts, a concrete base in which the barrier and strip specimens are placed and a 2-ft-diameter steel pipe, 10 ft high, in which the ground municipal refuse is compacted. This design allows for easy dismantling and recovery of the exposed specimens of liner materials.

2. The liner test specimen, 2 ft in diameter, is mounted in the concrete base, the interior of which is coated with epoxy resin.

3. The liner test specimens are sealed in place so that seepage can only be through the liner specimens

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and, thus, the permeability of the liner can be measured.

4. A 10-ft-high, 2-ft-diameter steel pipe, with a 2-in. flange of 0.25 steel at the bottom, was placed and sealed on the base, then filled with ground municipal refuse. The pipe is made of 12-gauge spiral weld steel.

5. The polyethylene that lines the pipe was selected over epoxy resins because of its low cost and its acknowledged inertness to degradation.

Figure 2. Schematic drawing of leachate generator and cell in which the liner materials are being exposed to leachate under conditions simulating sanitary landfills.

6. Collapsible plastic bags are used for collecting the leachate to avoid possible entry of air into the generators.

7. There is a system for collecting leachate both above and below the liner barrier.

8. To determine the level of leachate in these simulated landfills, standpipes were placed in four of them at the outlet above the liners.

Twenty-four of these simulated landfills have been constructed at the Sanitary Engineering Research Laboratory of the University of California, Berkeley. The site, at the Richmond Field Station of the University, on San Francisco Bay, has a moderate and uniform temperature over the entire year, mostly in the 55 to 60 F range.

The 12 liner materials were mounted in duplicate in the 24 generators. Twelve of these generators will be dismantled and the liners removed and tested after 12 mo exposure to leachate and the other 12 after 24 mo exposure.

Concrete Bases. The concrete bases were cast individually using a steel form in the top to give the interior shape of the base. This base has a horizontal ledge about 6 in. below the rim on which the liner specimens were mounted and sealed (Figure 3). The irregularities in the interior of the base were filled with an epoxy resin (Colma-Dur-Sika, Lyndhurst, New Jersey)--sand grout. In addition, because of the irregularity of the rim of the concrete bases, a 3-in.-wide ring of epoxy-sand was cast on the top to give a smooth mating surface against which the flange on the pipe could be placed. The interior surface of the bowls and the outside top surface of the bases were coated with an epoxy
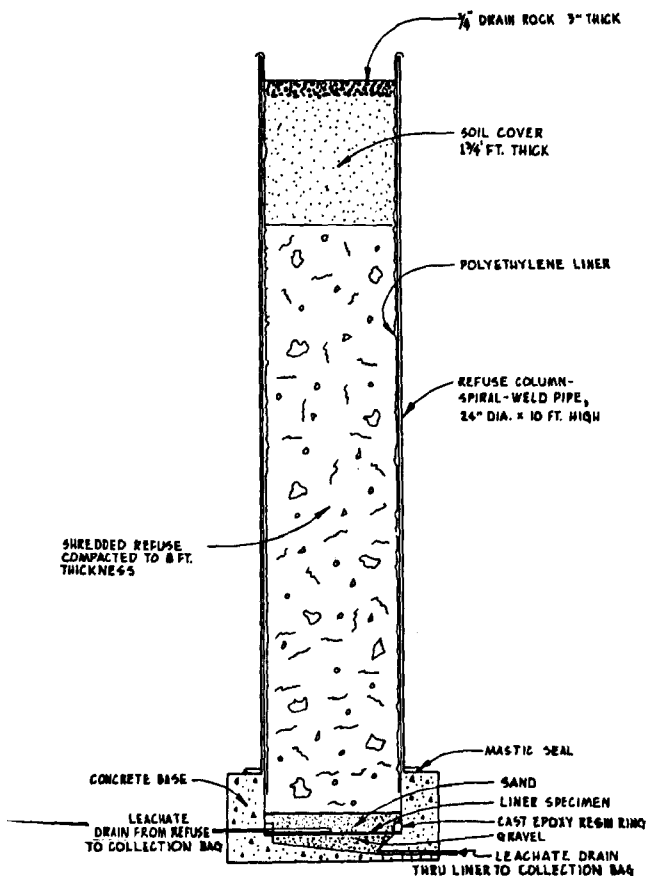
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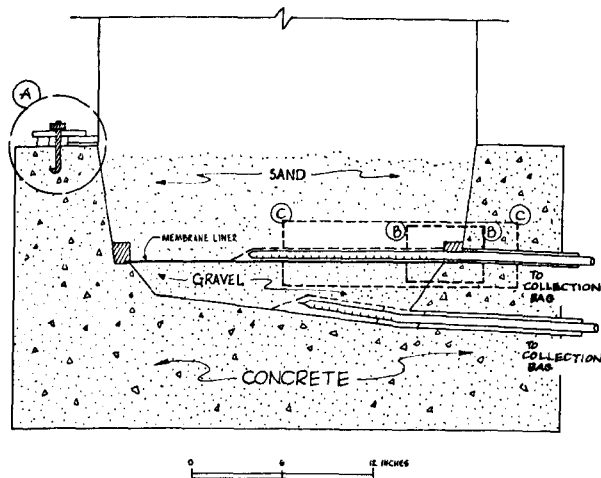
Figure 3. Base of leachate generator with membrane barrier.

DETAIL "A"

ATTACHMENT, STEEL REFUSE CELL TO CONCRETE BASE.

DETAIL "B"

EPOXY JOINT, FILM TO BASE AND DRAIN TUBE
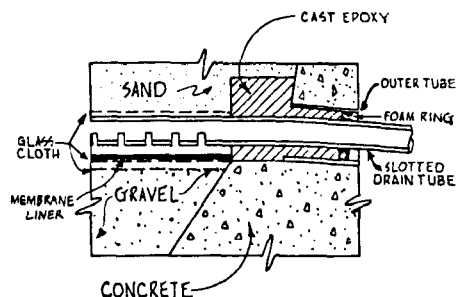
Figure 4. Details of construction.

resin (Concresive 1170--Adhesive Engineering Company, San Carlos, California) to ensure air and water tightness. Each completed base was spark and water tested to determine if there were any leaks through the coating.

Drainage of Leachate. A crushed, washed granite gravel, selected to give a stable base on which to compact the soil cement and the soil asphalt, and later the refuse, was placed in the lower part of all the bases. A piece of glass fiber cloth was placed above these stones to reduce the possibility of puncture during the compaction of the refuse on top of the membrane liners. Sand was placed over the membranes to protect them from possible damage by sharp pieces of refuse. Thin-wall PVC tubes cast into the concrete bases provided conduits for installation of the drainpipes. ABS resin pipe was used for drainage and sealed into place in accordance with detail B in Figure 4. The pipe was slotted, as shown in the drawing, and covered with a piece of glass fiber cloth to prevent sand from clogging the slots.

Liner Seal. All the liners were sealed into place by casting an epoxy ring around the periphery after they had been placed on the

bases. The membrane liner specimens were perforated around the periphery (Figure 5), and cast epoxy flowed into these perforations to form pins and, thus, prevent the liner from becoming dislodged during compaction of the refuse. Epoxy resin, Concresive 1217 (Adhesive Engineering Company, San Carlos, California), was mixed with a fine gravel and used to make these rings. The same procedure was also used in all the admix specimens. The admix liners, which were formed in place, have no annular space between concrete base and specimen.

Sealing the Steel Pipes to the Bases. Details of sealing the steel pipes onto the bases are shown in Figure 4. To make a smooth mating surface for the flanges, which had been welded onto the ends of the pipes, an epoxy ring was cast on the surface of the

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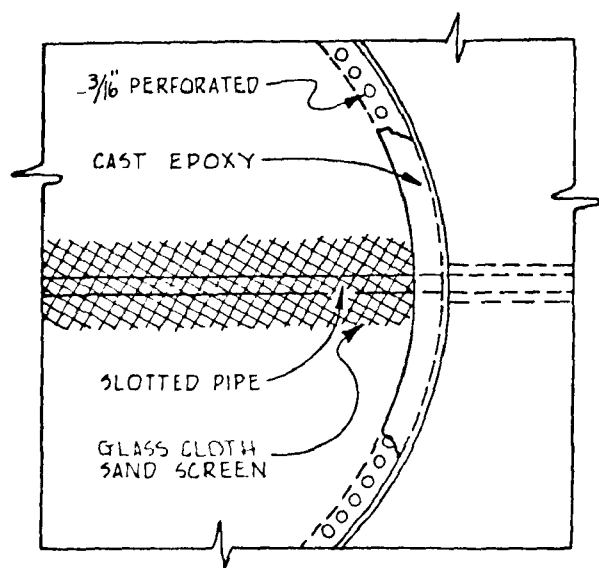
DETAIL "C"

SEAL, FLEXIBLE FILM IN BASE

Figure 5. Detail of mounting membrane liners.

concrete. Closed-cell foam gasket material of 1/4-in. neoprene was placed between the epoxy ring on the base and the flange. After the refuse had been loaded into the generators, a bead of mastic seal was placed around the periphery of the flanges.

Temperature Within the Generators. Two thermocouples were placed in each of four generators, one at the surface of the liners and another about 1 ft above the liners. (Measurements to date have shown temperatures in the generators to be less than 66 F (18.9 C) since the time the refuse was placed.)

Leachate Collection System. The system devised to collect leachate both above and below the liners and yet maintain the anaerobic conditions within the generators is shown in Figures 6 and 7. It features:

1. Polyethylene bags with two outlets.
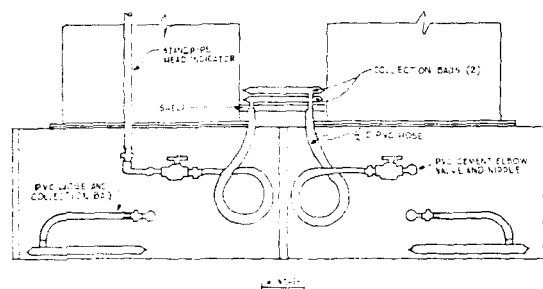
Figure 6. Leachate collection system.
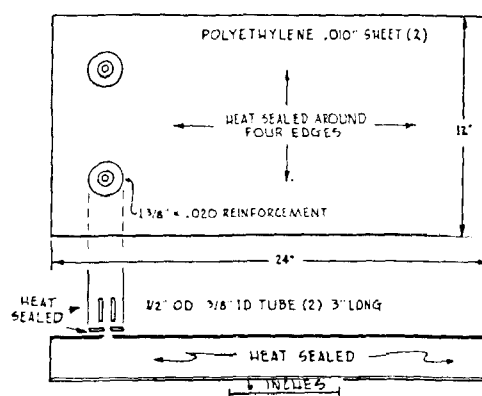
Figure 7. Leachate collection bag.

2. A valve on the outlet of the drainpipe from above the liner that is kept closed except at the time of draining of leachate. The capacity of the bag is about 2 1/2 gal. The drain from below the liner will be kept closed unless leachate passes through the liners, at which time a similar, smaller plastic bag will be mounted and the leachate will be allowed to drain continuously.

Collection and Grinding of Refuse And Loading of Simulated Landfills

Approximately 12 tons of refuse were collected by the Palo Alto Sanitation Company in the residential area of Palo Alto, California, and were ground by Combustion Power Company, Menlo Park, California, over a period of a week in an Eidal Mini-Mill Grinder (Model 100) without classification. The shredded refuse was delivered to the Richmond Field

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Station in three loads and was systematically loaded into the 24 generators on a rotating basis.

The residential refuse delivered by the packer truck to Combustion Power Company was obviously highly inhomogeneous. Yet, to make a good evaluation and comparison of the effects of leachate on the 12 different liner materials, the refuse in each of the generators should have the same composition, as should the leachate generated.

To accomplish this, good blending of the refuse and uniform loading and compacting of the refuse in the generators were required. Some blending of the contents of the packer truck occurred on the floor of Combustion Power Company and in the feeding and grinding of the refuse in the Eidal shredder. However, the grinding and delivery of the refuse took place in three different loads and each had a different overall appearance. To ensure that the same composition and amount of refuse was placed in each of the generators, we established the following plan for loading the refuse into the generators:

1. The refuse was weighed into 30-lb aliquots and dumped into the generators one after another in rotating order.

2. After each load, the refuse was compacted, using the two hand compactors designed for this purpose. One features dowel rods on a circular board to simulate a sheep's foot roller, such as is used in the compaction of soil, and the other was concrete cast in a polyethylene bottle.

3. Care was taken during the first few loads to avoid heavy compaction because of the possible damage to the liners and disturbing the test specimens buried

in the sand above the barriers. The refuse was difficult to compact, however, because of its dryness. Consequently, 1 gal of water was added to each load after weighing and before it was introduced into the generators. In addition, the weight of the individual loads per lift was reduced, after the fifth rotation, to 20 lb.

About 950 lb of refuse, having a water content of 12% to 15%, were added to each of the generators in 45 to 47 loads or lifts. This amount is equivalent to 1150 lb of refuse per generator of 30% water content, or about 1240 lb of refuse per cubic yard at 30% water content.

Screened topsoil was added and compacted to a depth of approximately 1.75 ft on top of the refuse, and 3 in. of 3/4-in. drain rock was added on top of the soil. In most of the generators, it was necessary to add the rock after the addition of water had been started to bring the refuse to field capacity. During this time the refuse consolidated and settled, allowing space for the rock to be placed on top of the soil. The top of the soil was given a saucer shape so that when the water was added it would pool in the center of the columns and not drain to the periphery and, thus, possibly channel down the edges near the walls.

Bringing the Refuse to Field Capacity and Regular Addition of Water. It was planned to bring the refuse in the generators to field capacity over a period of 30 days. Initial calculations, based on the moisture analysis of the refuse and the estimate of the water that had been added to aid in compaction, were that it would take approximately 1 gal water/day (5 days/week) to reach this objective. Several of the generators appeared to have produced leachate early; however, they did not continue producing,

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nor did the other generators produce leachate. It was, therefore, necessary to raise the addition of water to 2 gal/day.

All of the generators achieved field capacity within about 6 wk and have been producing leachate for the past 4 months. About 2 gal of water is being added every other week, which equals 1 in. on the top of the generators.

PRELIMINARY TEST RESULTS AND OPERATION OF SIMULATED LANDFILLS

Properties of the Liners and Seams Prior to Exposure to Leachate

Specimens of the liners, both the membranes and the admixed materials, were tested in accordance with the tests shown above. Most of these tests indicate general

adequacy of the liners and their meeting the specifications set forth by the suppliers. However, many of these properties do not reflect upon the actual performance of the material as a sanitray landfill liner. They may reflect on the quality of the particular liner with respect to other liners of the same type of raw material. The important factor will be the change in properties that will occur during exposure to the leachate. Properties of the unexposed liners are given in Appendices A and B.

At this point it seems that two properties could have a measurable effect on the performance of the liners during exposure to leachate, i.e., the swelling in water and the permeability of the liner material to water or water vapor. Results of these tests on the materials being exposed as barriers are presented in Tables 6 and 7.

Table 6. PROPERTIES OF LINER MEMBRANES BEFORE EXPOSURE TO LANDFILL LEACHATE

Type of membrane	Thickness, mils	Water absorption at room temp., %	Moisture vapor transmission, metric perms*	Permeability, cm/sec
Hypalon w/scrim	34	>6.77 at 26 weeks	0.057	0.0052
Butyl rubber	63	>0.88 at 26 weeks	0.0175	0.0029
Chlorinated poly-ethylene	31	>9.13 at 26 weeks	0.041	0.0033
Ethylene propylene rubber	51	>2.35 at 14 weeks	0.040	0.0053
Polyvinyl chloride	20	1.18 at 3 weeks(max.)	0.255	0.0135
Polyethylene	10	0.38 at 1 week (max.)	0.087	0.0023

*ASTM E96--66; metric perms: grams/24 hr/sq m/mm Hg; to convert to perms, multiply by 1.52.

Table 7. PROPERTIES OF ADMIX LINER MATERIALS BEFORE EXPOSURE TO LEACHATE

Material	Thickness, in.	Water swell, mil	Coeff. of permeability, cm/sec	Compression strength, % retained*
Asphalt concrete, paving (7.1 ph-agg)	2.2	1	1.2×10^{-8}	80
Asphalt concrete, hydraulic (9.0 ph-agg)	2.4	0	3.3×10^{-9}	86
Soil cement (10% type 5 cement)	4.5	0	1.5×10^{-6}	69
Soil asphalt (7.0 ph soil SC liquid asphalt)	4.0	17	1.7×10^{-3}	15
Bituminous seal	0.3	--	$<10^{-9}$	--
Asphalt emulsion	0.3	--	$<10^{-9}$	--

*After 24-hr immersion in water; asphalt concrete and hydraulic asphalt concrete at 60 C, and soil asphalt and soil cement at room temperature.

The polyethylene, polyvinyl cholride, and butyl show particularly low water adsorption. On the other hand, both polyvinyl cholride and polyethylene appear to have the greatest moisture vapor transmission.

In the case of the admixed materials, low water swell and good coefficient of permeability were observed for all but the soil cement and soil asphalt. To date, only the soil asphalt, with its relatively poor permeability and high swell, has allowed water to seep through the barrier. Preparation of this material on a larger scale would probably give better results than were achieved in the preparation of the test specimens. Difficulties were encountered in maintaining high temperatures during mixing and compacting the soil asphalt.

All of the splices met the specifications of the suppliers. However, the electronic and heat-sealing splices of the various membranes made yield the greatest strengths. As with the membranes themselves, the splices must retain their integrity during the exposure period and not allow leakage of leachate.

Performance of the Liners in Installation and Exposure to Leachate

All of the liners under test as barriers, except one, are performing satisfactorily. One of the liners, the soil asphalt, is allowing leachate to pass through. This was the most permeable of the liners being tested and was found to swell the most in laboratory tests. The leachate passing through has almost the identical composition as that being collected above the liner.

The polyethylene liner gave us problems when being placed in the bases of the generators because of small holes caused by creasing and/or punctures. No leak developed, even during compaction of the refuse,

but it appears that this type of liner material would be difficult to handle and install as a liner during landfill operations without developing holes and tears.

Variation in Liners of the Same Type

In this project we are studying 12 different liner materials and comparing them on the basis that each represents its respective type or class of material. Such an assumption is an oversimplification, because each liner material can vary considerably; this is particularly true of admix liners because of the inhomogeneity of the raw materials.

In the case of the polymeric membranes, there can also be considerable variation between materials based on a given polymer type, whether plastic or rubber. These variations can arise from the following:

1. Variation in the polymer type, e.g., grade, supplier.

2. Compound variation. Polymer suppliers may suggest recipes. Physical properties normally tested are not unique to a given composition. Therefore, individual liner manufacturers generally use different recipes, depending on both technical and economic factors. In the case of PVC there can be major variations in the amount and type of plasticizer and possibly in the use of various fillers; in the case of rubber, there can be variations in type and amount of filler, plasticizer, and curing agent (if any).

3. Variation in the techniques and the equipment used in forming the sheets.

These variations can show up in the various grades a manufacturer might supply.

Laboratory test data on all liners received and tested confirm the existence of significant variations in liners made of a given polymer. Five different companies supplied PVC sheeting. There is no indication as to the source of the PVC, but all suppliers are producers of PVC and, presumably, they use their own material as well as their own recipes in the manufacture of their respective liner sheetings. The following data show the range of properties reported for PVC sheeting:

Tensile strength, psi ... 1540 - 3400

Elongation, % .... 260 - 240

Set, % .... 20 - 110

Modulus @ 100% elong-
    ation, psi .... 980 - 1680

Hardness, Duro A .... 72 - 82

Tear, psi .... 270 - 390

Water absorption,
    7 days at 25 C, % .... 0.3 - 1.52

These data are primarily physical tests that characterize the compound but may not correlate with field performance, except possibly for water absorption. These physical properties are important in the installation of the liners, and these differences may indicate differences in the performance of these liners in a sanitary landfill environment involving long-term contact with leachate.

In the case of other liner sheetings, made of the same basic polymeric material, and supplied by different manufacturers, there have been significant variations in laboratory properties reflecting different compound recipes. Though

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made of the same material, they, too, may perform differently in a landfill environment.

The Hypalon liners have varied in water absorption in 2 hr at 100 C from 4.19 to 15.3 and in 70 days at room temperature from 4.52 to 8.66; and in hardness from 73 to 83. The variations in the other properties are largely due to the variations in the reinforcing fabric used. In the case of butyl, there are significant variations in tensile and water absorption and, in the case of EPDM, there are variations in tensile and tear.

We believe that the information obtained from the testing of all the samples, including the strip specimens, will give us information as to the importance of these variations on properties. This information will also be helpful in setting up performance and compositional specifications if such become necessary.

Characterization of the Refuse

It was the consensus of Combustion Power and the Palo Alto Sanitation Company personnel who saw the refuse before it was ground that it appeared typical of the refuse normally collected in the Palo Alto area and received at the Combustion Power laboratory, except that it appeared dry (see Table 8).

During the filling of the simulated landfills, a composite grab sample was accumulated by collecting one scoopful from each round when adding one lift to the 24 cells. The entire composite sample of 32.8 kg (72.2 lb) was separated into four size fractions by screening through a Sweco separator fitted with 25-mm (1-in.), 13-mm (0.5-in.), and 6-mm (0.25-in.) sieves. The entire 4.2-kg (9.3-lb), 6- to 13-mm fraction, and 4.2-kg aliquots of the plus 25-mm and 13- to 25-mm fractions were classified by hand sorting.

Table 8. CHARACTERIZATION OF COMPOSITE SAMPLE OF REFUSE (%)

Classification	Size of fraction and % of total				
	>25 mm (>1 in.) 27.0%	13-25 mm (0.5-1 in.) 36.8%	6-13 mm (0.25-.5 in.) 12.9%	<6 mm (<0.25 in.) 23.3%	% in total fraction
Water	11.9	11.8	4.3	17.3	12.2
Paper	69.6	58.1	50.5	29.8	53.6
Cloth	1.1	1.1	1.1	--	0.8
Plastic, rubber	9.8	5.4	2.1	--	4.9
Wood, garden, food waste	2.2	6.4	9.7	3.6	5.1
Oils and fats	--	--	--	4.0	0.9
Metal	5.4	15.0	4.3	--	7.6
Glass, rock, soil	--	2.2	28.0	45.3	14.9

Table 8 shows the classification of the composite sample. The content of identifiable food waste was low. It is assumed that the refuse must have been collected from a neighborhood where most food wastes are flushed into the sewers through sink disposal units. The actual content of putrescible organic material was somewhat higher than shown, as some pieces of paper and plastic in the larger size fractions were obviously saturated with fats, blood, etc.

Leachate Generation and Analysis

Table 9 summarizes leachate data. Leachate levels in excess of 2 ft have been observed in the four sand-pipes. Such levels are higher than expected and additional drainage

Table 9. DATA ON SIMULATED LANDFILLS AND LEACHATE; AVERAGE VALUES FOR 24 LANDFILLS

	Date/report number				
Determination	10 Dec. 2	6 Jan. 5	3 Feb. 8	3 Mar. 13	Breland* --
Amt. of leachate collected, liters	6.91	5.26	4.96	4.94	--
Temperature, C	11.5	10.5	11.5	--	--
Solids, %	3.49	3.38	3.58	3.54	1.25
pH	5.5	5.5	5.30	5.21	5.1
COD, mg/ℓ	46,106	58,375	45,075	43,520	18,000
TVA†, mg/ℓ	10,547	10,575	--	--	9,000
Organic acids:					
Acetic, mg/ℓ	1,446	--	--	2,000	5,160
Propionic, mg/ℓ	1,581	--	--	1,549	2,840
Isobutyric, mg/ℓ	330	--	--	497	--
Butyric, mg/ℓ	2,391	--	--	2,275	1,830
Isovaleric, mg/ℓ	124	--	--	339	--
Valeric, mg/ℓ	256	--	--	697	100
Caproic, mg/ℓ	0	--	--	0	--
Total organic acids	6,128	--	--	7,357	9,930

*Reference 26.
†Total volatile acids, as acetic acid.

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has been made to bring the levels down so that the level will be at 1 ft at the time a collection is made. Extensions are being placed on other collection bags, which are then raised to allow the extensions to perform as standpipes.

All of the 24 simulated landfills have been operating satisfactorily and producing leachate of quite similar composition for the past 4 mo. The analyses of the leachates averaged for the 24 simulated landfills are given in Table 9. They are compared with the results of Breland on a leachate from a control landfill at his highest concentration (26). The quality of the leachate appears to be satisfactory for testing the liner materials.

Some problems have been encountered with the polyethylene collection bags that were fabricated out of the same polyethylene film used in lining the generators. The bags failed at the seams and creases and have punctured easily. Polybutylene has been found to yield a much more durable bag, and a conversion to these will be made as soon as materials are available.

Except for the first few days of operation, the temperatures within all the simulated landfills have remained almost at ambient temperature of 11 to 19 C.

ESTIMATED COSTS OF POTENTIAL LINER MATERIALS FOR LANDFILLS

One of the objectives of this project is to determine the relative costs of the various liner systems. To compare the total real costs of liners for use in lining sanitary landfills, the following must be considered:

1. performance required, e.g., permeability

2. desired service life

3. costs of liner materials delivered to the site

4. costs of installation, adaptability to landfill operation

5. certain site preparation costs required for a given liner material

6. soil cover costs.

Obviously, overall effectiveness and service life will ultimately determine the cost that can be applied to a given liner. The information needed to make such estimates depends on actual experience and studies such as this.

Early in the project we assembled the then-current cost data on these various liner materials for comparative purposes. As the project progresses, these data will be reviewed and updated. The estimates made in October 1973 are presented in Tables 10 and 11; recognize that major cost increases probably have taken place.

Basically, those costs did not include the costs for site and surface preparation, nor the cost of ground cover which would be required in nearly all cases. The surfaces on which the liners are to be placed must be graded and smoothed for drainage and compacted to prevent settling of the ground below the liner and, in several cases, to give a firm table on which to compact the liner materials (i.e., soil asphalt, soil concrete, and the asphalt concretes). The cost of site preparation is essentially the same for all the liner systems, though it is possible that some of the liner systems may not require as much effort in surface preparation as others. A cover, preferably one which is somewhat porous, is needed as part of the liner system. Such soil covers will allow the large landfill equipment, e.g., caterpillar tractors and compactors, to operate on the liners.

Table 10. PRELIMINARY ESTIMATE OF COSTS* OF POTENTIAL LINERS FOR
SANITARY LANDFILLS: POLYMERIC MEMBRANES--PLASTICS AND
RUBBERS--UNREINFORCED

Item	Thickness, mils	Price of roll goods	Installed cost†
Butyl rubber	31.3 (1/32")	$2.25	$3.25 -$4.00
Chlorinated polyethylene (CPE)	20	1.58	2.43 - 3.24
Chlorosulfonated polyethyleneᵠ	20	1.66	2.88 - 3.06
Ethylene propylene rubber (EPDM)	46.9 (3/64")	2.42	2.65 - 3.42
Neoprene	62.5 (1/16")	2.97	4.41 - 5.40
Polyethylene film	10	0.36	0.90 - 1.44
Polyvinyl chloride	20	0.90	1.17 - 2.16

*Costs in dollars per square yard.
†Soil cover not included; membranes require some soil cover, cost of which
 can range from $0.10 to $0.50/sq yd per ft of depth.
ᵠHypalon, with nylon scrim.

EVALUATION OF LINER MATERIALS EXPOSED TO HAZARDOUS AND TOXIC SLUDGES

An urgent need also exists for information comparing various liner materials as barriers to hazardous and toxic wastes. Consequently, the Solid and Hazardous Waste Research Laboratory is sponsoring a study similar to the landfill study in which 12 liner materials will be exposed to six wastes over a period of 2 yr and the effects on properties of the liners will be observed. Work has been started on this project.

The performance requirements for liner materials for confining hazardous wastes will, of course, differ from those for sanitary landfills and are more stringent. In this program soils and clays will be included. At the present time, 11 of the 12 liner materials have been selected for exposure testing. Many of these will be the same as those tested in the landfill project. Table 12 lists the materials that will be exposed to such various hazardous and toxic sludges as acidic, alkaline, pesticide and cyclic hydrocarbon sludges; oil refinery tank bottom waste, and lead waste from gasoline tanks.

Figure 8 illustrates the type of exposure test cell that will be used for exposing a flexible membrane liner. Each of the flexible liner specimens will contain a field type splice and will be sealed in place with an epoxy resin. The same basic design will be used for the thick admixed specimens, except that a spacer will be used between the base of the cell and the upper part, which will contain the hazardous waste. In this case, a spacer will be used and will be sealed with epoxy. The sketch in the upper right shows the overall appearance of the exposure cell with a thick admixed liner mounted.

Table 11. ESTIMATES OF COSTS* OF POTENTIAL LINERS FOR SANITARY LANDFILLS SOILS, ADMIXTURE MATERIALS, AND ASPHALT MEMBRANES

Type	Installed cost
Soil + Bentonite 9 lb/sq yd (1 psf)	$0.72
Soil cement 6-in. thick + sealer (2 coats--each 0.25 gal/sq yd)	1.25
Soil asphalt 6-in. thick + sealer (2 coats--each 0.25 gal/sq yd)	1.25
Asphalt concrete--Dense-graded paving with sealer coat (Hot mix--4-in. thick)	2.35 - 3.25
Asphalt concrete--Hydraulic (Hot mix--4-in. thick)	3.00 - 4.20
Bituminous seal (catalytically blown asphalt) 1 gal/sq yd	1.50 - 2.00 (with earth cover)
Asphalt emulsion on mat (polypropylene mat sprayed with asphalt emulsion)	1.26 - 1.87

*Costs in dollars per square yard.

Table 12. LINER MATERIALS TO BE EVALUATED IN EXPOSURE TESTS TO HAZARDOUS AND TOXIC SLUDGES

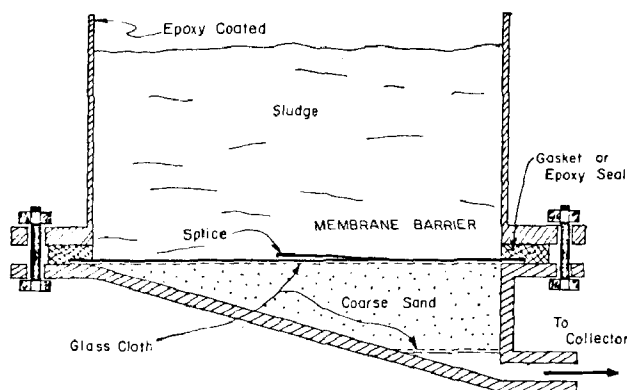|	
Polyvinylcholride	Bentonite clay seal
Butyl rubber	Asphalt emulsion on nonwoven fabric
Chlorosulfonated polyethylene (Hypalon)	Soil cement with seal
Chlorinated polyethylene (CPE)	Hydraulic asphalt concrete
Ethylene propylene rubber (EPDM)	Compacted native fine grain soil
Polychloroprene (Neoprene)	

Figure 8. Exposure cell for membrane liners.

The exposure cells are being fabricated and mounting of the specimens will begin about May 1, 1975. Properties of the test specimens will be measured after 6 mo and 1- and 2-yr exposures to the various wastes.

SUMMARY

Two specimens each of six admix and six polymeric membrane liner materials have been mounted as barriers in the bases of 24 simulated sanitary landfills for 1 and 2 yr exposure to leachate generated in these fills. These 12 barrier materials represent a wide range of compositions; they are currently being used in lining ponds, pits, lagoons, canals, etc., to prevent seepage of water or various wastes and appear promising for use as impervious barriers for lining sanitary landfills. In addition, 42 small membrane specimens, many of them incorporating splices, are being exposed to leachate by being placed in the sand above the barriers.

The 24 simulated landfills were uniformly filled with a shredded municipal refuse compacted to a density of about 1240 lb/cu yd at a moisture content of 30%. The simulated landfills are functioning properly; the conditions within the fills are anaerobic and there is no leakage around the liners. The leachate being generated appears to be representative of landfill leachate; it is quite uniform among the various generators so that all the liner specimens are being exposed to essentially the same type of leachate.

.At this time, only the soil-asphalt liner appears to be inadequate. Leachate is seeping through one specimen of this type liner, and indications are that the second liner of this type will also leak, although insufficient leachate has seeped through to be collected at this time.

The method of sealing the liners into the generator bases with a cast epoxy ring has worked out satisfactorily; except for the soil-asphalt liner, there has been no leakage or seepage of leachate into the lower compartment of the bases.

Although it has not failed as a barrier, the polyethylene film is sensitive to creasing and puncturing. Not only is it being exposed as barrier specimens in two of the simulated landfills, but it is also being used to line the 2-ft-diameter steel pipes containing the refuse and to fabricate the leachate collection bags. It appears, at this time, that polyethylene film would be difficult to handle in a landfill operation.

Heat sealing to splice the various membrane liners, such as polyvinyl chloride, polyethylene, chlorinated polyethylene, and Hypalon, yields particularly strong seams.

Laboratory tests of unexposed membrane liners of the same polymer indicate that there can be considerable liner-to-liner variation in liners of the same polymer type. These variations probably reflect differences in compounding and in fabrication of the liner materials.

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The first set of 12 simulated landfills will be disassembled in November 1975, at which time the barriers and buried specimens will be recovered and tested. The specimens will have been exposed to landfill leachate for 1 yr. Present plans call for the second set of 12 simulated landfills to be disassembled in November 1976 and the specimens tested after a 2-yr exposure to leachate.

REFERENCES

1. Brunner, D. R., and D. J. Keller, (1972). "Sanitary Landfill Design and Operation," U.S. Environmental Protection Agency Report SW-65ts.

2. Weiss, Samuel, (1974). "Sanitary Landfill Technology," Noyes Data Corporation, Park Ridge, New Jersey.

3. Asphalt Institute (1966). "Asphalt Linings for Waste Ponds," IS-136.

4. Asphalt Institute (1969). "Construction Specifications for Asphalt Concrete," SS-1.

5. Ellsperman, L. M., (1957). "Buried Asphalt Membrane Canal Linings," Third Congress of the International Commission on Irrigation and Drainage, San Francisco, Ca.

6. Ellsperman, L. M., and M. E. Hickey (1959). "The Uses of Asphalt in Hydraulic Construction by the Bureau of Reclamation," Third Annual Kansas Paving Conference, University of Kansas, Lawrence, Kansas.

7. Geier, F. H., and W. R. Morrison (1968). "Buried Asphalt Membrane Canal Lining," U.S. Bureau of Reclamation, Research Report No. 12.

8. Hickey, M. E. (1961). "Laboratory and Field Studies of Asphaltic Materials for Controlling Canal Seepage Losses," Conference on Use of Asphalt in Hydraulic Construction, sponsored by the Asphalt Institute, Bakersfield, Ca.

9. Hickey, M. E. (1969). "Investigations of Plastic Films for Canal Linings," Bureau of Reclamation, Research Report No. 19.

10. Hickey, M. E. (1971). "Synthetic Rubber Canal Lining Laboratory and Field Investigation of Synthetic Rubber Sheeting for Canal Lining--Open and Closed Conduit Systems Program," Bureau of Reclamation, Report No. REC-ERC-71-22.

11. Jones, C. W. (1971). "Laboratory Evaluation of Canal Soil Sealants, An Open and Closed Conduit Systems Investigation of Four Proprietary Materials Proposed as Canal Soil Sealants on Silty Sand in Laboratory Permeameters," Bureau of Reclamation, Report REC-ERC-71-1.

12. Lee, Jack (1974). "Selecting Membrane Pond Liners," Pollution Engineering, January.

155

13. Merton, F. K., and B. A. Brakey (1968). "Asphalt Membranes and Expansive Soils," The Asphalt Institute, IS-145.
14. Morrison, W. R., R. A. Dodge, J. Merriman, L. M. Ellsperman, Chung Ming Wong, W. G. Savage, W. W. Rinne, and C. L. Gransee. "Pond Linings for Desalting Plant Effluents," U.S. Dept. of Interior, Office of Saline Water, R & D Prog. Report No. 602.
15. NACE (National Association of Corrosive Engineering) (1966). "Chemical Resistance of Asphalt Coatings," Materials Performance 5, 81-83.
16. Phillips Petroleum Company (1972). "Asphalt Sealed Membranes for Pond Liners and Erosion Control," Handbook and Installation Guide for Petromat Fabric, Hydraulic Grade.
17. Portland Cement Association (1937). "Lining Irrigation Canals."
18. Portland Cement AsOociation (no date). "Soil Cement Linings for Water Reservoirs."
19. Rosene, R. B., and C. F. Parks. Environmental Quality Conference for AIME, Washington, D.C., June 7-9, 1971. "Preventing Loss of Industrial and Fresh Waters from Pits, Ponds, Lakes and Canals."
20. Smith, W. D. (1962). "Canal and Reservoir Lining with Asphalt," The Asphalt Institute, IS-121.
21. U.S. Bureau of Reclamation (1963). "Linings for Irrigation Canals," U.S. Government Printing Office.
22. U.S. Department of Agriculture (1972). "Asphalt Linings for Seepage Control: Evaluation of Effectiveness and Durability of Three Types of Linings," Technical Bulletin No. 1440, May.
23. Steiner, R. C., A. A. Fungaroli, R. J. Schoenberger, and P. W. Purdom (1971). "Criteria for Sanitary Landfill Development," Public Works, 103(3):77-79.
24. Vallerga, B. A., and R. G. Hicks (1968). "Water Permeability of Asphalt Concrete Specimens Using Back-Pressure Saturation," J. Materials 3(1): 73-86.
25. Ewald, G. W. (1973). "Stretching the Life Span of Synthetic Pond-Linings," Chemical Engineering, Oct. 1.
26. Breland, G. G., Jr. (1972). "Landfill Stabilization with Leachate Recirculation, Neutralization, and Sludge Seeding," Special Research Problem, Georgia Institute of Technology School of Civil Engineering, Sept.

APPENDIX A

PROPERTIES OF POLYMERIC LINER MEMBRANES INSTALLED AS BARRIERS

	1,19	2,20	3,21	4,22	5,23	6,24
Cell Number	1,19	2,20	3,21	4,22	5,23	6,24
Liner No.	21	17	7	6	16	12
Material	Polyethylene	Polyvinyl Chloride	Butyl Rubber	Hypalon, with nylon scrim	Ethylene-propylene-diene (EPDM) rubber	Chlorinated polyethylene
Thickness, mm (0.001 in.)	0.25-0.30 (10-12)	0.51-0.53 (20-21)	1.55-1.65 (61-65)	0.81-0.91 (32-36)	1.24-1.35 (49-53)	0.79-0.81 (31-32)
Coefficient of water permeability, cm/sec.		7.3×10^{-13}	1.1×10^{-11}	3.6×10^{-12}	2.3×10^{-11}	2.0×10^{-12}
Water absorption,%, 2 hr @ 100°C	-0.61	2.15	0.17	7.17	0.47	2.93
" " 7 days @ 25°C	0.38	0.95	0.18	2.04	0.61	1.43
" " 70 days @ 25°C			0.52	4.52	1.90	5.31
Puncture test,[1] 25 mm/min., max. force, N (lb.)			149 (33.5)	131 (29.5)	141 (31.6)	150 (33.8)
elongation, mm (in.)			29 (1.14)	26 (1.01)	35 (1.38)	26 (1.03)
Puncture test, 500mm/min., max. force, N (lb.)	61.9 (13.9)	115 (25.8)	199 (44.8)	146 (32.9)	175 (39.4)	209 (47.0)
elongation, mm (in.)	19 (0.76)	18 (0.69)	31 (1.22)	15 (0.60)	37 (1.44)	26 (1.04)
Splice strength, peel, kN/m (lb./in.)	2.73 (15.6)	0.70 (4.0)	0.66 (3.8)	5.25 (30)	0.44 (2.5)	1.75 (10)
" " shear, kN/m (lb./in.)	3.54 (20.2)	6.51 (37.2)	5.25 (30)	8.75 (50)	2.56 (14.6)	9.98 (57)
Hardness, Shore A, instantaneous	98	81	55	81	57	85
10 sec.	98	76	51	79	54	87

Grain direction

Property	Polyethylene with	cross	PVC with	cross	Butyl with	cross	Hypalon with	cross	EPDM with	cross	CPE with	cross
Modulus @ 100%, MPa (lb/in²)	8.76 (1270)	7.10 (1030)	8.69 (1260)	7.79 (1130)	2.41 (350)	2.02 (290)	6.90 (1000)	5.93 (860)	2.41 (350)	2.41 (350)	8.41 (1220)	3.59 (520)
Modulus @ 200%, MPa (lb/in²)	10.1 (1470)	7.24 (1050)	14.3 (2080)	12.8 (1850)	5.31 (770)	4.21 (610)	11.8 (1710)	9.17 (1330)	5.24 (760)	5.24 (760)	12.5 (1820)	5.79 (840)
Modulus @ 300%, MPa (lb/in²)	11.6 (1680)	7.72 (1120)			8.48 (1230)	6.90 (1000)			7.72 (1120)	7.72 (1120)	17.0 (2460)	8.27 (1200)
Tensile strength, MPa (lb/in²)	11.7 (1700)	17.9 (2590)	18.2 (2640)	17.4 (2520)	9.93 (1440)	9.86 (1430)	13.2 (1920)	11.1 (1610)	10.4 (1510)	9.93 (1440)	17.0 (2460)	14.3 (2080)
Elongation, %	320	690	270	290	360	430	250	250	420	400	300	520
Set, %	177	667	68	77	15	18	115	106	13	19	199	230
Tear resistance[2], kN/m (lb/in.)	72.6 (415)	63.0 (360)	61.6 (352)	55.5 (317)	31.5 (180)	31.5 (180)	56.0 (320)	49.0 (280)	31.7 (181)	31.7 (181)	47.2 (270)	42.0 (240)
Creep test[3], load, N (lb.)	7.78 (1.75)	7.78 (1.75)	14.0 (3.15)	12.7 (2.85)	14.0 (3.14)	12.5 (2.82)	25.3 (5.68)	29.8 (6.7)	8.18 (1.84)	6.89 (1.55)	22.2 (5.0)	14.1 (3.18)
hours to failure	>72 NF	>72 NF	26	12	>94 NF	>94 NF	>96 NF	2.2	>143 NF	>143NF	31	47
elongation, %	13.3	47.6	194	138	108	111	194	253	112	102	186	344
initial stretch, 1 min., %	5.7	7.7	73	51	84	83	15	17	79	78	62	129
set, %	9.5	19			5.4	5.2	100+		5.6	4.7		176
creep, cm/cm, 100 hr.	0.10	0.52	3.88	7.5	0.23	0.23	1.87	99	0.18	0.14	4.5	3.6

(1) Method 2065, Fed. Test Methods 101   (2) ASTM D624, Die C   (3) ASTM D674 - dumbbell specimens, restricted portion 51 mm (2 in.) long by 6.3 mm (0.25 in) wide. NF = no failure.

PROPERTIES OF ADMIX LINERS MOUNTED AS BARRIERS

	Asphalt Concrete	Hydraulic Asphalt Concrete	Soil Cement	Soil Asphalt	Bituminous Seal	Fabric + Asphalt Emulsion
Installed in Cell No.	7,13	8,14	9,15	10,16	11,17	12,18
Composition of Barrier Specimen	7.1 asphalt/ 100 aggregate	9.0 asphalt/ 100 aggregate	95 soil, 5 kaolin clay, 10 Type 5 cement, 8.8 water	7.0 SC-800 liq. asphalt/ 100 aggregate	Catalytically-blown asphalt, 4.7 kg/m^2 (8.7 lb/yd^2)	Asphalt (from emulsion) 4.8 kg/m^2 (8.9 lb/yd^2) on polypropylene non-woven fabric
Particle size distribution, %						
Passing 4.76 mm, (4 mesh)	90.7	89.4	88.9	79.2		
" 2.38 mm, (8 mesh)	61.0	67.1	70.8	55.8		
" 1.19 mm, (16 mesh)	45.1	50.9	53.7	39.9		
" 0.595 mm, (30 mesh)	30.1	33.7	38.8	27.3		
" 0.297 mm, (50 mesh)	19.4	21.5	29.2	18.5		
" 0.149 mm, (100 mesh)	11.2	12.4	20.8	13.4		
" 0.074 mm, (200 mesh)	6.6	7.2	15.0	11.4		
Sand equivalent			27	31		
Liquid limit			17.6	17.0		
Plastic limit			non-plastic	non-plastic		
Plasticity index			non-plastic	non-plastic		
Penetration at 25°C	68	68			45	
Penetration (extracted from barrier)	44	62				
Softening point, °C (°F)					89 (192)	
Penetration index					+5.2	
Viscosity, capillary at 60°C, cS				1101		
Viscosity, sliding plate at 25°C, at 0.05 sec^{-1}, MP	14.5	9.7		0.20	8.5	4.5
Viscosity, sliding plate at 25°C, at 0.001 sec^{-1}, MP	20.0	14.5		0.14	19.3	6.0
Microductility at 25°C, mm	40	76		7	2	29
Thickness of barrier specimen, cm (in.)	5.6 (2.2)	6.1 (2.4)	11.4 (4.5)	10.2 (4)	0.8 (0.3)	0.8 (0.3)
Density, g./cm^3 (lb/ft^3)	2.387 (149.0)	2.416 (150.8)	2.169 (135.4)(dry)	2.228 (139.1)		
Void ratio (vol. voids/vol. solids), %	6.4	2.9		10.4	0	
Water swell mm (0.001 inch)	0.03 (1)	0	0	0.43(17)		
Coefficient of permeability, cm/sec. (Ref. 21)	1.2 x 10^{-8}	3.3 x 10^{-9}	1.5 x 10^{-6**}	1.7 x 10^{-3}	< 10^{-9}	< 10^{-9}
Compressive strength, MPa (lb/in^2)	19.34 (2805)	18.70 (2712)	13.17 (1910)	8.40 (1218)		
Compressive strength after 24 hr immersion*	15.38 (2230)	16.05 (2328)	9.12 (1323)	1.27 (184)		
% retained	80	86	69	15		

* Asphalt Cement and Hydraulic Asphalt Cement immersed in water at 60°C, Soil Asphalt and Soil Cement at R.T.
** Measured on molded specimen

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LANDFILL MANAGEMENT WITH LEACHATE RECYCLE AND TREATMENT: AN OVERVIEW

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INTRODUCTION

Leaching of solid waste con-
stitutes from landfill disposal
of solid wastes has received con-
siderable attention in recent years.
Yet, definition of actual environ-
mental impacts has been exceedingly
elusive and fraught with controversy.
Much of this dilemma is related to
past landfill management procedures
which have generally failed to ade-
quately plan for and monitor opera-
ting conditions at a particular site.
In recognition of these deficiencies,
research on a new landfill management
concept employing leachate collection
and recycle was initiated at Georgia
Tech in 1970. This research has been
extended to include investigations on
leachate adjustment and control as
well as residual treatment alterna-
tives. An overview of the leachate
recycle/treatment concept is pre-
sented herein together with some
supporting data and their intrepre-
tation. Other details have been
presented elsewhere (1-3).

DESCRIPTION OF EXPERIMENTAL SYSTEMS

To accomodate the objectives of
the research effort, an experimantal
system (Figure 1) was devised to sim-
ulate landfill disposal of domestic-
type solid wastes but with opportuni-
ties for comparison of the character-
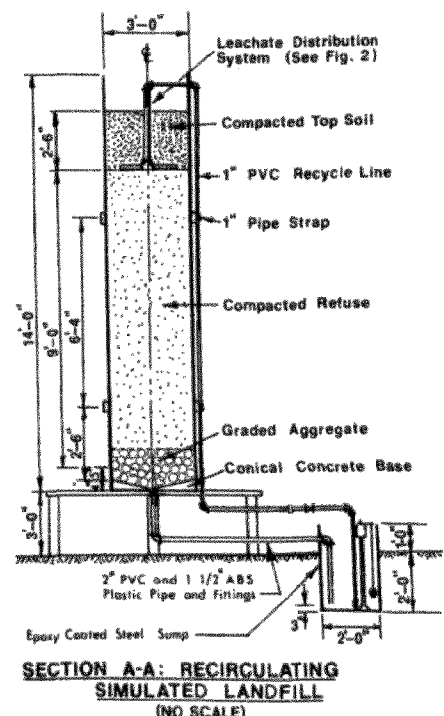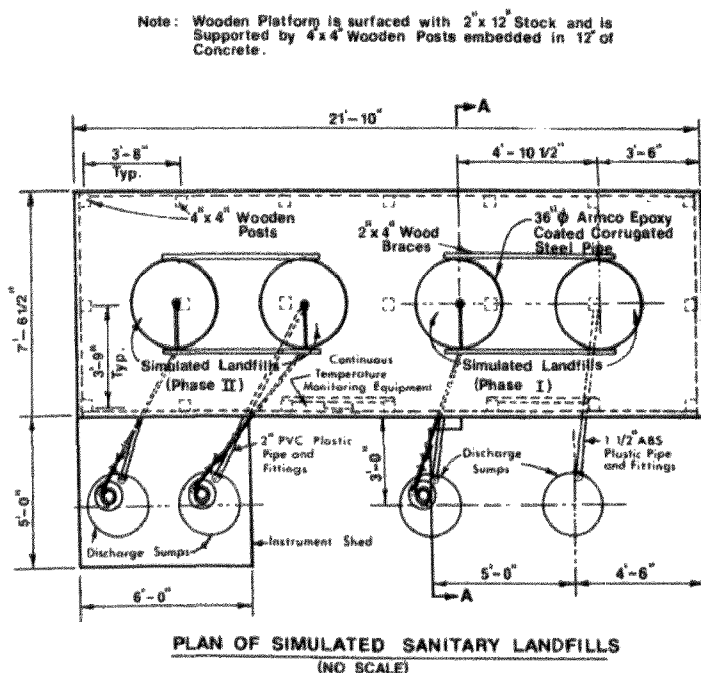
Figure 1. Simulated sanitary landfills.

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istics of normal leachate produced by intercepted rainfall with the leachate collected and recycled back through the landfill in a manner analogous to the operation of an anaerobic trickling filter. The basic experimental system illustrated in Figure 1 consisted of four 3-ft-diameter columns containing 10 ft of compacted solid wastes covered with 2.5 ft of soil with a sodded surface. The system was equipped to permit interception of incident rainfall and collection and analysis of subsequent changes in characteristics of the solid wastes, gas, and leachate generated over an experimental period of 3 yr.

Based on the premise that the acceptability of ultimate discharge of leachate accumulations into some receptor, after leachate recycle had attenuated the leachate constituents to residual concentrations, would be functions of environmental and/or regulatory requirements, the leachate recycle studies were complemented by separate physical-chemical and biological leachate treatment investigations. Batch ion exchange and activated carbon slurry systems were used as logical alternatives for physical-chemical treatment, and com-

pletely mixed, continuous flow reactor systems (Figure 2) but without solids recycle were used for both aerobic and anaerobic biological treatability studies.

EXPERIMENTAL TECHNIQUES AND DISCUSSION OF SELECTED RESULTS

Leachate Recycle Studies

The four simulated landfill columns were constructed in two phases. Phase I included operation of two fills (fills 1 and 2) for a period of 1063 days; phase II included operation of two fills (fills 3 and 4) concurrently with the last 747 days of the phase I studies. The fills of phase I differed in that fill 1 was constructed to permit collection of leachate without recycle; fill 2 had leachate collection with recycle. Similarly, the two fills of phase II provided for leachate collection and recycle but also initial pH control by neutralization with sodium hydroxide and, in fill 4, the addition of sewage sludge for initial seeding and nutrient supplementation.
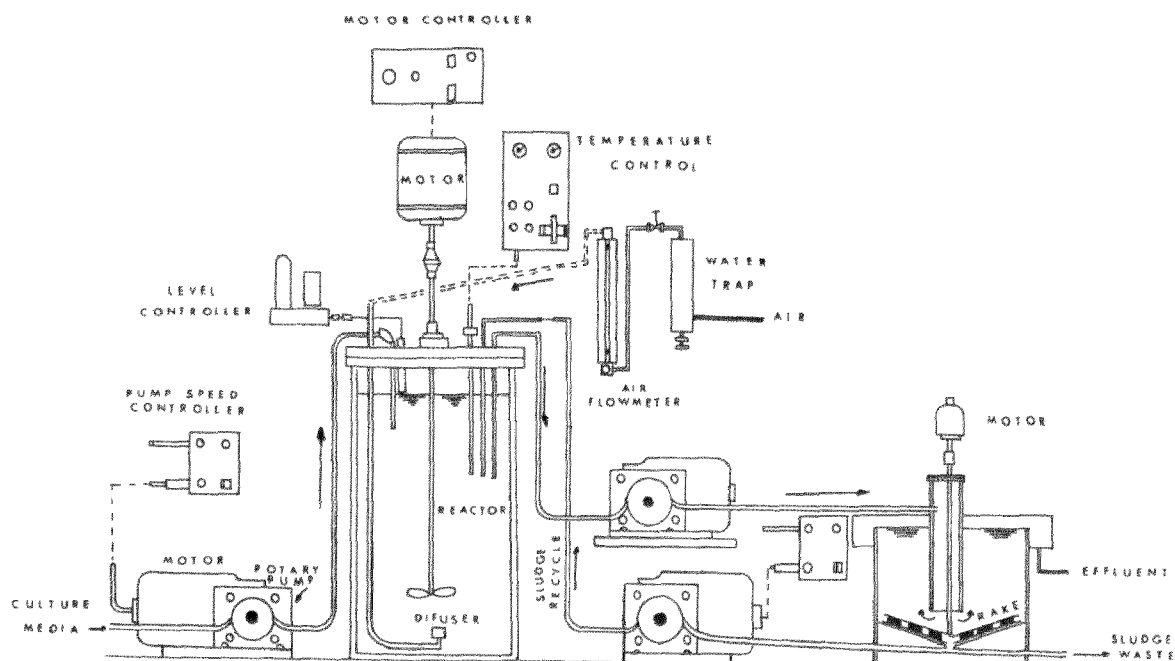
Figure 2. Completely mixed, continuous flow reactor system.

All columns were filled with coarsely ground test material chosen to simulate domestic solid wastes (Table 1). The material was manually compacted to a dry density of about 535 lb/cu yd. To allow for the immediate production of leachate (an experimental expediency), about 250 gal of tap water was added to each fill; primary sewage sludge replaced 30 gal of the tap water in fill 4.

The leachate accumulated as a consequence of rainfall was removed from the collection dumps, subjected to analysis, and then, for the control fill (fill 1) discarded; for fills 2, 3 and 4, it was analyzed and then intermittently pumped back through the distributor buried between the top of the solid wastes and the soil cover for the fills with leachate recycle. Automatic pH control was provided for the phase II fills until pH control was no longer required. An apparatus for collecting gas during phase II was also devised to provide information on gas evolution and quality.

Selected analytical data accumulated during the leachate recycle studies have been summarized and graphically displayed in Figures 3 and 4. Recognizing that those parameters commonly employed to measure pollutional potential are of particular importance in the identification of problems associated with the escape of leachate from

Table 1. COMPOSITION OF SIMULATED
SOLID WASTES

Constituent	Dry weight, %
Paper	50.0
Plastic	3.0
Glass	7.0
Garbage, garden debris	25.0
Rags	5.0
Stone, sand	5.0
Metal	4.0
Wood	1.0
Total	100.0

landfill operations, BOD_5 has been used herein to reflect the potential pollution derived from the decomposition of organic matter. As indicated in Figure 3, the control fill reached high BOD_5 concentrations and then slowly decreased in concentration with time, whereas the BOD_5 in the leachate from the fills with recycle rapidly decreased in concentration to much lower values.

The rapid decline in BOD_5 with time in the leachates from the fills with recycle was considered indicative of an initial acceleration of biological stabilization of the more readily available organics in the solid wastes with the addition of moisture and/or sludge. The recycle of leachate maintained an opportunity for continuous biological decomposition of the solid waste constituents, as well as those transferred to the leachate, whereas such biological action in the control fill was curtailed by the addition of moisture only during periods of rainfall and by the single-pass operation. Moreover, recycle with pH control permitted even greater rates of stabilization although sludge seeding initially appeared not as effective in improving leachate quality. This was considered due presumably to the conflict between pH control, which would abet anaerobic methane fermentation, and primary sludge seeding, which would and apparently did create an environment more beneficial to volatile acid forming organisms and therefore unfavorable to the methane formers because of reductions in pH. However, continued control of the pH at neutral and favorable methane fermentation eventually nullified the effect of excess volatile acid production in fill 4 and permitted an extent of stabilization similar to that achieved in the other recycle fills. Similar data on COD and TOC confirming the results observed for BOD_5 are presented elsewhere(3).

Since solid waste stabilization as achieved during landfill disposal is largely dependent on anaerobic activity, it is possible to further
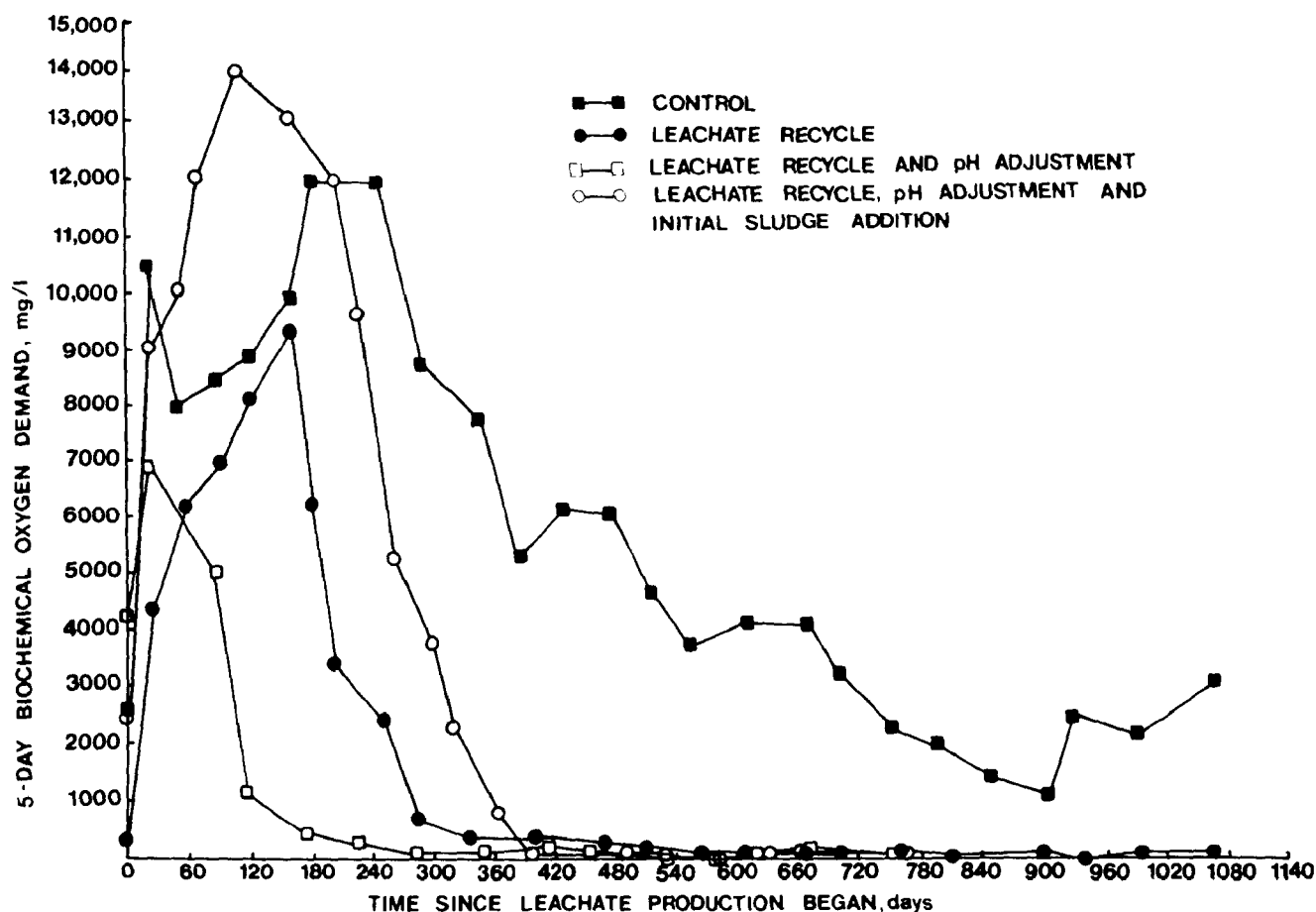
Figure 3. Biochemical oxygen demand of leachate.

interpret the observed changes in pollutional strength of the leachate from the fills with recycle. If the two-phase process of acid fermentation with the production of volatile acids followed by fermentation of these acids to methane and carbon dioxide is considered applicable, then the changes in pollutional charac-teristics should also be paralleled by an appearance and subsequent utilization of volatile acids. Inspection of Figure 4 indicates that this was the case. An initial rise in volatile acids was followed by their virtual elimination in the leachate from the fills with recycle, and elimination at a lesser and deferred rate in the control fill without recycle. Based on other data (3), decreases in volatile acids tended to proceed in a sequential pattern from higher to

lower homologues, and gas composition analysis confirmed their conversion to methane and carbon dioxide.

As could be expected, the pH decreased with an increase in volatile acid concentrations in the leachate of those fills without pH control until the acids began to be utilized. This suggested that the normal bicarbonate buffer established near neutral pH was initially replaced by that characteristic of the volatile acids (pk ≈ 4.5) and then was reinstated for the fill with leachate recycle as the volatile acids became less influential. In contrast with fill 2, the control fill did not exhibit a similar recovery although a gradual increase in pH was noted. The initial addition of caustic soda to the two fills with leachate recycle (fills 3 and 4) achieved
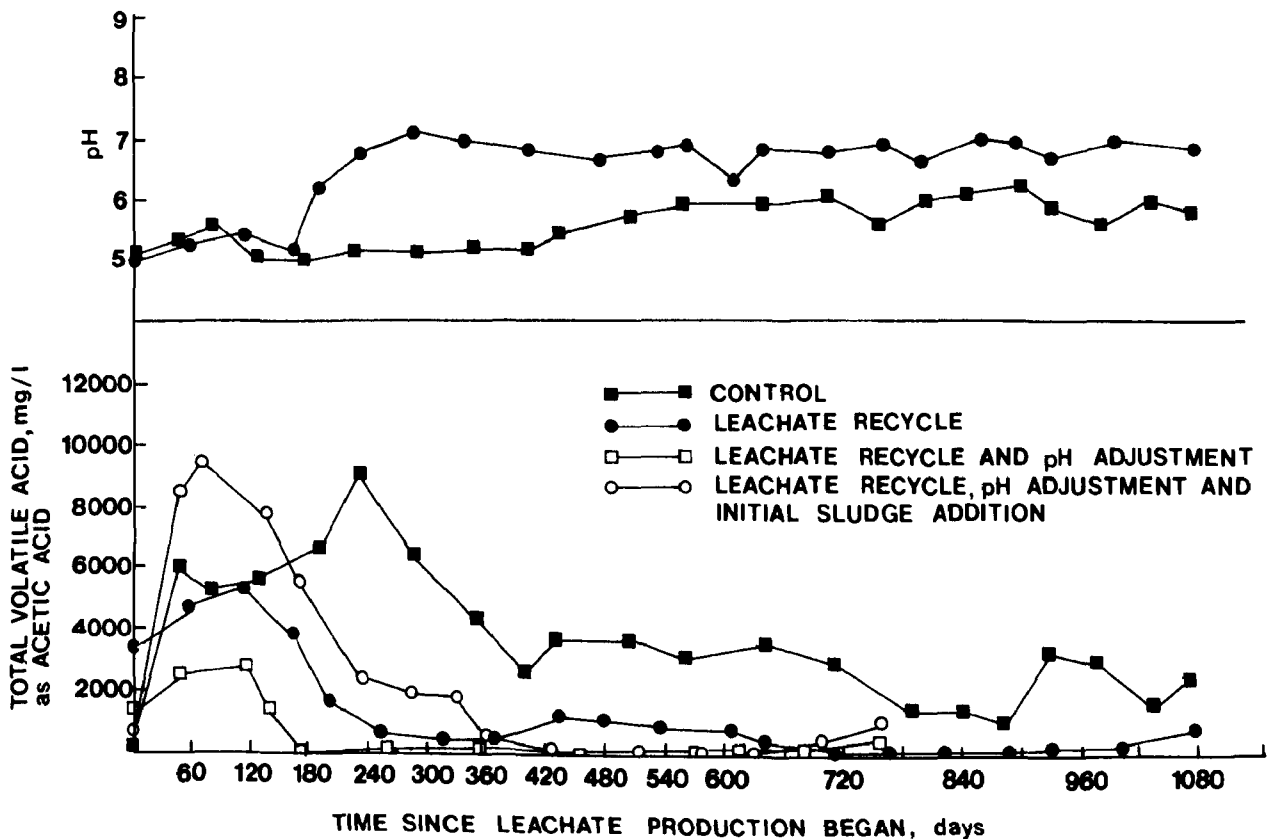
Figure 4. pH and total volatile acid concentration of leachate.

a similar acceleration in conversion of the volatile acids.

Separate Biological Treatment

The leachate used in both the anaerobic and aerobic biological treatment studies was a mixture of leachate accumulated in fill 1 and from a local landfill. The characteristics of the two leachate samples are indicated in Table 2. The pertinent results of these studies, although discussed in more detail elsewhere (3), have been summarized and included in Figures 5 and 6. The data indicate good removals of the pollutional components of the leachate as measured by COD, BOD_5, TOC, or volatile acids with an acceptable correlation between these parameters. Application of continuous culture theory analysis yielded the kinetic parameters indicated, with washout
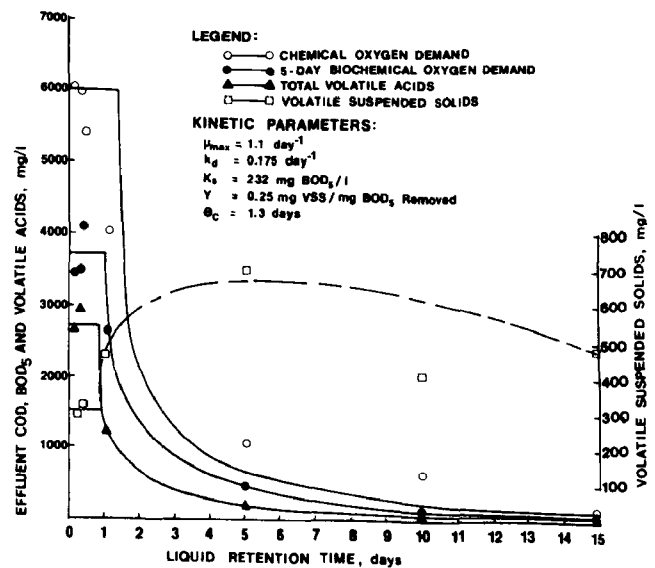
Figure 5. Anaerobic biological treatment of leachate in continuous culture.

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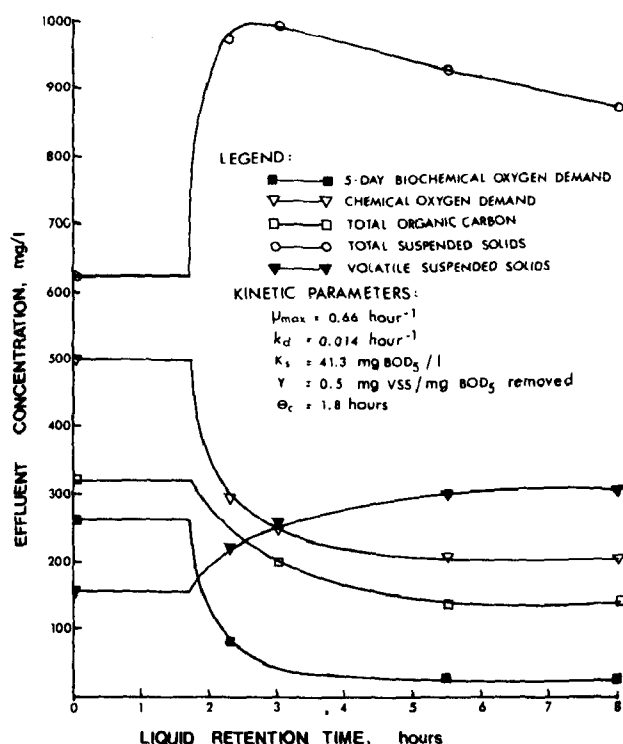
Figure 6. Aerobic biological treatment of leachate in continuous culture.

occuring at 1.3 days and 1.8 hr for the anaerobic and aerobic studies, respectively--a reflection of the relative differences in generation times between the anaerobic and aerobic organisms. Similarly, biological solids yield for the anaerobic system was half of the corresponding yield for the aerobic system--a consequence of less conversion of substrate to biomass in the former process. Once active anaerobic decomposition had been established, gas yields ranged between 9 and 17.4 cu ft/lb BOD_5 destroyed with a methane content of 70 to 80%. Although this concentration was higher than normally reported for anaerobic conversion processes, even at lower (60% to 70%) methane contents, energy recovery during anaerobic leachate stabilization would be an interesting possibility.

Separate Treatment of Leachate Residuals

Since inspection of the effluent quality data from the leachate recycle and/or separate biological

Table 2. CHARACTERISTICS OF LEACHATE USED DURING SEPARATE BIOLOGICAL TREATMENT

Leachate characteristic	Anaerobic treatment	Aerobic treatment
pH	5.1	7.0
COD, mg/ℓ	6,000	500
BOD$_5$, mg/ℓ	3,700	260
TOC, mg/ℓ	2,100	320
Suspended solids		
Total, mg/ℓ	1,100	625
Volatile, mg/ℓ	300	160
Calcium, mg/ℓ	200	100
Magnesium, mg/ℓ	64	35
Potassium, mg/ℓ	348	204
Sodium, mg/ℓ	313	425
Phosphate, mg/ℓ PO$_4$	--	0.7
Total volatile acids, mg/ℓ as acetic acid	2,700	410

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treatability studies indicated
organic and inorganic residuals
that could be unacceptable for
ultimate discharge, physical-chemical
processes including ion exchange
and carbon adsorption were applied.
As illustrated in Figure 7, effluent
from the separate aerobic biological
leachate treatment studies was
successfully treated with the
indicated cation exchange resin.
Of those cations measured, excellent
removals were achieved with the
divalent calcium and magnesium
preceeding the removal of the
monovalent sodium and potassium.

With mixed resin ion exchange
(Figure 8), effluent from the aerobic
leachate treatment studies was also
successfully treated for removal
of both cations and anions. The
data indicated that all measured
ionic impurities were capable of
removal again in order of resin
selectivity. Because of the opposite
influences of the resin reactions,
pH and alkalinity or acidity changes

Figure 8. Mixed resin ion
exchange treatment
of effluent from
aerobic biological
treatment of
leachate.

were not as dramatic with the mixed
resin ion exchange treatment as
with the separate cation exchange
treatment where ion replacement
released hydrogen ions in excess
and lowered the pH with an increase
in acidity to possibly unacceptable
levels (pH 2.5, 470 mg/ℓ acidity).
The impact of such changes would
necessarily be a function of ionic
concentration and degree of treatment
required.

To remove organic residuals,
effluent from the biological leachate
treatment studies was also subjected
to carbon adsorption as illustrated
by the isotherm developed in Figure
9. Again, the quantity of carbon
and degree of treatment required
would be a function of the leachate
character with respect to concen-
tration and types of materials
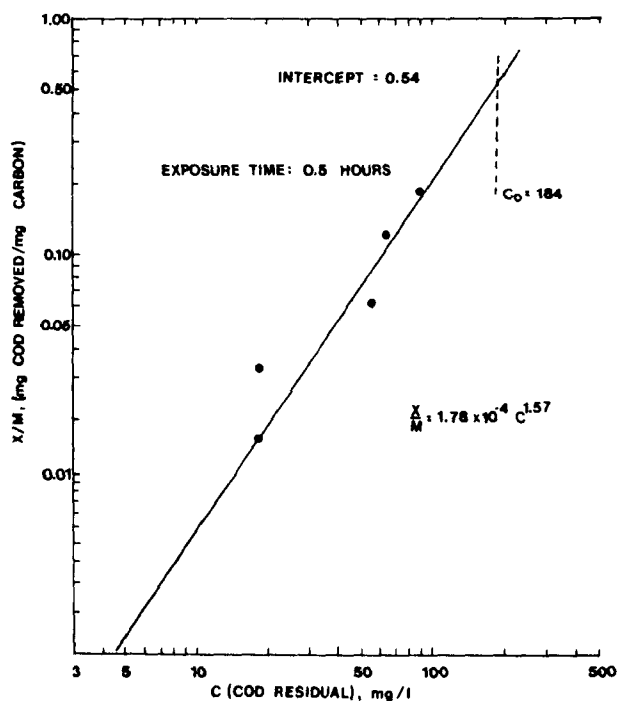present. Moreover, since other
data (3) indicated that certain

Figure 7. Removal of cations
from aerobic
biological treatment
effluent by cation
exchange.

Figure 9. Isotherm of carbon adsorption on effluent from aerobic biological treatment of leachate.

of anaerobic activity and methane fermentation, increase the rate and predictability of biological stabilization of the readily available organic pollutants in the wastes, dramatically decrease the time required for stabilization, and reduce the potential for environmental impairment. Moreover, leachate recirculation with pH control and initial sludge seeding may further enhance treatment efficiency so that the time required for biological stabilization of the readily available organic pollutants in the leachate can be reduced to a matter of months rather than years, with the opportunity for controlled final discharge and/or treatment of residuals as may be required.

Application of separate anaerobic and aerobic biological processes has proven satisfactory for leachate treatment; residual organics and inorganics in the effluent from these processes are removed well by carbon adsorption followed by mixed resin ion exchange. The degree of residual treatment is predictable and responsive to whatever effluent requirement may be imposed.

inorganics could be leached from the carbon if residual treatment with ion exchange and carbon adsorption was necessary, treatment should sequence carbon adsorption followed by ion exchange.

Based on the concept of leachate containment, collection, and treatment (either by recycle through the landfill and/or by separate biological and physical-chemical methods), the landfill of the future may well be conceived as a controlled process conducive to accelerated stabilization, environmental protection, and rapid realization of potentials for land reclamation and ultimate use.

SUMMARY AND CONCLUSIONS

The results of experimental studies on the treatment of leachate by recycle and/or separate biological and physical-chemical methods have indicated that a combination of these methods may be necessary to reduce the pollutional potential of leachate from solid waste disposal sites to a concentration acceptable for ultimate discharge. Recirculation of leachate through a landfill will promote a more rapid development

ACKNOWLEDGMENTS

REFERENCES

1. Pohland, F. G., and Maye, P. R., "Landfill Stabilization with Leachate Recycle," *Proc. 3rd Envir. Engrg. and Sci. Conf., Univ. of Louisville,* 389 (1973).
2. Pohland, F. G., "Sanitary Landfill Stabilization with Leachate Recycle and Residual Treatment," *Water-1974,* Ameri. Inst. of Chem. Engr., Symp. Ser. 145, 71, 308 (1975).
3. Pohland, F. G., "Sanitary Landfill Stabilization with Leachate Recycle and Residual Treatment," Final Report to EPA, Grant No. R-801397, 1975.

SOLID WASTE DEGRADATION DUE TO SHREDDING AND SLUDGE ADDITION

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INTRODUCTION

Among more recent methods of landfilling, the grinding, milling, or shredding of refuse and the addition of sewage sludge are becoming of increasing interest.

The shredding of refuse is done for a variety of reasons of which preparation for further processing, prepartion for long distance haul or handling, and preparation for landfilling are most common. In preparing refuse for land disposal, several advantages have been cited as justification for the cost of refuse shredding. Among the justifications are to make sites more acceptable to the public, to provide better day-to-day operational quality, especially under adverse weather conditions, to increase the density of refuse in pounds per cubic yard landfill space consumed, to promote changes in decomposition deemed desirable for a particular site, and to reduce cover requirements. Of special interest with regard to the purpose of this paper is the concept of changing the decomposition processes and, thereby, the products of decomposition, as a result of shredding refuse.

Sewage sludge may be added to the refuse for several reasons. The most common is to provide disposal by incorporating it with refuse in a landfill. An auxiliary purpose of adding sewage sludge to refuse, however, may be to change the degradation processes within the landfill. It is the latter point which is of special interest in this paper.

This paper will draw on an experimental program at Madison, Wisconsin, to describe the decomposition of shredded refuse in a landfill in comparison with that of unprocessed refuse. This program began in the late 1960's as a demonstration of the shredding process which was funded by a U.S. Environmental Protection Agency demonstration grant in cooperation with the City of Madison, The Heil Co. of Milwaukee, and The University of Wisconsin. One study initiated under this grant has been continued for purposes of additional monitoring of the decomposition of test refuse cells. The incorporation of sewage sludge in the landfills will be illustrated by descriptions of three plants using different methods of operating such a landfill and then by giving some general statements about a testing program carried out at Madison, Wisconsin, under the combined efforts of the Madison Metropolitan Sewerage District, the City of Madison, and The University of Wisconsin.

DECOMPOSITION OF SHREDDED REFUSE

The decomposition studies to be described are termed the Lysimeter

Studies. The first 2 yr of monitoring have been described previously (1). The lysimeters are each 30x60 ft in surface area. Six lysimeters, hereafter to be referred to as cells, were 4 ft deep and had 100 tons of refuse each. Two additional cells were 10 ft deep and had 215 tons of refuse each. All cells were constructed below grade and had vertical walls, of which three walls were made of cement and a fourth of wood. The bottoms of these cells were graded to carry leachate to a central collection reservoir. The bottom of each cell consisted of a bituminous layer covered with plastic that was then overlaid with crushed rock as a leachate carrying layer. Cell surfaces were sloped at a nominal 3% to one side where runoff was collected by a gutter arrangement for volume measurement.

Each set of cells was constructed simultaneously with residential and light commercial refuse to ensure equal composition. When cover was required, the cover was silty sand, commonly used for cover in the Madison area. The refuse was placed and compacted with regular sanitary landfill machinery and experienced operators were brought for this purpose from the city's sanitary landfill site.

The first four cells were constructed in September 1970, and cells 5 through 8 were constructed in October 1972. The cells were numbered as follows:

cell 1, unprocessed, covered immediately;

cell 2, shredded, covered immediately;

cell 3, shredded, covered after 6 months;

cell 4, shredded, not covered;

cell 5, unprocessed, covered with shredded refuse (66 tons unprocessed and 30 tons shredded);

cell 6, unprocessed, not covered;

cell 7, 10 ft deep, shredded, not covered;

cell 8, 10 ft deep, unprocessed, covered immediately.

Data include precipitation, leachate, and runoff quantity; leachate quality; gas composition; temperature; and settlement. Results from cells 1 through 4 will be presented in some detail in this paper, whereas the results from cells 5 through 8 will be presented qualitatively only. This is because the latter four cells have not reached a stable state of decomposition.

RESULTS AND DISCUSSION

The runoff data for the first four cells are given in Figure 1. It is noted that the covered cells, 1 and 2, produced approximately the same curves for the entire monitoring period. Cell 3 had about the same curve as 1 and 2 once it was covered. This cell was covered after 6 months, as indicated in the figure by stars. Cell 4 had no runoff for the first year, with increasing amounts of runoff since that time as the surface degraded to a soil-like surface.

Figure 2 summarizes the leachate volume data. All cells show a general increase in leachate volume with time, corresponding to generally wetter conditions in the latter half of 1972 and especially in 1973. Cells 1 and 2 compare throughout the reporting period, joined by cell 3 once it was covered. The starred points on cell 1 and 2 curves represent unnaturally large amounts of leachate caused by heavy rainfall and attendant physical damage to cell surfaces and runoff monitoring systems. The monitoring systems were extensively reworked during and after this period, and subsequent points are correct. The starred points should be discounted. Cell 4 produces slightly
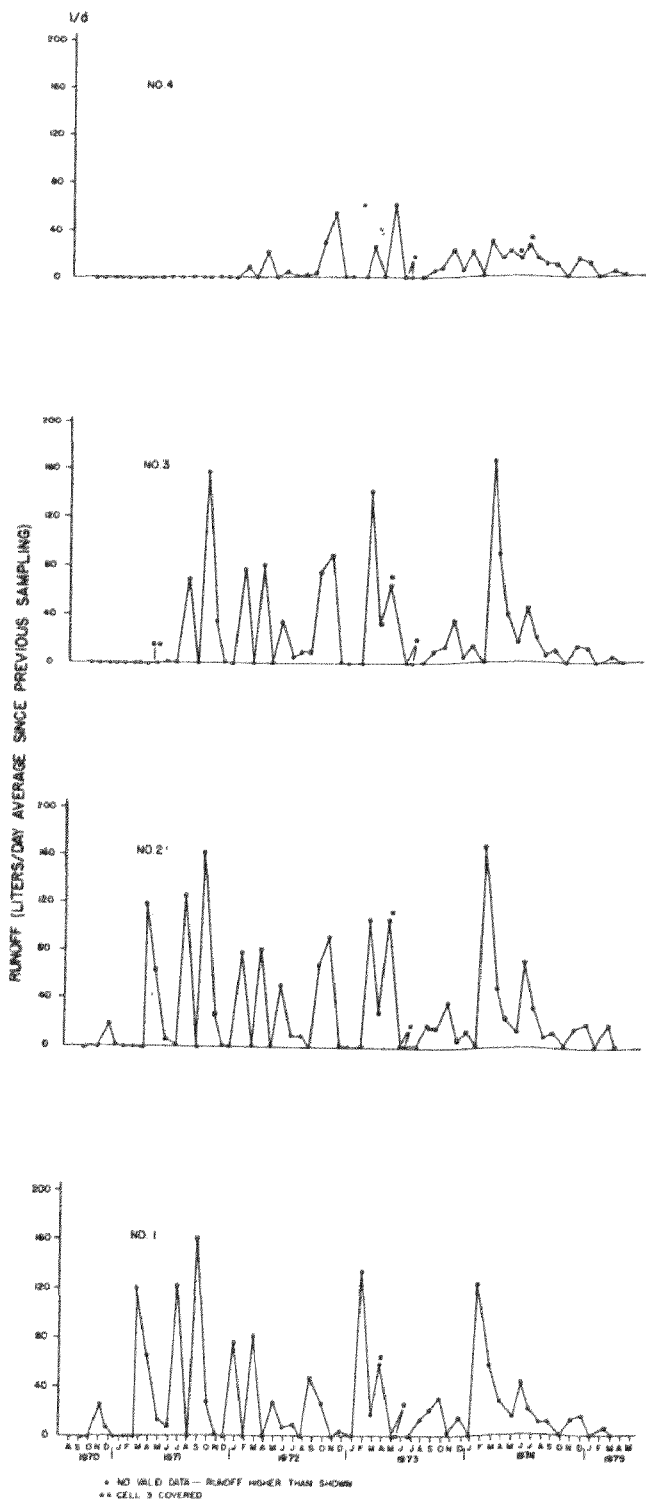
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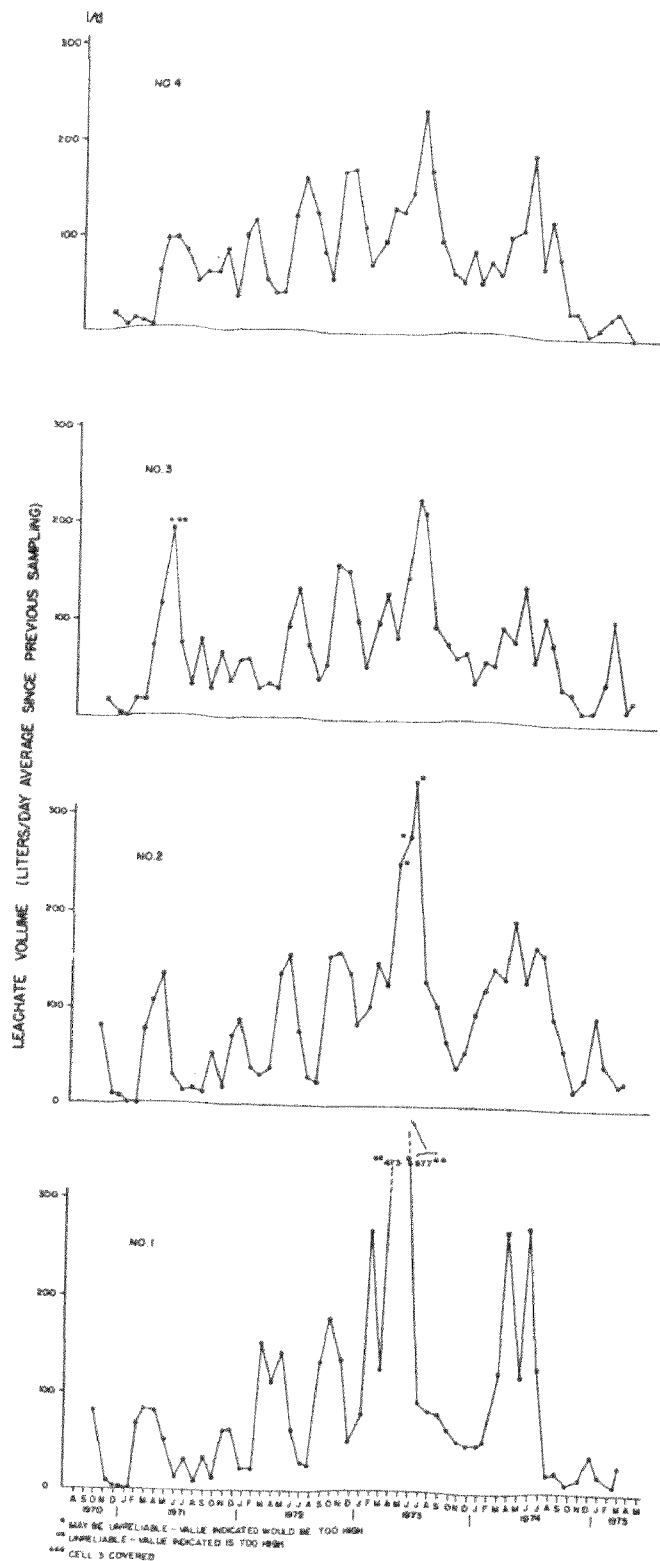Figure 1. Runoff volume for each cell.

Figure 2. Leachate volume for each cell.

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increasing leachate volumes with time. This was also in response to the weather conditions prevailing in 1972 and 1973.

The water budget over the first 4 1/2 yr of monitoring (excluding the period of cell damage) is summarized in Table 1 below. The conclusions to be reached from the water budget data are as follows: The effect of cover was to promote runoff, but the absence of cover promoted evapotranspiration. The net result was that the amount of leachate was approximately the same for all four cells whether the refuse was covered or not. It can also be concluded that the cells became more alike with time as the uncovered cell surface degraded to a rather soil-like consistency, and as all cells became covered with volunteer vegetation and so became more alike with time.

The COD concentration data are summarized in Figure 3. Note the distinct differences between the curve shapes of cells 1 and 4. Cell 1 produced COD concentrations which neither rose nor fell for 3 yr but, instead, fluctuated approximately 6,000 ppm according to weather conditions. In contrast, cell 4 produced a peak COD of 30,000 ppm but, after a few months, became relatively inactive with respect to COD concentration.

This occurred after approximately 10 months of decomposition. Except for a second summer rise, the COD's remained at well under 1,000 ppm for the final years of monitoring of this cell. The cell 2 curve is of the general shape of the cell 1 curve, but fluctuated at much higher COD levels. By comparing cells 1, 2, and 4, it can be concluded that the effect of shredding was to increase the concentration of COD approximately twofold, whereas the effect of covering was to prolong the period over which the COD concentrations remained at these levels. Cell 3 exhibited a curve generally of the same shape as cell 4 except for the tendency of this cell to produce COD concentrations substantially higher than those of cell 4 in the latter portion of the monitoring period.

The specific conductance data for the four cells are summarized in Figure 4. The curves are approximately the same shapes as were the COD curves for the respective cells, and the same general conclusions hold. This also applies to other specific chemical analyses not shown here.

The pH curve shapes generally were the inverse of the COD and specific conductivity curves, as shown in Figure 5.

Table 1. WATER BUDGET FOR PERIOD SEPTEMBER 1970 TO FEBRUARY 22, 1975*

Cell	% Runoff	% Leachate	% Evapotranspiration†
1	7.7	20.7	71.6
2	8.5	22.8	68.7
3	7.5	19.4	73.1
4	2.2	20.4	77.4

*Excluding March 6 to May 28, 1973. Total precipitation for this period: 540,000 liters per cell.
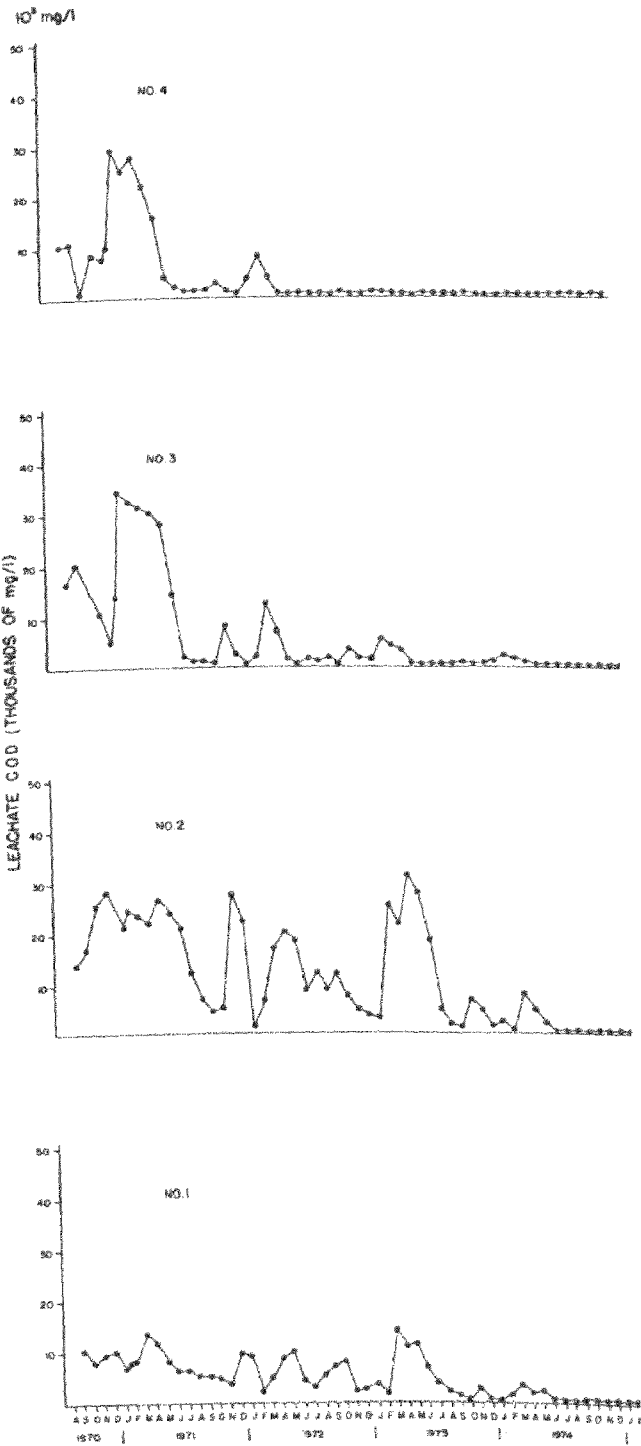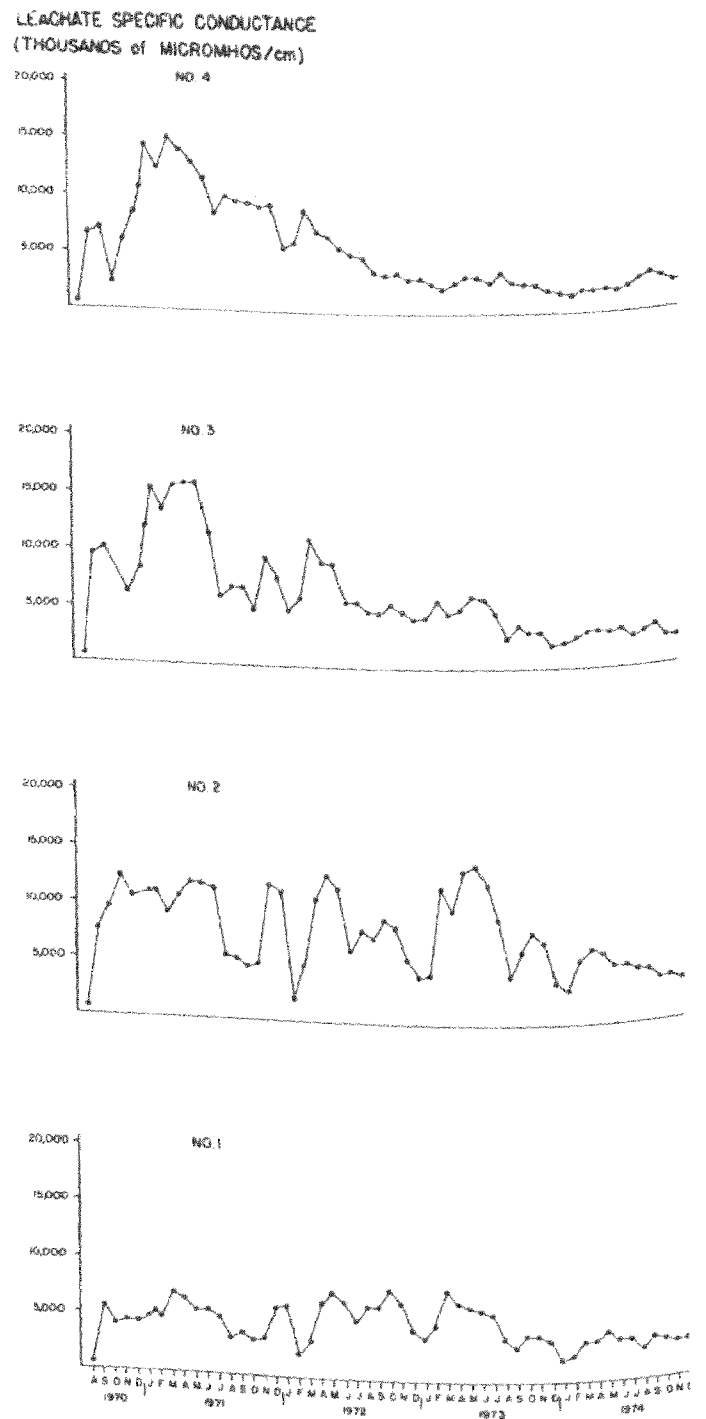†By difference.

Figure 3. Leachate COD.

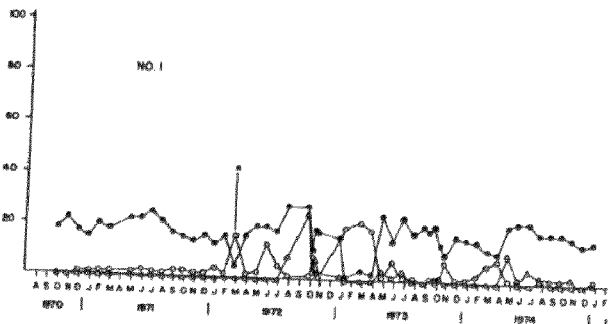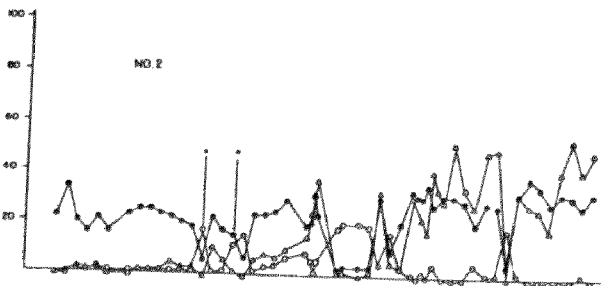Figure 4. Leachate specific conductance.

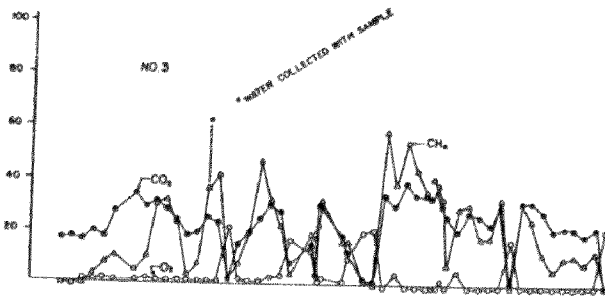Figure 5. Leachate pH.

Thus, cell 1 produced a generally low pH leachate, where the pH rose very slowly over the period of monitoring to levels near neutrality. In contrast, cell 4 had acidic pH levels for the first 8 to 9 months, after which the pH rose rapidly to neutrality and continued at that level. In comparing cells 1, 2, and 4, it is apparent that the effect of shredding was to lower the acidic pH levels attained, whereas the effect of cover was to prolong the period of acidic pH production.

The cumulative production of COD, in kilograms, for the period September 1970 to February 22, 1975, excluding the period of cell damage, was: Cell 1, 478.9; cell 2, 1106.6; cell 3, 680.8; cell 4, 417.4. It is observed that cell 4 has produced the least amount of COD over the monitoring period, followed in order by cell 1, cell 3, and finally cell 2. It is apparent that the covering of shredded refuse was detrimental as far as COD production was concerned since cell 2 produced over twice the total amount of COD-demanding substances in leachate over the period of monitoring in comparison with the production of the uncovered cell 4. Cell 3, covered after 6 months, also produced more COD than did cell 4; in cell 3, this difference is largely the result of the large amount of COD-demanding materials released during and immediately following covering operations on this cell.

The gas composition data are given in Figure 6, where the gas composition is presented on a volume percent basis. It is observed that methane production occurred more quickly with the shredded refuse cells and, in particular, with the two shredded refuse cells that were not covered immediately. Virtually no methane was produced in cell 1 even though oxygen levels were generally very low. The production of methane was observed to generally decrease in response to periodic increases in oxygen. Oxygen was apparently carried into the cells

GAS COMPOSITION AT 4 ft. DEPTH
(% VOLUME)

Figure 6. Gas composition data.

by infiltration or by decreased
biological activity due to
temperature or other changes. The
differences in methane levels between
the covered and uncovered shredded
cells are in part due to the ease
of transmission of methane out of
and atmospheric gases into the cells
that were not covered.

Conclusions to be made about
leachate quality from the first
four cells are that shredding refuse
promotes decomposition (resulting
in rapid oxygen use and methane
production) and stabilization of
leachate contaminant production.
Cover prolonged the period of acidic
leachate production and, hence,
postponed methane production. Cover
also seemed to postpone leachate
quality improvements, possibly due
to the unfavorable methane forming
condition present in those cells.

As stated earlier, the results
from cells 5 through 8 are not yet
complete, but some preliminary
conclusions can be given regarding
the data obtained thus far. The
degradation curves were generally
of the same shapes for the
unprocessed and unprocessed-covered-
with-shredded-refuse cells, and
these curves were generally the
same shapes as those for cell 4.
Serious problems were associated
with cells 5 and 6, both of which
had unprocessed refuse. In both
cases these cells experienced odor,
fly, and rodent problems, and cell
6, in particular, was visually
unacceptable. It is apparent that
the use of shredded refuse as cover
over unprocessed refuse is dangerous
and from this study could not be
condoned except for experimental
purposes. It is entirely possible
that use of carefully controlled
and monitored applications of greater
thicknesses of shredded refuse as
cover may prove acceptable, but
from this study and from observations
of other landfills where shredded
refuse is used as cover, it is
apparent that serious problems may
result.

The deep cells (numbers 7 and
8) produced higher leachate concen-

trations and are taking substantially more time to stabilize than the comparable 4-ft-deep cells. Additional work needs to be done on the effect of depth on refuse decomposition. Thus far, this study indicates that deeper refuse cells will result in increase in both the concentration and time over which highly contaminated leachates will be produced.

SEWAGE SLUDGE ADDITION

Among the reasons cited for adding sewage sludge to refuse are reasons associated with promoting decomposition of refuse. In particular, proponents of this system believe that the use of aerobic decomposition results in more rapid refuse stabilization. Once the refuse becomes stabilized, the potential for gas and leachate contaminant production is lowered, and compaction results in greater effective densities. Effective density means tons of refuse as received per cubic yard of landfill space consumed. In addition to promoting more rapid decomposition, sewage sludge is said to promote the production of heat, which may increase the rate of evaporation and, so, lower leachate production. Once heat is produced, an air circulation pattern will develop over the refuse that may aid continuing aerobic decomposition. Finally, the production of heat may aid the inactivation of pathogens.

Three examples of the use of this concept will be given. The first such example is at Giessen in West Germany where sewage sludge is added to refuse on the feeding conveyor to a Hazemag hammermill. The refuse is coarsely shredded and then placed on an aerobic windrow area for 6 months. After turning once, the refuse is landfilled without cover at a separate site. The objective in this operation is to provide sewage sludge disposal, reduce landfill volume requirements, and inactivate pathogens (2).

At Odense, Denmark, a similar process is in use. Refuse is shredded and sewage sludge is added on the shredded refuse takeaway conveyor. The nongrindable fraction, ballistically separated in the hammermill, is removed separately. The refuse-sludge mixture is placed on the landfill in windrows for 3 to 6 months. After the refuse has stabilized, the material is compacted and new windrows are placed over the area. The objective in this process is to limit the production of leachate, promote refuse stabilization, and increase the density of refuse in the landfill. This operation follows quite closely the concepts tested at the Kovik tip near Stockholm (3).

The third example is at Uttigen, Switzerland. The landfill is operated after the concepts of Professor Pircau of Berlin, who advocates aerobic landfilling to achieve greater density and limit leachate production. In this example, the refuse is not shredded and little or no sewage sludge is added. The concept is said to be applicable to the addition of sewage sludge. The refuse is placed in windrows for 3 to 6 months and covered with a foam material that is said to improve the site esthetically, limit (to some degree) rodent and fly infestation, and, in particular, promote evaporation of rainfall by holding rainfall at the surface of the landfill. After decomposition has more or less been completed, the refuse is compacted and a new windrow is built over the same area.

The examples cited above are only three of many similar operations in European countries. Because of the literature and the potential value of such methods of landfilling, a study was undertaken at Madison (Wisconsin) in cooperation with the Metropolitan Sewerage District, which has a sludge disposal problem, and the City of Madison, which has the shredded refuse. The study was modelled after the Kovick tip

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work, cited above. Twelve cells were constructed. The top of each cell was 20 by 20 ft in area. Leachate was collected from a 7- by 7-ft area with an underdrain system. The experimental results verified the design in that there was sufficient refuse around the leachate collection area that little or no edge effects were found to influence the data. Refuse was shredded and mixed with various amounts of sludge on a flat bituminous area with a front-end loader. The variables were as follows: depth, 3 ft or 6 ft; compaction, none or compacted; and sludge added, ranging from none to enough sludge to reach 50% moisture content, to enough sludge to reach 70% moisture content. The 70% moisture content figure is at or near field capacity.

The study is not yet complete, but some preliminary conclusions may be given (4,5). It was observed that the 70% water mixture was not workable during construction and could not be condoned for landfill purposes. Odors were observed, and perhaps even at the 55% water level, a sludge-refuse mixture would have sufficient odors to be of great concern. Flies were attracted to, and apparently hatched from, the refuse sludge cells. No flies were observed on the cells with no sludge. It was also observed that rodent activity developed on the non compacted cells. Little to no rodent activity was experienced on the compacted cells. It was of interest that good temperature development occurred even when the 3-ft cells received an additional 3-ft layer of refuse in February under extremely cold weather conditions. The production of leachate per ton of dry refuse increased markedly with initial percent water and was not a strong function of compaction or depth.

Preliminary conclusions from

this work suggest that one must be very careful in incorporating sewage sludge with refuse for landfill purposes. It may be that environmental and operational problems will be of great importance and could easily negate any advantages that could be obtained otherwise.

ACKNOWLEDGMENTS

REFERENCES

1. Anderson, C. R., and R. K. Ham, "Lysimeter Studies of the Decomposition of Refuse," Part I, Waste Age 5 (9):33, Dec. 1974; Part II, Waste Age 6:(1):30, Jan. 1975; Part III, Waste Age 6(2):38, Feb 1975.
2. Gotze, K., M. Budig, and E. Homrighausen, "The Giessen Model--Simultaneous Disposal of Solid and Liquid Wastes," Der Stadtetag, Vols. 4 and 5, 1969.
3. Yhland, E., and H. Karlsson, "Open Refuse Composting--Trials at the Kovik Tip 1970-71," A. Z. Sellbergs AB, Stockholm, Sweden, Nov. 1971.
4. Pickart, B. J., "Landfilling Milled Refuse Mixed with Digested Sewage Sludge," M.S. thesis, Civil and Environmental Engineering Department, The University of Wisconsin-Madison, 1974.
5. Boley, G. L., personal communication, 1975.

CASE HISTORY OF LANDFILL GAS MOVEMENT THROUGH SOILS

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This afternoon I am going to briefly describe three New Jersey underground landfill gas migration problems with which the New Jersey Cooperative Extension Service has been involved during the last half dozen years.

The New Jersey Cooperative Extension Service is charged with the responsibility of responding directly to the needs of the state's citizens; therefore, it becomes involved with various kinds of problems. One of these has been the underground generation and movement of combustible gases, carbon dioxide, and the odorous gases of sulfur reduction. Although there can be many different sources for these gases, those we have found that migrate the greatest distance laterally underground have arisen from the decomposition of organic matter in commercial refuse landfills located in old sand and gravel pits. It is our investigations into three of these problems that I will briefly describe this afternoon.

The members of the Rutgers Cooperative Extension Service faculty, who have been involved in these studies, include not only me, a specialist in environmental sciences, but also county agents, plant pathologists, entomologists, and soil scientists. In addition, whenever possible, we have involved the staffs of the state, county, and municipal health departments, the New Jersey Bureau of Solid Waste Management, the Federal Office of Solid Waste Management Programs, and the owners and operators of the various refuse landfills.

All three of these landfill gas migration problems were first brought to my attention by the respective county agents. After they, with the assistance of various specialists from Cook College, were unable to identify the cause of vegetation death, they asked me to look at the situation because I was listed as a refuse specialist and they knew that refuse was buried in extensive quantities not too far from the sites of the vegetation death.

The problems to be described include the death of peach trees in a commercial peach orchard in Gloucester County, the death of ornamental vegetation in Camden County, and the demise of commercial farm crops in Burlington County. In addition to the death of vegetation, the Camden County case involved a hazard to life and property because of the entrance of combustible gases into private residences adjacent to the landfill. However, even here, it was vegetation death that first brought the problem to our attention.

GAS MEASUREMENTS

All of our underground gas measurements were made in the field. For this purpose, we have adapted for our needs various gas measuring equipment that was originally

designed for making safety checks and measuring combustion efficiencies.

To determine whether or not foreign gases are present in the soil atmospheres, it is necessary in most cases to first make a hole in the ground. When we began these studies in 1969, we made these holes with a post-hole digger. After completing the hole, we covered it with a large garbage can lid and let the ground gas atmospheres "flow" for a specific time period into the hole from which we later drew our gas samples. This was a very time and energy consuming

Figure 2. Withdrawing a ground gas sample through an Explosimeter.

process. Later we drove a 1-in.-diameter steel rod into the ground with a sledge hammer and then sampled the ground gases directly from this bar hole. Finally, we have come to use a commercial bar hole maker to obtain a 3-ft-deep, 1/2-in.-diameter hole in the ground. This commercial instrument (Figure 1) incorporates the steel hole-making rod and driving weight into one convenient unit. This same type of unit is used by most gas utility companies when searching for leaks from their underground pipes. The handle of this bar hole maker is electrically insulated for safety to prevent a shock should you come in contact with a live underground electric wire.

Figure 1. Using 'Pogo Stick' (bar hole maker) to make a ground gas sampling hole.

The most convenient test (Figure 2) to make in checking for gases of

anaerobic decomposition of organic matter is for combustible gases with a combustible gas meter. A gas sample is drawn from the bar hole through an M.S.A. Explosimeter (Figure 2), which is the type of instrument used by the gas utility companies when looking for leaks in their underground lines. The Wheatstone's bridge principle is used within the instrument for determining the concentration of combustible gases. One leg of the bridge consists of a catalytic unit that burns the combustible gases-- changing its resistance, thereby unbalancing the bridge and giving a reading on the galvanometer (Figure 3). The sample is withdrawn from the bar hole by use of a 3-ft-long nonsparking probe. If desired, a nonconducting probe may also be used. A rubber stopper is placed over the upper end of the sampling probe to help seal the bar hole from the ambient air. However, the nature of the sampling method frequently incorporates large quantities of dilution air. These

combustible gas reading instruments indicate percent of the lower explosive limit of the gases for which the instrument is calibrated. The lower explosive limit for methane is a 5% dilution in air. However, it is possible to tell from the response of the meter whether or not the combustible gas concen- tration is between the lower and the upper explosive limits or above the upper explosive limit (15% methane in air). By the use of a dilution tube on the intake side of the meter, it is possible to theoretically determine the actual combustible gas concentration up to 100%. It is also possible with these instruments to separate heavy petroleum vapors from the lighter hydrocarbon gases by use of an activated charcoal filter on the sampling line.

The instruction manuals should be followed closely when using these meters and the instruments should be maintained and calibrated regularly. Calibration equipment

Figure 3. Close-up of Explosimeter showing a reading of about 75% of the lower explosive limit.

is available, and the instruction booklet will inform you as to the frequency and extent of routine maintenance.

The carbon dioxide and oxygen concentrations of the ground gases obtained from the bar holes are analyzed by the Orsat method, which is normally used to measure the efficiency of fossil-fuel-fired furnaces. In our field test work, we use the Bacharach Fyrite carbon dioxide and oxygen indicators. In the carbon dioxide indicator, the carbon dioxide is absorbed in a potassium hydroxide solution. In the oxygen indicator, a chromous chlorine solution is used. Carbon dioxide indicators are available for reading 0 to 20% and 0 to 60% concentrations. The oxygen indicators are for determining 0 to 21% concentrations (Figure 4).

Unpleasant ground gas odors are frequently an indication of the presence of the gases of anaerobic decomposition of organic matter. These odors can be checked for by withdrawing a soil sample from the ground and smelling the sample. If the unpleasant odors of the products of sulfur reduction are present, you will know it without having to receive any instructions.

We have occasionally used industrial hygiene dry tube indicators to check for the possible presence of sulfur gases and carbon monoxide. However, in general we find the field test for combustible gases to be the easiest, quickest, and simplest to make. Our next most frequently used field check is for carbon dioxide. Normally we would not expect to record the presence of combustible gases or carbon dioxide with these field test meters if there were not substantial gaseous productions of anaerobic decomposition present in the soil gases.

Figure 4. Close up of Fyrite oxygen tester showing a reading of about 6%.

THREE LANDFILL GAS MIGRATION CASES

The three New Jersey landfill gas migration cases that we have followed most extensively during the past half dozen years took place in Glassboro, Gloucester County; Cherry Hill, Camden County; and Cinnaminson, Burlington County. As I mentioned before, all of these cases were brought to my attention by the respective Rutgers County Agent who was responding to complaints of vegetation injury and death from unknown causes. Also, all three cases were associated with landfills where refuse had

been deposited in worked out sand and gravel pits.

Glassboro

The landfill in Glassboro covered about 6 acres. The refuse, which consisted of household and industrial wastes, demolition materials, and sewage sludge, was deposited to a total depth of 10 to 20 ft. A commercial peach orchard abutted the landfill along about 1000 ft of its outer periphery. The balance of the landfill was adjacent to open scrub vegetation land that was apparently not serving any commercial, agricultural, or residential use.

Landfilling at this site began February 1968. Refuse decomposition along the northeast line of the landfill adjacent to the peach orchard was completed in 1969. The peach trees nearest this line began dying during the summer of 1971. I made my first inspection of this site in September 1972. By that time, about 50 peach trees had died (Figure 5). Combustible gases and carbon dioxide were found along with low oxygen concentrations in the area of the root zones of most of these dead peach trees. Seventy feet was the greatest distance from the landfill for any of these dead peach trees.

In March 1974, landfill gases were found greater than 80 ft from the landfill. A total of about 70 peach trees had now died. In the same month, the peach farmer brought the operator of the landfill (the Borough of Glassboro) into the Chancery Division of the Superior Court of New Jersey. After a week of testimony, the case was settled out of court to the plaintiff's satisfaction.

I examined the landfill and peach orchard last week. The refuse landfilling is complete. It appears to have an adequate cover of bank run soil material (Figure 6). Some minor landfill settlement is taking place and causing surface water puddles to form following rain storms. More than half of the mature peach orchards that were adjacent to the landfill have been removed as part of the regular peach farming procedure. They were replaced more than a year ago by new peach trees. However, it was noted that along the row of trees nearest the landfill, the young trees had apparently died and had to be replaced again by new seedlings last fall or this spring. Apparently no corrective measures have been taken to reduce the lateral migration of these landfill gases from the landfill. Until lateral migration of these gases ceases as a result

Figure 5. A 1972 view of Glassboro peach orchard with dead trees adjacent to landfill.
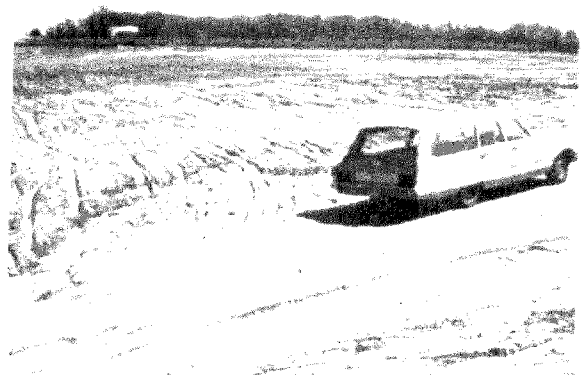

Figure 6. A view of completed refuse landfill at Glassboro, New Jersey.

of corrective measures or the cessation of their generation by the biodegradation of the organic matter, I expect that that the farmer will continue to experience the death of peach trees planted adjacent to the landfill.

Cherry Hill, Camden County

This 9- to 10-acre landfill is surrounded by 28 single family homes that were constructed prior to the refuse landfill. Refuse 10 to 60 ft deep has been deposited in this former sand and gravel pit. I was informed that when dumping began in the fall of 1963, only bulky wastes and demolition materials were being deposited. However, as time went on, the nature of the materials being deposited gradually changed until it was general municipal refuse and garbage that

were being deposited. Dumping was completed in 1970. Since then, Cherry Hill Township has been expending efforts to turn this former landfill into a municipal park. The fill has been placed in such a manner as to incorporate aesthetically pleasing hills and slopes within the park perimeter, topsoil has been brought in, and the area seeded.

We were first called to this area by the Camden County Agent in January 1969 to help determine the cause of vegetation death in the backyard of a home at 219 Rhode Island Avenue, abutting the landfill (Figure 7). We were told that much vegetation in their backyard had recently died, including a spruce tree, rhododendron, Japanese yew, azaleas, dogwood trees, flowering peach trees, Scotch brooms,

Figure 7. A 1969 view of backyard of 229 Rhode Island Ave, Cherry Hill, N.J.

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arborvitae and Douglas fir, as well as an area of lawn grass. Our ground gas tests indicated the presence of combustible gases, carbon dioxide, putrid ground odors, and the lack of oxygen in the soil gases. Periodic checks were made of this area, and it was noted that the vegetation sickness and deaths seemed to be gradually progressing from the area of the interface of the landfill and the backyard towards the house. In June 1971, a general inspection with the county agent of the whole area surrounding the landfill indicated that there were clumps of dead vegetation at various points around the total periphery and the landfill. At this time, it was recommended that the responsible governmental officials do a check of the complete periphery of the landfill to see if these landfill gases might be migrating from the landfill into other backyards and toward other homes. In the late summer and fall of 1971, landfill gas fires occurred in two homes adjacent to this former landfill. Tests of many homes at this time revealed the presence of very high concentrations of combustible gases in the soil beneath the crawl spaces of a number of the homes adjacent to the landfill. Many home owners were also complaining of unpleasant odors within various parts of their homes.

Between December 1971 and January 1973, four different 10- to 15-ft deep stone-filled trenches were installed in an effort to prevent the lateral migration of landfill gases from the landfill to the adjacent property (Figure 8). Some of these trenches were successful in stopping this migration; others were not. In April 1974, the legal case of about a dozen residents surrounding the landfill against Cherry Hill Township, the owner and operator of the landfill was heard in the Chancery Division of the Superior Court of New Jersey. After a week of testimony, the case was settled out of court. The plaintiffs received a total of $50,000 in settlement. In addition, the
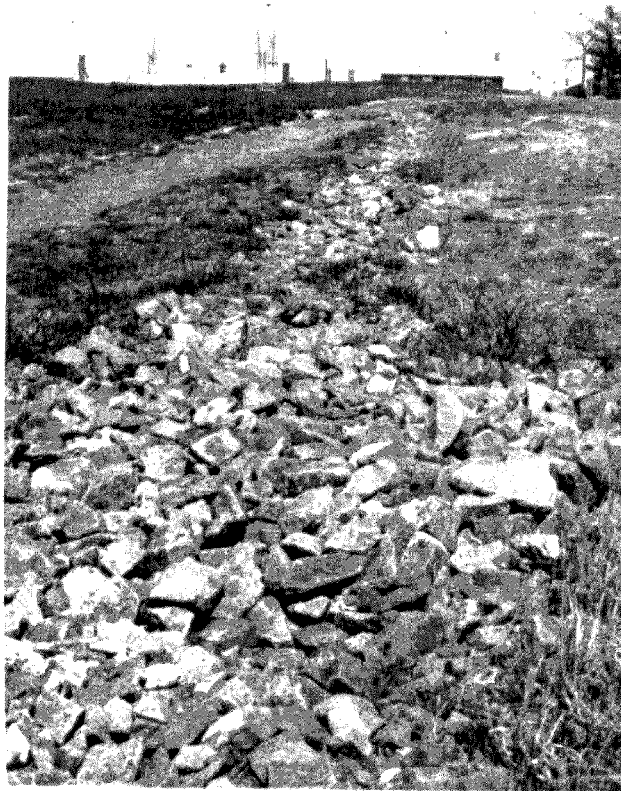

Figure 8. Trap rock fill trench in place for venting of lateral migrating gases, Erlton Landfill-Park, Cherry Hill, N.J.

township was required to vent the landfill gases from the periphery of the landfill.

In the fall of 1974, vertical venting pipes were installed on the periphery of the landfill (Figure 9). Many of the vent pipes along the east periphery of the landfill had been vandalized. Someone had taken the large rocks from the gas-venting trenches and used them as cannon balls to destroy the tops of the vents. However, it did not appear that the efficiency of these plactic pipes for gas venting had been decreased by this vandalism. No signs were noted in March of 1975 of the vandals having tried to ignite the gas coming from the pipes.

In April 1974, a single maple tree was planted in the lower

Figure 9. Vertical gas venting pipes on northwest side of Erlton Landfill-Park, Cherry Hill, N.J.

Figure 10. Newly planted trees and open shed on Erlton Landfill-Park.

elevations of this landfill park. However, no sign of this maple tree could be found in March 1975. Over the total park area, only one clump of naturally seeded trees was noted. This was a group of maple trees that has been growing for a number of years near the southeast corner of the landfill. Tests for ground gas indicated that the gases of anaerobic decomposition were not present in the root zone of these trees.

Examination of the landfill park last week revealed that a large number of deciduous and evergreen trees had been planted over much of the park area by a landscaper last fall. These included sweet gum, red oak, crab apple, Japanese poplar, white pine, Scotch pine, and fir, among others (Figure 10). No trees were planted on the tops of the high refuse hills.

As of March 1975, many of the evergreen trees appeared brown. There were also signs of vandalism of the trees as some had been pulled from the ground, others had limbs broken off, and some had apparently been stolen. The holes left after the trees had been pulled or dug from the ground appeared to be rather shallow and the soil beneath them appeared to be very well sealed since water puddles remained in them some days after a heavy rain. It will be interesting to observe the fate of this newly planted vegetation. A group from Cook College, including plant pathologists, will be examining the tree plantings in May to make an evaluation of their viability.

During the last half dozen years the vegetation in the backyard of the home of 226 Rhode Island Ave. has continued to die until only

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Figure 11. A 1975 view of the backyard of 229 Rhode Island Ave. adjacent to the former Erlton Landfill. Two newly planted evergreen trees are noted in the foreground.

that vegetation near the home remains (Figure 11). It will be interesting to see if the gas vents prevent further gas instrusion into these adjacent lands and permit vegetation to again be grown.

Cinnaminson in Burlington County

An operating landfill of about 100 acres exists on the northern side of Union Landing Road in which, we were informed, refuse has been placed to depths of 80 ft. A 600-ft-long road interface exits between the landfill and the farm on the southerly side of Union Landing Road. The distance between the edge of the farm and the edge of the landfill is 50 to 60 ft. This space is occupied by the Union Landing Road and its right of way. Figure 12 is a view of Union Landing

Road in Cinnaminson toward the southwest. The refuse landfill is on the right and the adversely affected farm fields are to the left of Union Landing Road.

Apparently all sorts of refuse materials have been accepted by the landfill, which began operation about 10 yr ago.

In the summer of 1970, the farmer experienced difficulty in growing tomatoes in his fields nearest the landfill. During the spring plowing of 1971, very unpleasant odors were noted by the farmer arising from the area of his field nearest Union Landing Road. The farmer now knew that something was very wrong with the soil in this field. At that time, the Burlington County Agent asked me to examine this field for possible problems associated with the refuse landfill. Examination revealed landfill decomposition gases in the farm soil atmospheres as far as 180 ft from the nearest edge of the landfill. In the fall of 1971, landfill gases were discovered 300 ft from the landfill. Since the spring of 1971, the farmer has not cultivated the 2 to 3 acres of his fields nearest Union Landing Road. The death of vegetation in the field resulted in erosion of the surface of the field until a weed cover crop developed.

By the fall of 1971, the landfill operator had recognized that his landfill was the source of the farmer's problem. He then installed a 600-ft-long, 10-ft-deep, 3-ft-wide, gravel-filled trench the total length of the interface between the landfill and the farm. This did not seem to alleviate the problem. Apparently the gases continued to flow from the landfill beneath the trench into the farm field.

In the spring of 1972, tests were made of the quality of the soils in the area of the farm field where vegetation was undamaged and where the vegetation had been killed by the landfill gases. The nutrient
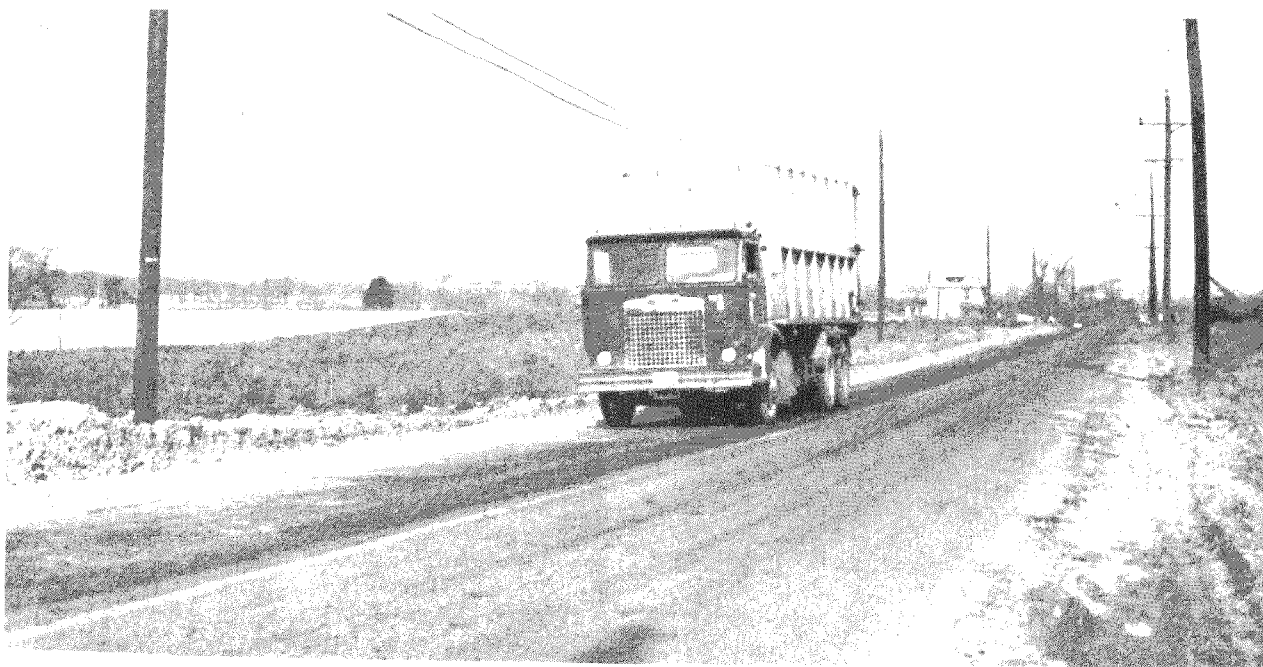
Figure 12. Looking southwesterly on Union Landing Rd., Cinnaminson, N.J. Refuse landfill on right of road and landfill-gas-affected farm fields on left of road.

quality of both soil areas was found to be the same. In March of 1973, the landfill operator installed 4-in.-diameter, plastic vertical venting pipes along the total length of the landfill farm field interface. These pipes were put in to a depth of 40 to 45 ft on 20- to 25-ft centers (Figure 13). In April of 1973, the farmer and the landfill operator developed an interim agreement relative to payments for damages claimed by the farmer to date and corrective actions to be taken to the landfill to alleviate the gas migrating problem. In December 1974, the farmer noted areas further from the field where his rye cover crop was dying (Figure 14). A check of the ground gases in these areas revealed the presence of combustible gases up to 600 ft from the nearest edge of the landfill. It appears that the venting pipes are not doing the job we had hoped they would although visual examination indicates that ground gases are being vented to the atmosphere by the pipe. One

suggestion that has been made to improve gas venting is to install evacuating pumps on the vertical pipes to withdraw the combustible gases from the landfill.

As indicated above, we found landfill gases 180 ft from the landfill in the spring of 1971, 300 ft from the landfill in the fall of 1971, and 600 ft from the landfill in December of 1974. However, the migration of these gases has not been a constant outward progression. Measurements taken at a number of other times indicated that the landfill gases had apparently retreated towards the landfill. Their outward progression seems to be at an uneven rate depending upon various factors, many of which we have incomplete understanding. Incidentally, the consultant hired by the landfill company reports that the degree of migrating gases has decreased with time. Obviously, our data are in conflict with his. Some of this disagreement might be due

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Figure 13. Vertical gas venting pipes along the Union Landing Road edge of refuse landfill, Cinnaminson, N.J.

to making measurements at different locations and times.

We will continue to follow this situation and determine the ultimate fate of the landfill gases and vegetation growth in the fields. Eight to ten acres of the farm field are now involved in problems of poor or no vegetation growth due to the adverse influence of gases from the landfill located to the north of the field. It is planned to have the Cook College Agricultural Experiment Station's weed specialist evaluate the weed growth on this field. Weeds seem to grow on landfill-gas-loaded soils. We would like to know if the species are atypical for the area and soils.

The landfilling directly north of Union Landing Road is coming to a close. Completion of the landfill will involve the filling

of the easterly corner of this former sand and gravel pit. This additional filling of the landfill area will bring the refuse in contact with the soil along Union Landing Road for an additional few hundred feet immediately opposite the Hunter Farm. This also includes an area of farm on which a farm house is located. It is possible that the refuse will be located against a soil bank within about 100 ft of the farm house. This farm house has a dirt floor cellar which is used to store various farm crops during the winter. Therefore, we have strongly recommended that permanent gas sampling stations be set up opposite the landfill area still to be filled. In addition to exposing this farm house to possible infiltration of landfill gases, another private home and at least one light industry building will also be within a couple hundred feet of the refuse upon completion of the landfill. Unless adequate protective measures, such as a gas barrier and/or adequate vents, are taken to prevent the migration of landfill gases, it is possible that these buildings may in time become involved with the entrance of combustible gases traveling underground from the landfill.

CONCLUSIONS

After a half dozen years of periodically surveying landfill gas migration problems, we have come to the following tentative conclusions:

1. Landfill operators can get themsleves into a lot of trouble with migrating landfill gases.

2. Injury and death of vegetation may be used as indicators of the presence of landfill gases in the surface soil layers. However, the gases can travel laterally below vegtation without injuring it and appear at the

Figure 14. Rye planting in farm field adversely affected by landfill gases 600 ft from nearest edge of landfill, Cinnaminson, N.J.

surface at a greater distance from the landfill.

3. It may take substantial time to note the effects of landfill gas migration upon vegetation.

4. Gases tend to travel laterally through permeable soils from refuse landfills.

5. Old sand and gravel pits are not good places to place refuse if you want to prevent lateral gas migration. If old sand and gravel pits are used as refuse landfills, gas vents and seals should be placed at the outer edge prior to refuse deposition.

6. Sometimes gas vents prevent the lateral migration and sometimes they do not. We still have quite a bit to learn about effective gas venting to prevent lateral gas migration.

7. Political and economic concerns can inhibit obtaining all the available accurate information relative to a problem such as landfill gas migration and vegetation death.

The above presents in brief outline form, to our best knowledge, the historical record of three landfill migrating gas cases in New Jersey and the associated vegetation death. We would very much like to know if others have

had similar experiences. We are particularly interested in knowing if others have experienced vegetation death associated with refuse landfill gases. If others have found that they can plant all kinds of deep-rooted vegetation over deep landfills in which uncombusted biodegradable refuse has been deposited during the last 20 years, we would also like to know about it. Much still needs to be learned about how to make vegetation planted over old landfills grow well. Information supplied by all concerning their experiences with landfill gases will be helpful in developing adequate protective measures.

16. ABSTRACT

A research symposium on sanitary landfills was held to bring together researchers, administrators, and other personnel to exchange state-of-the-art ideas and findings. This mechanism was utilized so that maximum coverage could be gained. Topic areas discussed were gas and leachate, their formation, collection, and treatment. The compilation of papers contained in this symposium presents the Solid and Hazardous Waste Research Division, Municipal Environmental Research Laboratory, research on sanitary landfills.